Stow

UNCLASSIFIED

ORNL MASTER COPY

External Transmittal Authorized
ORNL
Central Files Number
57-3-114
(Revised)

Cy 32

STATUS REPORT ON THE DISPOSAL OF RADIOACTIVE WASTES

U.S. ATOMIC ENERGY COMMISSION

NOTICE

OAK RIDGE NATIONAL LABORATORY
OPERATED BY
UNION CARBIDE NUCLEAR COMPANY
A Division of Union Carbide and Carbon Corporation
UCC
POST OFFICE BOX X · OAK RIDGE, TENNESSEE

UNCLASSIFIED

UNCLASSIFIED

ORNL
Central Files Number
57-3-114
(Revised)

STATUS REPORT ON THE DISPOSAL OF RADIOACTIVE WASTES

Prepared by
the

COMMITTEE ON DISPOSAL AND DISPERSAL
OF
RADIOACTIVE WASTES

For the

NATIONAL ACADEMY OF SCIENCES
NATIONAL RESEARCH COUNCIL

Date Issued

JUN 25 1957

Compiled and Edited by

Floyd L. Culler, Jr.
Oak Ridge National Laboratory
Oak Ridge, Tennessee
Operated by
UNION CARBIDE NUCLEAR COMPANY
For the
United States Atomic Energy Commission

and

Stuart McLain
Argonne National Labortory

UNCLASSIFIED

DISTRIBUTION

1-3.	Stuart McLain, Argonne National Laboratory
4.	W. A. Patrick, Johns Hopkins University
5-7.	C. I. Campbell, National Academy of Sciences
8.	Abel Wolman, Johns Hopkins University
9.	H. M. Parker, General Electric Company, Hanford Works
10-15.	F. L. Culler, Jr., ORNL
16.	Central Research Library, ORNL
17-31.	TISE, Oak Ridge Operations, AEC
32-150.	Laboratory Records, ORNL

MEMBERSHIP OF THE COMMITTEE ON DISPOSAL AND DISPERSAL OF RADIOACTIVE WASTES

Abel Wolman, Johns Hopkins University, Chairman
J. A. Lieberman, U. S. Atomic Energy Commission, Rapporteur
F. L. Culler, Oak Ridge National Laboratory
A. E. Gorman, U. S. Atomic Energy Commission
L. P. Hatch, Brookhaven National Laboratory
H. H. Hess, Princeton University
C. W. Klassen, Illinois State Department of Public Health
Sidney Krasik, Westinghouse Atomic Power Division
Stuart McLain, Argonne National Laboratory
H. M. Parker, General Electric Company, Hanford Works
W. A. Patrick, Johns Hopkins University
S. T. Powell, Consulting Engineer, Baltimore, Maryland
Leslie Silverman, School of Public Health, Harvard University
Philip Sporn, American Gas and Electric Company, New York City
Conrad P. Straub, U. S. Public Health Service, Cincinnati, Ohio
C. V. Theis, U. S. Geological Survey, Albuquerque, New Mexico
Forrest Western, U. S. Atomic Energy Commission, Washington

Consultants:
Paul C. Aebersold, U. S. Atomic Energy Commission, Washington
Karl Z. Morgan, Oak Ridge National Laboratory

Assistance in data calculation, compilation, analysis, and in the writing and editing of this summary report was given by:

E. D. Arnold	ORNL
J. O. Blomeke	ORNL
A. T. Gresky	ORNL
W. de Laguna	ORNL
R. J. Morton	ORNL
E. G. Struxness	ORNL
J. W. Ullmann	ORNL

This report has been assembled and edited by Floyd L. Culler, Jr., of Oak Ridge National Laboratory and Stuart McLain of Argonne National Laboratory.

PRELIMINARY TABLE OF CONTENTS
AND COMMENTS ON STATUS

**1.0 Introduction
  Definition of Problems in Radioactive Waste Disposal
  The Dual Nature of the Radioactive Waste Problem
    The Low Level, Distributed Hazard
    High Level Waste Disposal and Containment

2.0 Summary and Recommendations
  (Not yet prepared; portions available in preliminary copy form)

**3.0 The Nature of High Level Radioactive Wastes as Defined for Reactors and Chemical Process Techniques
  3.1 From Reactors
  3.2 Chemical Processing Cycle
  3.3 Nature of Wastes from Radiochemical Processes
  3.4 Nature of Fission Product Gases as a Waste
  3.5 Particulate Solids as Wastes in the Gas Phase
  3.6 General Comments About Handling Contaminated Gases
  3.7 Solid Radioactive Wastes

**4.0 Growth Predictions for Nuclear Reactor Capacity and the Magnitude of the Associated Fission Product and Transplutonic Waste Problem - Discussion of Present Knowledge
  4.1 Processing Requirements, Buildup of Fission Product Activity, and Liquid Waste Volumes in a Predicted Nuclear Power Economy
  4.2 Production of Transuranics and Transplutonics and Their Concentrations in Waste Streams
  4.3 Distribution of Fission Product and Heavy Element Activity Over the Nuclear Reactor Complex

**5.0 Relative Biological Hazards of Fission Products and Heavy Elements in Accumulated Radioactive Wastes

**6.0 Hazard Potentials Due to Accidents
  6.1 Major Release from a Reactor Accident
  6.2 Quantitative Description of Hazards in Ore and Feed Materials Processing of Virgin Fissionable and Fertile Materials

6.3 Potential Sources from Handling Fissionable and Fertile Material After Irradiation and Decontamination from Fission Products

6.4 Hazards in the Chemical Reprocessing Plant

**7.0 Transportation of Active Wastes

7.1 Estimation of Possible Magnitude of the Shipping Problem for Both Fuel and Wastes

7.2 Summary of Existing Shipping Regulations and Practices

7.3 Optimum Cooling Period for Wastes Before Possible Shipment

7.4 Possible Costs vs. Shipping Distances

7.5 Experience with Radioactive Waste Shipment

(All material contained in CF 57-2-20)

**8.0 Possibilities for Ultimate Waste Management and/or Disposal

(Author-Editor, J. O. Blomeke)

8.1 Introduction

8.2 Fixation of Wastes in Solid Form Prior to Ultimate Disposal

8.3 Effect of Cesium and Strontium Removal from Wastes

8.4 Ocean Disposal

8.5 Land Disposal

(1) Tanks or Lagoons

(2) Disposal in Salt Formations

(3) Disposal in Deep Wells

(4) Disposal in Dry Caves

(5) Surface Disposal of Liquid Wastes

9.0 Chemical Processes for Fission Product Concentration, Removal or Fixation

**10.0 Economic Considerations and Data

10.1 Rough Estimate of Allowable Costs of Waste Disposal

10.2 Costs of Evaporation of Radioactive Wastes

10.3 Waste Tank Costs

10.4 Cost of Collection and Disposal of Low-Level Liquid and Solid Wastes and Some Notes on Current Practices

10.5 Cost of Drum Drying Low-Level Wastes

General Bibliography

Appendix I

Description of Present Reactor, Chemical Processing, Waste Complex

Appendix II
Reactor Excursions

Appendix III
Criticality Hazard in Reprocessing Nuclear Fuels

1.0 Introduction

The new and as yet unsolved problems introduced by the production of large quantities of fission products and radioactive isotopes from fission or neutron capture present mankind a most complex technical, economic, and political problem. On one hand, the possibility of using the fission process to produce energy from an unexploited and abundant natural source is emerging from large programs of research and development. We are also beginning to see the promise of use of particulate and electromagnetic radiation for the good of man. On the other hand, we are presented with the problem of controlling the dangerous products of fission for periods of time measured in terms of many hundreds of years, periods longer than the effective tenure of any political state in history. We must not only devise ways of protecting ourselves in the present and for our lifetime but, in addition, we must establish the basic technical, social, and administrative control of vast quantities of artificial radioactivity that must remain effective for at least ten to twenty lifetimes.

✓

This status report on radioactive wastes has been prepared as a logical and necessary part of the Study of the Biological Effects of Atomic Radiation, sponsored by the National Academy of Science, the National Research Council, and the Rockefeller Foundation.(1)(2)

Radiation exposure to man and to members of man's ecological cycle comes from both natural and "manufactured" sources. The natural sources--cosmic rays and naturally occurring radioactive elements--have been with us for periods of time sufficient to have their effects integrated into the ecological and genetic equilibrium of mankind. The new source of radioactivity, the fission process, promises to produce sufficiently large quantities of radioactivity to effect drastically this equilibrium. Many segments of our scientific, industrial, and governmental establishments must participate in the definition and solution of the radioactive waste disposal problem.

The projected large-scale production of long-lived radioactive isotopes by an atomic power industry coupled with the diverse routes by which these elementary and highly toxic substances may traverse the whole of our physical, chemical, and biological environment presents us with an entirely new kind of problem in industrial pollution. The studies aimed at defining a means of managing this unparalleled problem must first extend deep into the basic life processes themselves. The need for measurement and knowledge of rates of spread in nature of these substances as waste products extends the problem of interest into the provinces of the physical sciences which deal with our environment. The interest and responsibility of industrial groups as operators of nuclear reactors and chemical plants is obvious. The necessity for penetrating and careful study of risks involved in atomic energy ventures by insurance and finance groups is equally a part of the whole. A regulatory function over radioactive wastes must be provided by agencies of guaranteed long tenure and by groups who clearly protect the welfare and physical well-being of all, who have foresight and wisdom to perpetuate this protection. County, state, national, and international regulation is implied.

In the atomic energy industry any waste containing levels of activity in excess of safe limits for human exposure is potentially hazardous throughout the period of its radioactivity. Ultimately, such wastes when released reach man or his environment through one channel or another. The integrated total of many small facets of release, each possibly of little consequence by itself, may be highly significant. The public interest requires that responsibility be placed for recording and integrating the cumulative effects of these sources of radiation. This is a joint responsibility of the industry and of public regulatory officials.

The United States Atomic Energy Act of 1954 places responsibility for disposal of radioactive wastes on the Atomic Energy Commission. Wisely, the Commission is seeking to carry this responsibility on a cooperative basis with established regulatory agencies in the various states and territories. It is known that many of these agencies feel strongly that they should exercise control in matters of public health and safety over industries using atomic energy as they do with other industries. Indeed, under existing laws unless they do so they may be charged with default in meeting their legal responsibilities. On the other hand, in Great Britain under the Radioactive Substances Act of 1948 the various ministries of Health concerned are charged with responsibility

> "to secure that any radioactive waste products resulting from such manufacture, production, treatment, storage or use as aforesaid are disposed of safely."

There is need of study as to how and by whom this responsibility could best be administered in the U. S. as the industry expands.

Because of the progressive changes in the technology of the industry based on research and experience there will be corresponding changes in processing and products and in use of nuclear energy. Progress in this direction will be reflected in the kinds and characteristics of wastes and in methods of treatment and disposal. Because of nuclear energy industry is unique and in such an early state of development its ultimate potentialities cannot now be measured. The dynamics of its development will, therefore, need more than normal scrutiny from the standpoint of its impact on man and his environment.

Solutions to the problem of radioactive waste control and disposal cannot be proposed at present because of lack of fundamental data. We are, therefore, presenting what information is possible on the more technical aspects of the problem. We have accomplished little more than the preparation of a summary of what has been said about the physical nature of radioactive wastes, their production and decay, and their equilibrium concentration in a nuclear power economy. We provide information on wastes currently produced by Atomic Energy Commission Operations, predict the nature of wastes resulting from new processes that will be required for economical power production, discuss relative biological hazards of fission products and transplutonic elements, discuss various processes proposed for isolating certain fission products, and review current work on processes for gross wastes that may precede ultimate regulated disposal. As background information we have included in appendixes summary information on the nature of reactors and chemical processes. Discussion of certain aspects of the economics of waste disposal is included.

We must consider this report to have the following purposes:

1. To provide a summary of present technical knowledge and data on problems of radioactive waste disposal.
2. To provide calculations of a general nature that will assist in defining a reference plant upon which to judge the over-all significance of the waste disposal problem and to measure the merits of suggested solutions.
3. To estimate the magnitude of the waste problem for the next forty years, based upon predictions of nuclear energy growth.
4. To discuss the few possibilities for permanent waste disposal.
5. To suggest areas of development and research.
6. To indicate those segments of our technology, business, and governmental structure that will be affected by production control and disposal of radioactive materials.

The report has been written for scientists and technologists who possess or will obtain background information on atomic energy; we have assumed familiarity with nomenclature, calculations, and materials involved in atomic energy work. However, since understanding of basic units used in the discussion of radioactivity will be required throughout this report, we include the following definitions:

1. Curie (c): The amount of radioactive material which disintegrates at the rate of 37 billion atoms per second (3.7×10^{10} disintegrations per second). Latest measurements of the half-life of Ra^{226} seem to indicate that a gram of Ra^{226} is slightly less than one curie.(8)
2. Millicurie (mc): The amount of radioactive material which disintegrates at the rate of 37 million atoms per second (3.7×10^{7} disintegrations per second).(9)
3. Roentgen (r): The quantity of x- or gamma radiation such that the associated corpuscular emission per 0.001293 gm of air (equal to 1 cc of air at 0°C and 760 mm Hg) produces, in air, ions carrying 1 esu of quantity of electricity of either sign.(9)
4. Roentgen equivalent physical (rep): The amount of ionizing radiation of any type that loses energy at the point in question in soft tissue to the extent of 93 ergs per gram. It is approximately equal to a roentgen of about 200 kv x-radiation in soft tissue.(8) This unit has been replaced by the rad.
5. Roentgen equivalent man (rem): The amount of ionizing radiation of any type that produces the same damage to man as one roentgen of about 200 kv x-radiation. 1 rem = 1 rad in tissue times RBE.
6. Millirem (mrem): (a) 1 milliroentgen, in the case of x-radiation of gamma radiation; (b) 1 millirad (0.1 ergs per gram) in the case of beta radiation; (c) one tenth millirad for protons of energy below 10 Mev. One twentieth millirad for alpha rays and heavy recoil particles.

7. Relative biological effectiveness (RBE) means the biological effectiveness of any type of energy of ionizing radiation in producing a specific biological damage (e.g., leukemia, enemia, sterility, carcinogenesis, cataracts, shortening of life span, etc.) relative to damage produced by x- or gamma radiation of about 200 kv. It is given frequently as an average value in the common energy range of a particular type of ion.(8)

8. Rad means an ionizing radiation unit corresponding to a loss of energy in any medium of 100 ergs per gram. (1 rad in tissue = 100/93 rep).(8)

Definition of Problems in Radioactive Waste Disposal

In this introductory section it seems appropriate to attempt to define the objectives of radioactive waste management and disposal. We chose as definition that the objective of managed disposal or containment of radioactive wastes is to prevent serious biological damage to man, or to the complex ecological, biological, and genetic equilibrium in which man exists with his environment, this to hold for the present and for as long as a radioactivity hazard exists from the waste materials. This definition, when considered in parts, points to certain important aspects of the waste disposal problem. First, for proper perspective of the waste problem, it must be realized that radioactivity release or release potential comes not only from the waste fission products, but from many other existing and potential sources, which we have divided categorically as follows:

1. Radiation which exists in the environment. This will consist of that which occurs naturally (cosmic rays, naturally radioactive elements) and that which has been and will be released in an uncontrolled manner so that it acts subsequent to release as part of the natural environmental radioactive potential.

   a. Natural radiation

   Cosmic rays ~0.6 mrad/week

Natural radioactivity	~1 - 2 mrad/week
K^{40} in the body	0.3 mrad/week
Total	2 - 3 mrad/week

(Natural tritium, C^{14}, and Ra, etc., in the body add insignificant contributions)

This is the radiation which results from natural causes (cosmic rays, naturally occurring radium, etc.) not under our control. Each person in the U. S. receives on the average a total accumulated dose of about 4.3 roentgens to the gonads over a 30-year period. At high altitudes this dose is greater, because of the increase of cosmic rays. Thus, this background is as high as 5.5 r in some places in the United States.(2)

b. Estimate of radioactivity released to the environment

Many sources have contributed, and probably will continue to contribute, to release of radioactivity to the environment.

(1) Atomic Energy Commission production and research operations and counterparts in other countries.

(2) Reactor accidents.

(3) Release of experimental radioisotopes.

(4) Nuclear weapons fall-out. In partial evaluation of this contribution, we quote from the NAS Summary Reports:(10)

> "With certain understandings [as enumerated in the reference] it may be stated that U. S. residents have, on the average, been receiving from fall-out over the past five years a dose which, if weapons testing were continued at the same rate, is estimated to produce a total 30-year dose of about one tenth of a roentgen; and since the accuracy involved is probably not better than a factor of five, one could better say that the 30-year dose from weapons testing if maintained at the

> past level would probably be larger than 0.02 roentgens and smaller than 0.50 roentgens.
>
> "The rate of fall-out over the past five years has not been uniform. If weapons testing were, in the future, continued at the largest rate which has so far occurred (in 1953 and 1955) then the 30-year fall-out dose would be about twice that stated above. The dose from fall-out is roughly proportional to the number of equal sized weapons exploded in air, so that a doubling of the test rate might be expected to double the fall-out."

(5) Operation of power reactors. As yet the general population has not received radiation from atomic power plants or from the disposal of radioactive wastes. These are future sources of radiation that might become dangerous.

2. Intentional and controlled low-level radiation exposure of limited numbers of people for medical diagnosis and treatment, for occupational purposes (reactor and chemical plant operators; nuclear-powered vehicle crews, waste disposal crews).

a. Medical and dental X-rays(10)

According to present estimates, each person in the United States receives, on the average, a total accumulated dose to the gonads which is about 3 roentgens of x-radiation during a 30-year period. Of course, some persons get none at all; others may get more.

b. Occupational exposures

It is our understanding that limits for occupational exposure may be set as follows: Individual persons should not receive a total accumulated dose to the reproductive cells of more than 50 roentgens up to age 30 years, and not more than 50 roentgens additional up to age 40. (About half of all U. S. children are born to parents under 30, nine-tenths to parents under 40.)(10)

The International Commission on Radiological Protection recently reviewed the regulations pertaining to radiation protection. The general recommendations of this group resulting from a meeting in April, 1956, have been summarized by Dr. Morgan as follows:(7)

1. The basic permissible absorbed dose rate will continue to be 0.3 rem in a week for occupational exposure. In exceptional cases, this weekly absorbed dose may be increased by a factor that might be as large as 10 provided the integrated absorbed dose during the 13 week period following the beginning of the higher rate is not greater than 3.0 rem.

2. The absorbed dose to each occupationally exposed individual is not to exceed 5 rem per year averaged over a 10-year period. This is intended to limit the absorbed dose to penetrating radiation to 50 rem by the age of 30 and to 200 rem by the age of 60.

3. The permissible exposure rates for prolonged exposure in areas in the neighborhood of the controlled areas are to be 1/10 of those permitted within the controlled areas.

4. Until more data are available and general agreement is reached, it is considered prudent to limit the permissible genetic-absorbed dose to large populations to be of the order of natural background in presently inhabited regions of the earth. It should be stressed that the foregoing statements are not the exact wording of the ICRP committee report but rather a paraphrasing of them with special emphasis on changes from recommendations previously given in publications of ICRP and NCRP.

The recommendation of the National Bureau of Standards for maximum permissible dose is as follows:

1. Accumulated dose. The maximum permissible accumulated dose, in rems, at any age, is equal to 5 times the number of years beyond age 18, provided no annual increment exceeds 15 rems. Thus the accumulated MPD = 5 (N - 18) rems where N is the age and greater than 18. This applies to all critical organs except the skin, for which the value is double.

2. Weekly dose. The previous permissible weekly whole-body dose of 0.3 rem, and the 13-week dose of 3 rems when the weekly limit is exceeded, are still considered to be the weekly MPD, with the above restriction for accumulated dose.

Experience with occupational exposure can be taken from carefully kept exposure records at all AEC sites. At Hanford, for example, Dr. H. M. Parker reports that a safety factor of five has been maintained under the previously used 0.3 rem per week maximum permissible exposure and annual exposure limited to 3 rem. The experienced average annual exposure is in the range of 0.1 to 0.2 rem. The average exposure probable in 12 years work at Hanford would therefore be 2 to 4 rem. Since current measurements do not determine the actual dose at the gonads from internal depositions of radioisotopes, this range might more properly increase to 3 to 5 rem.

Statistics on occupational exposure control at Hanford may be of interest. In attempting to control average exposures to an annual limit of 3r, it was found that:

1. If 0.05% to 0.2% of the force exceeds 3r in any one year, 3% to 5% will exceed 1r, and the annual average will be about 0.2r.
2. If 0.0% to 0.01% exceed 3r in any one year, about 0.1% will exceed 1r, and the annual average will be about 0.1r.

Dr. K. Z. Morgan summarized radiation exposure experience in 4000 employees at ORNL as shown in Figure 11.(7)

Comparative Summary of Accumulated Exposure of ORNL Employees to Ionizing Radiation

([illegible] revised)

1.6 rem = Average accumulated occupational exposure of all employees now
*(2.6 rem) at ORNL

49.1 rem = Accumulated exposure of the single employee at ORNL who has
*(76.8 rem) accumulated the highest recorded exposure

98 rem = Accumulated exposure the person would receive who worked at
*(196 rem) ORNL for the average employment period of 6.3 years and received the absorbed dose rate of 0.3 rem/wk to the entire body or 0.6 rem to the skin for the entire period as presently permitted by HB-59 and HB-52.

31.5 rem = Accumulated exposure the person would receive who worked at
*(63 rem) ORNL for the average employment period of 6.3 years and received the average absorbed dose rate of 5 rem/yr to the entire body or 10 rem/yr to the skin for the entire period as proposed by the ICRP.

*Values given in parentheses indicate the dose to the skin. The other values are for the penetrating component of dose.

---

3. High-level radiation exposure to large segments of the world's population from intentional or accidental release of activity from fission and fusion weapons (used either for test or warfare), stationary or mobile power reactors; radiochemical processing plants or fission product processors; transportation of irradiated fuel or concentrated fission product wastes; liquid waste tanks that are part of the reprocessing plant; or from the ultimate disposal of wastes to the environment.

One of the most significant questions to be answered early in the consideration of problems associated with radioactive wastes is whether or not <u>any</u> of the fission products and transplutonics produced by the growth of a nuclear power economy can be by plan freely released to the environment, in view of the radiation exposure potential from all sources other than high-level wastes as compared to the proposed maximum permissible dose for the general population. As a partial answer to this important question we must conclude, in view of the recommended general population radiation limits of 10 roentgens from conception to age 30(3), that high-level wastes cannot be released directly to the immediate environment in which man exists.

With this conclusion the definition of what is safe for ultimate disposal becomes difficult, since we are presented with the paradox of having only our environment in which to dispose of radioactive waste. The problem thus becomes one of (1) defining how much is safe in our immediate environment, allowing for possible accidents; and (2) finding either means of containment or remote natural sites that will retain radioactive wastes until their hazard no longer exists.

The Dual Nature of the Radioactive Waste Problem

We must consider the disposal of radioactive wastes as two separate problems: 1) the problem of management and dispersal of small quantities of radioactive materials that are greatly diluted and possibly widely distributed geographically; 2) the problem of almost perpetual containment of large quantities of radioactive elements that have high biological hazard and long radioactive half-life.

The first problem will involve the control of a large number of distributed small sources of radioactivity, such as result from the use of radioisotopes in research and medicine, the use of radiation sources, and the industrial application of radioactive materials. Radioactive isotopes will appear in highly diluted form in gases, liquids and solids from radiochemical separations plants, analytical laboratories and reactor cooling circuits. Control of the distributed low-level hazard may be difficult because of the large number of source and the number of people involved. A partially satisfactory control will exist, however, since the total quantity of radioactivity issued to these channels can be monitored.

The high level wastes clearly present a problem of containment. There is a faint possibility that certain radionuclides of low biological hazard and short half-life can be bled into easily accessible natural disposal systems, such as rivers, oceans and surface formations of the earth.

In the evaluation of the possibilities of routine discharge to accessible natural disposal systems, it will always be necessary to consider the effects of an accidental release of fission products and heavy elements from the large sources of radioactivity activity circuit such as power reactors, cooling systems, chemical processing plants and ultimate high-level disposal systems. A reserve potential must be maintained in the environment, and of course in the total exposure of people who receive an accidental release of large radiation sources.

The possibility of military use of fission and fusion weapons is another factor that may limit the quantities of activity that will routinely be discharged to the readily accessible environment.

The Problem of Low-Level Distributed Hazard

The presence of radioactive wastes in quantity will have a profound effect on certain non-nuclear industries and on other important segments of our lives which may be damaged by air or water contaminated with radioactive wastes. There are numerous wet-processing industries which are likely to be deterimentally affected by radioactivity, even in trace concentrations. Among this vulnerable group are those requiring water of the highest purity, such as for the manufacture of photographic film. Other industries which should be alerted to the problem are pharmaceutical manufacturers and food processing companies. It is not possible, at this time, to enumerate with assurance the industrial processes which can be completely eliminated as subjects of this potential hazard, without the assembly of extensive research and statistical data applicable to specific operations. For example, with respect to the photographic film industry, studies must include the effect of radioactivity upon all materials entering into the finished product such as gelatine, sensitizing chemicals, paper for the mapping of films, et cetera.

It will be necessary to catalogue those industries which appear to be most vulnerable to the presence of radioactivity and to define the level of activity which may be tolerated. In the preceding paragraph, there were mentioned industries which might be affected by trace concentrations of radioactivity. Other industries may be safe with respect to these levels of radioactivity, but may be seriously affected by accidental release of waste in greater concentrations. Such concentrations may be brought about by inadequate dilution or by an accidental discharge.

There is the constant threat that low-level concentrations of radioactivity may be intermittently raised beyond permissible tolerances because of concentrations of radioactive isotopes through the action of aquatic plant life. Such growths are known to be capable of accumulating radioactivity to a level of one thousand times that of the river water. Similar concentrations can be affected by selective adsorption or absorption.

It may be entirely impractical for the installation creating the waste to remove completely all radioactive material before discharge from the site. All that may be reasonably required is to reduce the wastes to levels assuring no environmental exposure of significance, assuming that significance levels can be defined. Industries' needs for water and air of specified quality can be met only by an informed management who should be made aware of the potential problems. The particular industries' needs can then be met by adequate monitoring and supplementary treatment within the industry.

It is quite possible (in fact, it seems probable) that there will be regulation controlling radioactivity for different parts of the country and world which vary greatly, possibly as much as several orders of magnitude. It is also probable that the regulations regarding allowable dispersal quantities of radioactivity will change from time to time, being dependent upon accumulations of activity in small geographical areas and exposure experience in limited population segments.

Because of the long-term nature of the hazard of any dispersed activity, it is obvious that city, county, state, and national records of all radioactive materials released to environment may become a part of our governmental control structure. Similarly, a record of individual radiation exposures for all people from birth may also necessarily become part of our governmental and social structure. It is probable too that responsible survey and control agencies which measure radioactivity in the environment routinely and by regular general surveys will be required.

As a part of the problem of control of low-level exposures to radioactivity, the development of instrumentation, sampling techniques, and measuring devices sensitive to the low levels of radioactivity that is of consequence is required.

It is most desirable that these dangers from low-level radioactivity be publicized, after mature analysis, to assure awareness and control of the problem. So far as industry is concerned, adequate dissemination of essential information can be accomplished best through group associations for specific industries. These educational programs should be guided and directed by the appropriate governmental agency.

The education of the general public to living safely with radiation will certainly be a much more difficult problem.

We shall proceed no further than to point out the necessity for considering the problem of disposal of the distributed low-level wastes in this report, but defer the analysis of the problem for later study by county, state, and federal regulatory agencies. Most certainly, careful consideration of local disposal factors and informed technical analysis will be required.

High-level Waste Disposal and Containment

To provide information on high-level wastes, we have prepared data based on uncertain predictions of the growth of our nuclear power economy; of the type of nuclear reactors; and particularly on the nature of chemical processes that discharge radioactivity in great quantity. The accuracy of many of the predictions and extrapolations is questionable, but the over-all waste picture obtained by so doing has real value.

Many factors must be considered to frame properly the waste disposal picture. In this report we will consider the following:

(1) The Chemical, Physical and Radiochemical Nature of Nuclear Wastes

- (a) Influence of reactor type
- (b) Wastes from operating reactors
- (c) Influence of radiochemical processes
- (d) Chemical and physical nature of highly active process wastes
  - 1) For liquids
  - 2) For solids
  - 3) For gases
- (e) Influence of specific fission products and their decay half-lives
- (f) Radiochemical nature
  - 1) Heat production
  - 2) Concentrations

(2) Magnitude of the Waste Problem in a Growing Nuclear Power Economy

- (a) Rate of nuclear power growth
- (b) Estimation of the magnitude of the radiochemical waste production for growing nuclear power economy
  - 1) Equilibrium quantity of each fission product
  - 2) Estimated physical volume of wastes
  - 3) Calculated decay rates for individual fission products and gross waste
  - 4) Heat production by radioactive wastes
  - 5) Estimated production of transuranics and transplutonics
  - 6) Distribution of fission product and transplutonic activity in the power reactor complex

(3) Relative Biological Hazard of Fission Products and Heavy Elements

(4) Hazard Potential Due to Accidents

(5) Processes Associated with Waste Disposal

- (a) Specific fission product removal and concentration
- (b) Processing of bulk wastes for disposal

(6) Transportation of Active Wastes

- (a) Optimum cooling period determination before shipment
- (b) Costs versus shipping distances
- (c) Estimation of transport for irradiated fuel and for high-level wastes

(7) Possibilities for Ultimate Waste Management or Disposal

- (a) Ocean
- (b) Land
    - 1) Salt deposits
    - 2) Dry caves and sealed faults
    - 3) Surface pits - retention in surface soils or solids burial
    - 4) Deep wells
    - 5) Tanks or lagoons
- (c) Space

(8) Economic Considerations and Data

(9) Absolute Hazard Potential of Wastes

- (a) Source strength, fission product spectrum, and heavy element concentrations
- (b) Chemical form of disposed wastes
- (c) Accessibility of gross waste deposit
- (d) The natural of the environment in which wastes are deposited
- (e) Accessibility of radioactive components

(f) Effect of decay on hazard reduction

(g) Probability of exposure of humans or human environment to waste

(h) Effect of radiation exposure on recipients

3.0 The Nature of High-level Radioactive Wastes as Defined by Reactors-and Chemical Process Techniques

The definition of the physical, chemical and radiochemical nature of wastes must start with a consideration of the types of reactors that may be represented in a nuclear power economy. Figure 1 graphically simplifies the reactor picture. A more thorough discussion of reactor types and an extensive bibliography on this subject appear in Appendix I.

3.1 Radioactivity Release from Reactors

(1) In circulating fuel reactors, such as the aqueous homogeneous reactor (Appendix I, page 10) the liquid metal fuel reactor (Appendix I, page 8) the primary fission gases may be obtained free of the circulating fuel as fast as they are formed by the fission process. These gases are (for U^{235} thermal fission) shown in Table 1. Since it is desirable for reasons of neutron economy to continuously remove Xe^{135}, all gases may be bled from the reactor circuit; Kr^{85} is discharged to the let-down systems along with Xe^{135}. Radioactive iodine isotopes present the most serious biological hazard to reactor operating personnel in case of even minor leaks in the reactor and in its protective enclosure.

(2) In solid fuel element reactors fission products are contained by protective metallic skins; gases are released only when ruptures occur, and then usually only in small quantity.

(3) In reactor cooling systems the following examples of induced activities

ORNL-LR-Dwg.-19982
UNCLASSIFIED

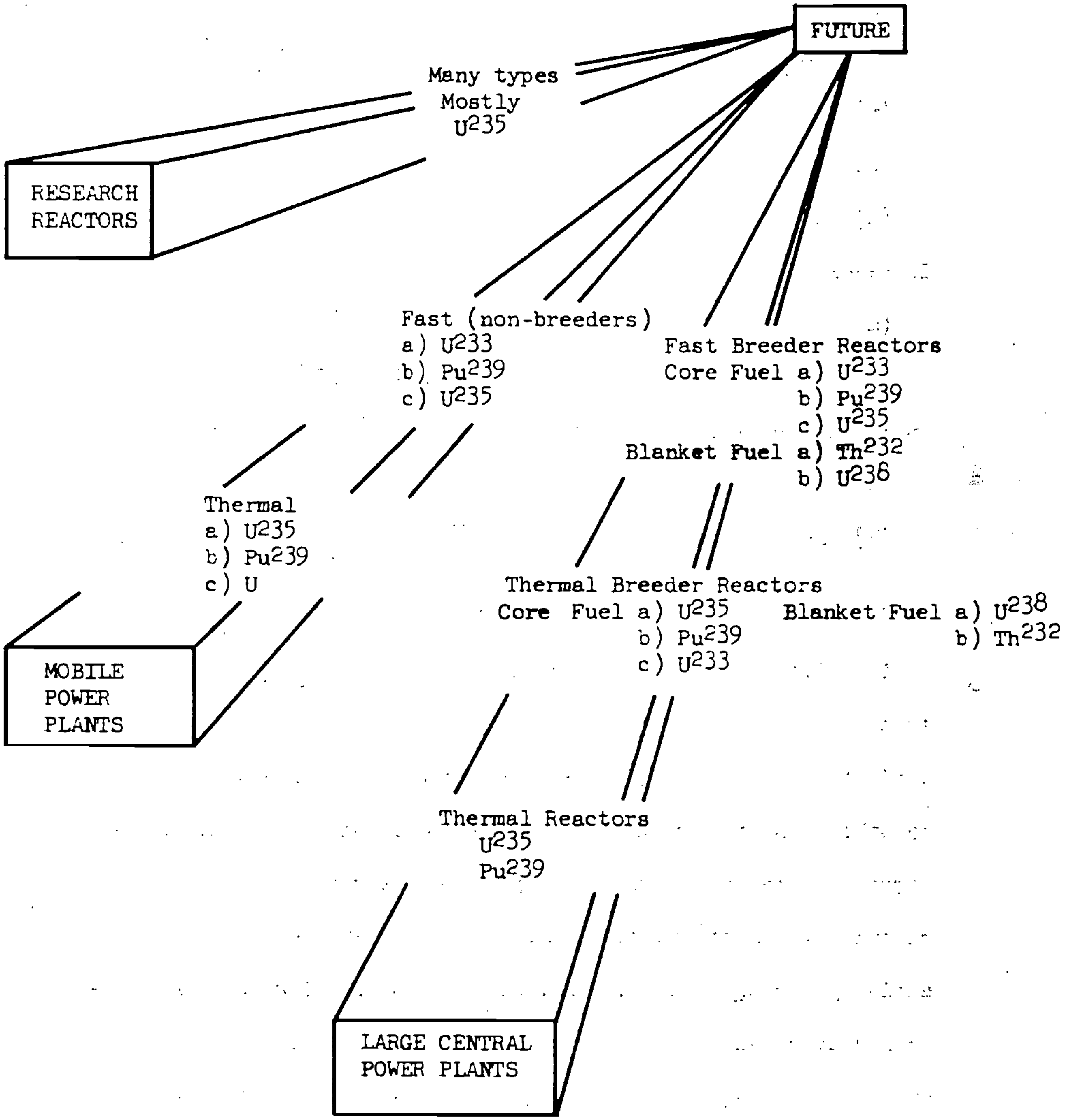


FIGURE 1

can be produced by neutron capture:

In H_2O:

Coolant	Active Isotopes Produced
(a) Na	15hr Na^{24}, 60s Na^{25}
(b) O_2	29s O^{19} (activity negligible)
(c) N_2	7.3s N^{16}
In gases:	
(a) Air	C^{14}, N^{16}, O^{19}, et cetera
(b) CO_2	5.58×10^3y C^{14}, 29s O^{19}
In liquid metals:	
(a) Na	Na^{24}, Na^{25}
(b) NaK	Na^{24}, Na^{25}, 1.8×10^9y K^{40}, 12.4h K^{42}, etc.
(c) Bi	138.3d Po^{210} (5.3 mev α, 0.78 mev γ)

Radioactivity induced by thermal neutron capture on water coolants is of relatively short half-life. After short decay periods, induced activity in water (e.g., due to activation of dissolved sodium, etc.) probably can be discharged to ground water under controlled conditions. Carefully treated reactor cooling water is recirculated in closed circuits for most reactors. Accidental release of fission product activity through ruptures in fuel element cladding is handled as an emergency condition, for which processing provisions are made. Induced activities in gas coolants are expected to be insignificant factors in disposal. A discussion of induced activity in reactor cooling water has been given by Moeller.(6)

However, activities produced in liquid metal coolants, e.g., Na, NaK or Bi, may provide rather severe problems due to 15hr Na^{24}, 138.3d Po^{210}, etc. Assuming a liquid metal fuel containing U^{235} at 600 ppm in bismuth, Po^{210} has an effective yield of 5.7% per fission. After 180 days Po^{210} activity would be 33 times that of Sr^{90}, and thus would constitute a very dangerous biological hazard in either the reactor or fuel processing cycles. This ratio would still be 9 to 1 even after three years operation.

(4) For all reactors, the potential hazard of accidental release of activity due to a catastrophic accident exists. As an indication of how much fission product activity is contained in a reactor, it has been pointed out(12) that a 500 MW (heat) electrical power unit which has operated for three years contains as much Sr^{90} as now exists in our stratosphere.

Table 1

Gaseous Fission Products from U^{235} Thermal Fission

Nuclide	Mass No.	Half-life t 1/2	Absorption Cross Section σ_a Barns	Yield %	Total β & γ Decay Energy Mev
$_{35}Br$	82	35.8h	-	3.8×10^{-5}	3.8
	83	2.4h	-	0.48	0.364
	84	30.0m	-	1.1	3.5
	85	3.0m	-	1.5	0.83
	other very short half-life				
$_{36}Kr$	85	10.27y	15	0.3	0.232
	87	78.0m	470	2.7	1.57
	88	2.77h	-	3.7	0.36
	89	3.18m	-	4.6	1.3
	other very short half-life				
$_{53}I$	129	1.72×10^{7}y	11	1.0	0.09
	131	8.05d	600	2.9	0.58
	132	2.4h	-	4.4	2.43
	133	20.8h	-	6.5	1.01
	134	52.5m	-	7.6	1.92
	135	6.68h	-	5.9	1.85
	other very short half-life				
$_{54}Xe$	131^{m2}	12.0d	-	0.03	0.163
	133^{m}	2.3d	-	0.16	0.233
	133	5.27d	-	6.5	0.196
	135^{m}	15.6m	-	1.8	0.52
	135	9.13h	3.5×10^{6}	6.5	0.570
	137	3.9m		5.9	1.33
	138	17.0m		(7.6)	1.0
	other very short half-life				

The Chemical Processing Cycle

A large chemical complex is required to supply fuel to reactors and to recover from them partially depleted and new fissionable material as illustrated by Table 2 showing various types of fuel processing which are either in operation or might be feasible based on present knowledge:

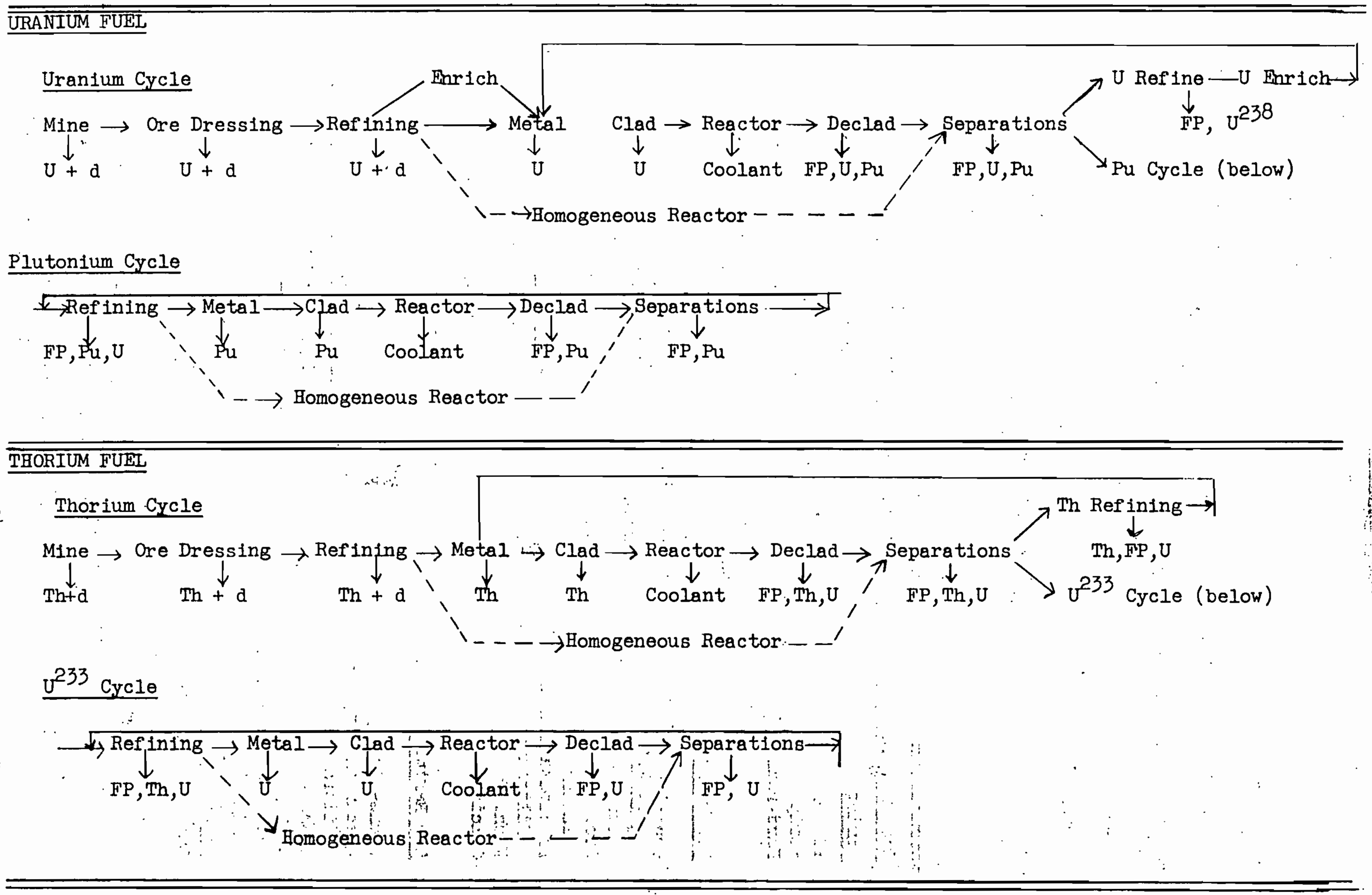
Table 2

PRINCIPAL TYPES OF FUEL PROCESSING

Ore Processing	Fuel Fabrication	Separations, Refining, Waste Treatment	
Mining		**Solvent Extraction**	
Placer	Recasting	TBP (Purex, Thorex)	
Strip	Shaping	Hexone (Redox)	
Hard-rock	Coating	Ethers (diethyl dibutyl,	
	Decoating	diisopropyl,	
	Pickling	Cellosolve)	
Ore Dressing	Canning	Diisopropyl Carbinol	
		Dibutyl Carbitol (Butrex)	
Crushing and grinding		Chelation (TTA)	
	Decladding	Triglycol Dicholride (Trigly)	
Flotation		Dioctyl Pyrophosphoric Acid (OPPA)	
Roasting	Dissolving		
Leaching	Machining		
Precipitation and filtration		**Precipitation**	
Washing		Bismuth Phosphate	
Calcining		Sodium Diiuranate	
		Scavenging ($Ni_2Fe(CN)_6$, $Ca_3(PO_4)_2$, $Sr_3(PO_4)_2$, MnO_2)	
Ore Refining			
Redissolving			
Precipitation and filtration		**Volatilization**	**Miscellaneous**
Washing		Liquid media	Ion exchange
Drying		Fused salts	Permeable membranes
Substitution (Cl, F NO_3, C_2O_4)		Fluorides	Electromagnetic
Electromagnetic			Evaporation
Gaseous Diffusion			Calcination
Reduction to metal		**High Temperature Processes**	Electrodeposition
		Slagging	
		Molten metal or salt extraction	
		Electrolysis	
		Distillation	
		Modified deBoer	
		Fusing (ceramic, glass)	

FIGURE 2

RECYCLE STEPS FOR EACH OF THE THREE FISSIONABLE ELEMENTS



The functioning of this complex can be affected at many points by changes in allowable radiation exposures to operating personnel; exposure potentials comparable or greater than those provided by a single reactor are possible. The steps in the recycle for a nuclear power economy for each of the fissionable elements is shown schematically in Figure 2.

The reprocessing of irradiated fuel elements produces the high-level fission product wastes as raffinates. The nature of chemical processes for accomplishing the simultaneous separation of fissionable and fertile material from fission products and from each other is discussed and referenced in Appendix I. Our processing experience, based on solvent extraction for plutonium production or for the recovery of enriched U^{235} from U-Al alloy used for MTR fuel elements, is not representative of processes that will be required for proposed reactor fuels. Because of the varied nature of reactor fuel elements for power reactors, adaptions of known process techniques or the development of entirely new processes will be required. The state of process development for reactors that have been proposed for power production does not allow an accurate estimate of the chemical and physical nature of wastes. Figure 3 shows the status of development for generalized fuel types.

Wastes from a chemical reprocessing plant appear as solids, liquids (fission product salts in aqueous solutions) or gases (fission product gases and/or particulates suspended in a non-radioactive carrier gas such as air or nitrogen). It is probable that for many years the high-level wastes will be aqueous raffinates from such basic processes as solvent extraction. Let us therefore turn our attention to high-level aqueous wastes produced as first cycle raffinates from solvent extraction processes.

Fig. 3. Status of Reprocessing Methods for Various Reactor Fuel Types

Process	Aluminum			Zirconium			Stainless Steel			Homogeneous			Ceramic			
	Th and < 3% U^{235} Hanford	10-40% U^{235} Geneva	~90% U^{235} MTR	Th and < 3% U^{235} Commonwealth	10-40% U^{235} APDA	~90% U^{235} STR	Th and < 3% U^{235} Consumers	10-40% U^{235} Holyoke	~90% U^{235} SIR	UO_2SO_4 HRT	ThO_2 PAR	U, Bi LMFR	UO_2, ThO_2	UO_2, PuO_2 FOB	UC_2 Graphite	UC_2 SiC_2
Aqueous with conventional refabrication	■	▨	■	▧	▧	▨	▧	▧	▨	▩	▧	▧	□	□	□	▧
Volatility with conventional refabrication	■	□	□	□	□	▩	□	□	▧	□	□	□	□	□	□	□
Pyrometallurgy with remote refabrication	□	□	□	□	▧	□	▩	□	□	□	□	▧	□	□	□	□

Key:

■ Has been operated on a significant scale with irradiated material.

▨ Ready for operation on a significant scale with irradiated material.

▩ Undergoing engineering-scale development.

▧ Undergoing laboratory-scale development.

□ Development not begun.

3.3 Nature of Wastes from Radiochemical Processes

The Idaho Chemical Processing Plant processes enriched U-Al fuel by a solvent extraction process using hexone. In addition to raffinates from processing of this type of fuel the Idaho Plant will produce wastes from other fuels as shown in Table 4. All wastes described will contain high concentrations of ions other than fission products (i.e. Zr(IV), Al(III)) which will limit their concentrations by evaporation. The processes which will produce the wastes described are those required for recovering highly enriched U^{235} from inactive diluent and cladding metals. A summary of the approximate nature of high-level wastes from other solvent extraction processes is given in Table 5.(28)

An attempt has been made to estimate the characteristics of the wastes emanating from the processes for proposed stationary power reactor fuels. First, an estimate was made of the number of reactors of a given type by the year 1980 for the United States only, based upon the major types of power reactors under consideration today. The distribution assumed (broken down by total power) was approximately 23% fast reactors of which the Detroit Edison type is used as an example; 23% homogeneous reactors, such as Wolverine and ORNL-TBR; 13% heterogeneous thorium breeders, such as Consolidated Edison; 9% seed and blanket type, such as the Rural Cooperative; and the remaining 32% were assumed to be slightly enriched heterogeneous reactors, such as that of Commonwealth Edison. Table 6 lists the distribution of reactor types by 1980.(29)

Although this distribution is arbitrary, it does cover the currently proposed major types of reactors and possible chemical processes which will yield aqueous wastes. Table 7(29) lists the waste volumes and waste characteristics for each of the reactor chemical process combinations which are under study for processing power reactor fuel elements by aqueous chemistry. Table 8(29) lists

TABLE 4

~~ICPP FIRST CYCLE~~ AQUEOUS WASTE FLOWSHEET COMPOSITIONS (UNCONCENTRATED)

		Hexone Extraction		TBP Extraction							
		Aluminum Alloy - A	MTR	Al Alloy (TBP)	Al Alloy (TBP)	H_2SO_4	Acid Stainless	HF Zircaloy		Sodium Graphite	PWR Seed
Specific Volume liters/kg 25(1)	M	515	825	592	545	447	223	2500	415	7.2	960
H^+	M			1.06	0.96	3.37	0.45	2.14	0.70	1.37	2.1
Al^{+++}	M	1.42	1.50	1.5	1.51		0.70	0.75	0.70	0.43	0.75
Zr (IV)	M							0.55		0.03	0.55
NH_4^+	M	1.31	0.82	0.05			0.39		0.78	1.96	
Hg^{++}	M	0.012	0.0012	0.011	0.005						
Other Metals	M				0.1	0.108		0.01			0.007
NO_3^-	M	5.34	5.07	5.63	5.50	2.73	2.59	3.59	2.88	4.5	4.3
F^-	M							3.00		0.18	3.0
Acid Deficiency	N	0.25	0.25								
$SO_4^=$	M					0.47					
Sp. G.	M	1.255	1.250			1.094		1.216	1.15		

---

(1) Per kg U^{235} after burnup

Table 5

CHARACTERIZATION OF FIRST CYCLE HIGH LEVEL AQUEOUS WASTES FROM SELECTED SOLVENT EXTRACTION PROCESSES

	Chemical Properties, Constituents (M), Approximate, Exclusive of Fission Products and Heavy Elements																		Waste Volume, gal/g U^{235} Consumed	Approximate Specific Gravity	Approximate Concentration of U in Feed g/liter	Waste Activity$^{(2)}$		
Process	H	Al	Fe	Cr	Ni	Zr	Na	NH_4	NH_3	Sn	Mn	Mg	Hg	NO_3	F	SO_4	PO_4	Cl				curies/gal	Total watts/gal$^{(3)}$	BTU/hr/gal
Redox	-0.2	1.2	*	*	*		0.46							4.1					0.27	1.18	450	1720	8.6	29.4
Purex	2.5 to 7.0		*	*	*		*							2.5 to 7.0		*			0.34	1.07	300	1320	6.7	22.7
Thorex	-0.1	0.5	*	*	*		*						0.01	1.4	0.05		*		0.34	1.12	350 (Th)	81	0.4	1.37
Hexone-"25"	-0.2	1.5	*	*	*			0.5					0.01	4.5					0.31 to 0.13	1.23	2-5	1620 to 3940	8.1 to 20.2	27.7 to 69.0
TBP-"25"	0.5	1.6	*	*	*								0.01	5.5		0.02	*		0.22 to 0.11	1.29	3-6	2580 to 5160	12.9 to 26.0	44.1 to 88.6
Zirconium-HF for enriched U	0.8 to 2.0	.8		0.1		0.5				0.02				2.3	3.0				~1.50	~1.1	0.3	350	1.75	6.0
Stainless Steel-H_2SO_4 for enriched U-235	2.6	0.25	0.05	0.01	003		*				0.001			2.4		0.30			~0.35	~1.3	2-3	1500	7.5	25.0

Notes: (1) Wastes are untreated; they are essentially as they leave the solvent extraction plant and are subject to further treatment such as evaporation, neutralization, chemical treatment for fission product removal, etc.

(2) Basis for activity numbers: Irradiation period 4000 Mwd/t for natural uranium
5×10^{13} n/(cm)^2(sec)
4000 grams U^{233} chain per ton of thorium
53% burn-up for U^{235} in enriched fuel elements
100 days decay cooling from time of reactor discharge

(3) After 100 days' decay, the distribution of energy is approximately 50% γ and 50% β.

(4) Waste volume per gram U^{235} consumed is an inverse function of burnup; i.e., for hexone-25 at 20% burnup, the gal/g U^{235} = $(\frac{53}{20})(0.14)$.

(5) Waste activity varies approximately as the (irradiation level)$^{0.2}$

TABLE 6

DISTRIBUTION OF REACTOR TYPES BY 1980

IN UNITED STATES ONLY

BASED ON POWER BUILDUP CURVE OF J. A. LANE

Reactor Type	Power Level for Each (Heat Mw)	No. of Each Type	Total Power for Type (Heat Mw)	Processing Rate for Type (tons/yr)	Burnup or Irradiation Level	Initial Enrichment
Consolidated Edison	560	25	14,000	200 (Th)	57% U^{235}	>90%
Thorium Breeder					49% U^{233}	--
					(10,000 g/t)	Th
Commonwealth Edison	720	25	18,000	364	12,200 Mwd/t	1.5%
Detroit Edison	400	62	24,800	342 (c)	~ 4% U^{235}	27%
Fast Breeder				2040 (b)	Ax.76 Mwd/t	Dep.
					Rad 984 Mwd/t	Dep.
Consumers Public Power	300	25	7,500	610	4,250 Mwd/t 16.2% U^{235}	2.27%
Yankee Atomic Electric	555	15	8,000	330	7,000 Mwd/t 24% Bu	2.49%
Seed and Blanket Type (Modified Version of Rural Cooperative Reactor)	360	25	9,000	1.10 (c) 182 (b)	40% U^{235} ~10,000 Mwd/t	>90% Nat.
Homogeneous						
Wolverine Electric						
Modified	480	25	12,000	2.2 ("25")	400% U^{235}	48% (Eq.)
ORNL-TBR	480	25	12,000	550 (Th)	6,000 g/t	THO_2 (b)
				1.3 ("23"C)		~33% "23"C)
Total Power			105,300			
Total Th Processing Capacity				750	7,000 g/t	
Total Nat. or Sl. Enriched Uranium Capacity				3,526	3,600 Mwd/t	
Total Uranium Core Capacity						
(Highly Enriched)				4.6		
(10-30% Enriched)				342		

TABLE 7
VOLUMES AND CHEMICAL COMPOSITION OF WASTES FROM VARIOUS REACTOR FUEL PROCESSES

Reactor Type	Fuel Element and Sub-Assembly Type	Process Description	Feed Conditions to HA Column	Waste Vol From HA Col gal/kg U or Th	Waste Vol After Evap. gal/kg U or Th	Waste Vol After Evap. gal/gm "25" consumed	Waste Conditions for Final Disposal	Estimated Vol of Final Waste of Each Type gal/yr By 1980	Comments
Consolidated Edison Thorium Breeder	Core & Blanket Plates Core - U-Zr Blanket Th Clad - Zr-2 Alternate Plates 32 Core - 33 Blanket Irradiate to 10,000 g/t Th	Dissolution 1) HCl Gas Phase 2) HNO_3 Modified Int 23 ORNL-MR-HEP	420 g/l Th 12 g/l U 0.5 M HNO_3	0.94	0.94	0.10**	280 g/l Th 0.33 M $Al(NO_3)_3$ 0.3 M HNO_3	188,000*	*Either of these or any combination - not all three ** gals/gm "23" produced. Has additional $ZrCl_4$ waste
		Thorex ORNL-MPP	350 g/l Th 10 g/l U -0.1 M HNO_3 0.56 M Al	1.36	0.394	0.04	Long Cooled No Pa 2.0 M Al -0.38 M HNO_3	79,000*	
				0.59	0.394	0.04	Pa Recovery - Final Waste -0.64 M HNO_3 2.5 M Al	79,000*	
Commonwealth Edison	UO_2 Pellets 1.48% "25" Tube Sub-Assembly 25 Sub-Assemblies per Assembly 3600 g Pu/T U Zr Clad-SS ends	Dissolution 1) HCl Gas 2) HNO_3 Purex ORNL-MR-HEP	324 g/l U 1.15 g/l Pu 2 M HNO_3	1.24	0.124	0.0135	~ 7 M HNO_3	45,000	Purex Type Waste gives advantage of 10:1 volume reduction
		(Alternate) Idaho FAN 0.5 M HF 0.7 M ANN 3 M HNO_3	24 g/l U 0.093 M Zr 2.4 M H^+ 0.5 M F^-, 0.75 M Al 4.15 M NO_3^-	13.2	5.0	0.55	1.6 M Al 1.1 M F^- 6.3 M H^+ 0.21 M Zr 10.8 M NO_3^-	1,850,000	Conditions based on optimistic volume reduction may have extensive evaporator corrosion

Reactor Type	Fuel Element and Sub-Assembly Type	Process Description	Feed Conditions to HA Column	Waste Vol From HA Col gal/kg U* or Th	Waste Vol After Evap. gal/kg U* * or Th	Waste Vol After Evap. gal/gm "25" consumed	Waste Conditions for Final Disposal	Estimated Vol of Waste of Each Type by Year 1980 gal/year	Comments
Detroit Edison Fast Breeder									
Core	Pin (Swaged at ends) 144 Pins Sub-Assembly 2.16 Kg Mo 20.16 Kg U(27% "25") 0.86 Kg Zr 4.8 Kg SS	ORNL-MR-HEP 1) Dissolution 1-6 M H_2SO_4 2) HF & HNO_3 Mo- Centrifugation	12 g/1 U(a) 0.048 g/1 Pu 6 M HNO_3 Some ANN	33.6 1.06 H_2SO_4 Waste	33.6	3.1	5.8 M HNO_3 0.3 - 0.5 M ANN 0.03 M F^-+ Zr	11,500,000	Has SS Waste from H_2SO_4 dissolution (a)Criticality Limitations gives low concentration
Core Alternate Scheme to above Not in addition		ORNL-MPP Dissolve with HNO_3(13 M) 846 L $Al(NO_3)_3$, 13.12 Kg H_2O 15.4 l KF 1.74 Kg (18 Assemblies) day 364.5 kg U	237 g/1 U 2.44 g/1 Pu 5 M HNO_3 0.05 M Zr 0.35 M $Al(NO_3)_3$ 0.3 M F	1.71	1.71	0.158	3.97 M HNO_3 0.033 M Zr 0.23 M $Al(NO_3)_3$ 0.2 M F^-	585,000	Indicate advantages of designing process in terms of criticality limits
Axial Blanket	Rod (ends recessed) 16 Pins/Sub Assembly 13.97 Kg U 0.03 Kg Na 3.8 Kg SS	ORNL-MR-HEP Aq. Regia Dissolution	324 g/1 U 88 g/1 SS 2 M HNO_3 0.13 g/1 Pu	1.22	1.22	0.113*	2.3 M HNO_3 58 g/1 SS	2,040,000	High SS content makes volume reduction difficult
Radial Blanket	Rod 25 Pins/Sub Assembly 71.25 Kg U 0.15 Kg Na 16.3 Kg SS	ORNL-MR-HEP Aq. Regia Dissolution	324 g/1 U 1.3 g/1 Pu 2 M HNO_3 74 g/1 SS	1.22	1.22	0.113*	2.3 M HNO_3 48.6 g/1 SS		*Based on Core burnup since blanket is depleted Uranium

Reactor Type	Fuel Element and Sub-Assembly Type	Process Description	Feed Conditions to HA Column	Waste Vol From HA Col gal/kg U or Th	Waste Vol After Evap. gal/kg U or Th	Waste Vol After Evap. gal/gm "25" consumed	Waste Conditions for Final Disposal	Estimated Vol of Waste of Each Type by Year 1980 gal/year	Comments
onsumers ublic Power eactor	Slug - 0.455" 2.27% "25" 2500 g Pu/T(Final) 10 elements per Sub-Assembly NaK bond 19 Sub-Assemblies per Assembly SS Structure.	Remove ends by Sawing Aq. Regia Dissolution	324 g/l U 0.81 g/l Pu 28 g/l SS 2 M HNO_3	1.24	1.24	0.354	2.3 M HNO_3 18.4 g/l SS	757,000	Has Solid SS Waste ~ 34 tons/yr on assumed basis
Yankee omic Electric Reactor	Cylindrical Pellet Sintered UO_2 2.49% "25" 128 Sub-Assemblies per Assembly Pellet in tubular Sub-Assembly 131.6 Kg U, 0.7 Kg Pu 27 Kg SS	Sawing Aq. Regia Dissolution	324 g/l U 67 g/l SS 1.72 g/l Pu 2 M HNO_3	1.24	1.24	0.182	2.3 M HNO_3 44 g/l SS	410,000	Has Solid SS Waste
		(Alternate) Idaho SS 0.4 T/Day SS Dis. in 6 M H_2SO_4 44.6 g SS/l in 4.8 M H_2SO_4 UO_2 Dis. in 3 M HNO_3	35.6 g/l U 3 M HNO_3 0.75 M H_2SO_4 7.3 g/l SS	9.15	2.30	0.34	~ 7 M HNO_3 ~ 2.5 M H_2SO_4 24 g/l SS	760,000	Only Approximate waste condition This Volume is Alternate to that above - not additional

Reactor Type	Fuel Element and Sub-Assembly Type	Process Description	Feed Conditions to HA Column	Waste Vol From HA Col gal/kg U or Th	Waste Vol After Evap. gal/kg U or Th	Waste Vol After Evap. gal/gm "25" consumed	Waste Conditions for Final Disposal	Estimated Vol of Waste of Each Type by Year 1980 gal/year	Comments
Seed and Blanket Type (Rural Cooperative Reactor)	Plate Type Seed Highly Enrich. "25" Plate Type Nat U Blanket Both Clad in Zr 6 Seed Plates/ Assembly 4 Blanket Plates per Blanket Assembly Based on 1.25 Kg Seed per ton Nat. U	Separate Streams, Seed: HCl Gas Dissolution - $ZrCl_4$ ZrO_2 Waste Removal	(Seed) 3 g/l U 6 $\underline{M}$ HNO_3	106	10.6	0.026	7.0 $\underline{M}$ HNO_3 1.2 $\underline{M}$ Al^+ 0.03 $\underline{M}$ $Fe(NH_2SO_3)_2$	11,700	
		Blanket HCl Gas Dissolution - $ZrCl_4$ & ZrO_2 Waste Removal (10,000 Mwd/t) ORNL-MR-HEP	(Blanket) 324 g/l U 1.3 g/l Pu 2 $\underline{M}$ HNO_3	1.08	0.11	0.046*	7.0 $\underline{M}$ HNO_3 (Purex)	20,500	*Based on Core Burnup
Homogeneous									
Wolverine Electric Co.	UO_2SO_4 in D_2O 7 g U^{235}/Kg D_2O Highly Enriched (Initially) Ship in SS as slurry in light water	Dissolution of U & SS Container in Aqua Regia	0.28 $\underline{M}$ H_2SO_4 6 g/l U 6 $\underline{M}$ HNO_3	18.2 (U^{235})	18.2 (U^{235})	4.5*	~ 0.2 $\underline{M}$ H_2SO_4 5.3 $\underline{M}$ HNO_3 0.003 $\underline{M}$ $Fe(NH_2SO_3)_2$	40,000	Have Solid Waste Sludge *Equivalent to 400% Burnup
ORNL TBR (2-Region)	UO_2SO_4 in D_2O (C) Th O_2 in D_2O (B)	HNO_3, F^- Dissolution of Th O_2 Add Hydroclone underflow from core (ORNL-1761)	1.5 $\underline{M}$ Th 0.5 $\underline{M}$ Al -0.1 $\underline{M}$ HNO_3 2.8 g U/l ~.2 g Pa/l	0.48	0.48	0.10	1.87 $\underline{M}$ Al -0.35 $\underline{M}$ HNO_3	264,000	This type best suited for on site processing Processing of single region slurry reactor would yield similar waste conditions

Reactor Type	Fuel Element and Sub-Assembly Type	Process Description	Feed Conditions to HA Column	Waste Vol From HA Col gal/kg U or Th	Waste Vol After Evap. gal/kg U or Th	Waste Vol After Evap. gal/gm "25" consumed	Waste Conditions for Final Disposal	Estimated Vol of waste of Each Type by gal/year	Comments
search									
MTR Type	Plate Type U-235 in Al MRT Highly Enriched	HNO_3 Dissolution of U-Al Alloy ORNL-MPP	5 g/l U (avg) 1.0 M HNO_3 1.8 M Al $(NO_3)_3$	135 (68 Avg)	135 (68 Avg)	0.54	1.5 M HNO_3 1.625 M $Al(NO_3)_3$	108,000	Based on 800 Kgs Enriched U/Yr + 400 Kgs "25" 20% Enrich/Yr
Foreign	Foreign 20% "25" (35% U in Al)		Ratios and Through-put proportioned to Al Content.	3	3	0.06		30,000	

TABLE 8

ACTIVITY OF WASTES FROM VARIOUS REACTOR FUEL PROCESSES AS A FUNCTION OF DECAY TIME

Reactor Type	Flux ϕ_F = fast flux ϕ_T = thermal flux (n/cm^2/sec)	Irradiation Level (and Irradiation Time) (1std units)	Activities (1) Curies/kg or gal (2) total watts/kg or gal (3) Btu/hr/kg or gal At Decay Times of (Units/kg U or Th) 1 day	10 days	Activities (1) Curies/kg or gal (2) total watts/kg or gal (3) Btu/hr/kg or gal At Decay Times of (Units/gal of waste) 60 days	200 days	500 days	1000 days	2000 days	3000 days	Waste Volume gal/kg U or Th for Basis (see Table 2)
Consolidated Edison	$\phi_F = 2.2 \times 10^{14}$	10,000 g/t	(1) 1.13×10^{5}	(1) 7.0×10^{4}	(1) 3.29×10^{4}	1.10×10^{4}	4.38×10^{3}	1.98×10^{3}	9.1×10^{2}	6.0×10^{2}	0.94
Thorium Breeder	$\phi_T = 6.5 \times 10^{13}$	57% Bu (25)	(2) 566	(2) 350	(2) 164	55	21.6	9.9	4.5	3.0	
		(360 d)	(3) 1940	(3) 1200	(3) 564	188	75	33.8	15.5	10.3	
Commonwealth Edison	$\phi_F = 1.1 \times 10^{14}$	11,400 Mwd/t	(1) 1.1×10^{4}	(1) 6.3×10^{3}	(1) 2.45×10^{4}	1.08×10^{4}	4.95×10^{3}	2.79×10^{3}	1.62×10^{3}	1.20×10^{3}	
	$\phi_T = 1.6 \times 10^{13}$	62% (25)	(2) 48	(2) 26	(2) 91	36	14.4	6.9	3.8	2.9	0.124
		(1020 d)	(3) 164	(3) 89	(3) 310	123	49	24	13	10	
Detroit Edison Fast Breeder											
Core	$\phi_F = 5 \times 10^{15}$	~ 4% Bu	(1) 1.3×10^{5}	(1) 8.1×10^{4}	(1) 1.52×10^{4}	3.9×10^{3}	1.01×10^{3}	300	219	137	1.71
		(104 d)	(2) 660	(2) 410	(2) 77	19.7	5.1	1.55	1.11	0.69	
			(3) 2200	(3) 1370	(3) 257	66	17.1	5.1	3.70	2.29	
Axial Blanket	$\phi_F = 3 \times 10^{14}$	400 g/t	(1) 640	(1) 400	(1) 105	27	7.0	2.1	1.51	0.95	1.22
		(104 d)	(2) 3.2	(2) 2.0	(2) 0.53	0.14	0.035	0.011	0.0076	0.0048	
			(3) 11.0	(3) 6.8	(3) 1.8	0.46	0.12	0.036	0.026	0.016	
Radial Blanket	$\phi_F = 3 \times 10^{14}$	3-4000 g/t	(1) 870	(1) 600	(1) 252	117	57	33	18	11.9	1.22
		(1435 d)	(2) 4.4	(2) 3.0	(2) 1.3	0.6	0.30	0.17	0.093	0.062	
			(3) 14.8	(3) 10.2	(3) 4.3	2.0	0.97	0.57	0.31	0.20	
Consumers Public Power Reactor	$\phi_T = 2 \times 10^{13}$	16.2% (25)	(1) 1.96×10^{4}	(1) 1.1×10^{4}	(1) 3.66×10^{3}	1.3×10^{3}	4.78×10^{2}	2.2×10^{2}	1.10×10^{2}	78	
		(395 d)	(2) 87.3	(2) 42.5	(2) 13.7	4.55	1.44	0.56	0.25	0.17	1.24
			(3) 298	(3) 145	(3) 47	15.5	4.9	1.9	0.86	0.58	
Yankee Atomic Electric Reactor	$\phi_T = 2 \times 10^{13}$	8400 Mwd/t	(1) 2.2×10^{4}	(1) 1.2×10^{4}	(1) 4.0×10^{3}	1.44×10^{3}	5.25×10^{2}	2.37×10^{2}	1.21×10^{2}	86	
		(365 d)	(2) 96	(2) 46.6	(2) 15.1	5.0	1.58	0.61	0.27	0.19	1.24
			(3) 328	(3) 159	(3) 51.5	17	5.4	2.1	0.93	0.65	
Seed & Blanket Type	$\phi_T = 3 \times 10^{13}$		(1) 1.19×10^{6}	(1) 6.4×10^{5}	(1) 3.14×10^{4}	9.44×10^{3}	3.5×10^{3}	1.55×10^{3}	7.4×10^{2}	5.2×10^{2}	
	(Seed)	40% Bu	(2) 5470	(2) 2620	(2) 96.5	33.2	10.6	4.1	1.73	1.2	10.6
	(365 d)		(3) 18,700	(3) 9000	(3) 330	114	36	14	5.9	4.1	
	$\phi_T = 3 \times 10^{13}$	10,000 Mwd/t	(1) 1.02×10^{4}	(1) 6.2×10^{3}	(1) 2.98×10^{4}	1.75×10^{4}	8.64×10^{3}	5.1×10^{3}	2.98×10^{3}	2.4×10^{3}	
	(blanket)		(2) 46	(2) 24	(2) 86	46	23	12.5	7.5	5.6	0.11
	(6-8 Yr)		(3) 157	(3) 82	(3) 293	160	79	43	25.6	19.1	
Homogeneous											
Wolverine Type	$\phi_T = 1 \times 10^{14}$	400% Bu	(1) 7.0×10^{5}	(1) 3.5×10^{5}	(1) 9.6×10^{3}	6.4×10^{3}	3.2×10^{3}	1.6×10^{3}	1.0×10^{3}	800	
	(3000 d)		(2) 3500	(2) 1750	(2) 48	32	16	8	5	4	18.2
			(3) 12,000	(3) 6000	(3) 164	110	55	28	18	14	
ORNL-TBR Type	$\phi_T = 3 \times 10^{14}$ (c) $\phi_T = 6\text{-}8 \times 10^{13}$	(c) 260% Bu (190 d-c)	(1) 8.0×10^{4}	(1) 5.6×10^{4}	(1) 2.8×10^{4}	7.0×10^{3}	3.0×10^{3}	1.4×10^{3}	820	475	
		(b) 6000 g/t (320 d-b)	(2) 400	(2) 280	(2) 140	35	15	7.1	4.1	2.4	0.48
		(Th Basis)	(3) 1370	(3) 956	(3) 475	120	52	24	14	8.2	

the activity level of 1) the material to be processed from each reactor type and 2) the chemical process waste streams as a function of decay time. Table 7 also lists research reactors from which fuel probably will be processed by aqueous methods. No mention has been made of liquid metal reactors, fused salt reactors or reactors which might be processed by fused salt methods, since there is insufficient data on the nature of these wastes.

These tabular values of concentrations, volumes or activities are not firm but are only estimates of the conditions which would occur if groups of these several reactors were processed by those methods listed. Process flowsheets are either in the laboratory development stage or are extrapolations of existing technology. None of the flowsheets have been demonstrated on an engineering scale with the assemblies for which they are proposed.

3.4 Nature of the Fission Product Gases as a Waste

From a chemical reprocessing plant the fission product gases appear in the off-gas systems of sealed processing vessels, highly diluted with diluent gases such as air, nitrogen, water vapor and oxygen. A list of radioactive gaseous fission products from U^{235} thermal fission would include the elements previously listed in Table 1.

The important gases from a long term waste disposal standpoint are Kr^{85}, and much less significantly I^{129}. From the standpoint of hazard during reactor operation and short-cycle processing, I^{131} is by far the most significant. It will always be necessary to remove iodine continuously from the gas discharge streams which can be accomplished by processes previously mentioned (4)(5), but possibly not to a complete enough degree for use in areas adjacent to highly productive land or metropolitan living areas. The solution to the problem of complete iodine removal and its isolation will require further development and research.

Hazard from the release of xenon and krypton is slight and confined to the vicinity of the reactor. However, because of its long half-life of 10.27 years Kr^{85} builds up to a significant concentration as a result of an expanding nuclear power economy, possibly to 90 megacuries by 1980 when 10^5 megawatts of heat may be produced by fission. In Table 9 a rough estimate has been made of the buildup of rare gases in the atmosphere by the year 2000 A.D., assuming the growth curve for a nuclear power economy predicted by Lane in which 700,000 Mw of nuclear heat generation capacity is predicted.

The effects of release of the noble fission product gases to the atmosphere are roughly as shown below:(13)

TABLE 9

ESTIMATE OF Kr^{85} AND Xe^{133}

RELEASE FROM POWER REACTORS TO THE ATMOSPHERE, 2000 A.D.(13)

Assumptions		
1. Kr^{85} production, U.S. alone		700 megacuries
2. Xe^{133} production, U.S. alone		37,000 megacuries
3. Entire world product, 3 times above		
4. Uniform distribution of the atmosphere		
5. Mass of air in atmosphere	or	4×10^{21} grams 3.1×10^{18} cubic meters STP
6. Mixing time for complete mixing		2 weeks
7. Background radiation, atmosphere at sea level		0.015 mr/hr
Calculations		
1. Kr^{85} activity		7×10^{-10} curies/meter^3
2. Xe^{133} activity		3.6×10^{-8} curies/meter^3
3. Dose from Kr^{85} for which biological tolerance of 2×10^{-6} c/cc is assumed (for total body irradiation)	or	6.3×10^{-4} mr/hr 4.2% of background
4. Dose from Xe^{133} for which tolerance, based on total body irradiation, is 4×10^{-6} c/cc of air	or	1.6×10^{-2} mr/hr 107% of background
5. Total dose Kr^{85} and Xe^{133} =		1.66×10^{-2} mr/hr

Thus, even with complete mixing, the contribution of released fission gases is appreciable. If mixing is not complete, and assuming adverse meteorological conditions, concentrations can be high. The contribution to general atmospheric activity by Xe^{133} can be eliminated by trapping and storing this gas for about 60 days or ten decay half-lives. Kr^{85} would require storage for much longer periods of time since it decays with an approximate ten-year half-life.

We conclude that Kr^{85} probably will require isolation and containment for decay before release to prevent a slow-build-up of atmospheric background count. 700 megacuries of Kr^{85} accumulated in the air surrounding the earth to a height of ten miles, could increase air background by 4.2 per cent or greater (background assumed to be 0.015 mr/hr).

The release of noble fission product gases to the atmosphere without decay may be possible for the early periods of nuclear power generation, but a maximum cut-off allowable quantity to be released must be established, a quantity which probably will be lower than the equilibrium accumulation of Kr^{85} by 1975 and later.

3.5 Particulate Solids as Wastes in the Gas Phase

There are problems of control of particulates from reactor cooling circuits; from all phases of the preparation of uranium and thorium and intermediates for reactor fuels; from radiochemical reprocessing; from analytical chemical and low-level isotope use; and particularaly from the handling of highly alpha active materials such as plutonium polonium, americium, curium, neptunium, uranium-233 and possibly thorium. Much attention has been devoted to particulate activity; careful study and active research have been sponsored by the AEC as a result of programs initiated by the Stack Gas Committee.(11) The classified literature contains much data and discussion which we shall not review.

3.6 General Comment About Handling Contaminated Gases

Since the maximum permissible concentrations for radioactive substances in air are very low, it is necessary that essentially all active gases and particulates be removed from reactor and chemical plant effluents. Installations and processes to remove active gases can be very expensive, require heavy shielding, remote operation, and careful and difficult analytical control.

The removal of radioactive particulates from a gas stream is also very expensive, requires shielding, remote operation and without present techniques is difficult to control either by continuous monitoring or by sampling and chemical analysis.

The cost of gas cleaning facilities can be significant in the construction and possibly the operating costs of any radiochemical facility, particularly a radiochemical reprocessing plant. For example, in the Idaho Chemical Processing Plant, the following costs were incurred for the off-gas and ventilation facilities in 1950-1952.(14)

(1)	Dissolver off-gas decontamination system	$ 619,300
(2)	Dissolver off-gas collection system	35,400
(3)	Sampler off-gas system (ind. filters)	46,700
(4)	Vessel off-gas system (ind. filters)	89,500
(5)	Cell ventilation system (no filters installed), including $194,000 for 250 ft. acid brick lined stack	440,300
(6)	Bldg. and cells to house air cleaning and process equipment, prorated share of base waste bldg. cost of $1,330,000 total	897,500
	TOTAL	$2,128,700

(7) Total construction cost of entire plant, exclusive of engineering, start-up, certain service facilities—$21,400,000

NOTE: Laboratory hoods and certain special filters not included,

These costs do not include filters for general processing cell ventilation. At Hanford such cell ventilation air cleanup facilities have been installed in the form of extensive deep bed sand or glass wool filters. One of the questions that must be answered in each processing installation, or in fact in any facility designed to handle radioactivity, is whether or not all ventilation air must be filtered before discharge.

Another fact that may be overlooked is that most gas and ventilation cleaning or processing systems produce relatively large volumes of low-level liquid waste from such equipment as scrubbers, electrostatic precipitators, silver nitrate regeneration solution for I_2 removal towers; or difficultly handled solid filters. Facilities must be provided for handling these liquid or solid wastes from air or gas cleaning operations.

3.7 Solid Radioactive Wastes

Solid radioactive waste materials have been produced in great variety in the AEC program. Others will be produced in the future. A partial list follows:

(1) Solids of a Low-level of Contamination

- (a) Laboratory combustibles - incinerated under controlled conditions, usually with high efficiency scrubbers or filters on flue gas streams.
- (b) Radioactive particulate contaminated fibrous or granular filters.
- (c) Contaminated glassware - experimental equipment, sample bottles, etc.
- (d) Inactive portions of fuel rod and control rod assemblies.
- (e) Experimental animals and animal residues from destructive disposal.
- (f) Certain reactor fuel cladding materials which can be mechanically separated from fissionable or fertile material in the fuel.
- (g) Certain reactor coolants.
- (h) Contaminated processing equipment.
- (i) Crucibles, molds, and recasting furnaces.
- (j) Gaskets, filter elements, glove boxes, etc.

(2) Solids of High-level of Contamination

- (a) Resudues containing fission products from such processes as the oxidative slagging of molten uranium; dissimilar metal or fused salt raffinates from high temperature fuel element reprocessing.
- (b) Solid residues and scums, along with filters, filter aids, etc., from aqueous-organic fuel element reprocessing.
- (c) Metal components of reactor fuel elements made active by parasitic neutron capture which can be removed by mechanical processing.

(d) Absorbents and adsorbents for process of off-gas streams circulating fuel reactors or chemical reprocessing plants.

(e) Fission product and transplutonic bearing fused salts from fuel element reprocessing using such techniques as fluoride volatility.

(f) Oxides for such reactors as aqueous homogeneous breeder which uses ThO_2.

(g) Precipitated concentrated fission product mixtures from aqueous homogeneous reactors, circulating fuel reactors like the liquid metal fuel reactor; and from fission product isolation processes.

(h) Fixed gross fission products in any of the many proposed disposal, chemical and physical forms.

Solid radioactive wastes, such as machine turnings, useless contaminated equipment and contaminated trash which are generated in all operations, have not constituted a serious technical problem as yet. The levels of activity associated with solid wastes for which disposal has been attempted have varied from a few times background to those requiring shielding or remote handling. To date, burial of such wastes under known, controlled conditions and, in specific isolated AEC owned locations, disposal at sea have successfully handled the low-level problem. Relatively small quantities of radioactivity (estimated in the range of hundreds of curies, with the bulk coming from UCRL and BNL) have been disposed of at sea.

Established burial grounds exist only at large atomic energy production and research sites such as Oak Ridge, Savannah River, Idaho, Los Alamos, and Hanford. Solid wastes, however, originate at all locations where radioactive materials are used. At areas other than those noted above which usually encompass comparatively small areas and are near or in densely populated sections, it is the general policy not to dispose of solid wastes on site, but to ship them off site for final disposition.

Of some concern here is the problem of locating suitable burial grounds to facilitate and reduce the cost of handling and disposal of these solid wastes. This problem is particularly acute in the Northeastern United States where, at present, the only available receivers for these wastes are Oak Ridge and the Atlantic Ocean. In view of shipping and sea disposal costs, it is obvious that a more centralized burial facility is required. There is, of course, every indication that future operations both in and out of AEC will greatly accentuate this need.

Experience with handling highly active solids as a disposal problem, with its associated problems of heat generation and removal, particulate protection, container design, receiving site preparation, and disposal media have received little attention experimentally. Much work remains.

4.0 Growth Predictions for Nuclear Reactor Capacity and the Magnitude of the Associated Fission Product and Transplutonic Waste Problem

Many predictions have been made concerning the growth of a nuclear power economy in the United States and for other parts of the world. We have been somewhat at a loss to predict rates of nuclear reactor power buildup, but have attempted an estimate of the magnitude of the waste disposal problem based upon predictions of J. A. Lane.(15)

Before presenting the results of fission product buildup calculations, we will present other estimates and data along with which our estimates can be considered.

Reactors for Central Station Power Generation

At the present time there are no nuclear reactors in operation in the United States whose principal purpose is the generation of electrical power. The power reactor demonstration program of the Atomic Energy Commission does provide for the building of a number of such plants, however. The earliest of these is to be in operation in 1957, while the last will be in operation by 1962. In addition to these plants, there is a significant group of reactor power plants to be constructed in the same period which are financed in whole or in large measure by private capital. The total electrical generating capacity of all of these planned plants is somewhat over 800,000 kilowatts. As a first point of reference, therefore, it may be noted that there are firm plans in existence to place in operation in the five years from 1957 to 1962 electrical generating capacity of over 800,000 kilowatts powered from nuclear reactors.

It has been estimated in the McKinney report(16) that the installed central station electrical generating capacity in the United States powered from nuclear reactors will amount to 3 to 4 million kilowatts by 1965 and will range from 20 to 45 million kilowatts by 1975. The wide variation in the predictions of the amount of nuclear power to be expected in the next two decades is due to differences in judgment as to the speed with which technical obstacles can be overcome and competitive costs achieved.

Based on present technology, the cost of nuclear power is very high indeed compared to the cost of power derived from fossil fuel in a modern, favorably located station. The over-all costs -- as they are known today -- favor fossil fuels by a factor of from five to ten. It is certain that the development programs presently planned will reduce the cost of nuclear power sharply. Despite this fact, the cost of nuclear power is so high today that it is by no means clear that nuclear power will become competitive with power derived from fossil fuels for at least ten to twenty years.

Because of this situation, it seems reasonable to project the nuclear power capacity for the next ten years on the basis of large experimental plants built principally to study the technical problems of such units. These plants will be built not only with government subsidy but by private capital as well, since many utility companies will wish to study the operational problems of nuclear power plants at first hand. On this basis, the planned electrical generating capacity of 800,000 kilowatts by 1962 can grow to 3,000,000 kilowatts by 1965 with little acceleration in the present rate of growth. An upper estimate based on an accelerated rate of growth would be 5,000,000 kilowatts of installed generating capacity by 1965. It might be noted that it is dubious whether the industrial capacity for supplying plant equipment could grow sufficiently, rapidly to provide 5,000,000 kilowatts of installed capacity by 1965 for large, experimental plants.

The growth in installed plant capacity from 1965 to 1975 is entirely dependent on the technical progress made in the next ten years. It seems most likely that the technological problem will be overcome slowly. For this reason the growth in the decade 1965 to 1975, which will be motivated primarily by economic considerations, will likewise be relatively slow. An estimate, primarily one of technical judgment, is that the installed electrical generating capacity based on nuclear power will grow to 20,000,000 kilowatts by 1975.

The electrical generating capacity figures must be translated for purposes of this study into average fission heat release rate by the reactors. The overall efficiency from reactor heat release to electrical output for nuclear plants of the size contemplated will range roughly from 25 to 33%. The duty of these plants, i.e., the equivalent fraction of time at which they are operating at capacity, is estimated to be a maximum of 75%. An upper estimate of the average reactor heat generation rate, using an overall plant efficiency of 25% and a duty of 75%, is three times the installed electrical generating capacity. Using installed electrical generating capacity figures of 3,000,000 kilowatts by 1965 and 20,000,000 kilowatts by 1975, the average rate of heat release from nuclear reactors supplying central generating stations is estimated to be 9,000,000 kilowatts in 1965 and 60,000,000 kilowatts in 1975.

Reactors for Marine Propulsion

There has been some Congressional discussion concerning the application of nuclear power plants for commercial marine propulsion. Some steps are being taken in this direction by the Maritime Administration. While the application is technically feasible today, the economic feasibility is questionable. According to the McKinney report, the introduction of nuclear propulsion for specialized application, such as tankers, could lead to the installation of reactors capable of 15,000,000 kilowatts of heat in the 1980 period. It does not seem that such installations could change the estimates made here for the next two decades in any substantial manner. For this reason this application will not be taken into account in the estimated totals.

Reactors for Locomotive Propulsion and Other Vehicles

There seems to be little interest at this time in application of nuclear reactors for locomotive propulsion. While technically the application is probably feasible, the economics do not appear to be favorable.

The application of nuclear power reactors to the propulsion of automotive vehicles seems highly unlikely. For much the same reason, application to military vehicles, such as tanks, gun carriers and prime movers, seems equally unlikely for the immediate future.

Research Reactors

Nuclear reactors for research purposes have already had widespread application in development of nuclear energy. Such reactors are now being utilized by several universities and institutes of technology as teaching and research aids. Industrial laboratories have announced plans for research reactors for a variety of research and development purposes.

The power level of research reactors generally ranges from 100 to 1000 kilowatts. At the present rate of growth, there will probably be fifty or more installations by 1965 and perhaps several hundred by 1975. Without considering the fraction of time that such reactors may be operating at full power, it is clear that they can contribute only a small increment to the fission heat release rate of power reactors.

Test Reactors

Test reactors are used principally for isotope production, tests of developmental fuel elements for power reactors, as well as for general research purposes. The power rating of such reactors ranges from 10 to 50 megawatts. Several such reactors will be in operation by 1957 and it is probable that 10 to 20 may be in operation by 1965. At an average power level of 30,000 kilowatts, this might represent 300,000 to 600,000 kilowatts of reactor heat release by 1965. These reactors have, in general, a very high duty. It is not likely that there will be a spectacular growth of test reactors from 1965 to 1975 because of the specialized nature of their application. It is possible that the number of test reactors may double in this period. Such reactors will represent then, at most, a small fraction of the heat rate of power producing reactors.

Reactors for Chemical Processing

Nuclear reactors have a possible application in chemical processing to supply heat or nuclear radiation. At the present time, nuclear reactors do not appear to be economically attractive as a source of heat. Some development work is being carried on at present in the application of nuclear radiation to chemical processing. No industrial application is presently planned even on a pilot plant basis. While such applications could lead to very substantial reactor plants, there is no basis at present on which to estimate the possible magnitude of such plants. It seems likely that there will be no significant installations for this purpose by 1965.

A related use of nuclear reactors is for the production of fissionable materials for weapons purposes. There is no basis available for estimating the installed reactor capacity presently devoted to this purpose or likely to be devoted at some future time.

Package Power Plants

A military application of nuclear power which is receiving attention at this time is the development of package power plants for remote military bases. At least two such are under development and this will probably lead to the construction of a number of such plants. The capacity of the plants is fairly low -- of the order of 1,000 to 10,000 kilowatts. The total number of such plants which might be installed will probably be limited.

Summary

The preceding discussion indicated the principal source of fission products from nuclear reactors in the next two decades will arise from the generation of electricity at nuclear power central stations. Other sources are comparatively small and amount to substantially less than the uncertainty in the estimates of the principal uses.

Zeitlin, Arnold and Ullmann have predicted processing requirements and the buildup of fission product wastes.(19) The following calculations and data are taken wholly from their report.

4.1 Processing Requirements, Buildup of Fission Product Activity, and Liquid Waste Volumes in A Predicted Nuclear Power Economy

Based on a predicted nuclear power economy growth rate made by J. A. Lane(15), calculations have been made to determine as a function of time the magnitude of several quantities of prime importance to the radiochemical processing and waste disposal industry. Included are such quantities as required processing capacity, buildup of activity of important fission products, and accumulated volume of high activity liquid wastes.

If it is assumed that the installed electrical plant capacity of the United States will increase eight-fold during the next 50 years, that there will be 500 MW of installed nuclear electric capacity in 1960, and that half of all new plants built in the year 2000 will be nuclear plants, the installed nuclear plant capacity, N, in megawatts, at time T, in years after 1960, will be given by the expression:

$$N = 5{,}800 \quad (1.09^{T} - 1) + 5000 \qquad (1)$$

Assuming a thermal efficiency for the reactor system of 25 per cent, the above equation becomes, for heat power requirements:

$$N(\text{MW heat}) = 23{,}200 \quad (1.09^{T} - 1) + 2000 \qquad (2)$$

A curve representing this equation for the period from 1960 to 2000 A. D. is given in Figure 4.

Differential equations were set up for the simultaneous growth and decay of various fission products being produced by the expanding power economy. The solutions give the total number of curies of a fission product in existence at any time between 1960 and 2000. It should be emphasized that the activities plotted are not only the activities to be found in waste disposal tanks but include fission products present in the reactors and fuel being stored prior to processing. This is especially important for the shorter-lived isotopes such as Ba^{140}.

UNCLASSIFIED
ORNL-LR Dwg. 9239

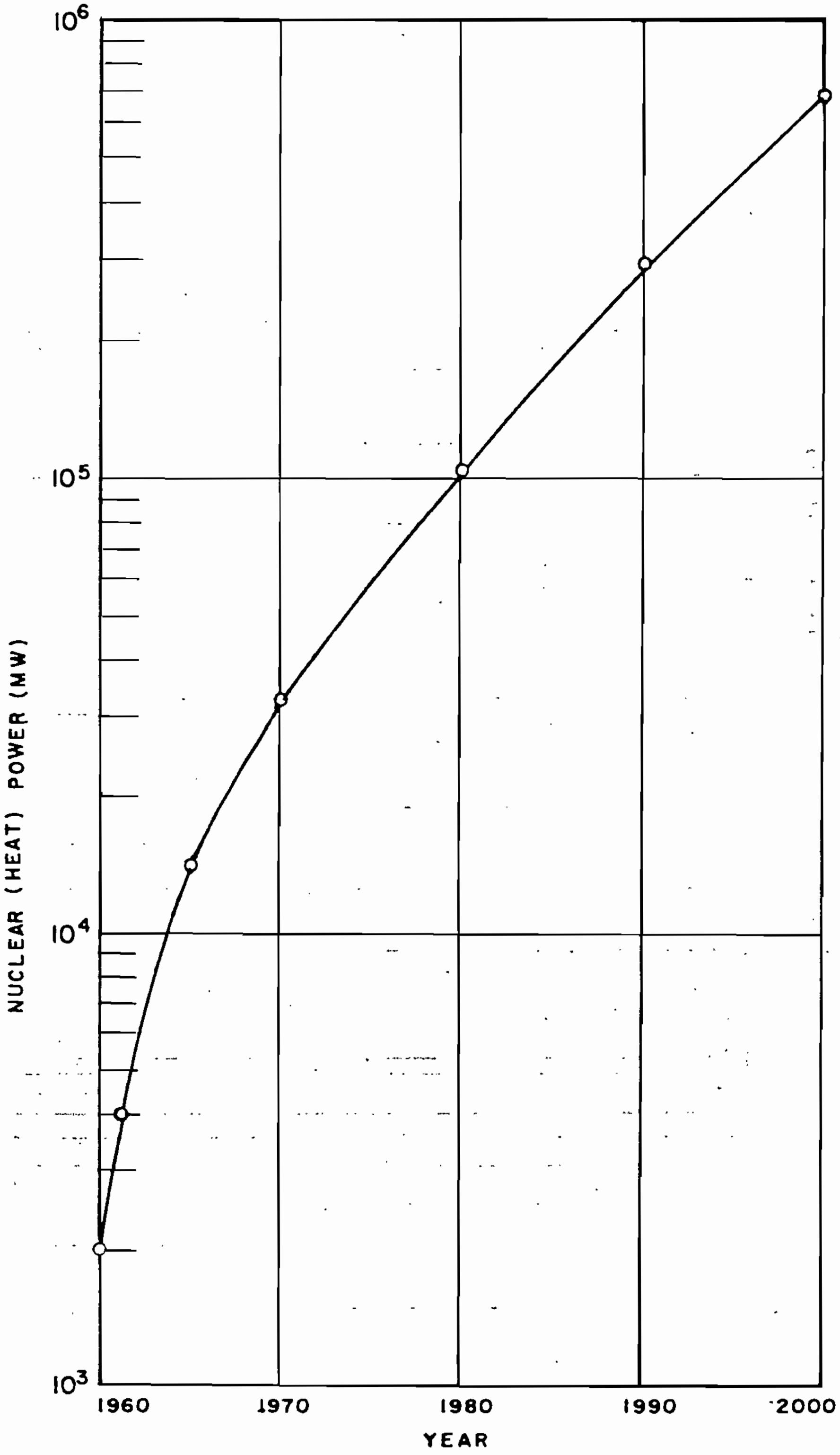


FIGURE 4 PREDICTED NUCLEAR (HEAT) POWER GENERATION FOR THE UNITED STATES

UNCLASSIFIED
ORNL-LR-DWG 73418R

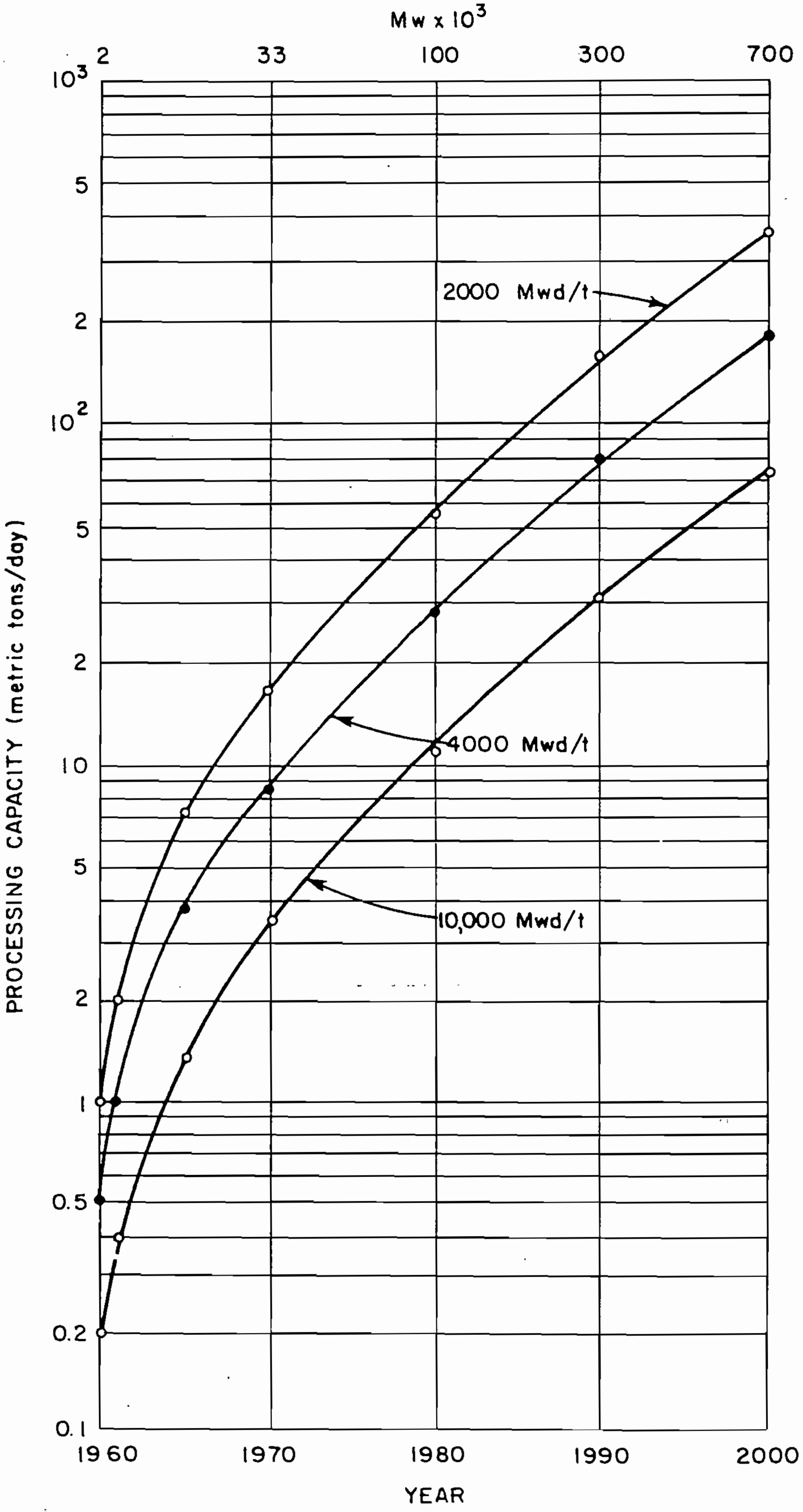


Fig. 5 Radiochemical Processing Capacity Requirements in a Predicted Nuclear Power Economy.

The following items were plotted as a function of time:

(1) Processing capacity required (Figure 5)

Note: In Figures 5, 6, 7, the average irradiation level of the spent reactor fuel is taken as a parameter.

(2) Total spent fuel processed (Figure 6)

(3) Accumulated volume of high activity waste (Figure 7)

(4) Total accumulated activity (Figure 8)

Note: The dashed lines in Figures 8 through 13 indicate the decay of activity from various selected points along the primary accumulation curve. These lines can be used (as in Figure 8) to determine the accumulated amounts of activity following any specified cooling period. By "accumulated activity" is meant the integrated production of fission product activity with time minus loss by decay over a like period of time minus loss by neutron capture over that fraction of the period of time during which the fission product was in the reactor (the last item being negligible and making the curves essentially independent of the reactor design and operating conditions). The activity can be considered as being dumped into and accumulated in a common "sink".

(5) Accumulated activity of important fission products (Figure 10)

Note: The dashed lines indicate the decay of gross activity from various selected points along the primary curve. An infinite family of curves can be generated from these decay lines to indicate the total accumulated activity in the "sink" for any cooling period preceding discharge of the activity into the "sink". Selected members of this family of curves have been plotted.

UNCLASSIFIED
ORNL-LR-Dwg 7344 R

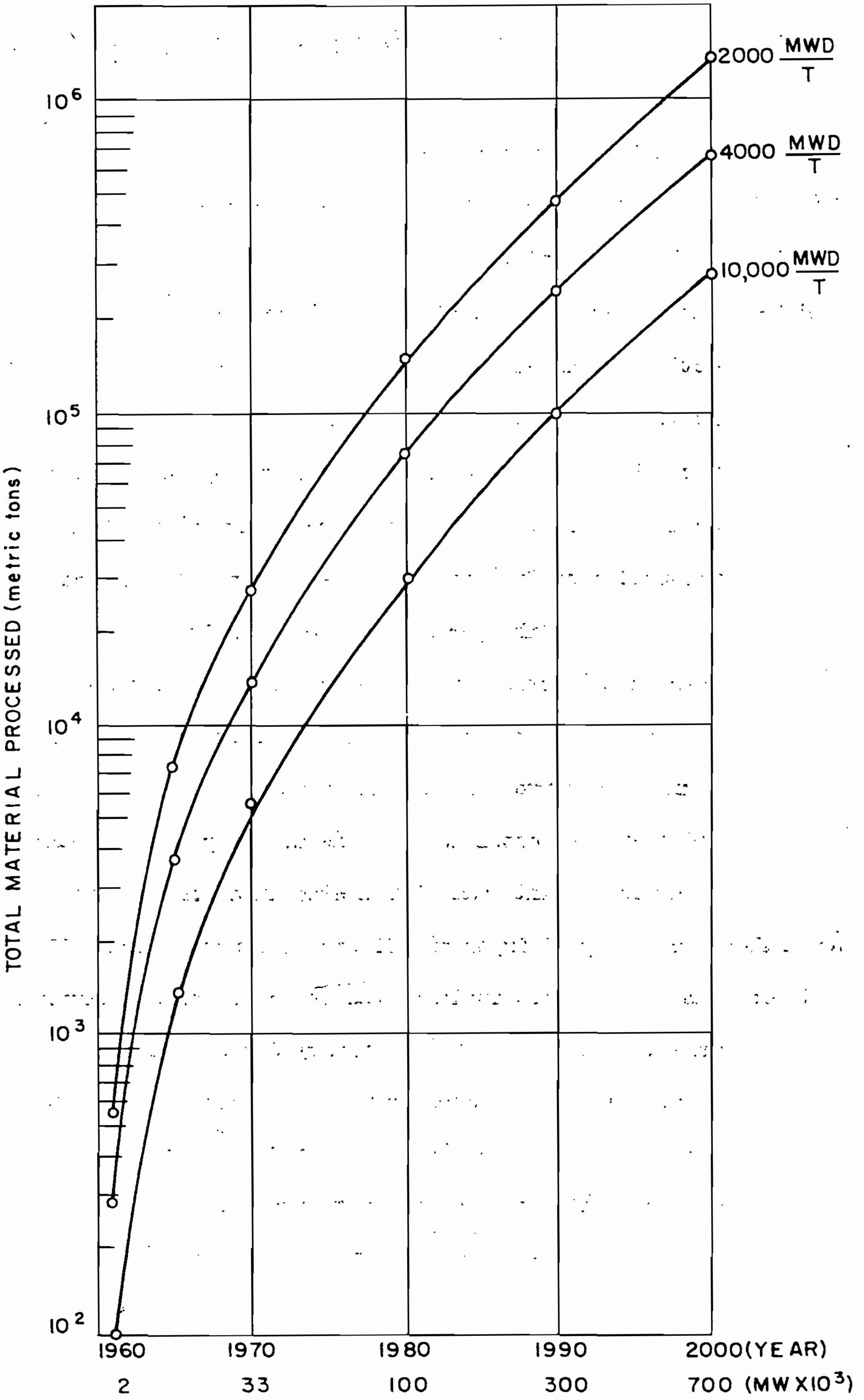


Fig. 6 TOTAL SPENT FUEL PROCESSED

UNCLASSIFIED
ORNL-LR-Dwg 7343 AR

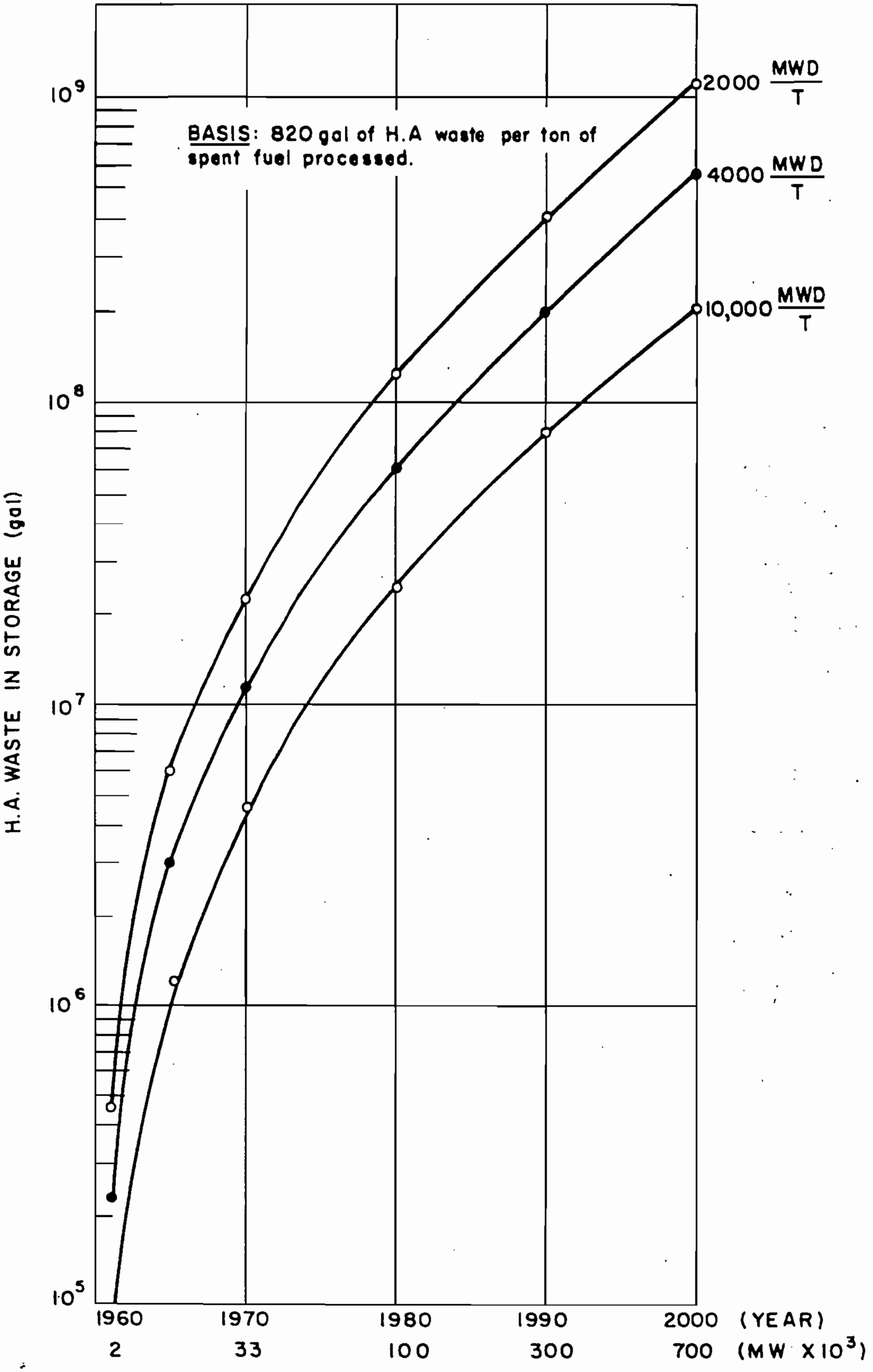


Fig. 7 ACCUMULATED VOLUME OF HIGH ACTIVITY WASTE

UNCLASSIFIED
ORNL-LR-DWG 73458

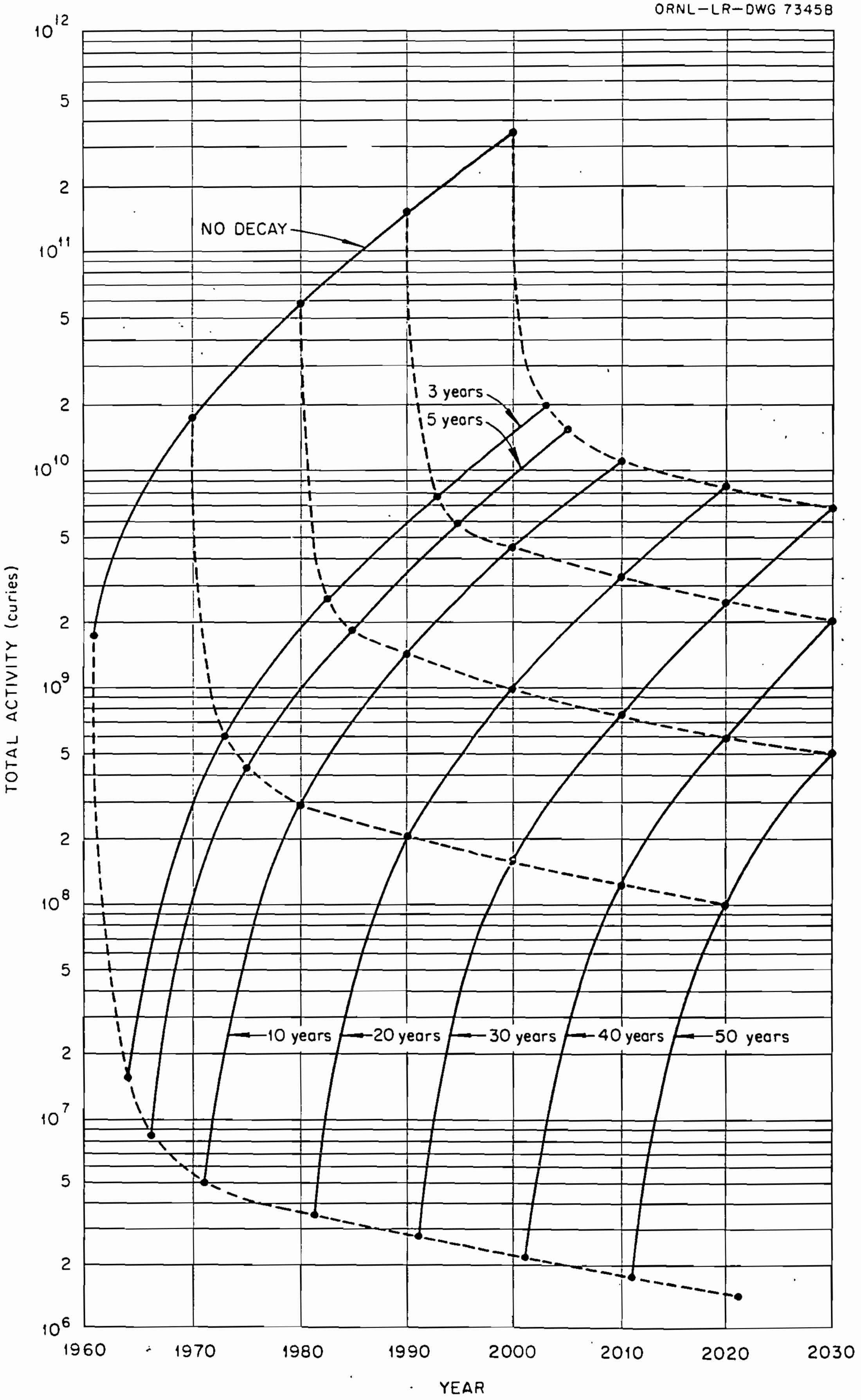


Fig. 8 Total Accumulated Activity Resulting from a Predicted Nuclear Power Economy.

UNCLASSIFIED
ORNL-LR-DWG 73468

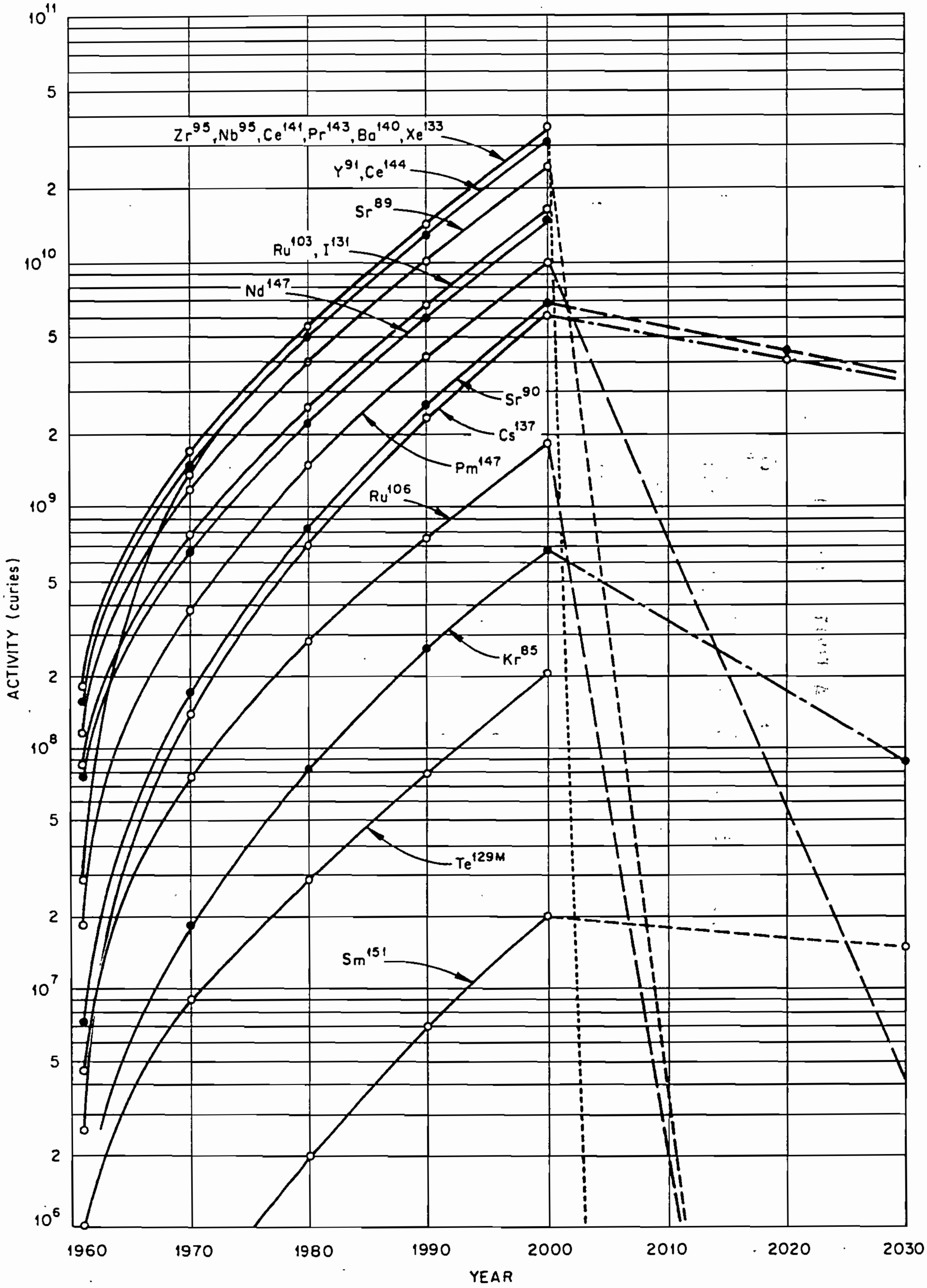


Fig. 9 Accumulated Activity of Important Fission Products Generated in a Predicted Nuclear Power Economy.

UNCLASSIFIED
ORNL-LR-Dwg 7347

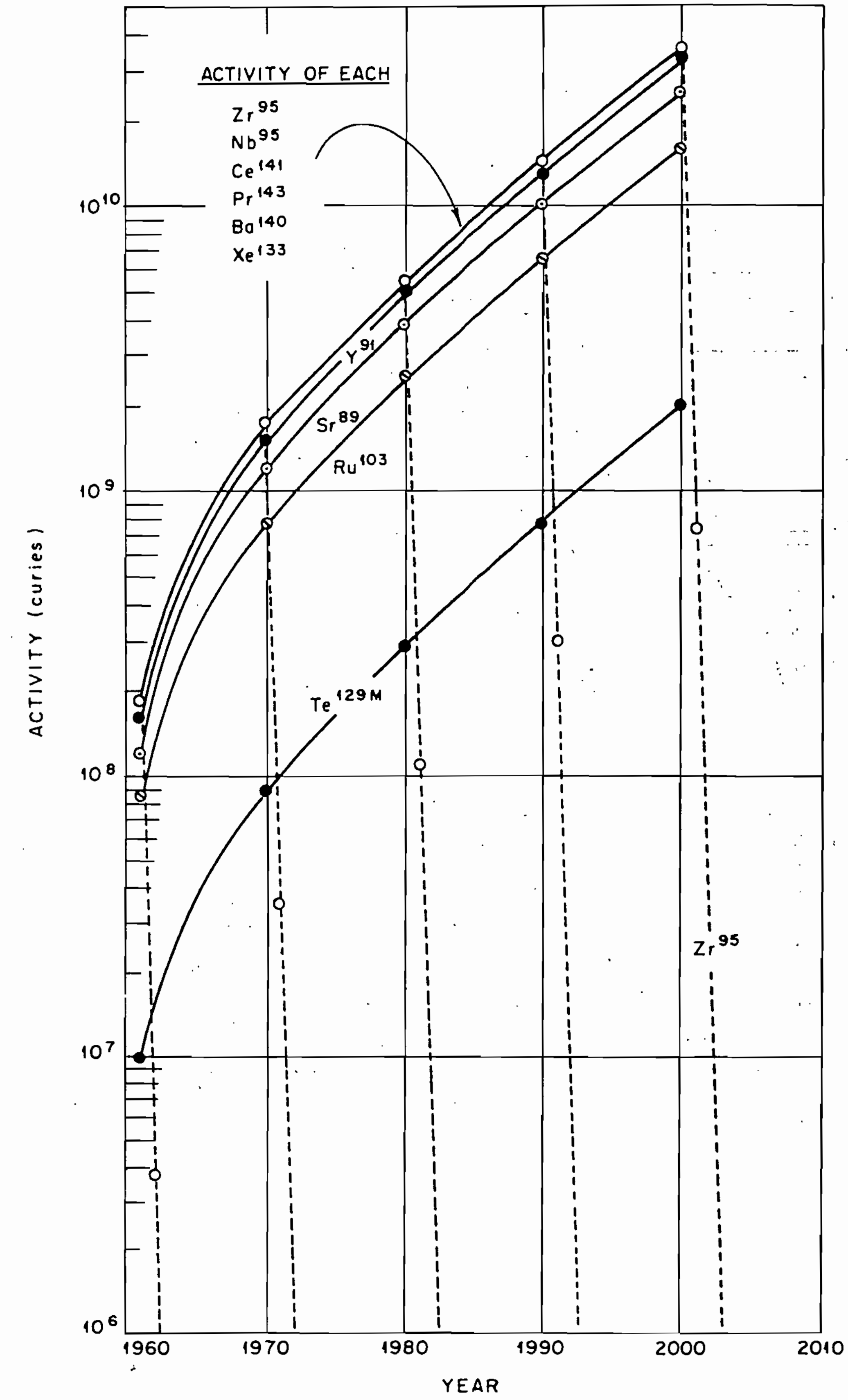


Fig. 10 ACCUMULATED ACTIVITY OF SPECIFIC FISSION PRODUCTS ($T_{1/2}$ <100 DAYS)

UNCLASSIFIED
ORNL-LR-DWG 7349A

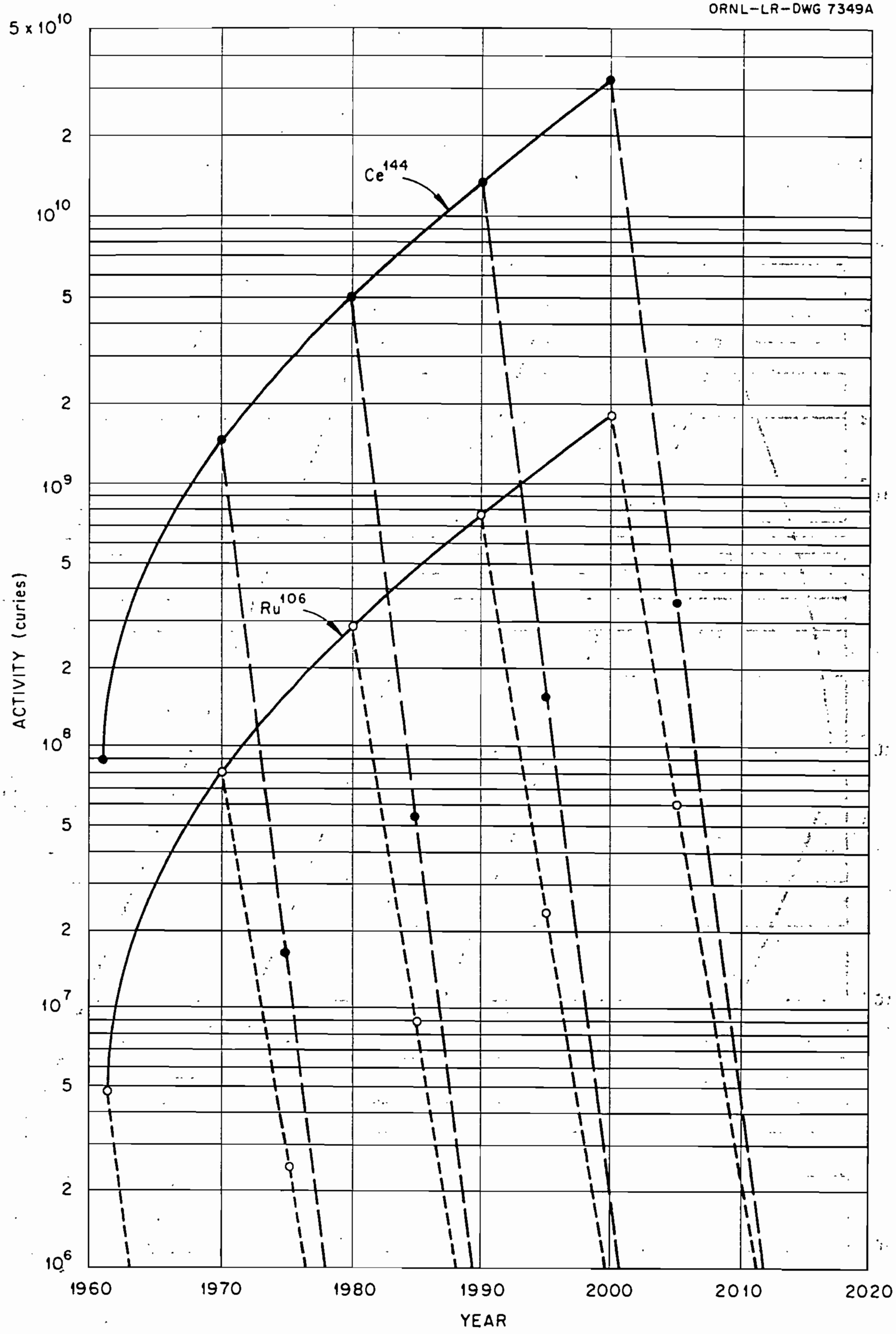


Fig. 11 Accumulated Activity of Ce^{144} and Ru^{106}

UNCLASSIFIED
ORNL-LR-DWG 7348A

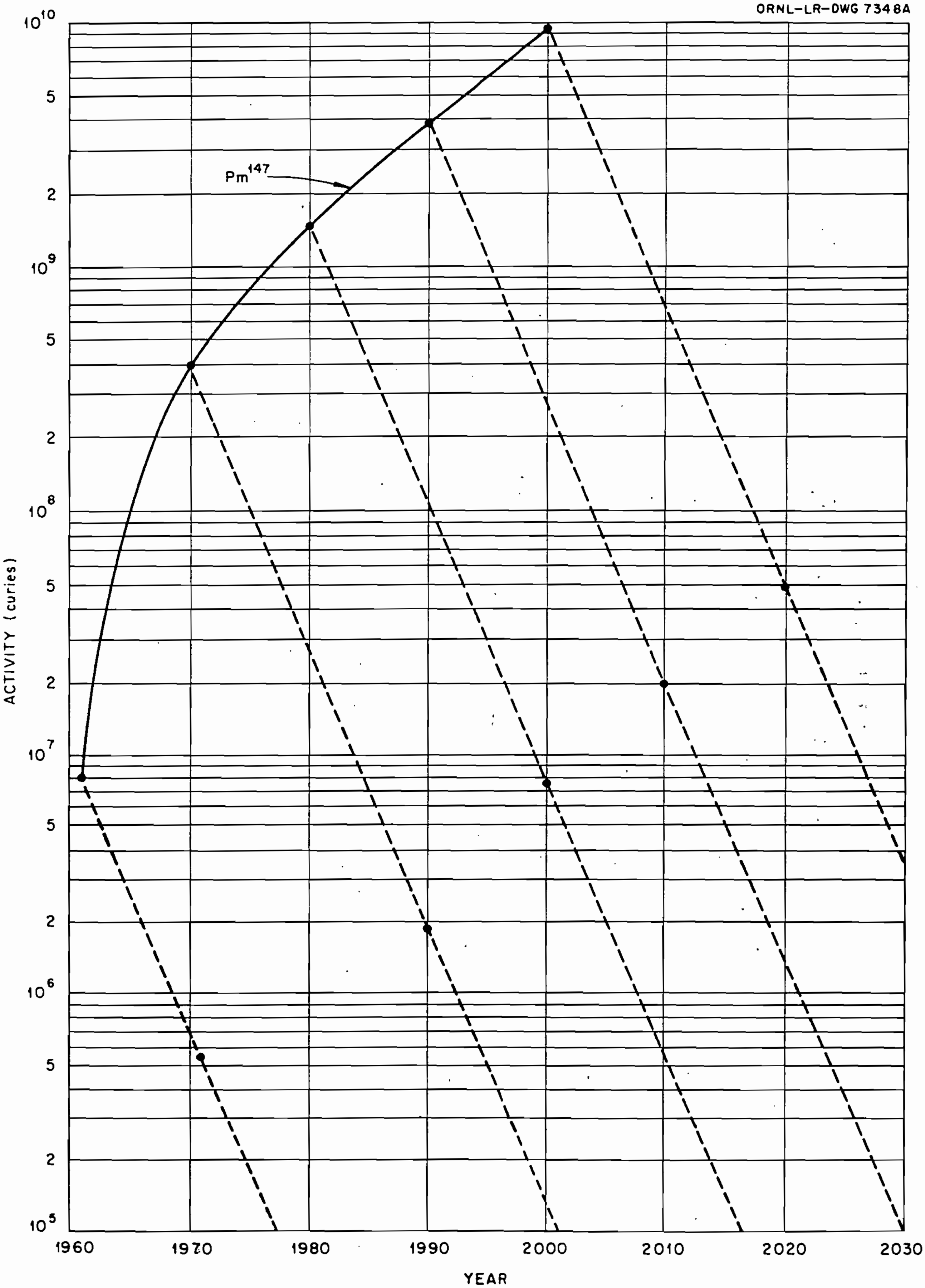


Fig. 12 Accumulated Activity of Pm^{147}.

UNCLASSIFIED
ORNL-LR-DWG 7350A

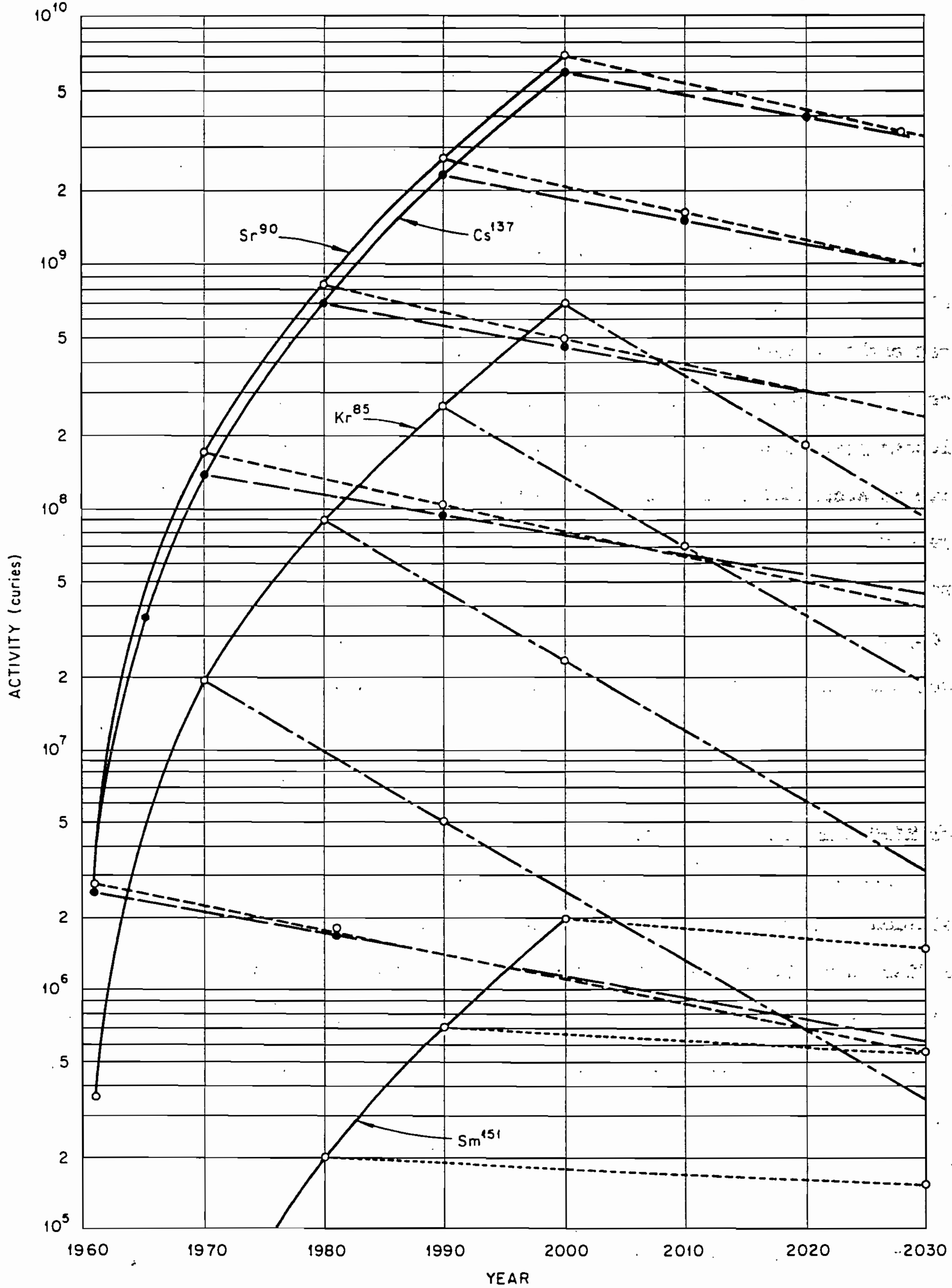


Fig. 13 Accumulated Activity of Kr^{85}, Sr^{90}, Cs^{137}, and Sm^{151}.

(6) Accumulated activities of specific nuclides (Figure 10 - 13)

Note: For explanation of dashed lines, see note for Figure 9 above.

Table 10 presents the growth of fission product activity in tabular form for each of the important fission products. Table 11 presents tabular data on gross activity decay after varying periods of accumulation.

The curves obviously show predicted buildups which result from a larger nuclear power economy than discussed in the first part of this section, where the first eight to twenty years of nuclear power growth in the United States are discussed. Although all buildup curves (Figures 5 through 13) are plotted with the calendar year as the abscissa, the results shown would be usable (although not quite as accurate) if predicted nuclear heat generation estimates obtained from Figure 4 were used. Actually, the true variable, and that which establishes the equilibrium value of fission product activity, is the total nuclear heat generation; or, assuming 25% thermal efficiency and full time operation as we have cone, the true variable is installed heat generation capacity, as shown in Figure 4.

Heat Generation in Nuclear Wastes

A portion of the energy released in nuclear fission takes the form of radiations from the fission products and ultimately appears as heat. For example, the fission products from a reactor operating for one year at a heat output of 1,000 KW after a period of 100 days would produce approximately 3,400 B.T.U. per hour. This is equivalent to an activity of about 140,000 curies and would drop to about 20 per cent of its 100 day decay value in one year. The heat produced by the wastes of a power reactor operating at 500 MW heat output, a reasonable power level from the economic standpoint, would amount to nearly 2×10^6 B.T.U. per hour after a decay period of 75 days.

[redacted]

TABLE 10

EFFECT OF PERIOD OF ACCUMULATION ON TOTAL ACTIVITY IN WASTE SYSTEM

Nuclide	Fission Product Activity (Megacuries) at Accumulation Period (Years)				
	1	10	20	30	40
Cs-137	2.6	140	710	2,300	6,000
Sr-90	2.8	170	810	2,600	6,900
Y-91	160	1,500	5,100	13,000	33,000
Sr-89	120	1,200	3,900	10,000	25,000
Zr-95	180	1,700	5,400	14,000	35,000
Nb-95	190	1,700	5,500	14,000	36,000
Ru-103	86	770	2,500	6,500	16,000
Ru-106	4.7	81	280	730	1,800
Te-129	10	89	290	750	2,000
I-131	89	730	2,300	6,300	16,000
Ba-140	200	1,700	5,400	15,000	35,000
Ce-141	190	1,600	5,100	14,000	34,000
Pr-143	190	1,600	5,300	14,000	35,000
Ce-144	88	1,400	4,700	13,000	31,000
Pm-147	7.8	390	1,500	4,000	10,000
Kr-85	0.37	19	90	270	700
Xe-133	210	1,800	5,600	15,000	37,000
Nd-147	79	690	2,200	5,900	15,000
Sm-151	0.00	0.3	2.0	7.0	20
Total	1810	17,200	56,700	152,000	376,000

TABLE 11

EFFECT OF DECAY TIME ON ACCUMULATED ACTIVITY

Period During Which Activity Is Accumulated in Waste System (Years)	Fission Product Activity (Megacuries) at Decay Time (Years)					
	0	3	5	10	20	30
1	1,810	15.57	8.32	5.07	3.59	3.72
10	17,200	593	412	284	204	158
20	56,700	2,523	1,879	1,351	998	763
30	152,000	7,470	5,690	4,250	3,170	2,430
40	376,000	19,370	15,080	11,330	8,410	6,440

J. O. Blomeke has calculated buildup and decay curves for thermal fission for all fission product chains under many conditions of flux.(20) From these data he has prepared a heat decay curve shown in Figure 14 for the aqueous wastes, assuming a natural or slightly enriched uranium reactor operated to 10,000 Mwd/ton uranium, and assuming 800 gallons of wastes per ton of uranium processed. In addition to gross fission product heat, the contributions of Cs^{137} and Sr^{90}, rare earths, and rare gases are given.

4. Production of Transuranics and Transplutonics and Their Concentration in Waste Streams

After irradiation in a reactor none of the fertile or fissionable materials exist as pure isotopes; they are always accompanied by isotopes produced by parasitic neutron capture. The production of and the fate of parasitically produced isotopes has a profound effect upon the activity levels to be encountered in the recycle of fuel and fertile material; any lowering of radioactive exposure limits must consider the effects upon what are now considered to be "cold" operations.

In addition, the heavy elements produced by neutron capture which appear in the wastes from a processing plant, will be among the most controlling hazardous activities in long term waste disposal because most are alpha emitters of high energy, short decay half-life, and many with long biological half-lives. Measurements of basic nuclear constants, production calculations, chemical separations development, and hazard evaluation for these materials is of great importance ot the evaluation of hazards from waste disposal, and may be of possible controlling significance in certain steps in the recycle of reactor fuel.

The buildup scheme for these isotopes both during reactor irradiation and decay may be represented by the reaction chart shown in Figure 15. (LR Dwg. 13205) Alpha decay is shown only for the important biological hazards.

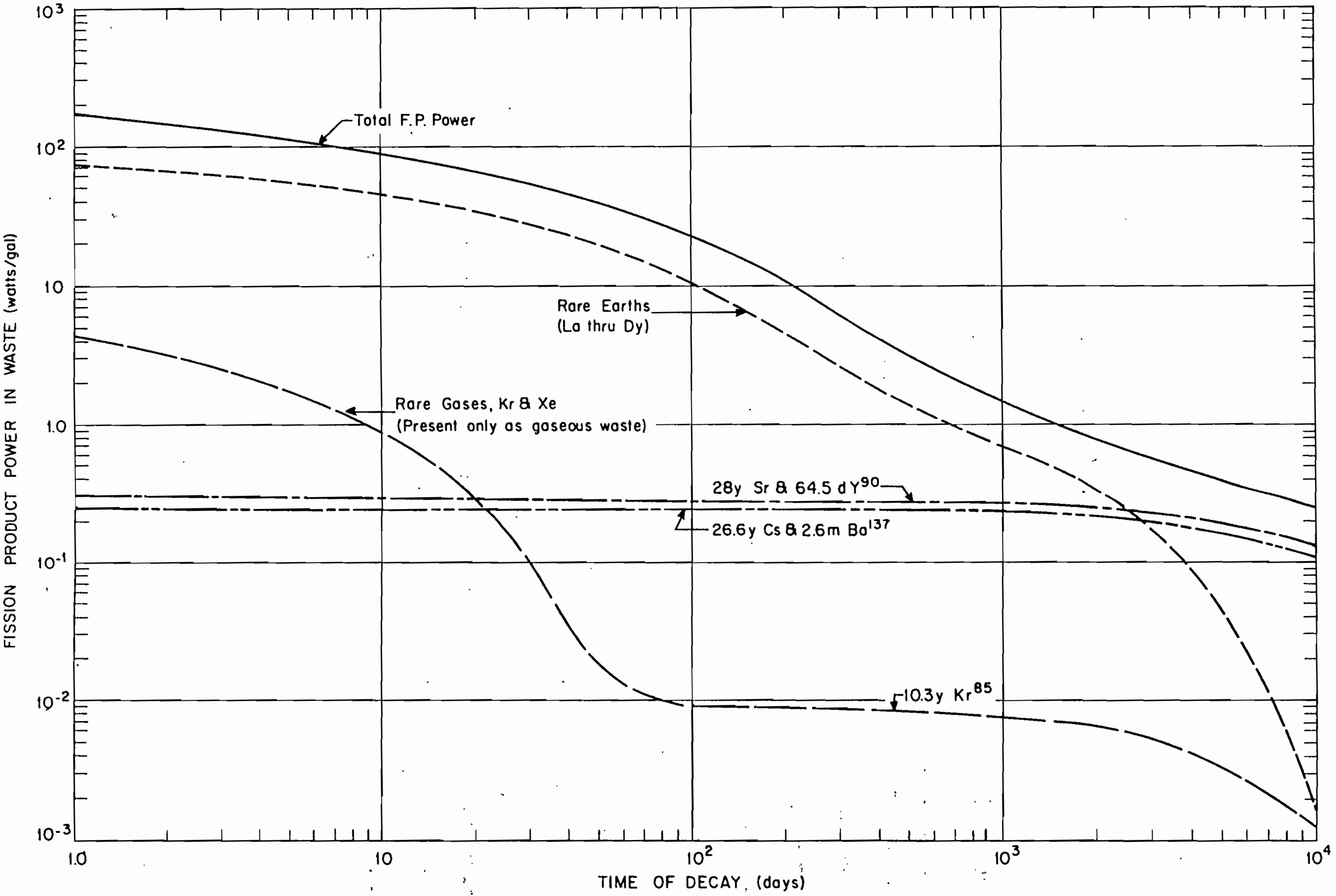
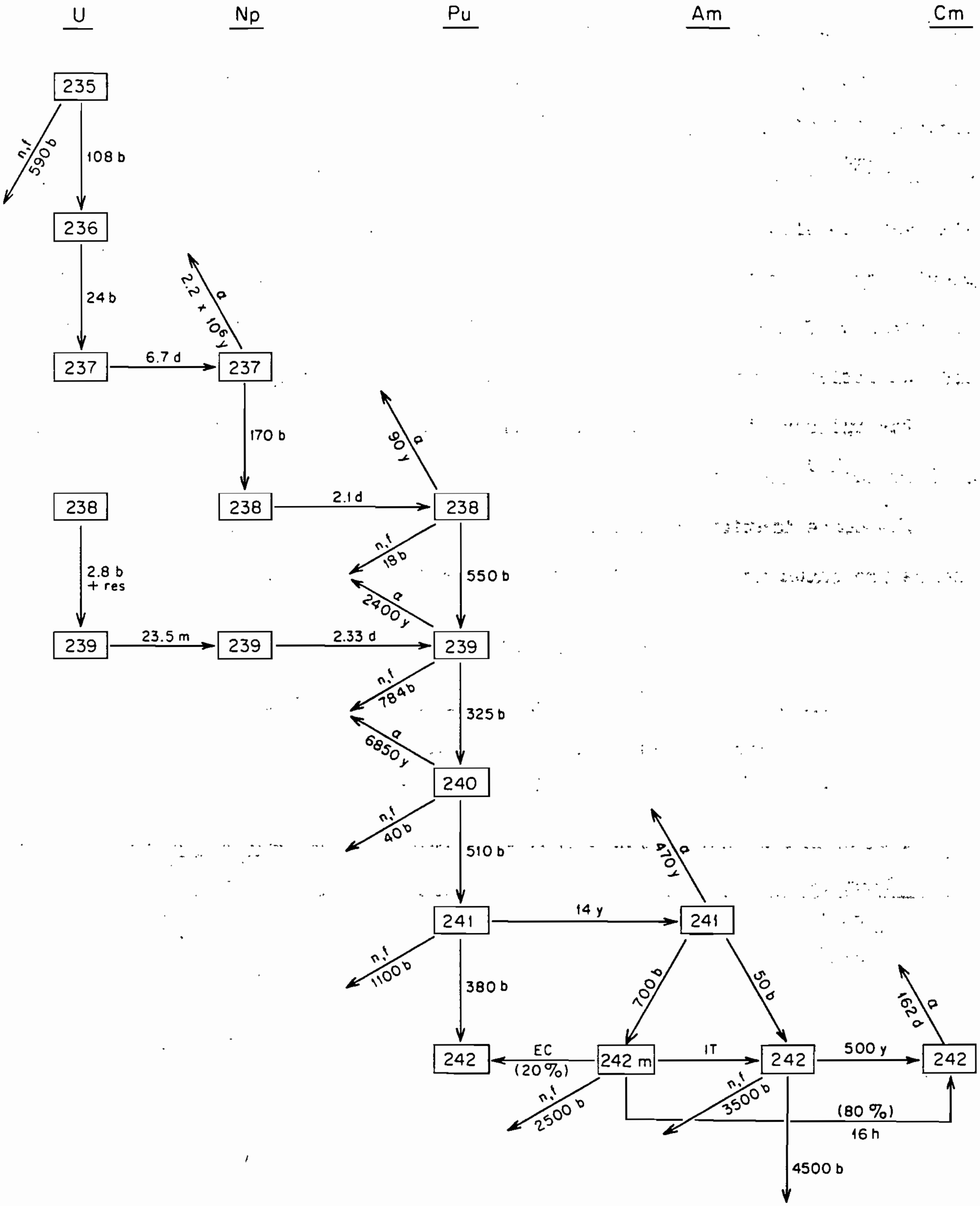


Figure 14. Fission Product Power in Waste as a Function of Decay Time. Basis: 800 gal waste per metric ton uranium, irradiated 10,000 Mwd/ton at 33 Mw/ton specific power.

ORNL-LR-Dwg.-19983
UNCLASSIFIED

UNCLASSIFIED
ORNL-LR-DWG 13205



Nuclear Reaction Chart.

FIGURE 15

The buildup of transuranics and transplutonics calculated by E. D. Arnold was based on 4000 Mwd/t irradiation of uranium having an initial enrichment of 1% U^{235}. Plutonium losses to the waste stream were calculated on the losses of 0.1% of total plutonium production, while Np^{237} losses to the waste stream were assumed at 100% of the production (700 g Np^{237}/ton U) based on infinite recycle and no U^{236} removed in a diffusion plant. The concentration of the heavy elements will vary considerably with changes in irradiation level, enrichment, diffusion plant recycle and flux, and of course with the chemical processes. With the exception of fluxes between $3 \times 10^{14} - 10^{15}$ n/cm^2/sec, the variation of fission product activity with reactor parameters is insignificant.

The buildup of transplutonics reported herein was obtained from the data of Blomeke(22) and Arnold.(21)

The concentrations of important heavy elements at time of reactor discharge are shown in Table 12 for thermal neutron flux of 10^{14} m/cm^2/sec.

TABLE 12

HEAVY ELEMENT CONCENTRATIONS AT REACTOR DISCHARGE

FOR 4000 Mwd/t 1% U^{235} INITIAL ENRICHMENT

$\phi = 10^{14}$ n/cm^2/sec

Isotope	Concentration g/t U
Np^{237}	700 (oo recycle of U^{236})
Np^{239}	163
Pu^{239}	2920
Pu^{238}	30 (oo recycle of U^{236})
Pu^{240}	339
Pu^{241}	69
Am^{241}	2.4×10^{-1}
Pu^{242}	6.3
Am^{242}	1.3×10^{-3}
Cm^{242}	3.1×10^{-2}

These values tabulated in Table 12 were multiplied by the total quantity of uranium processed during a thirty-year period and then corrected for decay during a thirty-year accumulation and growth period. The only significant activity affected by the decay correction during this thirty-year period is Cm^{242}. This effect can be illustrated by the following example:

(a) Cm^{242}/Pu^{239} activity ratio in discharged 4000 Mwd/t material = 558.

(b) Cm^{242}/Pu^{239} activity ratio after 30 years accumulation = 16.

After accumulation, the Cm^{242} activity decreases rapidly. Thus, it may be seen that Cm^{242} is a much more important biological hazard for short-term storage than it is for long-term storage.

The total number of curies of transuranic and transplutonic elements which could accumulate in high-level wastes by 1990 as a result of the above calculations are shown in Table 13. The effect of thirty years' decay following the thirty-year accumulation growth period is shown, also.

TABLE 13

ACCUMULATED TRANSURANICS AND TRANSPLUTONICS IN WASTE SYSTEM BY 1990

(30 YEARS ACCUMULATION TIME)

AND AFTER 30 YEARS DECAY
ASSUMING NO ADDITIONS TO THE 30 YEAR TOTAL VOLUME

Isotope	Activity in Waste by 1990 (curies)	Activity in Waste After 30 Years Decay (2020) (curies)
Np^{237} (oo recycle)	1.1×10^5	1.1×10^5
Pu^{238} (oo recycle)	3.3×10^5	2.6×10^5
Pu^{239} -Pu^{240}	1.2×10^5	1.2×10^5
Pu^{241*}	2.4×10^6	5.4×10^5
Am^{241}	4.8×10^5	5.1×10^5
Cm^{242}	1.42×10^6	9.7×10^3
Am^{242*}	1.2×10^4	9.7×10^3
TOTAL	8.0×10^{10}	2.4×10^9

* Not biologically important compared to Pu^{239} and Pu^{240} except for decay to their respective daughters which are biologically significant.

Pu^{238} formed by successive neutron captures from U^{235} could also influence the overall biological hazard of the wastes, especially from highly enriched fuel. For the case of 1% U^{235}, loss of 0.1% Pu^{238} to the waste would increase the total plutonium hazard about 5-10% for once through (pure U^{235} + U^{238}) material. Infinite recycle and no removal of U^{236} in a gaseous diffusion plant would increase the total plutonium hazard 3-5 fold. However, 25% removal of U^{236} per pass through the gaseous diffusion plant would only increase the hazard of plutonium 20-40 per cent.

Np^{237} is not a significant hazard even in the worst case of infinite recycle and no U^{236} removal in the diffusion plant.

Distribution of Fission Product and Heavy Element Activity Over the Nuclear Power Reactor Complex

From Figure 4 the prediction was made that by the year 2000, assuming 700,000 MW of nuclear heat generating capacity, approximately 4 x 10 curies of total fission product activity would exist in equilibrium. Arnold(23) calculated that the nuclear power complex and waste system will contain 5 x 10^6 curies of important transuranics (Pu^{239}, Pu^{240}, Am^{241}, Cm^{242}) by that year as shown in Table 13. Using these data we have calculated the distribution of the important radioactive elements among reactors, decay cooling systems, chemical processing plants and waste disposal or containment systems for the forty year accumulation period ending in the year 2000 A.D.

Although the buildup of fission product activity in an expanding nuclear economy depends only upon the rate of buildup of power with time (the reactor parameters are negligible), the distribution of radioactivity is strongly dependent upon the choice of reactor operating conditions and recycle assumptions. In this section we have calculated the activity distribution for one set of such conditions and assumptions to provide an approximate basis for estimating the relative hazard due to wastes to be assigned to each of the general divisions of the overall nuclear reactor complex. The assumptions that have been used in preparing this breakdown are as follows:

(1) The buildup of nuclear power will follow the curve estimated by J. A. Lane, with the nuclear heat power being 7×10^5 Mw in 2000 A.D.

(2) The average irradiation level is 4000 Mwd/ton of 1% enriched uranium.

(3) The average specific power of the reactors is 20 Mw/ton of uranium.

(4) Decay cooling period for discharge fuel elements is 200 days.

(5) Inventory in radiochemical processing plant is 20 days.

(6) Loss of 0.1% Pu + 100% of the transuranics and transplutonics to the high-level waste stream.

(7) Number of reactors = 1000 at 700 Mw of heat production capacity each.

(8) Number of processing plants = 20 at 7 tons/day each.

(9) Number of waste disposal or containment sites = 6 to accept 7×10^6 gals/yr each.

Based on these assumed conditions, the calculated activity distribution shown in Table 14 indicates that 80-90% of the total activity due to primary long-lived fission products (Cs^{137}, Sr^{90}, Kr^{85}, plus 95% of total Am^{241}) would exist in the waste systems (with the exception of Kr^{85}, which would probably be in the atmosphere). Almost 100% of the total short-lived activities (I^{131}, Ba^{140}, La^{140}) would exist in the reactors. Only 3-6% of the long-lived radioactive elements would exist in reactors, while the short-lived activities would be almost non-existent in the waste. Only 1.7% of total Pu^{239} + Pu^{240} would exist in the waste since only 0.1% is lost to the waste stream. The remaining fission products would distribute much more uniformly over the system as shown in Table 14. Table 15 provides an estimate of the total curies of each important isotope by 2000 A.D. in each part of the power reactor complex.

TABLE 14

PERCENTAGE OF TOTAL ACCUMULATED ACTIVITY BY YEAR 2000 A.D. IN VARIOUS PARTS OF THE REACTOR COMPLEX

Assumptions: 7×10^5 MW Heat Reactor Power, 4000 Mwd/ton, burnup, 1% enrichment, Lane curve
20 MW/ton specific power, 200 days decay prior to processing
20 days in processing plant

	Percent of Total Activity of Each Isotope at the Following Points			
Isotope	Avg. Activity In Reactor	Avg. Activity In Decay	Avg. Activity in Chem. Plant	Accumulated Activity in Waste
Cs^{137}	3.63	7.22	0.73	88.32
Sr^{90}	3.71	7.33	0.73	88.23
I^{131}	100.0^-	1.8×10^{-2}	--	--
Ba^{140}	100.0^-	0.43	--	--
Kr^{85}	5.14	9.95	0.97	83.94
Ce^{144}-Pr^{144}	26.78	37.75	2.82	32.65
$Pu^{239}+Pu^{240}$	46.89	46.88	4.70	1.73
Am^{241}	2.48	2.47	0.25	94.8
Cm^{242}	39.60	25.80	1.60	33.0
Pm^{147}	12.00	22.10	2.00	63.9

TABLE 15

EXTENT OF ACTIVITY IN VARIOUS PARTS OF THE REACTOR COMPLEX

Assumptions:	Year-2000 AD, 7 x 10^5 MW Heat Reactor Power, Lane growth curve 4000 Mwd/t average irradiation at 20 MW/ton specific power 200 days decay prior to processing, 20 days in processing plant

Isotope	Curies of Each Activity At The Following Points						
	Activity in Reactor		Activity in Decay		Avg. Activity Inventory Chemical Plant	Total Accumulated In Waste	Total Activity
	Avg.	ATD	Avg.	At End			
Cs^{137}	2.18 x 10^8	4.35 x 10^8	4.33 x 10^8	4.30 x 10^8	4.29 x 10^7	5.30	6.00 x 10^9
Sr^{90}	2.56 x 10^8	5.11 x 10^8	5.06 x 10^8	5.04 x 10^8	5.03 x 10^7	6.09 x 10^9	6.90 x 10^9
I^{131}	1.60 x 10^{10}	1.60 x 10^{10}	2.93 x 10^6	4.8 x 10^2	10	--	1.60 x 10^{10}
Ba^{140}	3.5 x 10^{10}	3.5 x 10^{10}	1.52 x 10^8	6.93 x 10^5	2.33 x 10^4	--	3.50 x 10^{10}
Kr^{85}	3.6 x 10^7	7.1 x 10^7	6.97 x 10^7	6.84 x 10^7	6.80 x 10^6	5.875 x 10^{8*}	7.00 x 10^8
Pm^{147}	1.14 x 10^9	2.26 x 10^9	2.10 x 10^9	1.95 x 10^9	1.92 x 10^8	6.07 x 10^9	9.5 x 10^9
Pu^{239} + Pu^{240}	3.8 x 10^6	7.6 x 10^6	7.6 x 10^6	7.6 x 10^6	7.6 x 10^5	2.8 x 10^{5**}	16.2 x 10^6
Am^{241}	--	2.90 x 10^4	2.90 x 10^4	2.90 x 10^4	2.90 x 10^3	1.1 x 10^6	11.16 x 10^6
Cm^{242***}	--	3.96 x 10^6	2.58 x 10^6	1.68 x 10^6	1.54 x 10^5	3.3 x 10^6	1.0 x 10^7
Ce^{144} - Pr^{144}	8.3 x 10^9	1.48 x 10^{10}	1.17 x 10^{10}	9.16 x 10^9	8.7 x 10^8	1.013 x 10^{10}	3.10 x 10^{10}

---

* Probably in atmosphere.

** Assumes 0.1% loss of Pu to first cycle aqueous wastes.

*** Cm-242 decays rapidly; its activity will decrease by factor of 100 in 5 years following accumulation period termination.

The distribution of radioactivity per unit in the total structure of the complex of an assumed 1000 reactors, 20 processing plants, 6 disposal sites, is shown in Table 16. This table indicated that the radioactivity in each 700 MW heat capacity reactor would be much less (with the exception of I^{131} and Ba^{140}) than the total radioactivity existing in each of 20 decay canals, or 20 processing plants, or the 6 waste systems. In all cases, except for the very short lived isotopes and plutonium, one waste system could contain as much as 4000 times as much radioactivity as exists in one 700 MW reactor, as is the case for Sr^{90}.

We are the first to recognize that it is possible to argue with the growth estimates and distribution calculations and to question the assumptions made concerning the number of units in parts of the reactor-recycle complex. However, it is certain that potential long-term radioactive hazard (to people other than those in the immediate vicinity of a reactor catastrophe) is far greater in, 1) decay cooling systems for fuel and wastes associated with large multi-purpose radiochemical reprocessing plants; 2) in the chemical plants themselves; and 3) particularly in any ultimate waste disposal site than in any single reactor.

For this reason we urge that overall hazard analyses, similar to those in progress for single reactor accidents, be made for the decay cooling, radiochemical plants, waste disposal site system is urgently required. Such an analysis has the possibility of drastically affecting the wisdom of manufacturing large quantities of fission products and heavy elements. The cost of insuring against single total catastrophe risks for chemical plants or for a waste disposal area may be far greater than the value of power produced.

TABLE 16

ACTIVITY LEVEL IN ANY ONE UNIT OF THE NUCLEAR POWER COMPLEX BY 2000

Assumptions:
1) One thousand - 700 MW (heat) reactors operated to 4000 Mwd/ton of 1% enriched uranium
2) 140 tons U/day processed in 20 - 7 ton/day chemical plants
3) 43×10^6 gals waste/year sent to 6 integrated waste disposal sites, each handling 7×10^6 gals/year

	Average Activity (Curies) in a Single Unit			
	In Each of 1000 Reactors (Average)*	In Each of 20 Chem. Plants		In Each of 6 Waste Sites
		In Decay** (Average)	In Process	
Cs^{137}	2.18×10^5	2.17×10^7	2.15×10^6	8.83×10^8
Sr^{90}	2.56×10^5	2.53×10^7	2.52×10^6	1.0×10^9
I^{131}	1.60×10^7	1.47×10^5	0.5	--
Ba^{140}	3.50×10^7	7.6×10^6	1.16×10^3	--
Kr^{85}	3.60×10^4	3.49×10^6	3.40×10^5	9.8×10^7
Pm^{147}	1.14×10^6	1.05×10^8	9.6×10^6	1.0×10^9
$Ce^{144}-Pr^{144}$	8.30×10^6	5.09×10^8	4.4×10^7	1.69×10^9
Am^{241}	29	1.45×10^3	1.45×10^2	1.84×10^5
Cm^{242}	3.96×10^3	1.29×10^5	7.7×10^3	5.5×10^5
$Pu^{239} + Pu^{240}$	3.8×10^3	3.8×10^5	(at 4000 Mwd/t) 3.8×10^4	4.66×10^4

---

* Assumed condition of normal distribution for irradiation level in reactors.

** Part of decay may take place at reactor or during transit.

5.0 Relative Biological Hazards of Fission Products and Heavy Elements in Accumulated Radioactive Wastes(24)

In this section we shall estimate the relative contribution to the overall radiation hazard to man of the various fission products and parasitically produced heavy elements that will accumulate as a result of a growing nuclear power economy. To understand the results of this study, it is necessary that the terms used be defined.

Relative Hazard - The relative hazard of any radioisotope as compared to another is directly proportional to the quantity of material present, inversely proportional to its biological tolerance as measured by the maximum permissible concentration in air or water; and finally inversely proportional to its half life. This relationship may be expressed mathematically by the following equation:

$$\text{Relative hazard} = \frac{N_i \lambda_i}{3.7 \times 10^{10} \frac{\text{d/sec}}{\text{curie}}} \Big/ \text{MPC} \qquad (1)$$

where N_i = the number of atoms of a specific radionuclide existing at any instant per gram of fissionable materials (as charged to a reactor)

λ_i = radioactive decay constant, sec^{-1}

MPC = maximum permissible concentration as given in references (7) (36) for water or air.

In the consideration of long-term accumulation and storage of radioactive wastes, this relative hazard can be used to define a more useful quantity, which we shall call potential hazard. Relative hazards, in conjunction with an estimate of the buildup rate of nuclear power and the counterbalancing natural decay of fission products and parasitically produced heavy elements, defines an integrated hazard which results from the total quantity of radionuclides produced in any time interval. Potential hazard is more carefully defined below.

Potential Hazard - The potential hazard due to radioactive elements in a given system is determined by the total accumulation of activity divided by the MPC. Thus, the value of the potential hazard is really the quantity of air or water (depending upon the basis used) necessary to dilute the total accumulation of each isotope to the accepted value of the maximum permissible concentration.

Thus

$$\text{Potential hazard} = \frac{A_i}{\text{MPC}}$$

where

$$A_i \text{ (curies)} = 8.012 \times 10^5 \lambda_i y_i \left\{ \frac{\alpha-\gamma}{\lambda_i} \left[1-e^{-\lambda_i t} \right] + \frac{\gamma}{\lambda_i + \beta} \left[e^{\beta t} - e^{-\lambda_i t} \right] \right\} \quad (2)$$

where α, β, γ are constants for an assumed equation used to estimate nuclear power buildup, such as that proposed by Lane(15). For our work we have used Lane's estimated buildup equation in which the constants are:

$\alpha = 2000$

$\beta = \ln 1.09$

$\gamma = 23{,}200$

A_i thus gives the total accumulated quantity of any radionuclide existing in the entire reactor complex of reactor, decay cooling systems, chemical plant, and waste systems. We have assumed full time operation of the installed nuclear power plants.

Absolute Hazard - The absolute hazard associated with a waste system cannot be defined by any mathematical relationship at this time since only rough estimates have been made and a particular waste disposal method or site has not been established. However, the absolute hazard will depend on how much of the potential hazard (accumulated activity) can be released, what is the probability of release, what is the mechanism of release, how many people will be exposed by what mechanism, and what effects will the direct radiation and residual contamination have on biological systems exposed.

The relative hazard of the accumulated waste products was based upon the dose delivered following a single exposure. We chose this approach because of the assumption that any catastrophe involving the waste system will contaminate the air and water of a given area for a short time only, and that if necessary, the population will be evacuated shortly after an accident in order to limit the ingestion or inhalation of radioactivity from the environment. We have assumed, without too much justification, that a comparison of hazards on the basis of long-time (assumed to be 70 years) continuous equilibrium exposure may not be justified at present, since it is doubtful that wastes per se will be added continuously to the environment. Wastes will probably not be added directly to streams or oceans until the most hazardous isotopes have been removed, if at all. Also, wastes that are stored by proposed surface or subterranean methods will still be carefully contained under normal conditions and presumably will not contaminate the natural water table. The principal hazard will therefore be that of accidental release of large quantities of activity, which is most difficult to evaluate.

The maximum permissible concentration for air contamination is that quantity of a radioisotope in μc/cc of air, which will, when this air is breathed at a rate of 2×10^7 cc/day (normal breathing rate) for one day (or at an equivalent rate-time relationship if taken for a period shorter than one day) give a 15.7 rem dose to the critical organ indicated over the following year. In a like manner, the maximum permissible concentration for water contamination is that quantity of a radioisotope in μc/cc of water, which will, when ingested at a rate of 2200 cc/day for one day or for an equivalent rate-time relationship give a 15.7 rem dose to the critical organ over the following year. A dose of 15.7 rem is the accepted allowable dose for one year as determined by (0.3 rem/wk) x 52 wks/yr. (However, this dose should not be used as the allowable dose for each year during the working years for people employed in atomic energy installations.)

The maximum permissible concentration of a radioisotope depends on such factors as the critical organ involved and its size; fraction of ingested radioisotope going to the organ in question, energy and relative biological effectiveness of the radiation emanating from the radioisotope, and effective half-life. If it is assumed that radionuclides are eliminated exponentially, the effective half-life T, is determined by the radioactive half-life, T_r biological half-life T_b, as follows: $T = T_r T_b/(T_b+T_r)$.

The maximum permissible concentration values listed in Table 17 are for a single exposure to an individual. These values do not reflect any genetic effects and at present there is no basis for translation from these values to allowable concentrations for the population as a whole in terms of genetic effects due to internal exposure. The Oak Ridge National Laboratory Health Physics Division is investigating by means of spectrographic analysis of tissue the distribution of trace elements in human tissue. Particular emphasis is being placed on the genetic organs and those organs in close physiological proximity to the genetic organs. The results of this investigation should permit a more precise evaluation of maximum permissible concentrations in air and water in terms of genetic damage. This investigation should be completed during 1957.

The relative biological hazard as used in this study is, in truth, the number of cubic meters of air or water necessary to dilute the total accumulation of activity to their corresponding maximum permissible concentration for a one-day intake which will then give a 15.7 rem dose during the year following exposure.

The following tables list data and results of our calculations:

Table 17 is a summary of the radioisotopes which constitute the important biological hazards in waste. This table lists the isotopes, the critical organ(s) involved and the maximum permissible concentration in both air and water for a 15.7 rem dose following a one-day intake.

Table 18 lists the total accumulated activity in the total nuclear economy waste systems by 1990, and the activity thirty years later assuming no new activity is added to this waste system.

Table 19 lists the relative biological hazard of each important isotope in the waste system. Figures 17 and 18 describe graphically the magnitude and decay of the important hazardous isotopes for both air and water contaminations.

The major biologically hazardous radioactive elements in the waste after a 30-year accumulation period and a 5-year decay following accumulation are shown to be arranged in the following order of decreasing hazard: Sr^{90}, Cs^{137}, Ce^{144}, -Pr^{144}, Pm^{147}, Am^{241}, Pu^{239} and Pu^{240} (+ Pu^{238} in some cases), Np^{237} and Cm^{242}.

Several assumptions other than those already were necessary before an evaluation of relative biological hazards could be determined. The hazards of the heavy elements were based on 4000 Mwd/t irradiation of uranium having an initial enrichment of 1% U^{235}. Plutonium losses to the waste stream were calculated on the basis of 0.1% of total plutonium production, while Np^{237} losses to the waste stream were assumed to be 100% of the production (700 g Np^{237}/ton U), based on infinite recycle and no U^{236} removal in a diffusion plant. The concentration of the heavy elements will vary considerably with changes in irradiation level, enrichment, diffusion plant recycle and flux. The variation in the concentration of the radioactive fission products will vary much less with these variables. In fact, with the exception of fluxes between 3×10^{14} - 10^{5} n/cm^{2}/sec, the variation of fission product activity with reactor parameters is insignificant.

TABLE 17

SUMMARY OF RADIOISOTOPES CONSTITUTING IMPORTANT BIOLOGICAL HAZARDS IN WASTE

Isotope	Critical Organ	Maximum Permissible Conc. in Air for 15.7 rem dose in 1 day (curies/cubic meter)	Maximum Permissible Conc. in Water for 15.7 rem dose in 1 day (curies/cubic meter)
Sr^{89}	Bone	2×10^{-6}	2×10^{-2}
Sr^{90}	Bone	2×10^{-7}	2×10^{-3}
Y^{91}	Lungs, Bone	4×10^{-6}	
	GI Tract		1×10^{-1}
Nb^{95}	Lungs	6×10^{-6}	
	Bone		4×10^{-1}
I^{131}	Thyroid	8×10^{-7}	6×10^{-3}
Cs^{137}	Lungs	10^{-6}	
	Muscle		6×10^{-1}
Ba^{140}-La^{140}	GI Tract	4×10^{-6}	6×10^{-2}
Ce^{144}-Pr^{144}	Lungs, Bone	8×10^{-7}	
	GI Tract		4×10^{-2}
Pm^{147}	Lungs	10^{-5}	
	GI Tract		8×10^{-1}
Np^{237}	Bone	2.9×10^{-8}	8.4×10^{-2}
Pu^{239}, Pu^{240}	Bone	6×10^{-9}	
	GI Tract		10^{-3}
Am^{241}	Lungs	10^{-8}	
	GI Tract		10^{-3}
Cm^{242}	Lungs	2×10^{-8}	
	GI Tract		9×10^{-4}

TABLE 18

ACCUMULATED ACTIVITY IN WASTE SYSTEM BY 1990
(30 YEARS ACCUMULATION TIME)
AND AFTER 30 YEARS DECAY

Isotope	Activity in Waste By 1990 (curies)	Activity in Waste After 30 Years Decay (2020) (curies)
Sr^{89}	1.0×10^{10}	
Sr^{90}	2.6×10^{9}	1.2×10^{9}
Y^{91}	1.3×10^{10}	
Nb^{95}	1.4×10^{10}	
I^{131}	6.3×10^{9}	
Cs^{137}	2.3×10^{9}	1.2×10^{9}
Ba^{140}-La^{140}	1.5×10^{10}	
Ce^{144}-Pr^{144}	1.3×10^{10}	
Pm^{147}	4.0×10^{9}	1.3×10^{6}
Np^{237}(oo recycle)	1.1×10^{5}	1.1×10^{5}
Pu^{238}(oo recycle)	3.3×10^{5}	2.6×10^{5}
Pu^{239}-Pu^{240}	1.2×10^{5}	1.2×10^{5}
Pu^{241}*	2.4×10^{6}	5.4×10^{5}
Am^{241}	4.8×10^{5}	5.1×10^{5}
Cm^{242}	1.43×10^{6}	9.7×10^{3}
Am^{242}*	1.2×10^{4}	9.7×10^{3}
TOTAL	8.0×10^{10}	2.4×10^{9}

---

*Not biologically important compared to Pu^{239} and Pu^{240} except for decay to their respective daughters.

TABLE 19

RELATIVE BIOLOGICAL HAZARD

	Hazard in Waste by 1990		Hazard in Waste After 30 years decay	
	In Air	In Water	In Air	In Water
Isotope	cubic meters x 10^{-15}	cubic meters x 10^{-11}	cubic meters x 10^{-15}	cubic meters x 10^{-11}
Sr^{89}	5	5		
Sr^{90}	13	13	6	6
Y^{91}	3.25	1.3		
Nb^{95}	2.34	0.35		
I^{131}	7.9	10.5		
Cs^{137}	2.3	3.83 x 10^{-2}	1.2	2.0 x 10^{-2}
Ba^{140}-La^{140}	3.75	2.5		
Ce^{144}-Pr^{144}	16.2	3.25		
Pm^{147}	0.4	5 x 10^{-2}	1.3 x 10^{-4}	1.6 x 10^{-5}
Np^{237*}	3.8 x 10^{-3}	1.3 x 10^{-5}	3.8 x 10^{-3}	1.3 x 10^{-5}
Pu^{238*}	5.5 x 10^{-2}	3.3 x 10^{-3}	4.3 x 10^{-2}	2.6 x 10^{-3}
Pu^{239}-Pu^{240}	2 x 10^{-2}	1.2 x 10^{-3}	2 x 10^{-2}	1.2 x 10^{-3}
Am^{241}	4.8 x 10^{-2}	4.8 x 10^{-3}	5.1 x 10^{-2}	5.1 x 10^{-3}
Cm^{242}	7.2 x 10^{-2}	1.6 x 10^{-2}	4.9 x 10^{-4}	1.07 x 10^{-4}

*Based on oo recycle of U^{236}

UNCLASSIFIED
ORNL-LR-DWG 13176

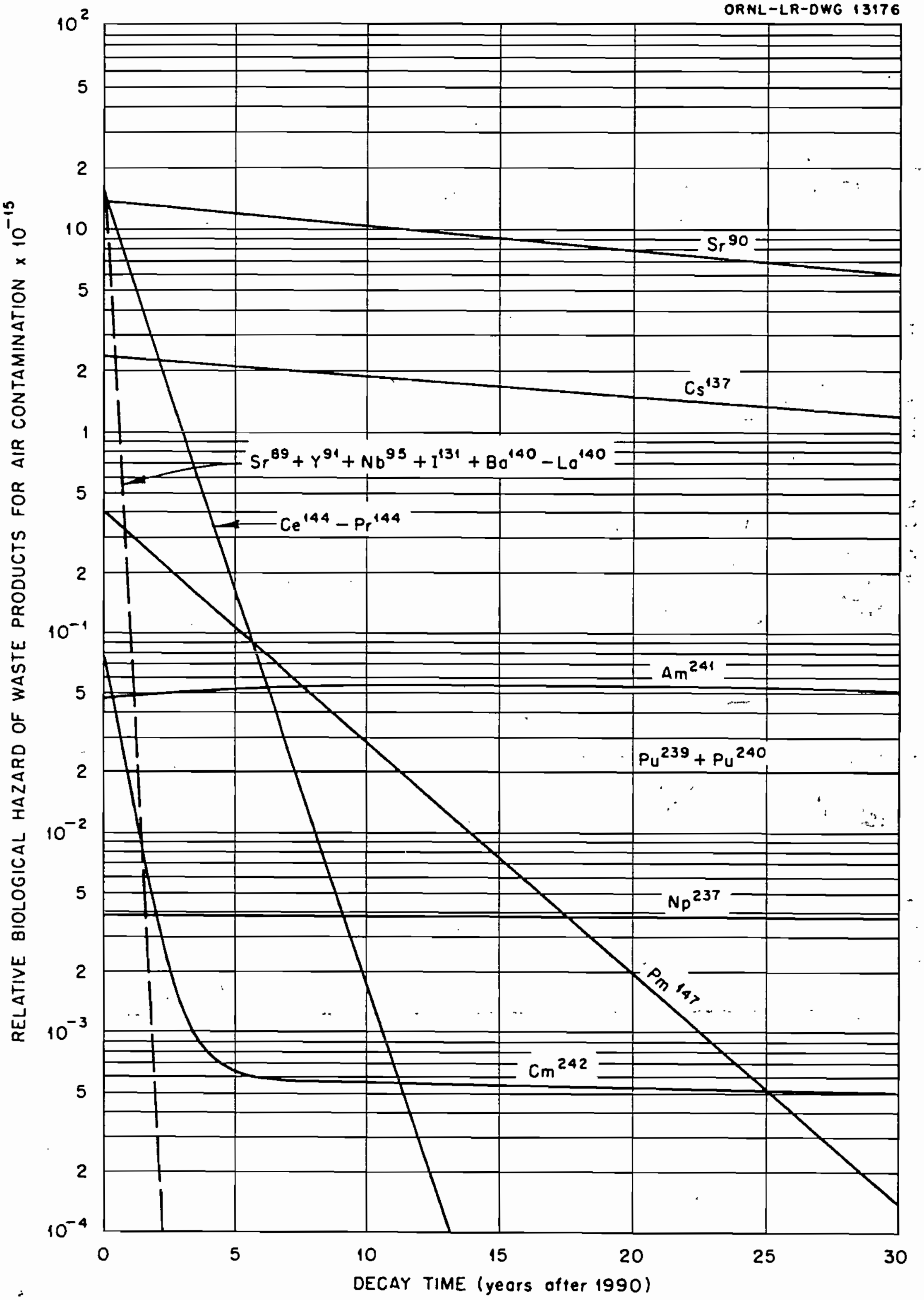


Fig. 17 Effect of Decay Time after 1990 on Biological Hazard of Accumulated Waste (1960–1990) Based on Air Contamination.

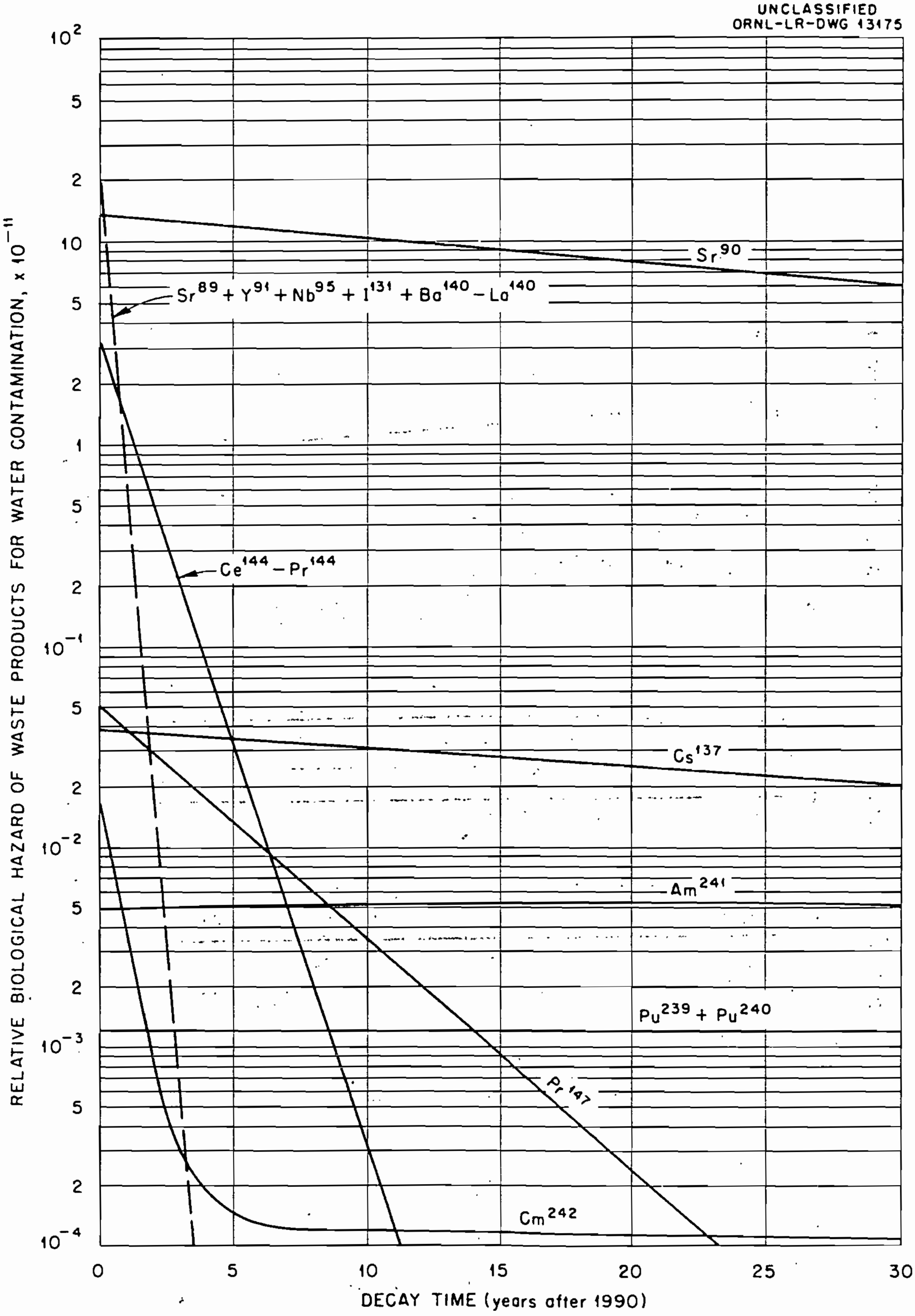


Fig. 18 Effect of Decay Time after 1990 on Biological Hazard of Accumulated Waste (1960–1990) Based on Water Contamination.

It is obvious that Sr^{90} and Cs^{137} present the most serious long-term waste hazard in both air and water. In air the Sr^{90} is about 300 times more hazardous than the third most hazardous isotope, Am^{241}. In water Sr^{90} is 3000 times more hazardous than Am^{241}. As has been suggested by all workers and commentators on waste disposal it is desirable under some circumstances to remove Cs and Sr from bulk waste streams, provided:

(a) That there is a safe way to permanently store Cs and Sr once they are removed.

(b) That satisfactory heat removal techniques can be developed to remove heat from the fission product concentrates.

(c) That the hazard of the bulk waste streams be sufficiently reduced to employ disposal or containment measures that require less control and are less expensive. This will be true if:

1) The decontamination factors for Cs and Sr removal are high enough ($DF = 10^4$, or greater) to allow release to certain chosen portions of our environment. W. A. Rodger(26) at Argonne has observed that the removal of strontium must be quantitative to materially affect the disposal picture for the bulk of fission product wastes. His calculations are reproduced in summary in the following Tables 20 and 21.

TABLE 20

50-YEAR ACCUMULATION OF LONG-LIVED ISOTOPES AND REQUIRED DISPERSAL VOLUMES

Basis: 2.2×10^6 MW Installed Reactor Capacity = 3 Tons Fission Products/Day

Isotope	Accumulated Quantity in 50 Years, curies	Maximum Permissible[c] Concentrations in		Volume Required to Dilute to Tolerance	
		Water uc/ml	Air uc/m	Water cubic miles	Air cubic miles
Zr^{95}	1.3×10^{11}	4×10^{-3}	4×10^{-7}	7.8×10^{3}	7.8×10^{7}
Ce^{144}	1.1×10^{11}	4×10^{-2}	7×10^{-9}	6.6×10^{3}	3.8×10^{9}
Ru^{106}	1.0×10^{11}	0.1	3×10^{-8}	2.4×10^{2}	8×10^{8}
Pm^{147}	5.1×10^{10}	1	2×10^{-7}	12	6×10^{7}
Sr^{90}	8.6×10^{10}	8×10^{-7}	2×10^{-10}	2.6×10^{7}	1×10^{11}
Cs^{137}	8.1×10^{10}	1.5×10^{-3}	2×10^{-7}	1.3×10^{4}	9.7×10^{7}
Tc^{99} [a]	2.0×10^{7}	3×10^{-2}	3×10^{-6}	0.2	1.6×10^{3}
Pu^{239} [b]	2.8×10^{6}	1.5×10^{-6}	2×10^{-12}	4.5×10^{2}	3.4×10^{8}

---

* Taken from IDO-14363 (Pt I), p. 328, by W. A. Rodger, Argonne National Laboratory.

a Decay Neglected

b Based on a Loss of 0.1% in Processing

c From National Bureau of Standards Handbook 5t (1953).

TABLE 21

EFFECT OF RESIDUAL Sr^{90} ON DISCARDABILITY OF WASTE

Per Cent Sr Remaining	Decontamination Factor	Years' Storage Required
0		13 years
0.00001	10^7	13 years
0.0001	10^6	90 years
0.001	10^5	180 years
0.01	10^4	270 years
0.1	10^3	360 years
1.0	10^2	450 years
10.0	10	540 years
100.0	1	630 years

---

* Taken from IDO-14363, (Pt I), p. 329, by W. A. Rodger, Argonne National Laboratory.

a To point where 1 cubic mile water will dilute activity to tolerance.

Rodger further observes that processes that achieve separation factors of the order of 10^6 to 10^7 such as solvent extraction have been developed for plutonium and uranium. He states that it is possible for cesium-strontium removal, but that the costs of such processing may be comparable to those incurred by the initial separation of uranium and plutonium from fission products. It is possible that income from sale or use of fission products could partially offset this separation cost.

2) That the nuclides which remain decay rapidly to levels safe for disposal. This condition may not be possible because of the presence in bulk wastes of transuranics and transplutonics, assuming that these trace elements report to the high-level wastes.

The most significant fact revealed by the plots of relative biological hazard is the marked significance of Am^{241}, Pu^{239}, Pu^{240}, Np^{237}, and Cm^{242}. We should note here that the production calculations for these heavy elements have just been made, and that a full evaluation of their significance has not been completed. It is also significant that the best recovery processes allow sufficient plutonium to report to the waste streams to control hazards in waste after Sr and Cs. There are no developed processes for removal of Am, Np or Cm.

Another factor that should be recognized in discussion pertaining to relative biological hazard is that the determination of the relative biological effectiveness, MPC values, and the critical organs for almost all radioisotopes are not well established, nor is sufficient research in progress to establish better understanding. Very little experimental data exist to define the effects of the simultaneous bodily retention of two or more radioisotopes. A definite cynergistic effect has been observed for radioisotopes that produce the same sytomatic result by affecting different organs in the body: i.e. blood cell production impairment by effects on bone marrow simultaneously with damage to the spleen. Dr. K. Z. Morgan currently estimates that this cynergistic effect, where it exists, may not change the MPC values by more than a factor of two.(27) We must expect this effect to be important in considerations involving potential exposures to mixed fission products and heavy elements.

SECTION 6.0 Hazard Potentials Due to Accidents

An accidental hazard occurs whenever radioactive materials are unintentionally released in dangerous amounts to the environment. The hazard is due to possible biological effects caused by irradiation of the human body by neutrons, alpha, or beta particles, and by gamma rays emitted by the radioactive materials as they undergo various nuclear or decay processes.

Potentially, accidents involving release of radioactive materials may occur whenever such materials are present. Accidents may occur with release of hazardous amounts of radioactive substances from nuclear reactors, plants manufacturing fuel elements, plants decontaminating and recovering unburned fertile and fissionable materials removed from nuclear reactors, handling and transportation of all types of radioactive materials, manufacture and use of all types of radioisotopes, and waste storage areas. Thus, accidental release of radioactive materials may occur wherever natural radioactive substances, such as radium, or materials irradiated in a nuclear reactor or accelerator are utilized.

The accidental release of long-lived hazardous isotopes such as Sr^{90}, Cs^{137}, Am^{241}, and Cm^{242} while possibly confined to limited areas (and to limited numbers of people) at the time of the accident, can become generally distributed over the period of the hundreds of years during which their hazard persists. The probability of population exposure to such a distributed activity varies markedly with the system in which activity is contained, i.e. whether it is in the reactor, a chemical plant, or a waste disposal system.

We will not discuss at length the aspects of accidents involving radioactive materials, but it is very evident that the accident potential of all parts of the reactor complex must be better defined. This rather obviously cannot be done by collecting statistics on accident frequency, since the effects of one major accident are long lasting and so serious.

For example, a flash flood which flushed 1 megacurie of mixed fission product activity from a waste storage tank could easily deposit this over a square mile of land. One megacurie of 2 year old fission product wastes per square (assume average of 0.7 Mev gamma energy) would give a dose rate of between 4.2 r/hr and 10.6 r/hr, depending upon the roughness of the ground. People in this flood area would necessarily be required to evacuate in great haste if dose rates were this high, since an exposure of several hours would equal the allowable lifetime dose. It is probable that the results of such a deposition of radioactivity over an inhabited area would prevent land use for many years, and that effective cleanup would be very costly.

6.1 Major Release from a Reactor Accident

Accidental release of activity has been considered primarily from the standpoint of reactor accidents. The magnitude of an accident which occurs with a large nuclear reactor may be catastrophic due to the possible release and dissipation of hundreds of pounds of both fission products and uranium-233 or plutonium. Aside from what reactor technologists say or think can be done to insure higher probabilities of normal reactor safety per se, one hazard study group points out that an abnormal "major disaster" may occur despite all human efforts - and that the probability for same cannot be proven as <u>zero</u>. One might "guess" that its probability may range from a lower limit of perhaps 10^{-4}/reactor-yr. (sabotage) to an upper limit of 10^{-2}/reactor-yr (present statistics). Thus the "major release" potential must exist for a reactor economy.(31)

The few reactor accidents which have occurred have involved reactors of relatively low power generation capacity. In Appendix III we have extracted reports of the following reactor excursions:(30)

1. Borax Destructive Experiment(32)

2. Canadian NRX Reactor Incident in 1952(33)

3. Experimental Breeder Reactor Incident(34)

Since problems of reactor safety and reactor site selection are covered rather extensively in the declassified literature,(37)(38)(39)(40) we will not discuss this subject further.

6.2 Qualitative Description of Hazards in Ore and Feed Materials Processing of Virgin Fissionable and Fertile Material

6.21 Natural uranium - the hazards associated with radioactivity from the processing of natural uranium to all the classes or population will be slight since the activity associated with the decay chain of U^{238} and U^{235} is not great. The activity exposure to plant operating personnel will be easily controlled by a minimum of protection from ingestion and inhalation of particulates. In the case of a serious accident, such as an explosion, in a natural uranium processing plant the exposure to the surrounding and general populace will be slight and the area of contamination limited to the plant site itself. Transportation hazards with natural uranium are negligible, assuming normal handling precautions. Wastes will contain products in the decay chains of U^{238}, U^{235} and U^{234}.

6.22 Natural uranium - the decay chain of thorium contains β and γ emitters that will increase the hazard of handling natural thorium above that for natural uranium. If very low exposure limits are established, some problems of control of exposure to plant operating personnel will arise, although limited exposures can be obtained by proper handling and operation. Wastes will contain small quantities of the decay chains of Th^{232}.
In the event of an accident in a thorium processing plant, exposure of the surrounding populace would be slight, and to the general populace, negligible. The transportation of natural thorium can be accomplished with normal packaging control.

6.3 Potential Sources of Hazard from Handling of Fissionable and Fertile Materials After Irradiation and Decontamination from Fission Products

6.31 Natural, depleted and slightly enriched uranium - Handling uranium after irradiation in a reactor is made more hazardous by the production of U^{237}. The U^{237} activity depends upon irradiation history and decay cooling period, assuming complete decontamination from fission products, plutonium and other heavy elements. For 10,000 Mwd/ton irradiation and infinite recycle, approximately 180 days decay cooling are required to allow U^{237} to decay to the background activity of natural uranium. For short decay cooling periods U^{237} activity will make limited thickness shielding necessary for operations subsequent to the fission product separation step. Transportation of uranium containing appreciable concentrations of U^{237} will require shielding. An accident in a plant processing uranium containing U^{237} will greatly increase the immediate exposure potential to operating personnel and to the populace in the immediate vicinity of the plant. No long-term radiation hazard greater than that for natural uranium or slightly enriched uranium will result, since U^{237} decays with a 6.8 day half-life.

6.32 Thorium - The recycle of irradiated thorium after decontamination from fission products is complicated by the presence of Th^{234} and Th^{228}. Th^{234} concentration is directly dependent upon the irradiation period; it decays with a 24.1 d half-life with the emission of beta and gamma energy. A decay period of about 300 days is required for the decay of Th^{234}.

Th^{228} presents a much more serious problem. It is the decay daughter of U^{232}, produced by an (n, 2n) reaction on Th^{232}. The decay chain of Th^{228} contains very high energy α , β and γ. In addition, the activity of thorium containing Th^{228} (and all irradiated thorium will contain a quantity determined by the fast neutron flux-time product of the irradiation) will increase for ten years after solvent extraction separation. The quantity of Th^{228}, which follows the separated thorium product, can be reduced by processing reactor blankets with very short cooling (about 30 days), but the thorium product is then very radioactive because of the presence of undecayed Th^{234}. It appears that the thorium recycle will always provide radiation exposure potential to operating personnel. The wastes from thorium recycle plants may contain certain elements in decay chain of U^{232}, which will undoubtedly dictate their controlled storage and disposal. The hazard of an accident in a thorium recycle plant to operating personnel and to the population in the surrounding area is significantly greater than for natural uranium or unirradiated thorium. A relatively long-term exposure hazard will result from the dispersion of recycle thorium. Transportation of thorium at any time after exposure in a reactor may require some shielding.

6.33 Enriched U^{235} - The activity of parasitically produced U^{237} will govern the cooling period for this class of material, but this period in no case will exceed 180 to 200 days to allow decay to background. In some reactor cycles the inventory cooling charges may be of such magnitudes as to require shorter decay cooling, in which case, the uranium cycle subsequent to solvent extraction will be slightly to moderately active. The cycle will offer no long-term population hazard, affecting only operating plant personnel, shipment, and the immediate environs of the processing plant. Lower radiation tolerance levels can increase the cost of uranium recycle.

6.34 U^{233} - The decay of parasitically produced U^{232} makes the hazard involved in U^{233} recycle greater than for any other source or fissionable material except possibly plutonium. After separation by solvent extraction from fission products and thorium, U^{233} product is at its natural background activity, but after separation it increases in activity level, reaching maximum activity level after approximately ten years. From processing to reactor fuel it is possible that no marked problem will exist if final fuel element or material preparation is accomplished within several months after separation. U^{232} decay daughters can be removed at any time by reprocessing through solvent extraction. Considerable care and control of U^{233} will be required to keep U^{233} recycle from being a hot operation, which requires heavy gamma shields. Wastes from steps subsequent to solvent extraction could contain the decay daughters of U^{232}, and possibly would require a limited decay period. The lowering of radiation tolerance levels will have a serious economic effect on certain U^{233} cycles. Accidents from a U^{233}-thorium plant can have a serious long-term effect on plant personnel and the population in the immediate vicinity of the plant. Transportation of U^{233} must be considered from the long-term hazard viewpoint.

6.35 Plutonium - Processing and subsequent recycle of plutonium presents the most serious plant and general population hazard from the fissionable and fertile material recycle. In concentrated form, plutonium must be carefully handled in absolutely sealed systems. Irradiation of Pu^{239} fuel elements will produce higher isotopes of Pu that increase the hazard. All operations in which plutonium is handled must be carefully controlled; wastes must be contained and plutonium isolated. Plutonium presents a serious long-term hazard to plant operating personnel, population in the immediate vicinity of the processing plant, and to the general populace.

6.4 Hazards in the Chemical Reprocessing Plant

Hazards due to radiation and chemical toxicity in a chemical processing plant under conditions of normal operation can be controlled. General radiation levels for radioactive fuel processing equipment can be reduced with proper shields. Ingestion and inhalation of radioactivity can be controlled.

6.41 Hazardous Operations - the most hazardous operations from a radiation exposure standpoint are:

a. Sampling of radioactive solution and solution transfer to analytical laboratories
b. Analytical chemical analyses of radioactive solutions
c. Product removal operations conducted on a warm basis, particularly for such materials as thorium, U^{233} and plutonium
d. Maintenance of equipment
e. Exposure to accidentally released fission product gases or radioactive particulates
f. Handling of radioactive feeds to the chemical plant
g. Storage and disposal of liquid wastes

These obviously are hazards to plant operating personnel. The population surrounding a chemical plant can be exposed to hazard from uncontrolled discharge as waste gases containing fission product gases and particulates. Similarly, the controlled or accidental discharge of liquid wastes to ground water can expose a relatively large number of people.

6.42 Accidents - Accidents in a chemical processing plant can be of very serious consequence to operating personnel and possibly to people living in the environs of the chemical plant. The type of accidents that can occur in a plant are:

a. Leaks due to equipment failure and corrosion

b. Chemical explosions from such materials as organic solvents; hydrogen evolved from acid dissolution; steam explosions; oxidation of such materials as molten metals; fluorine or interhalogen reactions; uncontrolled dissolution of metals.

c. Criticality. Since this subject is not too well covered in the open literature, a separate discussion of criticality control has been included in Appendix III, prepared by J. W. Ullmann from declassified data.

(Any explosion in the chemical plant inself probably will be contained within the plant and, at most, in a limited area surrounding the plant. Any single accident will involve only several pieces of process equipment from which relatively limited quantities of activity could be discharged.)

d. Enemy action in time of war. Results of bombing of a radiochemical plant with its necessary waste tanks could provide a very serious hazard to a large population group in an area surrounding the chemical plant. The hazard would be of long duration.

e. Natural catastrophies, such as earthquakes, windstorms, and particularly floods.

The radiochemical processing plant and its associated waste storage facilities can be considered as an accumulator of fission products and heavy element transmutation products. Because of economic considerations, it is probable that one chemical processing station will serve many power reactors. A study of processing costs versus processing plant capacity, based on extrapolation of our existing process technology, indicates that a central chemical processing plant may be of economic necessity large enough to process the fuel from reactors producing about 30,000 megawatts of heat.(15) Such a chemical processing plant will have a constant "inventory" of approximately 10^8 to 10^9 curies of fission product activity.

7.0 Transportation of Active Wastes

7.1 Requirements for the Shipment of Fuel and Waste(1)

A nuclear power economy will require the shipment of large quantities of radioactive material, first as irradiated fuel from reactors, then possibly as waste. In 1980, using Lane's build-up data, and assuming that stationary power reactors will average approximately 1000 Mw heat production capacity each, about 700 reactors will be in operation. Fuel from these 700 reactors may be shipped after an estimated 100 days decay cooling, to 20 large chemical plants. After chemical reprocessing, the wastes can economically be stored for five to ten years before shipment to an ultimate disposal site.

In order to provide a rough approximation of what the transportation of fission products and fuel element will do to "spreading the hazard", J. W. Ullmann has prepared the analysis shown in the following table.

TABLE 1

ANALYSIS OF SHIPMENT OF RADIOACTIVE FUEL AND WASTE

Fuel Shipped after 100 days Cooling
Wastes Shipped after 2000 days Cooling

Build-up and Production Data	In 1980	In 2000
Megawatts of heat from stationary reactors (Lane)	1.1×10^5	7×10^5
Specific power assumed, megawatts/ton U	20	20
% of reactor power as fission products		
after 100 days cooling	0.13	0.13
after 2000 days cooling	0.02	0.02
Watts of fission product heat per ton U		
after 100 days cooling	26,000	26,000
after 2000 days cooling	4,000	4,000

(Table continued)

	In 1980	In 2000
Fission products, curies per watt after 100 or 2000 days cooling	200	200
Fission products, curies per ton U		
after 100 days cooling	5.2×10^6	5.2×10^6
after 2000 days cooling	8.0×10^5	8.0×10^5
Megawatt days per ton, burnup	4,000	4,000
Tons U per day processed	27	175
Gallons liquid waste per ton of U processed	1,200	1,200
Shipments of Fuel		
Tons of U in transit	189	1,225
Fission products, curies in transit	9.8×10^8	6.4×10^9
Tons of fuel per carrier (assumed)	2	2
Shipping time elapsed (days)	7	7
Number of carriers in transit	95	613
Fission products per carrier, curies	1.0×10^7	1.0×10^7
Watts heat per carrier (Cooling required)	5×10^4	5×10^4
Shipments of Waste		
Gallons of waste in transit	227,000	1,470,000
Fission products, curies in transit	1.5×10^8	9.8×10^8
Fission products, curies per gallon of waste	660	660
Gallons of waste per carrier (assumed)	450	450
Fission products per carrier, curies	3.0×10^5	3.0×10^5
Number of waste carriers in transit	505	3,270
Watts heat per carrier (Cooling probably not required)	1.5×10^3	1.5×10^3
Thickness of lead shielding	4 inches	4 inches
Probable carrier weight	12,000 lbs.	12,000 lbs.

The significance of this estimate is that is points out a very important problem, that of the distributed hazard resulting from the required movement of irradiated fuel and radioactive waste to chemical plants and disposal sites. The fact that 10^8 to 10^9 curies of radioactive fission products (plus many millions of curies of the alpha emitters, Pu^{239}, Pu^{238}, Pu^{241}, Am^{242}, Cm^{242} and others) are in motion as fuel elements and waste at any instant presents a distributed hazard that has not yet been evaluated. Without the benefit of experimental data, we may find that it is necessary to decide whether the shipment of large quantities of radioactivity will be allowed at all; and certainly we must early establish the controls under which shipments of radioactive materials can occur; and to provide emergency regulations to be used when activity is accidently released by accident in transit.

Shipment of radioactive materials on a large scale may be a necessary part of a nuclear power economy that is competitive with fossil fuel.(2) The uneconomical alternate to large central chemical processing plants with capacity to process fuel shipped from many remote reactors is a multiplicity of small chemical plants, each located with a single or small group of reactors. The dispersal of radiochemical plants in turn spreads the hazard due to wastes to possibly the same or greater extent than the distributed hazard presented by transported fuel and wastes. Such a coupling of small chemical plants with a few reactors may also make recycle costs too great for the production of competitive power in the United States.

Wastes can be shipped from the chemical plant to disposal sites as either solids or liquids, with the latter form probably being the most hazardous and the most expensive, assuming that solid wastes can be shipped without external cooling. They can and probably should be shipped in small packages to limit the total quantity of activity that could be released in case of accident. Packaged and shielded shipments can be made (in increasing order of cost) by water, truck, rail and air.

Arnold(3) has pointed out both the economic attractiveness of a protected pipeline for radioactive wastes and the possibility that the pipeline may actually be the cheapest and safest method of waste movement. The total volume of wastes shipped in 1980 can be pumped through a 2" diameter pipeline.

A double concentric pipe in a concrete trough surrounded by earthen shielding with an ion exchange capacity, and of course with appropriate monitoring, pumping automatic block systems and cooling systems, might be as safe as other waste transport methods.

7.2 Regulations Applicable to Shipments of Radioactive Materials

The following information was taken from a report prepared by A. L. Ayers, Phillips Petroleum Company:(4)

A very useful source of federal regulations is the "Handbook of Federal Regulations Applying to the Transportation of Radioactive Materials" obtainable from the United States Atomic Energy Commission, Division of Construction and Supply, Traffic Management Section, Washington, D. C. Transportation of radioactive materials in interstate commerce by land or water is subjected to regulations of the Interstate Commerce Commission. The regulations applicable to radioactive materials are not issued separately but are included in the complete regulations covering the packaging, labeling, and transportation of dangerous articles published as Title 49, Part 71 to 78 of the Code of Federal Regulations. Between revisions, annual pocket supplements are issued. Ammendments subsequent to the period covered by the most recent revision or annual supplement may be obtained from the daily issues of the Federal Register. All of these are for sale by the Superintendent of Documents, U. S. Government Printing Office, Washington 25, D. C.

The ICC Regulations are published also by the Bureau of Explosives of the Association of American Railroads, H. A. Campbell, Agent, 30 Vesey Street, New York 7, New York, as "Tariff No. 9, Publishing Interstate Commerce Commission Regulations for Transportation of Explosives and Other Dangerous Articles", and by the Tariff Bureau of the American Trucking Association, Inc., F. G. Freund, Agent, 1424 16th Street, N. W., Washington 6, D. C., as "Motor Carriers Explosives and Dangerous Articles Tariff No. 7".

Transportation of radioactive materials by water is subject to regulations prescribed by the Commandant of the United States Coast Guard.

Current regulations applicable to radioactive materials appear in Title 46, Part 146, of the Code of Federal Regulations as ammended. The United States Coast Guard Regulations covering transportation, storage or stowage of dangerous articles on ships are also published by the Bureau of Explosives, H. A. Campbell, Agent, 30 Vesey Street, New York 7, N. Y., as "Water Carrier Tariff No. 6".

Transportation of radioactive materials in interstate commerce moving by rail, water, or public highway is regulated by the Interstate Commerce Commission. Some states extend the ICC regulations to include intrastate shipments while in others specific and sometimes more restrictive regulations apply to shipments within the state. Additional regulations upon this transfer of radioactive materials may be made by local authorities as in the case of movement through tunnels, port areas, etc.

The interstate commerce regulations covering transportation of explosives and other dangerous articles include a part of Title 49 of the Code of Federal Regulations as follows:

PART 71 -- General Information and Regulations
PART 72 -- Commodity List of Explosives and Other Dangerous Articles
PART 73 -- Regulations Applying to Shippers
PART 74 -- Regulations Applying Particularly to Carriers by Rail Freight
PART 75 -- Regulations Applying to Carriers by Rail Express
PART 76 -- Regulations Applying to Rail Carriers in Baggage Service
PART 77 -- Regulations Applying to Shipments Made by Way of Common, Contract or Private Carriers by Public Highway
PART 78 -- Shipping Container Specifications

Regulations in Parts 71 to 78 cover preparation of explosives and other dangerous articles for transportation common carriers by rail freight, rail express, rail baggage, highway or water, construction of containers, packing, weight, marking, labeling when required, billing, and shipper's certificate of compliance with these regulations; also cars, loading, storage, billing placarding, and movement thereof by carriers by rail.

The regulations define that anyone knowingly not conforming to these regulations is subjected to fine or imprisonment, or both.

The Commission has been given power by Congress to formulate regulations for the safe transportation of these materials. The Commission or any other interested party may initiate requests for changes in regulations. The Bureau of the Safe Transportation of Explosives and other Dangerous Articles may at the request of the Commission accumulate data from all available sources to determine the most effective and logical regulations.

Generally, a notice of ninety days is given before a new or modified regulation becomes effective. However, a shorter time may be authorized if special and peculiar circumstances justify it. Periodic public hearings are held in which evidence may be produced concerning proposed changes. The Commission may act without hearing and without notice, although usually twenty days notice of proposed changes or additions are given.

The regulations show the kind of label when required on shipment of explosives and other dangerous articles and also lists those which are prohibited for transportation. A list of the materials to which these regulations apply are also given. Items are listed in alphabetical order and for each item there is given the proper shipping name, the class of hazard, cross references to sections specifying exemptions and packing, color or label required, if not exempt, and maximum quantity in one outside container for shipment by rail express. All radioactive materials are classed as poison, Class D, and are properly shipped as "radioactive materials". A blue or red label is required as specified.

It is the duty of the shipper to follow the regulations. All the radioactive materials, that is Class D hazards, which have also another hazardous characteristic are subject to this specific regulation for both hazards. As an example, radioactive sodium would be classed as A flammable solvent as well as a radioactive material. Shipments of radioactive materials made by the Atomic Energy Commission under escort are exempt from these regulations. However, the regulations with the AEC specify the ICC regulations as a standard of safety for transportation of radioactive materials without exception. Escort may be provided for safety reasons as well as security.

The consignees must report promptly to the Bureau of Explosives all instances of improper staying and broken, leaking, or defective containers. The Bureau of Explosives, upon receipt of such reports from consignees, should promptly report to the shipper full particulars covering all such cases.

The empty containers previously used for the shipment of radioactive material must have all openings including removable heads, filling and vest holes, tightly closed before being offered for transportation. Small quantities of the material may be allowed to remain in empty containers and when the vapors remaining are unstable it is permissible to add sufficient inert gas to render the vapors stable. Empty containers must be leak free. The empty containers must not contain more than 1/10 millicurie of radium, polonium, or more than 0.135 millicuries of strontium-89 or strontium-90 or barium-140, or more than 1.35 millicuries of any other radioactive substance. There must be no significant alpha, beta or neutron radiation emitted from the exterior of the package. Gamma radiation at the surface of the package must be less than 10 milliroentgens per 24 hours. When shipments of radioactive materials are loaded on the cars by shippers or unloaded from cars by the consignee, the cars must be placarded and unloaded according to regulations. A radioactive material is any material or combination of materials that spontaneously emit ionizing radiation.

Radioactive materials, Class D, poison, are divided into three groups according to type of rays emitted at any time during transportation, as follows:

GROUP I - Radioactive materials that emit gamma rays only or both gamma and electrically charged corpuscular rays.

GROUP II - Radioactive materials that emit neutrons and either or both types of radiation characteristic of Group I materials.

GROUP III - Radioactive materials that emit electrically charged corpuscular rays only, that is alpha, beta, etc., or any other that is so shielded that the gamma radiation at the surface of the package does not exceed 10 milliroentgens for 24 hours at any time during transportation.

As far as the shipments of which we are discussing, spent fuel will always fall in Group I, and recovered fissionable materials may fall in either Group I or III, depending upon the effectiveness of the processing in removing gamma emitters.

The purpose of classifying radioactive material by group is to facilitate the statement of regulations covering labeling and handling. Group I or II materials require special precautions in transit and in storage to protect personnel and photographic film. The stipulation "at any time during transportation" is made necessary due to the possibility of an increase in gamma radiation during transit due to the formation of gamma emitting daughter products of the material being shipped.

Not more than 2000 millicuries of radium polonium, or other members of the radium family of elements and no more than 2700 millicuries of any other radioactive substance may be packed in one outside container for shipment by rail freight, rail express or highway, except by special arrangements and under conditions approved by the Bureau of Explosives.

Radioactive materials such as ores, residues, etc. of low activity packed in strong tight containers are exempt from specification packaging and labeling requirements for shipment in carload lots by rail freight only provided the gamma radiation or equivalent will not exceed 10 milliroentgens per hour at a distance of 12 feet from any surface of the car and that the gamma radiation or equivalent will not exceed 10 milliroentgens per hour at a distance of 5 feet from either end surface of the car.

The term "low activity material" is not defined by the ICC. However, it is implied that any gamma emitting material, a full carload of which does not produce radiation in excess of 10 mr/hr at a distance of 12 feet from any surface of the car, may be considered low activity material for this purpose. The limitation on the radiation from either end of the car at 10 mr/hr at 5 feet may be achieved either by spacing or by the use of shielding.

Although no exact provision is given, it would appear that ICC approval for shipment of tank cars of low enrichment recovered uranium solutions may be possible on the basis presented here for full car shipments.

In the event of breadage of container, wreck, fire or unusual delay involving radioactive shipments as covered by the regualtions under discussion, the car containing loose radioactive materials must be isolated as far as possible from danger of human contact and no persons must be allowed to remain close to the car or contents until qualified persons are available to supervise handling. The shipper and Bureau of Explosives should be notified immediately.

Cars, building, areas, or equipment in which Class D poisons have been spilled must not be again placed in service or occupied until decontaminated by qualified persons.

Any box car or motor vehicle which, after use for the transportation of radioactive material in carload or truckload lots, is contaminated by radioactive material shall be thoroughly cleaned to the extent that a survey of the interior surface shows that beta-gamma radiation is not greater than 10 milliroentgens physical equivalent in 24 hours or that average alpha contamination is not greater than 500 disintegrations per minute per 100 square centimeters. A certificate to that effect must be furnished to the local agent of the carrier or to the driver of the motor vehicle. Cars and vehicles which are used solely for the transportation of radioactive materials are exempt from the provisions of this section.

Containers of radioactive material must not be placed in vehicles, terminals, and other places closer than 3 feet to an area which may be continuously occupied by passengers, employees or shipments of animals. Materials of undeveloped film must not be placed closer than 15 feet to this type of shipment. No more than 40 units of radioactive material shall be transported in any vehicles or stored at any location at any one time. One unit of radiation equals one milliroentgen per hour at one meter for hard gamma radiation or any amount of radiation that has the same effect on film as 1 mrhm or hard gamma rays of radium filtered by 1/2 inch of lead.

Permissible Radiation Levels

The carrier must be designed so that there will be no significant radioactive surface contamination on any part of the container. All outside shipping containers must be of such design that the gamma radiation will not exceed 200 milliroentgens per hour or equivalent at any one point or readily accessible surface. Containers must be equipped with handles and protective devices when necessary in order to satisfy this requirement. The outside of the shipping container must be at least equivalent to a heavy wooden box or a fiber-board box. However, in the case of the shipments under consideration, only metal containers are efficient. Radioactive materials of Group I, liquid powder, or gaseous, must be packed in suitably packed containers, completely surrounded by a shield of lead or other suitable material of such thickness that at any time during transportation the gamma radiation at 1 meter from any point on the radioactive source will not exceed 10 milliroentgen per hour. This shield must be so designed that it will not open or break under conditions incident to transportation. The minimum shielding must be sufficient to prevent the escape of any primary corpuscular radiation to the exterior of the outside shipping container. In this regulation, at 1 meter from any point on the radioactive source is interpreted to mean at 1 meter from the nearest point on the source.

Radioactive materials Group III, liquid, solid, or gaseous, must be packed in suitable inside containers completely wrapped and/or shielded with such material as will prevent the escape of primary corpuscular radiation to the experior of the shipping containers, and secondary radiation at the surface container must not exceed 10 milliroentgens per 24 hours, at any time during transportation. Acceptable instruments for measuring gamma radiation packages include the gamma survey meters listed in the SIC series of the AEC Instruments Catalogue. There must be no loose radioactive material in the car, and the shipment must be braced so as to prevent leakage or shift of lading under conditions normally incident to transportation. Except when handling is supervised by the Atomic Energy Commission, shipments must be loaded by the consignor and unloaded by the consignee.

The regulations covering transport by water are in most cases identical with those prescribed by ICC. The vessel must be loaded so that a gamma radiation or equivalent at any spare point in any space or area continuously occupied by passengers, crew, or shipment of animals will not exceed 40 milliroentgens per 24 hours at any time during transportation.

ICC approval must be obtained for shipping containers not specified by regulations. This is obtained by certification through the Bureau of Explosives.

7.3 Optimum Cooling Period for Wastes Before Shipment

Zeitlin, Ullmann and Arnold(5) have published a study of storage plus transportation costs in which they determined optimum cooling periods for solvent extraction first-cycle raffinates prior to shipment as liquids to a remote ultimate disposal site such as might be provided by a deep well or salt bed. Their study was aimed at determining the optimum cooling time for wastes at the chemical plant site assuming the worst conditions of: 1) shipping liquid wastes (800 gallons/ton of U) in small 250 gallon capacity shielded carriers; 2) initial radiation burnup to 10,000 Mwd/T of fuel; 3) several storage costs as shown in Table 3; and 4) varying decay cooling times and shipping distances.

After calculating the shielding required and determining overall carrier rate using rail freight costs in the United States were: 1) $1.40/hundred lbs round trip for one way distance of 200 miles; 2) $2.60/hundred lbs round trip for 500 miles one way and $4.50/hundred lbs round trip for one way distance of 1000 miles, they determined optimum cooling periods as shown in Table 2.

TABLE 2

UNIT STORAGE COSTS ($/gal/yr)

$0.25/gal Initial Investment for Tanks

Rate for Fixed Charge	15%	12%	4%
Fixed Charge	0.038	0.030	0.010
Direct Charge	0.003	0.003	0.003
Total	0.041	0.033	0.013

$2.00/gal Initial Investment for Tanks

Rate for Fixed Charge	15%	12%	4%
Fixed Charge	0.30	0.24	0.08
Direct Charge	0.003	0.003	0.003
Total	0.30	0.24	0.08

Assumptions:

(1) Lifetime of underground storage tank of 50 years
(2) Purex-type waste
(3) Tank farm operating personnel of 2 men/shift

Table (continued)

(4) Fixed charges were calculated for initial investments of $0.25 and $2.00 per gallon at three different annual rates. 12% and 15% per year represent the range used by utility companies to write off investments inclusive profit, taxes, and interest on capital. 4% per year might be the rate for a government-owned burial site (2% amortization plus 2% average interest).

(5) The cost of land $\frac{\$5000/\text{acre}}{1{,}000{,}000\ \text{gal/acre}} = \$0.005/\text{gal}$ was neglected compared to the initial cost of tankage

(6) Direct operating costs (based on a 20,000,000 gallon form which has reached steady state) will be:

$$\frac{2\ \text{men/shift} \times 4\ \text{shifts} \times \$4500/\text{man-year}}{20{,}000{,}000\ \text{gal}} = \$0.0018/\text{gal/year}$$

Allowing for 67% overhead, the direct charge will be $0.003/gal/year

TABLE 3

OPTIMUM WASTE COOLING TIMES AT CHEMICAL PLANT SITE

Storage Cost $/gal/yr	0.013	0.041	0.30
$ Gallon Tank Cost	$0.25	$0.25	$2.00
Tank Amortization Rate	4%/year	15% Private	15% Private
Optimum Cooling Time (years) for One Way Shipping Distance of:			
200 mi	9	6-1/2	0
500 mi	11	7-1/2	5-1/2
1000 mi	14	9	6-1/2

From this we might conclude that the chemical plant may always utilize waste storage tanks to economic advantage to allow for the decay of activity prior to possible shipment to an ultimate disposal site. The same cooling period would be advantageous in reducing shielding requirements and costs for waste processing operations leading to recovery of useful fission products or to fixation of gross activity in non-leachable soluable form.

7.4 Possible Costs vs. Shipping Distances

In order to check the economic feasibility of shipping wastes for less than a 0.05 to 0.1 mill/kwh of electricity, we have estimated shipping charges as shown in Table 4, which also includes the costs of storage of wastes for optimum cooling periods discussed in Section 7.3. Shipment of liquid wastes in a 250 gallon lead shielded carrier was assumed.

We venture to draw several general conclusions from this study:

1) 500 mile shipping costs alone for wastes, cooled seven years or longer are of an order of magnitude less than the allowable costs for radiochemical separations to meet the requirements of 8 mills/kwh electricity; i. e. allowable radiochemical costs may be 0.5 mill/kwh vs. approximately 0.025 mill/kwh for shipping.
2) Combined costs of shipping plus interim decay storage prior to shipment can be kept below 0.05 mill/kwh of electricity if tank investment costs are lower than possibly $0.60/gallon.
3) Waste tanks at the radiochemical separations plant for decay cooling of wastes are economically justified. This brings about a corollary advantage of permitting the accumulation of wastes in tanks during the first few years (possibly as long as ten) of operation of a radiochemical separations unit without significant economic disadvantage.

7.5 Experience with Radioactive Waste Shipment

At sites where large quantities of radioactive wastes are produced,

Table 4

ESTIMATED COST OF INTERIM STORAGE AND SHIPPING OF HIGH LEVEL WASTES

Assumptions:
1) 500 mile shipping distance, rail freight no escort, round trip base rate $2.60/cwt.
2) Fuel irradiate to 4000 Mwd/T uranium.
3) Reactor operates at 25% thermal efficiency.
4) Storage costs taken from Table 3
5) 800 gallons waste per ton of uranium processed.

Years Cooling Time	Interim Storage Costs $/gal or mills/kwh				Shipping Costs		Total Costs	
	$0.041/gal/year		$0.30/gal/year				0.041/gal/yr	0.30/gal/yr
	$/gal	mills/kwh	$/gal	mills/kwh	$/gal	mills/kwh	mills/kwh	mills/kwh
0.5	0.02	<0.001	0.15	0.005	2.73	0.091	0.091	0.096
5	0.20	0.007	1.50	0.050	1.19	0.039	0.046	0.089
7	0.29	0.010	2.10	0.070	0.79	0.026	0.036	0.096
10	0.41	0.013	3.00	0.10	0.71	0.024	0.037	0.124
20	0.82	0.027	6.00	0.20	0.66	0.022	0.049	0.222

Conversion factor; Multiply $/gal by 3.3×10^{-2} to get mills per kwh of electricity.

such as the Idaho Chemical Processing Plant, Savannah River and Hanford, liquid waste raffinates from solvent extraction are piped to the waste storage tanks using stainless steel pipe in a protective pipe and encased in a concrete trough. Monitoring systems for leak detection are used, along with cathodic protection (in areas where ground eddy currents require it) and provisions for maintenance. The experience with piping wastes has been uniformly good. At Oak Ridge National Laboratory, where some high level liquid waste lines have been in service for 15 years, lines are buried directly in the ground without protection. Very few leaks have occurred, and where they have appeared it has been possible to repair and replace piping with maintenance procedures only slightly different from normal practice. Earth surrounding the leak has been removed by using a drag bucket on the end of a long crane boom. For limited distances pipe lines have been uniformly satisfactory, but the area through which lines have been run has always been within the confines of the processing site. The problem of piping wastes for great distances across a right of way of limited area has not yet been attempted.

Liquid wastes of high activity have been shipped in sealed, shielded carriers from the Idaho Chemical Processing Plant to the Multicurie Separations Pilot Plant at Oak Ridge.(6) The carrier for this purpose was designed and built as a prototype of tanks to be used to transport aqueous solutions of radioactive fission products. The tank has been used in test runs between Arco, Idaho and Oak Ridge National Laboratory to check the performance of design features.

The spherical shape of the container was chosen because of the optimum ratio of volume to weight obtained in this shape. The total weight of the empty tank with shipping skid is 28,200 lb. The working volume is 210 gal of liquid weighing 2,000 lb. The total capacity of the tank is 250 gal.

The inner tank, containing the liquid, is a 48-in.-diameter sphere, made by welding together two hemispheres pressed from 3/8-in. thick type 347 stainless steel plate. Surrounding the inner tank is a layer of lead shielding 5-1/2 in. thick. A sphere of 13/16-in.-thick steel plate, clad with stainless steel on the outside, encloses the lead-shielded inner container to provide protection against external forces or internal pressures.

Two pipe connections to the inner tank provide for filling, emptying and venting the tank (Fig. 1). The tank is filled by first pumping a vacuum on the tank, closing off the vacuum connection on the short leg, and allowing the tank to fill through the long leg without additional pumping. This eliminates the danger of overflow.

A Teflon-lined plugcock is provided in each of the two pipes. Quick-opening couplings on the ends of the pipes are of the valved type, so that they automatically seal against pressure from within the tank when the couplings are disconnected. The entire external piping assembly is enclosed in a cylindrical cupola (Figs. 2 and 3), which is shielded with 2 in. of lead and which is closed by a shielded cover seating on a corrugated stainless steel gasket. The inner tank is thus sealed against leakage to the outside by two seals.

The liquid level in the tank is measured by conductivity probes which indicate volumes of 125, 200, and 210 gal.

The total of fabrication of the shielded transfer tank was $22,726 of which $12,791 was for material, $6,624 for labor, and $3,311 for overhead. An additional $2,500 was expended for engineering.

Two shipments of IAW waste from the Idaho Chemical Processing Plant have been made. This is the aqueous raffinate from the first column of the solvent extraction process for the recovery of U^{235} from exhausted MTR fuel. This waste contained 2.76 curies of Cs^{137}, and a total of 30.3 gross beta curies, per gallon. The radiation reading on the outer surface of the container was 6.2 mr/hr.

ORNL-LR-Dwg. -21098
UNCLASSIFIED

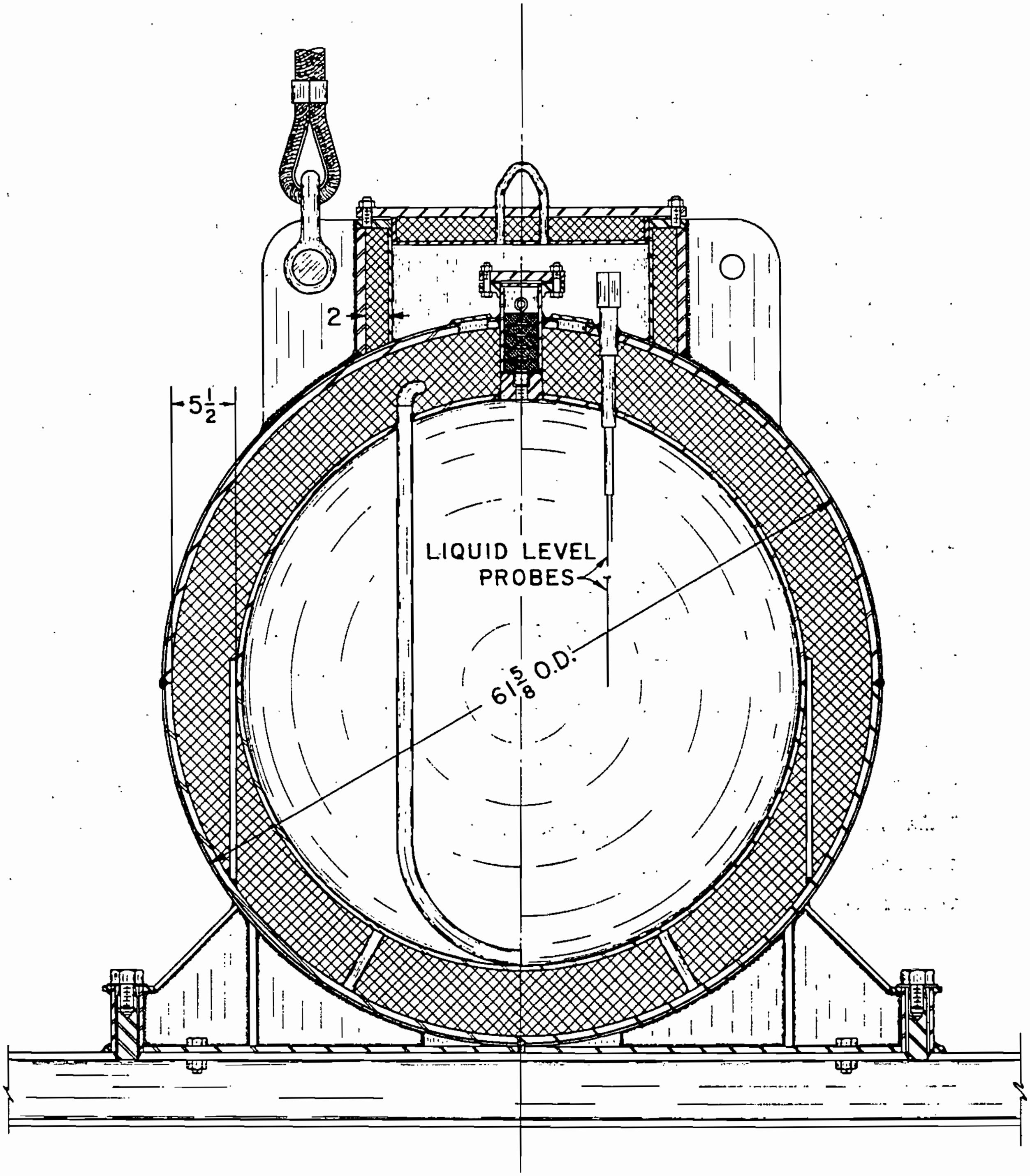


Figure 1

SOLUTION CARRIER

(SHIELDED TRANSFER TANK)

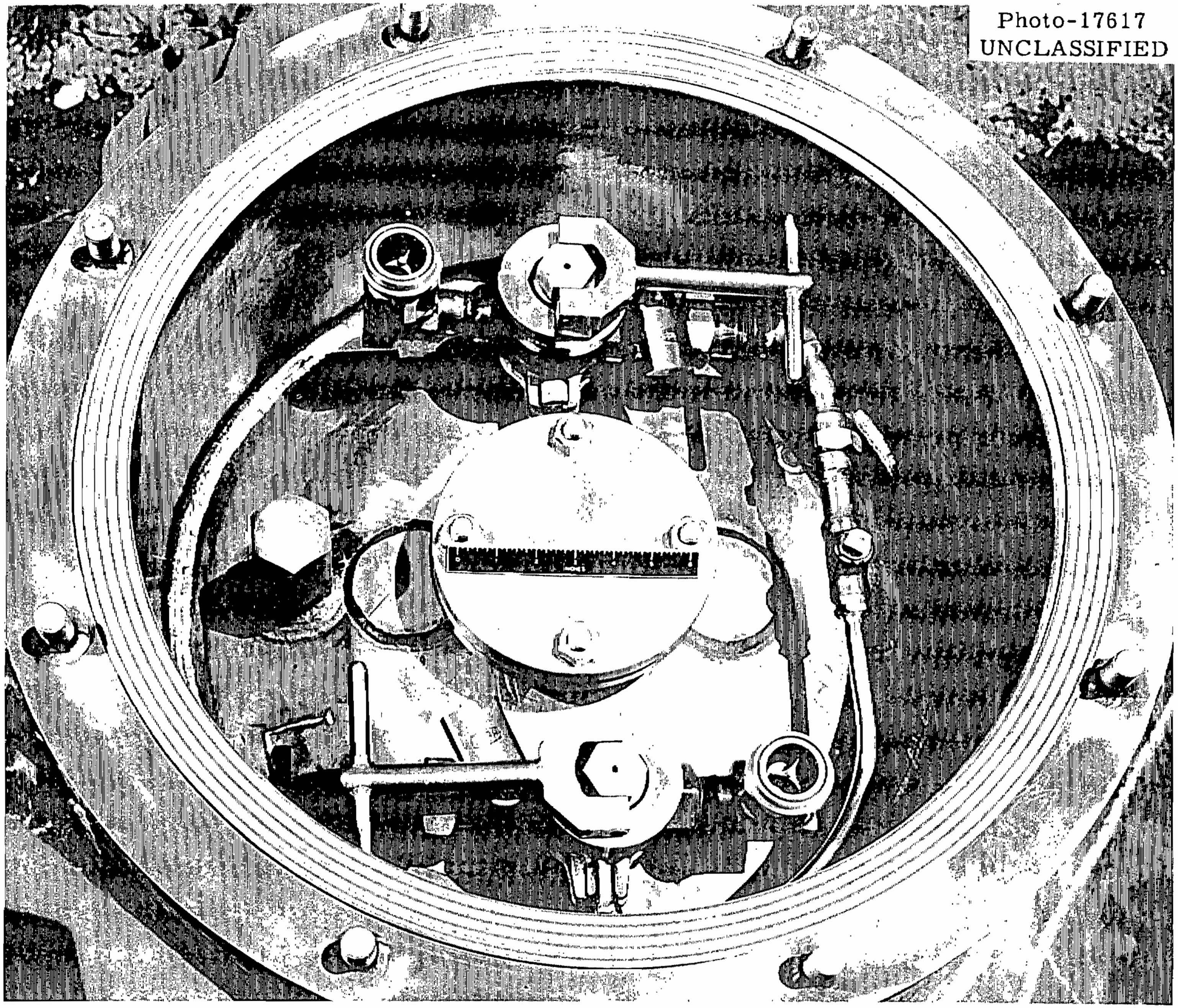


Photo-17617
UNCLASSIFIED

Figure 2

Photo-15102
UNCLASSIFIED

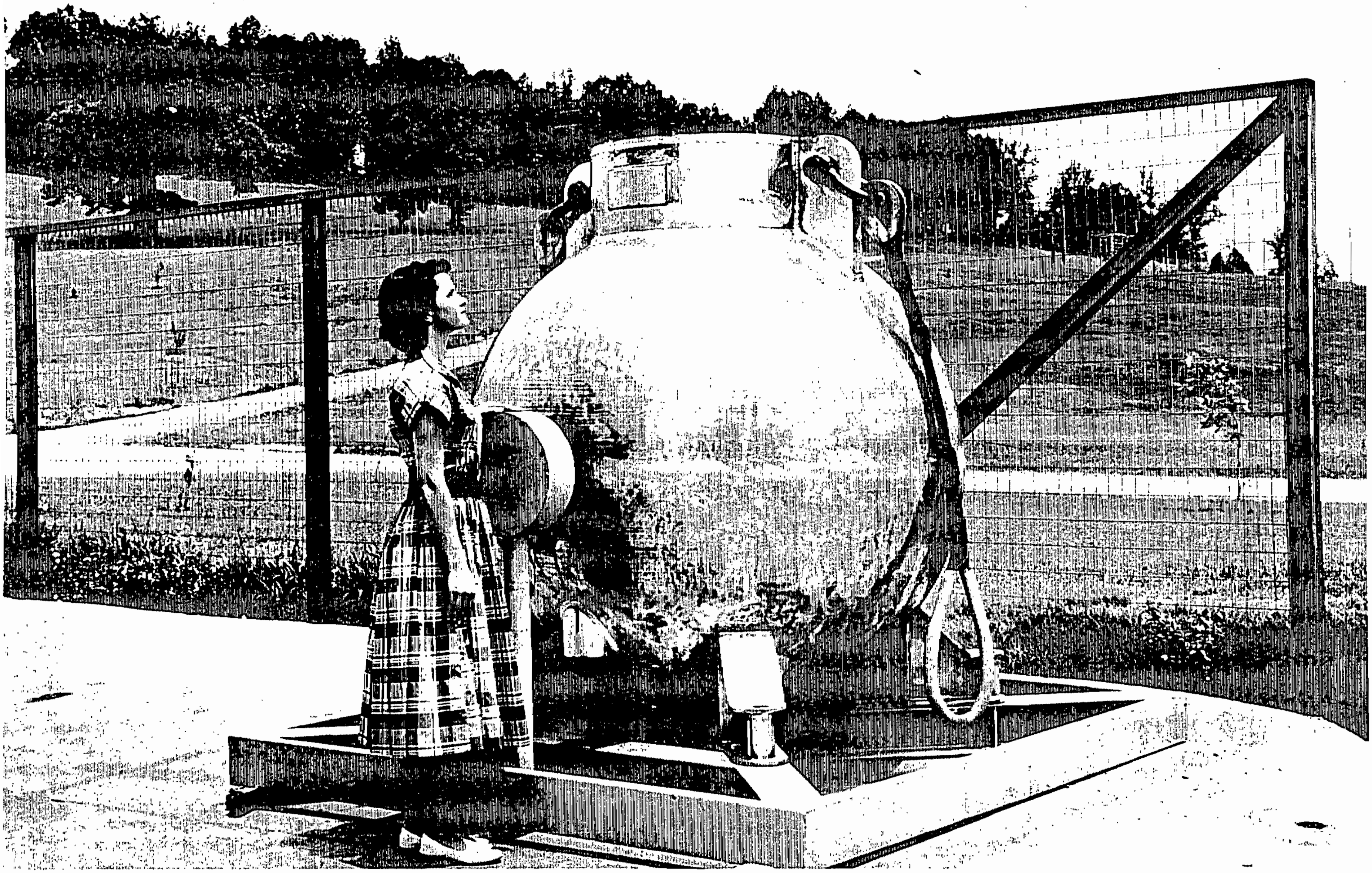


Figure 3

REFERENCES

1. F. L. Culler, "Comments on the Transportation of Irradiated Fuel and Radioactive Wastes", ORNL CF 57-5-24, pp 14-16, Unclassified, May 6, 1957.

2. F. L. Culler, "General Economics of Chemical Reprocessing for Solvent Extraction Processing", ORNL CF 57-4-100, May 20, 1957, Unclassified.

3. E. D. Arnold, "Certain Aspects of the Economics and Hazard Potential of Radioactive Waste Disposal", Presented June 11, 1957, American Nuclear Society, Pittsburgh. Available at ORNL.

4. A. L. Ayers, "Requirements and Costs of Shipping Irradiated and Recovered Fuels", IDO-14363, Part I, Jan., 1956, pp 155-194. (AEC document) and ORNL CF 57-5-24, Unclassified.

5. H. R. Zeitlin, J. W. Ullmann, E. D. Arnold, "Economics of Waste Disposal", Nucleonics, 15(1), 58-62 (1957) (Taken from ORNL CF 55-6-162, ORNL CF 55-10-101, ORNL CF 56-1-162.

6. W. E. Unger et al, "Auxiliary Radiochemical Equipment", ORNL CF 57-5-17, pp 2-7, May 15, 1957.

8.0 Possibilities of Ultimate Waste Management and/or Disposal

8.1 Introduction

Perhaps the most significant problem in radioactive waste disposal is that of determining the final repository for radioactive materials. It appears from surveying both the classified and the unclassified literature that the research and development programs aimed at providing information on safe ultimate disposal (and corollary efforts in environmental effects) are most urgently requiring investigation. Research and development leading to the selection of satisfactory disposal sites and to the undertaking of significant experimental programs to define the health and safety aspects of ultimate disposal methods must be selected to give significance to any chemical steps taken to reduce volumes, mobility of radioactivity, costs, heating problems, etc.

A rather fundamental question that probably must be answered without the enlightenment of much development data is whether or not any large quantity of long-lived fission products and heavy elements can be placed in some remote natural sink without surveillance. If the answer to this question is no, then it is necessary to seriously question the alternate solution, which implies control by some agency of man's design, an agency which should be self-perpetuating for possibly a thousand or so years.

In either case the implication is that long-term controls for either the release of or the retention of radioactivity will be required on a worldwide basis with an unfailing constancy.

Actually, much thought has been given to the possibilities that exist for permanent or ultimate disposal. We shall review some of the more interesting possibilities.

The disposal of conventional industrial wastes and sewage usually encompasses methods of returning them to the environment in such form and concentration that they do not represent hazards to existing plant and animal life. The disposal of radioactive wastes presents a different and more difficult problem in that the radiotoxicity of these wastes cannot be destroyed or diminished by any known treatment. Furthermore, the limits of biological tolerance of radioisotopes in the environment are so restrictive that the problem of adequately dispersing large quantities is almost insurmountable. Most of the radioisotopes appear in aqueous effluent streams from chemical processing plants and the practice has been to store this material in large tanks interred at the surface. Only a relatively minor portion, including certain of the gaseous wastes, have been released to the environment. The advent and growth of a nuclear power industry portends a great increase in the volumes or radioactive wastes to be encountered during the next half century, and the present practices which are at best only temporary expedients, cannot be expected to meet the requirements for ultimate or permanent disposal.

In recognition of these facts there has been both speculative and serious consideration given to methods which might serve the purpose of ultimate disposal. It has been proposed that the oceans, by virtue of the tremendous dilution factor they offer, might serve as a medium for dispersal. Other possibilities have been seen to exist in the various types of underground geological formations where permanent isolation of the wastes from the natural environment might be achieved. A realistic evaluation of all such proposals, even in a preliminary sense, requires the careful study and consideration of experts from a number of highly specialized fields. Under auspices of the Atomic Energy Commission, groups of qualified persons have met for the purpose of considering the possibilities of sea disposal and land disposal of radioactive wastes. While there is, admittedly, insufficient information to form a positive and final evaluation of any proposed method at the present time, it was, nevertheless, possible to evaluate the potentialities in a qualitative sense and specify the research yet remaining to be done before significant field-scale experiments can be performed.

8.2 Fixation of Wastes in Solid Form Prior to Ultimate Disposal in Ground

Although the occurrence of radioactive wastes as aqueous solutions is convenient from the standpoint of transport within a processing facility and for efficient removal of decay heat during storage, the attendant properties of mobility and chemical reactivity render liquid wastes especially hazardous to dispose of permanently. A substantial reduction in the long-term hazards associated with disposal could be achieved by combining or "fixing" the radioisotopes in solids from which they could not easily be removed. These solids could then be disposed by storage in suitable areas.

The basic requirement of any process for this purpose is that it economically produces a thermally and mechanically durable solid which will retain the activity should it be exposed to water or brines. The maximum allowable costs cannot be strictly specified until the other requirements in the overall waste disposal complex such as shipping, interim storage, and ultimate disposal methods have been defined. However, it should be pointed out that the high-level liquid waste streams are as large in volume as the primary product streams in the chemical processing plants and that, consequently, multi-step processes can be expected to approach prohibitive costs rather quickly unless one can rely on commercial utilization of some of the by-products.

It is equally impossible at this time to impose limits of leachability on the final products. While it would be desirable to produce solids from which the fission products could not be leached within limits of detection, it would seem more realistic to accept greater leachability if substantial savings in process simplicity and costs resulted. There is reason to believe that partially leachable solids could be either packaged economically or stored without packaging in dry spaces like salt cavities without unacceptable hazard.

There is a number of processes for converting liquid wastes to solid form currently under development. None of these have been carried to the stage of pilot plant testing with high level wastes but enough basic information has been acquired in some cases to warrant demonstrations at higher activity levels in the near future.

8.21 Ultimate Disposal Utilizing Montmorillonite Clay

Possibly the earliest work on fixation of radioactive wastes was initiated by L. P. Hatch at Brookhaven National Laboratory.(1)(2) This process involves as a preliminary step, evaporation of the wastes to dryness and decomposition of the nitrates to oxides. Upon discharge from a calciner, the dry, granular oxides of aluminum or zirconium are contacted with water or weak acid for removal of leachable fission products. The resulting leach solution is passed over extruded montmorillonite clay which sorbs essentially all the fission products in solution (except ruthenium) and these sorbed activities are subsequently "fixed" by firing the clay at 1000°C. Fission product bearing clays are heated in silicon carbide or inconel containers.

8.22 Fixation with Nepheline Syenite(3)

The nepheline syenite process as studied at Chalk River, Canada, consists of mixing nepheline syenite (a low-melting silicate) with acid wastes. A gel is formed which is porous and can be dried with little entrained activity. When heated to 1200°C, it fuses to a glass from which the only leachable activity is that apparently resulting from surface contamination. Work is currently underway to convert the process from a batch to a continuous operation and to reduce the temperatures by adding fluxes to promote the formation of non-corrosive, lower-melting glazes.

8.23 Self-sintering in Insulated Pits with Shale as Ultimate Disposal Possibility and Extremely Fired Sintering Prior to Ultimate Disposal

The self-sintering process is designed, as its name implies, to make use of the heat evolved by fission product decay to obtain the temperatures required for fixation. Work on this process has been performed by Struxness et al(4) at Oak Ridge National Laboratory. Liquid wastes are mixed with definite proportions of shale or clay, limestone, and soda ash, then are allowed to stand in well-insulated pits until the fission product heat has evaporated the mixture to dryness, decomposed the nitrates, and finally, elevated the temperature of the resultant cake to the region of 900°C. This process has the potential economic advantages of requiring no chemical pretreatment and relatively minor process equipment, but suffers the disadvantage of being limited in a practical sense by heat requirements to only the most concentrated wastes with heat evolution requirements of at least several watts per gallon.

Tests were made with aluminum nitrate simulated wastes and shale in the following proportions in a heated and insulated pit:

720 gallons of 2.2 M. $Al(NO_3)_3$ waste
2405 lbs. of 200 mesh Conasauga shale
720 lbs. of limestone
720 lbs. of soda ash

The sintered product was hard and durable. Laboratory studies using specific fission products as part of the sinters (by external firing) indicated that static water leach would remove only small tracers of activity.

8.24 British Process for Fixation of Highly Active Wastes

The British have been working on the fixation concept since 1953. Amphlett and co-workers(5) have studied all the approaches being considered in the United States and Canada without carrying any of them into the engineering or equipment stage. They have obtained excellent fixation and higher capacities by disregarding the ion-exchange effect and mixing their wastes as solutions or slurries with clays, soils, and fluxes and firing at temperatures near 1000°C. Although they agree that self-fixation should be feasible in those cases of very high concentrations of activity, they appear to favor at this time mechanical heating of their own wastes and are ready to begin tests of the equipment and remote handling devices required.

8.25 Fluidized Bed Calcination

A concept quite closely akin to fixation is that of simply calcining the aluminum or other high-salt-content wastes without the addition of other solids. While fixation of most or all the activities is desired, clay or other solids are not added specifically for that purpose. Use of the fluidized bed technique for this process has been studied both by Jonke(6) at Argonne National Laboratory and by Grimmett(7) of Phillips Petroleum Company, National Reactor Testing Station, Idaho. The concentrated aluminum nitrate waste is injected into a vigorously fluidized bed of aluminum maintained at about 500°C. The fission product and aluminum nitrates decompose to their respective oxides and accumulate in the form of agglomerated spherical Al_2O_3 particles which are continuously withdrawn from the bottom of the column. A volume reduction of about 6 is achieved and the technique appears to be applicable to zirconium-type wastes as well. A hot pilot plant capable of handling a maximum of 200 curies of 1 Mev gamma radiation has been constructed at ANL and will process 2 to 3 gal/hr of aqueous waste. This will probably be the first of these processes to be tested with significant radioactivity.

8.26 The Brookhaven Waste Calciner

Manowitz and Hittman(2)(8) have proposed calcination of aluminum nitrate wastes in a screw calciner. The presence of sodium nitrate in these wastes serves as a flux, and a free-flowing solid, melting at 300°C, is produced. The solid offers a volume reduction factor of 3 over concentrated aqueous wastes and can be cast in desired sizes and shapes for efficient heat removal during storage.

8.3 Separation of Strontium and Cesium Prior to Disposal

The major long-lived contaminants and biological hazards in radioactive wastes from reactor fuel processing are 26y Sr^{90} and 26.6y Cs^{137}. Furthermore, after a decay period of about eight years, these isotopes and their daughters account for virtually all the heat being evolved in the wastes. Qualitative separation of these species would greatly reduce the thermal problems that may be associated with ultimate disposal in salt formations or deep wells, but decontamination by factors of 10^6 to 10^7 would be required before the wastes could be safely released to the environment. In most cases, the additional decontamination of plutonium and the transplutonics by factors of 10^2 to 10^3 would have to be achieved before release could be permitted. It can be expected that separations of such high order would be very difficult to attain and probably would not be economically feasible.

Possibly the greatest experience in separating fission products from waste streams exists at Oak Ridge where the production of radioisotopes for commercial purposes has been underway since the war. Rupp(9) has described the processes currently in use for separating cesium and strontium from aluminum nitrate wastes. The cesium is removed first by co-crystallization as alum, from which very pure sources of cesium chloride are prepared. The rare earths are next separated from the waste by precipitation as the hydroxides with ammonia gas, and the strontium is then removed by precipitation as the carbonate.

There are processes under development at Hanford(10) and Idaho based on metal ferrocyanide scavenging which have demonstrated greater than 99% removal of strontium and cesium. While such processes cannot of themselves serve the purpose of ultimate disposal, they do serve to reduce both the hazard and the heat production to levels where more economical storage and disposal might be effected. In addition, they could provide economical production for purposes of commercial utilization.

8.4 Ocean Disposal

The oceans have been used only to a very limited extent for disposal of certain low-level wastes. In the United States wastes from laboratories and other research use have been carefully packaged and dumped at sea. The British have carried the practice further by dumping liquid wastes off-shore in the Irish Sea.(11) In both instances only inconsequential quantities were involved compared to the large-scale disposal operations required by a nuclear power economy. The conclusions of a number of qualified specialists who have considered the longer-range aspects of ocean disposal have been summarized by Renn.(12)

It has been proposed that radioactive wastes might be disposed in a number of ways in the ocean. One possibility is by pumping the dense, saline wastes into any of a number of deep holes where large bodies of stagnant water are known to exist. It is expected that the wastes would remain in such locations until their activity decayed to safe levels. However, there appears to be sufficient evidence based on temeprature and oxygen content of the waters in such deeps, to conclude that there is a more frequent overturn than had originally been assumed. Prolonged cooling cycles and other types of surface weather conditions probably cause vertical mixing in cycles of every century or so.

A second possibility has been to dump packaged wastes into canyons on the North American continental shelf, the advantage being that such areas are well-defined and close to shore. Submarine geologists have pointed out, however, that these canyons are produced by local instabilities and are scoured periodically by submarine mudslides which reach velocities of 15 to 20 miles per hour. It seems unlikely that economical containers of the structural strength required to withstand such treatment could ever be developed.

Proposals to deposit packaged wastes in deep sea muds and oozes where they would become buried have in many cases been unrealistic. Many areas where such oozes are known to exist contain such deposits only in superficial depth. The fines are generally underlain with consolidated putty and clays making any degree of useful penetration extremely unlikely. There are two general areas where natural containment of packaged wastes may be possible, however. One of these is in the Gulf of Maine which is also an area of commercial fishing and deep-sea trawling operations. A second, yet more distant, area exists in parts of the Gulf of Mexico.

A subject of great importance and uncertainty is the degree of assimilation of radioactivity one can expect of plankton and organisms in the sea. Marine biologists and ecologists are concerned over the potential hazards associated with assimilation and concentration of fission products by plants and animals in the sea. Little is known about what the rate and form of concentration of long-lived strontium and cesium would be, but a careful examination of all the important variables that enter into the marine environment would be required.

Many proposals for disposal of radioactive wastes at sea are based entirely or partially on the concept of dilution by the ocean waters. Experience has shown, however, that the mechanism of mixing in large masses of water is very unpredictable. Cases have been studied where dense, saline wastes were dumped in the ocean and it was found that movement occurred horizontally at much greater rates than vertically. Such a phenomenon greatly restricts the volume of water available for dilution and emphasizes the necessity for discharging liquid wastes directly into the stratum where dilution is desired.

Such considerations as the above, when taken with the problems of developing economical methods of transporting wastes to selected disposal sites and reliable methods of monitoring such areas, present a very formidable and not encouraging picture of the prospects for disposing of significant quantities of wastes at sea. A vast amount of work remains to be done before the necessary degree of confidence in such an operation can be established.

8.5 Land Disposal

The National Research Council under contract to the Atomic Energy Commission formed a Committee on Waste Disposal to evaluate all suggestions and research to date on disposal methods that involve land, surface, or underground sites and recommend programs of research that should be carried out. The Committee offered the following specific recommendations on disposal:

1) Disposal in tanks is <u>at present</u> the safest and possibly the most economical method of storing waste.

2) Disposal in salt is the most promising method for the near future. Research should be pushed immediately on the structural problem of stability vs. size of cavities at a given depth; on the thermal problem - getting rid of the heat or keeping it down to acceptable levels - and on the economics of such disposal.

3) Next most promising seems to be stabilization of the waste in a solid and preferably non-leachable form such as a ceramic material. This could be followed by controlled storage in dry mines, surface sheds or large cavities in salt.

4) Disposal of waste in deep porous beds interstratified with impermeable beds in a synclinal structure is a possibility for the more distant future. This is of particular interest for disposal of large volumes of waste. The reaction of the waste with connate waters or constituents of the rocks soluble in the waste solution will have to be studied. The composition of the rocks and the connate waters are both variable as will be the composition of the waste solutions so that an almost infinite variety of circumstances result. In general such highly salted wastes as acid aluminous waste, in undiluted form, would almost certainly tend to form precipitates which would clog pore spaces. The problem would have to be solved first for a given bed at a given site for a given waste solution.

5) The removal of Cs^{137} and Sr^{90} from the waste would make disposal somewhat easier for the waste free of these isotopes, but does not change the recommendations made in the report qualitatively.

6) Disposal even of low level waste in the vadose water zone, above the water table, is of limited application and probably involves unacceptable risks.

In the following pages, a review of the potentialities and problems of land disposal leading to the above recommendations is given.

8.51 Tank Storage

The early decision for holding radioactive wastes in tanks was no "out of sight, out of mind" policy. A great deal of study and planning has preceded the building of these "tank farm" systems, and to date there have been no instances of important structural failure. Reliable monitoring and leak detection systems have been developed. In some cases a second line of defense against the ever-present danger of leaks and breaks developing in the tanks was conceived through interpretation of results of extensive studies of the geology and geochemistry of the local regions, the theory being that an insight into the probably natural course which the active wastes would follow, in the event that a leak or break occurred, would permit a calculation of the ensuing hazard. An overall factor of safety might thus be foreseen in a higher degree than would be permissible from a consideration of the physical and chemical stability of the tanks and their supporting structures alone. However, there can be no sound basis for calculating the useful storage life of tanks until much more is known about the important factors of corrosion. Consequently, tanks will, in all probability, be used not as a means of ultimate disposal, but as a storage or holdup medium to allow fission products to decay to safe disposable levels.

8.52 Disposal in Salt Formations

One of the most attractive possibilities for the disposal of radioactive wastes is its underground storage within deposits of rock salt. Large deposits of salt exist in many well-defined and accessible locations within the United States and commercial mining operations create annually, spaces which are greatly in excess of the expected volumes of high-level waste production at the end of this century. These spaces possess many desirable attributes for radioactive waste storage. In addition to offering an isolated and relatively uniform chemical and mineralogical environment, salt is plastic under load and deposits are impervious to water. Cavities can be mined in such a manner as to be structurally safe and accessible to personnel and equipment. Because of its plasticity, salt deposits are considered to possess immunity to earthquake hazard to a unique degree. Heroy(13) has made a preliminary study of the use of salt formations for the purposes of radioactive waste disposal and has described its availability and characteristics in some detail.

Occurrence of Salt

The principal areas underlain by salt in the United States are shown in Figure 1. The major deposits occur in the north central states and in the

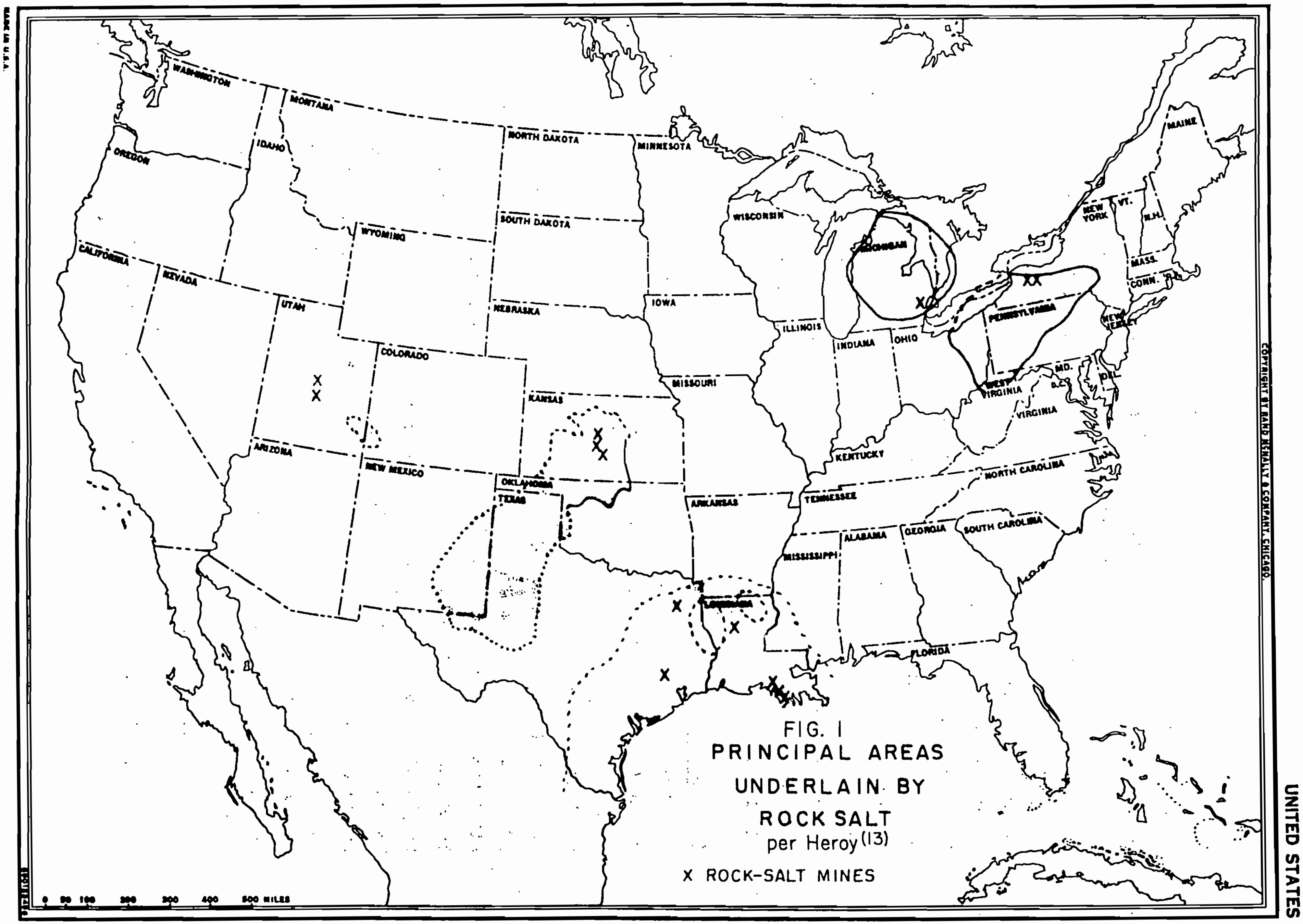


FIG. I
PRINCIPAL AREAS
UNDERLAIN BY
ROCK SALT
per Heroy (13)

X ROCK-SALT MINES

southern states along the Gulf Coast. The salt formations of New York, Michigan, Ohio, and Kansas underlie many thousands of square miles and extend from surface outcroppings to depths of more than 5000 ft. They are frequently several hundred feet thick. In New York salt occurring in the Salina formation crops out along a band extending from the Mohawk Valley on the east to the Niagara River on the west. The beds dip southward at a low angle, averaging from 50 to 100 ft. per mile and extend into southern New York and northern Pennsylvania. At its maximum, the salt is about 1000 feet in thickness and in northwestern Pennsylvania it is found at depths of from 1500 feet to more than 8000 feet in the deepest part of the syncline.

The Salina beds extend westward into Ohio where they underlie an area of over 15,000 square miles. The salt is all below the surface at depths of from 1000 to more than 4000 feet and have a thickness over most of this area of more than 100 feet.

It is estimated that an area as great as 35,000 square miles in Michigan is underlain by salt-bearing formations. The formations are found within the Michigan basin at depths ranging from a few hundred feet near Detroit to 2000-2300 feet in the western part of the basin near Ludington and Mainstree. Thicknesses as great as 1800 feet have been penetrated.

About 30,000 square miles in the central and southeastern parts of Kansas are underlain by salt-bearing formations. This salt dips from its outcrop in Salina and Sumner Counties to depths of 650 feet at Hutchison, 1000 feet at Lyons, 1700 feet in Kiowa County, and 2000 feet in Clark County. Its thickness is usually from 200 to 300 feet. Salt of similar thickness occurs at depths of 1000 to 1600 feet in the southwestern part of the state.

In the Gulf Coast area of Louisiana and Texas, salt often occurs in the form of domes lying anywhere from a few hundred feet to as much as 10,000 feet below the surface. It is believed that such formations resulted from flowage of salt under pressure upward through overlying beds. The location of as many as 200 of these domes is known, ranging in size from nearly circular domes, one-half of two miles in diameter, to elongated masses several miles in length. Thicknesses of 500 to several thousand feet are normal.

Rock salt also occurs in eastern Utah and western Colorado. Its extent has not been fully determined, but it has been estimated to underlie at least 10,000 square miles. It has been penetrated in some test wells to a thickness of over 3000 feet.

In the southwest salt occurs in the Delaware Basin of New Mexico and Texas to an extent that may underlie approximately 76,000 square miles. The beds vary in thickness up to 3500 feet and lie within about 500 feet of the surface. In part of this area a zone of potash salts is present which has been extensively developed near Carlsbad, New Mexico during the past 25 years. The salt is not mined, however, except as a by-product of the potash, and it is marketed to only a very limited degree.

Mining and Production of Salt

Salt is mined commercially both in its solid form and by dissolution in water and removal as brine. It is also produced as natural brines which are pumped to the surface from porous formations and evaporated. The total annual national production currently exceeds 20 million tons, about 60% of which is produced as natural or artificial brines, 20% by underground mining of salt deposits, and the remainder as evaporated salt. Table I, from Heroy, presents 1953 salt production according to states and to the form in which it was produced.

Rock salt was mined at 14 sites in the United States in 1953 with locations in New York, Michigan, Kansas, Louisiana, Texas and Utah. The production by states as estimated by Heroy is given in Table II. The total space mined in 1953 was 1547 acre-feet (67.4 million cubic feet), and based on reported production, a volume of 21,250 acre-feet has been mined during the last 20 years. The deepest mines extend to depths in excess of 1000 feet and are connected to the surface by shafts large enough to accommodate power equipment. From 50 to 60 per cent of the salt is extracted and the remainder is left as pillars for structural support. These mined spaces are quite level and are extremely dry. Inspection has shown them to be frequently devoid of faults, indicating a geological history of stability.

Production as artificial brines is accomplished by pumping water into beds of rock salt under pressure, and as salt is dissolved, returning the solution to the surface. Although this is a more economical process than mining, the cavities resulting from such operations have been known to fail structurally due to the unsupported weight of overlying rocks. Greater

TABLE I

SALT - PRODUCTION BY STATES - 1953 - SHORT TONS
per Heroy(13)

	Rock Salt	Salt in Brine	Total	Evaporated Salt	Total	Value	
						Total	Per Ton
California	-	273,365*	273,365*	850,000*	1,123,365	$6,263,059	$5.58
Kansas	534,658*	-	534,658	370,569	905,227	7,480,556	8.27
Louisiana	1,338,997*	1,600,827*	2,939,824	121,410	3,061,234	9,189,526	3.01
Michigan	1,000,000*	3,306,727*	4,306,727	860,660	5,127,387	22,171,988	4.31
New Mexico	-	-	-	62,087*	62,087*	216,364	3.48
New York	1,200,000	1,589,735*	2,789,735	532,924	3,322,659	17,351,111	5.22
Ohio	-	2,541,799*	2,541,799	498,438	3,040,237	7,484,795	2.48
Puerto Rico	-	-	-	13,692	13,692	131,490	9.60
Texas	400,000*	2,333,339*	2,733,339	111,851	2,845,190	5,010,624	1.76
Utah	5,000*	-	5,000	149,088	154,088	772,035	5.00
West Virginia	-	419,907*	419,907*	-	419,907	1,490,592	3.55
Others(1)	-	542,344*	542,344*	171,586*	713,930	714,527	1.00
TOTALS	4,478,655	12,608,043	17,086,698	3,702,305	20,789,003	$78,276,667	$3.77

* Estimated

(1) Alabama, Hawaii (evaporated); Oklahoma (salt in brine); Virginia (salt in brine).

TABLE II

ROCK SALT

ESTIMATED PRODUCTION BY STATES - 1953 - SHORT TONS

per Heroy(13)

	Production	Value	Per ton	Equivalent space, acre-foot(1)	Average thickness mined	Acres mined out	Depth to salt
Kansas	534,658	2,194,751	$4.10	185	10	37	600-1000
Louisiana	1,338,997			462	80	10	600-800
Michigan	1,000,000			346	30	25	1000
New York	1,200,000			414	10	68	1000
Texas	400,000			138	60	5	700;1500
Utah	5,000			2			
TOTALS	4,478,655	23,777,527	$5.34	1,547		145	

---

(1) Specific gravity, 2.15; 134 lbs. per cu. ft.; 15 cu. ft. per ton; 2900 tons per acre-foot.

(2) Assuming 50% or 60%, according to locality, left as pillars.

experience with these techniques, however, has provided an increased measure of control over the size and shape of the cavities and some are currently being used for storage of liquified petroleum gas products under pressure.

Utilization of Salt Space for Waste Disposal

In the light of the characteristics and availability of salt deposits, it appears that under the proper circumstances they could be used for storage of both solid and liquid wastes. As is discussed in Section 9.2, there are a number of processes under development designed to convert high-level liquid wastes to less mobile, solid forms. Excavations in rock salt would seem to be especially suitable for storage of these packaged or solid wastes. Before such disposal practices could be initiated, however, a very thorough study of the availability and cost of the desired space should be made with particular emphasis being placed on the structural properties of the salt deposit under consideration and the effect of temperature on these properties. In addition, the thermal problems arising from decay heat during storage will have to be defined and any necessary cooling and ventilation equipment designed. Finally, engineering studies must be made of the best methods and equipment for handling and conveying radioactive solids of the type to be disposed.

The disposal of liquid wastes in salt offers the advantages of a maximum of control over the disposed wastes with the possibility of ultimate recovery if desired. It can be anticipated that wastes already near saturation with dissolved chemicals could be stored in contact with salt without incurring serious chemical or physical changes. The success and safety of such an operation will depend in large measure, however, on the severity of the thermal problems occurring from the heat emitted by radioactive decay. Unless this heat could be dissipated by conduction in the salt without undue rise in the liquid temperature, it would be necessary to extract it by some mechanical means designed to operate on a long-term basis.

The temperature of the wastes could be maintained at a desired level by submerged cooling coils; however, the presence in solution of both chlorides and nitrates would impose severe corrosion problems. If, on the other hand, heat were removed by allowing the wastes to boil and refluxing condensed vapor, a somewhat greater hazard with less control over the system would be accepted. It is probable that either operation could be accomplished more safely and economically in steel tanks near the surface, and that, consequently, disposal of liquid wastes in salt should only be considered in those cases where subsequent cooling is not required.

Hydraulically mined cavities offer some attractive features for liquid storage; They can be excavated in a variety of sizes and shapes with great precision which should make possible the attainment of structurally safe spaces possessing large surface-to-volume ratios for efficient heat dissipation. Access to them would be by a shaft to the surface permitting the location of all auxiliaries above ground.

8.53 Disposal in Deep Wells

An attractive possibility for ultimate disposal of radioactive wastes appears to be the utilization of deep wells probing into subterreanean geological formations. The feasibility of such a concept is suggested by the techniques of brine injection as practiced by the petroleum industry. For a number of years great volumes of brines have been successfully injected either for the purpose of disposal or for the secondary recovery of oil.(14) With such a technology already established, it seems reasonable to expect that applications to radioactive waste disposal may exist.

Analyses of the anticipated problems associated with the disposal of radioactive wastes in deep wells have been made by de Laguna,(15) Theis,(16) Roedder,(17) Kaufman, et al,(18) and Pecsock.(19) Attempts were made to define the attributes of an underground formation suitable for containment of these wastes and preliminary consideration was given to the most likely site locations. While many of the arguments presented are speculative and therefore controversial to some degree, it is of interest to note that none of the problems so far envisioned appears insurmountable.

Hazards

In a category by itself, separate and distinct from considerations of technical feasibility, is the primary requirement that the disposal method meet those specifications required for the protection of this and future generations of man. These specifications are far more rigorous for radioactive wastes than for brine or other chemically toxic substances. Sodium chloride is dangerous only when present in concentrations of several hundred parts per million and dilution can be relied upon as a practical and effective means of control. Radioactive wastes, on the other hand, would have to be diluted by factors of 10^{10} and greater before they could be considered potable. Proportionately greater care would have to be taken during the preparation and operation of a radioactive waste injection system to thoroughly seal the well below the potable water bearing formations and to maintain a completely leak-proof system. Injection must be made into formations where there is maximum assurance that migration to ground or surface water does not occur, and in areas where it is least likely that valuable petroleum or mineral deposits exist. There must, furthermore, be assurance that no other wells - new or abandoned - pierce the injected formation within the area to be contaminated.

Chemical Compatibility

Experience with brine injection has shown that if plugging of the wells is to be avoided, care must be taken to ensure chemical compatibility between the waste and the residual liquids and solids of the aquifer. With brines, plugging is minimized by such pretreatment as sedimentation, filtration, and the addition of certain chemicals for control of objectional bacteria and algae. Because of their diverse and complex chemical nature, it seems likely that the radioactive wastes will also require treatment prior to injection. In all likelihood this will be a more severe problem than for brines since their chemical nature will differ radically from that of typical connate waters. Treatment by dilution and addition of complexing agents are likely avenues of approach to chemical compatibility, but a very thorough chemical and mineralogical knowledge of the aquifer will be required before compatibility with any particular waste can be assured. Roedder has discussed the severe problems to be expected should aluminum nitrate wastes be injected. While wastes containing other chemical constituents may be more amenable, these considerations could, nevertheless, impose limitations on the types of wastes suitable for injection.

Heat Evolution

A problem entirely unique to radioactive wastes is that of heat generation. The energy of the radiations from fission product decay ultimately appear as heat which must be effectively dispersed to the environment if intolerably high temperatures are to be avoided. Although fission product heat is of a very low quality and decreases with time, its production continues inexorably as long as the radiation persists. The severity of this problem as it relates to deep well injection will depend on such factors as the age and concentration of fission products in the waste, the heat transfer characteristics of the storage aquifer and contiguous geological formations, and whether or not any tendency exists toward reconcentration of the fission products through precipitation or sorption on the solids of the aquifer. Although such a physical system would be very difficult to simulate mathematically, it seems reasonable to believe that the effects of heat evolution can be controlled by the proper combination of aging, treatment, and dilution of the wastes before injection.

Table III presents the thermal conductivities of a number of sedimentary rocks selected by Theis from a more extensive compilation by Birch et al.(20) The rocks selected are among the more prominent species to be considered in ground disposal of radioactive wastes.

Hydrologic Considerations

While it is not expected that the hydrologic problems associated with deep well disposal will be severe, a detailed study will be required for the purpose of accurate control. The volumes of wastes to be disposed will range from several millions of gallons per year at first to an anticipated several hundreds of millions of gallons per year in the year 2000. The petroleum industry is currently injecting comparable volumes of brine. In the case of radioactive waste disposal, however, injection pressures must be held to a minimum for assurance that upward leakage will not occur, thus both the transmissibility and the capacity of the storage aquifer must be well defined. While high transmissibility and large capacity are desirable from the standpoint of large injection rates at low pressure, their advantages may be compromised to some extent by greater and more rapid distribution of the contaminated waters.

It has been suggested that an outer ring of wells would be required for monitoring the flow within the aquifer. For efficient monitoring, such wells would be pumped, and could thus serve as a source of water for dilution of the injected wastes as a means of reducing pressure within the aquifer.

TABLE III

THERMAL CONDUCTIVITY OF ROCKS
per Theis(16)

Rock	Temperature (Degrees C)	Conductivity, $\left(\frac{\text{Cal.}}{\text{Sec. cm.deg.}}\right)$
Limestone, dolomitic, Queenston Ontario	123	3.4 x10^{-3}
	177	3.4
	254	3.3
	332	3.2
Marble (17 varieties)	30	7.7-5.0
Proctor, Vermont, Parallel to bed	0	7.36
	100	6.0
	200	5.2
Perpendicular	0	7.2
	100	5.7
	200	5.1
Quartzitic sandstone		
Parallel to bed	0	13.6
	100	10.6
	200	9.0
Perpendicular	0	13.1
	100	10.3
	200	8.7
"Recrystallized sandstone"	30	11
"Hard sandstones"		10.8-6.2
Sandstone, Boreland Bore	17	10
"Soft sandstones"		4
Slate, Wales		
Parallel to schistosity		6.7
Perpendicular to schistosity		3.9
Slate	30	4.7
Shale		4.1-2.4
"very fine-grained"	17	1.4
"with sand"	17	2.8
Gerhardminnebron Bore(Witwatersrand)		
from 6457 feet	25	6.6
from 4190	25	4.4
Silty clay	17	3.7
Silt, Hankham (borehole)	17	4.4
"uncemented"	17	5.3
"micaceous, argillaceous"	17	2.5
Fireclay, Boreland Bore	17	4.4
Red marl, Holford	17	5.25
Gray marl, Holford	17	2.2 3.5
Rocksalt, Holford	17	17.2

The Ideal Aquifer

Based on his interpretation of the considerations and problems associated with disposal by deep well injection, de Laguna has summarized the requisities of a desirable aquifer as follows:

1) The transmissibility should be high, preferably ten thousand gallons a day per foot, or more, although limiting values cannot be specified.

2) The hydrologic properties should be sufficiently uniform so that quantitative values for the movement of liquid through the aquifer can be determined and applied with confidence.

3) The aquifer should have a considerable extent, but not so extensive that it creates a potential hazard at distant points.

4) A depth of a very few thousand feet is probably sufficient, particularly if the cover is known to be highly impermeable. Great depth is likely to make drilling and monitoring expensive and so reduce the safety that may be attained with a given expenditure of funds.

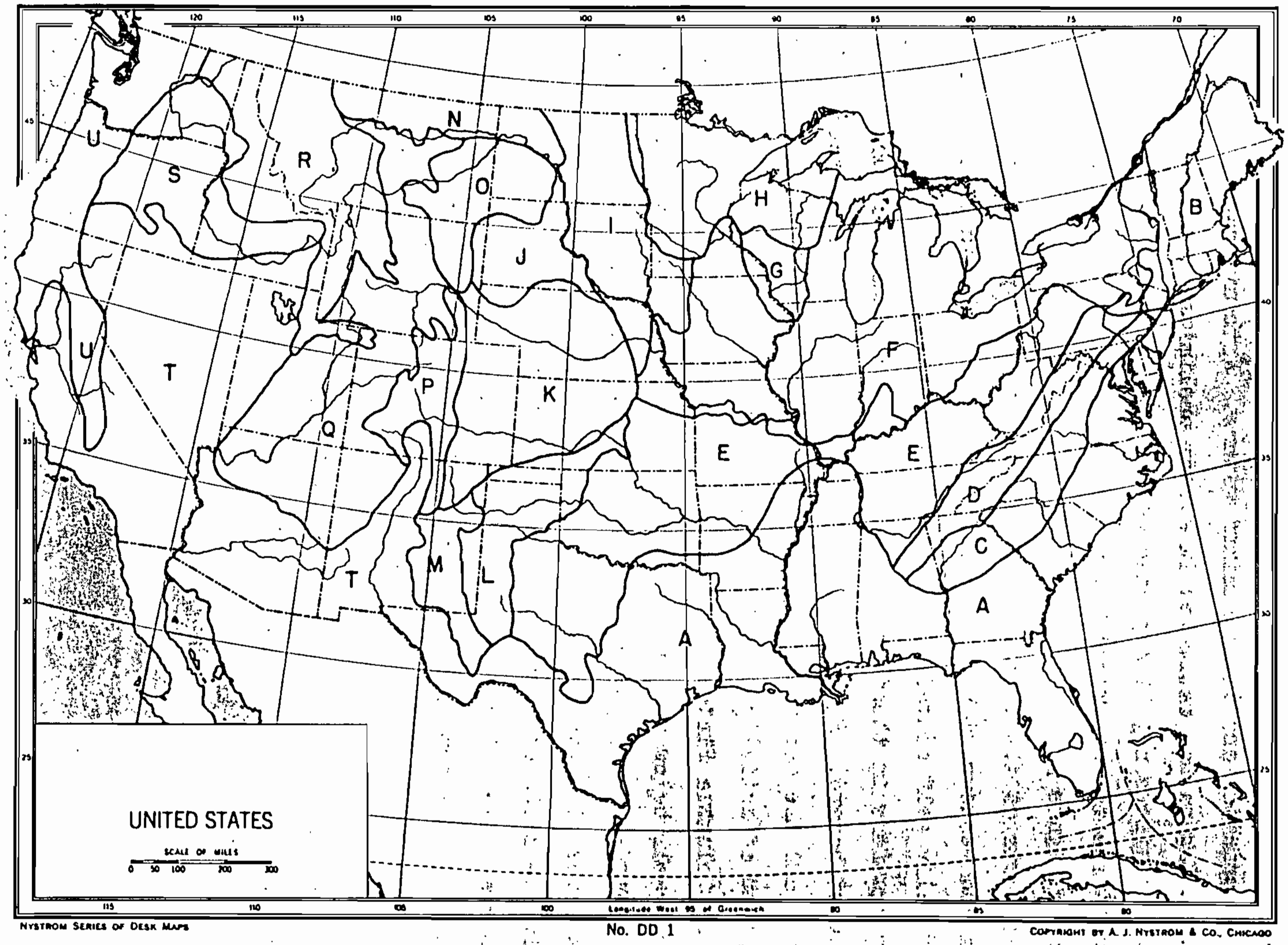
5) High porosity and coarse texture are in general desirable, but are secondary considerations.

6) A simple mineral composition is desirable. Assuming a dominately quartz sand, iron oxide, clay and glanconite are likely to be annoying adsorbents; sulfate, and to a lesser extent carbonates, may promote undesirable precipitation; chlorides are no problem. The so-called heavy minerals and fresh feldspar are not likely to cause trouble.

7) A series of individually thin permeable beds separated by less permeable material, rather than a single thick aquifer, may serve to reduce the problem of dissipation of heat.

Site Location

The choice of the most suitable location for injection of radioactive wastes will be based principally on two geological considerations. First, the location will be restricted to those areas where large, permeable aquifers, geologically isolated, might be expected to exist. Second, the regional hydrology of the area must be such that the hazards of inadvertent contamination of the ground water would be minimal. If two or more areas meeting the above requirements to an equal extent are found, it is possible that economic considerations can determine the ultimate choice. However, recent studies by Zeitlin, Arnold and Ullmann(25) and by Wolff and Rekemeyer(26) have shown that the optimum costs of shipping irradiated fuels from reactors to a single



GROUND-WATER PROVINCES

A	Atlantic Coastal Plain province
B	Northeastern Drift province
C	Piedmont province
D	Blue Ridge-Appalachian Valley province
E	South-Central Paleozoic province
F	North-Central Drift-Paleozoic province
G	Wisconsin Paleozoic province
H	Superior Drift-Crystalline province
I	Dakota Drift-Cretaceous province
J	Black Hills Cretaceous province
K	Great Plains Pliocene-Cretaceous province
L	Great Plains Pliocene-Paleozoic province
M	Trans-Pecos Paleozoic province
N	Northwestern Drift-Eocene-Cretaceous province
O	Montana Eocene-Cretaceous province
P	Southern Rocky Mountain province
Q	Montana-Arizona Plateau province
R	Northern Rocky Mountain province
S	Columbia Plateau lava province
T	Southwestern Bolson province
U	Pacific Mountain province

Fig. II.

processing plant and shipping wastes from the plant to an ultimate disposal site are not greatly affected by the relative locations of the plant and disposal site.

The search for an area possessing the characteristics desired for deep well disposal must be based initially on a very thorough study of pertinent geological information already in existence. The information acquired from ground water surveys would be expecially relevent while the knowledge of deeper formations possessed by the petroleum industry would be equally vital. After a general area has been chosen, detailed seismographic exploration will be required, followed by experimental drilling, sampling, and monitoring of the proposed storage formation and its contained waters. Without resort to a detailed study, de Laguna has estimated in a very preliminary and general fashion where suitable aquifers may be expected to exist. In describing these areas, reference was made to the ground water provinces defined by Meinzer(27) and illustrated in Figure 2.

In parts of the Southwestern Balson province, particularly in much of Nevada and western Utah, there exist many intermountain basins which are hydrologically self-contained. There is a possibility of finding deep, permeable aquifers in these areas where injection could be accomplished with a minimum of hazard. Probable disadvantages are the limited extent of these aquifers and the occurrence of clay and weathered rock which would promote adsorption of activity near the wells.

The Columbia Plateau lava ground-water province may possibly contain deep aquifers well below the main drainage of the area. It would be expected that these aquifers would possess very high permeability and low ion exchange properties. One disadvantage would be that the rock is very hard and would, consequently, be difficult and expensive to drill.

Large scale brine disposal by deep well injection is currently being practiced by the petroleum industry in parts of Kansas, Nebraska and Texas. These areas lie in the Great Plains Pliocene-Cretaceous, Great Plains Pliocene Palcozoic, and the South-Central Palcozoic provinces.

Advantages of using some of these same aquifers for radioactive waste disposal would accrue from close association with a well established practice and from the detailed knowledge that exists of the local geology.

In the South-Central Palcozoic and North-Central Drift-Palcozoic provinces there are large, deep aquifers containing highly mineralized water. For purposes of disposal these aquifers possess the advantages of simple, uniform structure and hydrologic properties and they are relatively well defined geologically. Over wide areas, however, these formations contain fresh water which would have to be maintained safe from contamination.

Aquifers in the Atlantic Coastal Plain Province are coarse beds composed of sand or sand and gravel. It is conceivable that deep aquifers covered with beds of low permeability and containing stagnant salt water could be used for radioactive waste disposal. There would have to be assurance, however, that overlying aquifers or landward extension of the injection aquifer would not be useable for water production.

8.54 Storage in Dry Mines or Caves

It has been suggested that abandoned mines or caves could be used for storage of high-level radioactive wastes. To ensure adequate containment and minimize the hazards, such areas would necessarily be restricted to storage of solid or packaged wastes. An additional requisite for safe storage would be the absence of water or moisture since leachability of activity from solids and corrosion of container materials by water could be serious problems over periods of centuries.

Although there have been reported instances of mines which were dry, particularly at great depths, the consensus is that the vast majority of mines and caves are quite the opposite. The possibility of finding a suitable area within a reasonable distance of a likely chemical processing site seems remote.

8.55 Surface Disposal of Liquid Wastes

In the processing of irradiated reactor fuels, large volumes of liquid wastes are produced which, while not containing the bulk of the fission products, are nevertheless of sufficient toxicity to preclude their release to the environment. Because of their dilution, the expense of concentrating and storing these wastes in underground tanks would be very formidable. The practice has been at both Hanford and Oak Ridge to utilize the absorptive and ion exchange properties of the local soils for the purposes of disposal.

Brown et al(24) have described the ground disposal of radioactive wastes at Hanford where the wastes are discharged into gravel-filled pits, or cribs, and allowed to seep into the soil. The soil has an exchange capacity of about 0.05 milliequivalents per gram and is used to retain the radioisotopes above the ground water table which lies between 300 and 400 feet below the surface. Monitoring wells are used to determine the presence and extent of the radionuclides in the soil and when trace contamination is detected in the ground water, use of the affected crib is discontinued and operation of a new facility is initiated. Laboratory and field studies have determined the soil capacity under various conditions of waste acidity and salt content for the most important constituents of the wastes and it has been found that, of these, plutonium is most strongly adsorbed, followed, in decreasing order of affinity, by the rare earths, strontium, cesium, ruthenium, and nitrate.

Ground disposal at Oak Ridge has been summarized by Struxness et al.(25) Three, one million gallon surface pits, obelisk in shape, have been used to dispose of 4.2 million gallons of waste containing 50,000 curies of Cs^{137} and 12,000 curies of Ru^{106} through June, 1956. Unlike Hanford, the ground-water table at Oak Ridge is very near the surface and reliance is placed on the Conasauga shale formations of that region to retain the radioactive species. This shale has an exchange capacity of about 0.25 milliequivalents per gram, is of reasonably uniform, although low permeability, and has been found to retain all the radionuclides in the wastes to a high degree with the exception of ruthenium. The ruthenium, together with the nitrate which is also not retained by the shale, eventually finds its way into the ground waters of the area where it is diluted to acceptably safe levels.

Since the choice of Hanford and Oak Ridge as sites for radiochemical processing was not predicated on the suitability of those areas for ground disposal of radioactive wastes, it is largely due to good fortune and careful handling and monitoring techniques that disposal operations of this nature have been possible. Every potential site must be evaluated in the light of local problems. Brown et al have outlined the most important factors to be considered in determining the feasibility of ground disposal. They include:

1) The chemical and radiochemical content of the waste.
2) The effectiveness of retention of the radioisotopes in the available soil column above the ground water table.

3) The degree of permanence of such retention, as influenced by subsequent diffusion, leaching by natural forces, and additional liquid disposal.

4) The natural rate and direction of movement of the ground water from the disposal site to public waterways, and possible changes in these characteristics from the over-all liquid disposal practices.

5) Feasibility of control of access to ground water in the affected region.

6) Additional retention, if any, on sands and gravels in the expected ground water travel pattern.

7) Dilution of the ground water upon entering public waters.

8) Maximum permissible concentrations in public waters of the radioelements concerned.

The basic disadvantages of ground disposal are concerned primarily with the hazard of disposing of dangerous products in a manner that leaves them in unrecoverable form, yet does not fix them in a permanent sense with assurance that they can never become dispersed in the environment. Furthermore, these operations carried out on a continuing or expanded scale would obviously render large surface areas uninhabitable for centuries.

9.0 Chemical Processes for Fission Product Concentration; Removal or Fixation

(very little done)

REFERENCES

1. W. S. Ginell, J. J. Martin, L. P. Hatch, "Ultimate Disposal of Radioactive Wastes", Nucleonius 12, No. 12, 14-18 (1954). See also, report BNL-1781, Feb., 1954.

2. L. P. Hatch, W. H. Regan, B. Manowitz, F. Hittman, "Processes for High Level Radioactive Waste Disposal", International Conference on the Peaceful Uses of Atomic Energy, 9, P/553, pp. 648-658, United Nations, New York, 1956.

3. J. M. White, G. Lahie, "Ultimate Fission Product Disposal. Disposal of Curie Quantities of Fission Products in Siliceous Materials", Report CRCE-591, March 1955. See also, entries under waste disposal or fission product disposal in progress reports PR-CM-1, PR-CM-2, PR-CM-3, PR-CM-4, PR-CM-5A, and PR-CM-6A, PR-CM-7A.

4. E. G. Struxness et al, "Ultimate Disposal by Self-Sintering at Elevated Temperatures", ORNL-CF-57-2-20, pp. 323-337 in "Compilation and Analysis of Waste Disposal Information", Feb. 11, 1957.

5. C. B. Amphlett, "Treatment of Highly Active Wastes", Atomics 8, No. 4, 116-120 (April, 1957). See also, Harwell Reports AERE CR-1862, AERE CR-1861, AERE CR-1686, and AERE CR-1294.

6. A. A. Jonke et al, "Fluidization Studies", in Chemical Engineering Summary Reports: ANL-5422, 5466, 5529.

7. E. S. Grimmett et al, "Treatment of Process Wastes by Calcination", Technical Progress Reports: IDO-14364, pp. 55-63 - 14385, pp. 97-102 - 14391, pp 30-40 - 14395, pp. 57-62.

8. B. Manowitz, F. Hittman, "Proposal for Waste Processing at Idaho Chemical Processing Plant", BNL-329, Feb. 1, 1955.

9. A. F. Rupp, "Fission Product Processing", Reactor Fuel Processing Costs: Papers presented at the Idaho Symposium, Jan. 18-20, 1956, IDO-14363 (Pt. III).

10. G. B. Barton, J. L. Hepworth, E. P. McClannahan, Jr., R. L. Moore, H. H. Van Tuyl, "Fission Product Recovery from Chemical Processing Plant Waste Solutions", Paper presented at American Chemical Society, Miami, Florida. April, 1957.

References (continued)

11. H. Seligman, H. J. Dunster, D. R. R. Fair, A. S. McLean, "The Discharge of Radioactive Waste Products into the Irish Sea", International Conference on the Peaceful Uses of Atomic Energy, 9, P/418, P/419, P/420, pp. 701-717, United Nations, New York, 1956.

12. C. E. Renn, "Disposal of Radioactive Wastes at Sea", International Conference on the Peaceful Uses of Atomic Energy, 9, P/569, pp. 718-721, United Nations, New York, 1956.

13. W. B. Heroy, "Disposal of Radioactive Waste in Salt Cavities", Report prepared for the Committee on the Disposal of Radioactive Waste Products in Geologic Structures; Subcommittee on Deep Disposal, National Research Council, Division of Earth Sciences, March 11, 1957.

14. "Salt Water Disposal, East Texas Field", prepared by the staff of East Texas Salt Water Disposal Company and issued by the Petroleum Extension Service, University of Texas, March, 1953.

15. W. de Laguna, "Disposal of Radioactive Wastes in Deep Wells", in "Compilation and Analysis of Waste Disposal Information", ORNL-CF-57-2-20, pp. 268-300. See also ORNL-2266, pp. 34-74, Feb. 11, 1957.

16. C. V. Theis, "Problems of Ground Disposal of Nuclear Wastes", International Conference on the Peaceful Uses of Atomic Energy, Vol. 9, P/564, pp. 679-683, United Nations, New York, 1956.

17. E. Roedder, "Atomic Waste Disposal by Injection into Aquifers", Paper 57-NESC-11, presented at Nuclear Engineering and Science Conference, Philadelphia, Pa., March 11-15, 1957.

18. W. J. Kaufman, R. G. Orcutt, G. Klein, "Underground Movement of Radioactive Wastes", AECU-3115, 1955.

19. D. A. Pecsock, "Disposal of Nuclear Power Reactor Wastes by Injection into Deep Wells", ORNL-CF-54-10-64, October 6, 1954.

20. F. Birch et al, "Handbook of Physical Constants", Geol. Soc. Am. Special Papers No. 36, pp. 251-258, 1942.

21. H. R. Zeitlin, E. D. Arnold, J. W. Ullmann, "Economics of Waste Disposal", Nucleonics 15, No. 1, 58-62, Jan., 1957.

References (continued)

22. R. W. Wolff, P. C. Rekemeyer, "Economics of Waste vs. Spent Fuel Shipping", MIT Engineering Practice School, Memorandum EPS-X-295, Jan. 18, 1957.

23. O. E. Meinzer, "The Occurrence of Ground-Water in the United States with a Discussion of Principles", U.S.G.S. Water-Supply, Paper 489, Washington, 1923, p. 309.

24. R. E. Brown, H. M. Parker, J. M. Smith, "Disposal of Liquid Wastes to the Ground", International Conference on the Peaceful Uses of Atomic Energy 9, P/565, pp. 669-675, United Nations, New York, 1956.

25. E. G. Struxness, R. J. Morton, C. P. Straub, K. E. Cowser, "Disposal of Intermediate Level Radioactive Liquid Wastes in Terrestrial Pits", pp. 145-159 in "Compilation and Analysis of Waste Disposal Information", ORNL-CF-57-2-20, Feb. 11, 1957, see also Geneva Conference Paper P/554.

10.0 Economic Considerations and Data

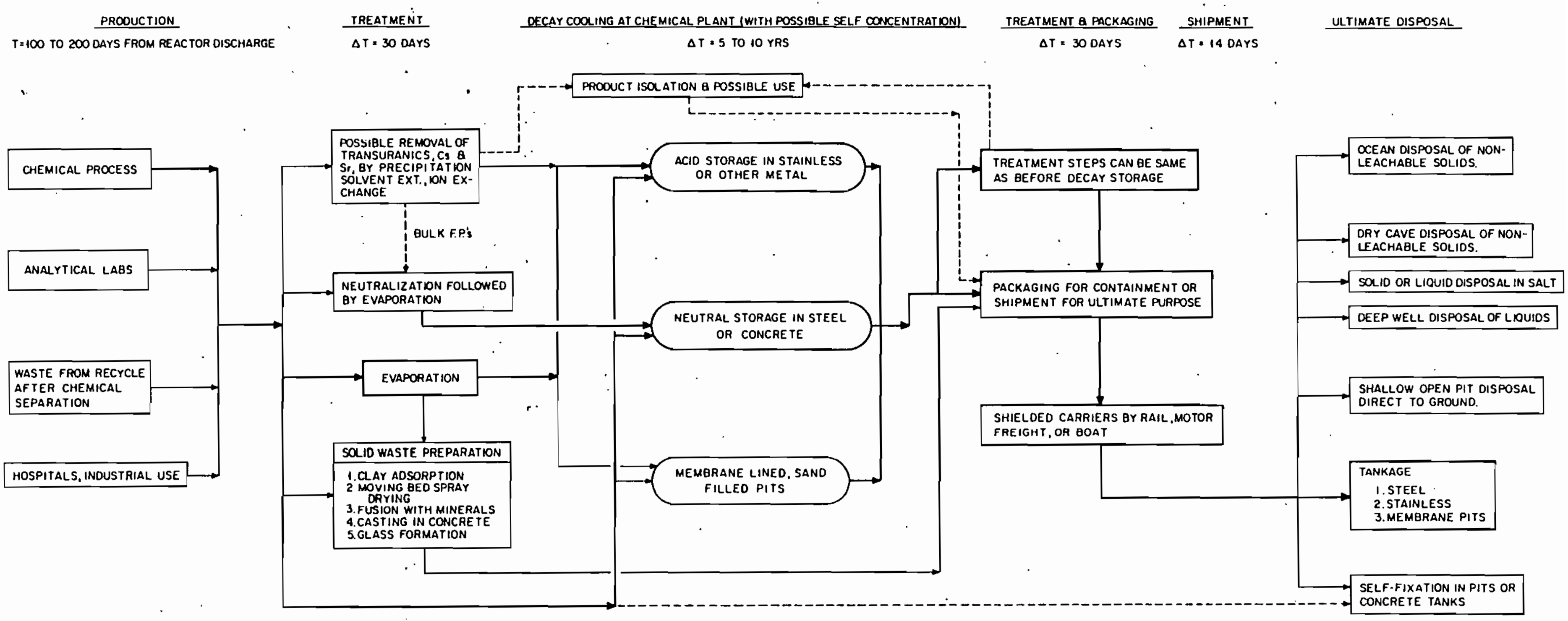
10.1 Rough Estimate of Allowable Costs of Waste Disposal

It is impossible to predict the exact chemical procedures and steps that will lead to safe radioactive waste disposal; equally uncertain is the choice of the nature of the ultimate disposal container and environment. However, it is possible to define in a more or less general fashion the steps that will lead to ultimate disposal of waste, and to suggest possible means of accomplishing each of the generalized steps, basing the suggestions on experience, development work now in progress, or in opinion. Having done this much, it is then possible to place costs on the better understood stages of the general scheme, to thereby determine how much might remain for steps as yet undeveloped.

A generalized scheme for waste disposal flowsheet is given in Figure 1, which we shall use as a guide for collecting costs. Costs have been accumulated or estimated for certain steps in this overall waste disposal scheme. However, the costs have not been made on any consistent basis, nor have the important economic effects of plant capacity and many other variables been considered. Costs in this report may best serve as a general guide as to what can be expected. More thorough cost studies will be required as development progresses.

The assumptions that the economy of the United States require are: 1) the production of electricity at 8 mills/kwh; 2) that the overall cost of fuel recycle cannot exceed 1 mill/kwh (and probably 0.75 mill/kwh) of electricity; 3) that reactors operate with an average of 25% thermal efficiency; and 4) that total waste costs, through final disposal, cannot exceed ten per cent of the recycle cost (or 1% of the total cost of electricity), establish a rough guide to allowable costs for waste operations and disposal. Since many costs have been reported as costs per gallon of wastes, Zeitlin(2) has prepared a set of "conversion" charts incorporating the variables of fuel burnup, gallons of waste per ton of uranium processed and allowable cost, one of which is given in Figure 2. The shaded area of this curve represents the probable liquid waste volume produced

UNCLASSIFIED
ORNL-LR-DWG 13943-A



HEAVY LINES INDICATE ROUTE AGAINST WHICH COSTS MUST BE COMPARED.
TIMES GIVEN ARE TIME FROM REACTOR SHUTDOWN & ARE ADDITIVE THRU THE DISPOSAL CYCLE.
BROKEN LINES INDICATE ALTERNATE ROUTES.

Fig. 1 Schematic Flowsheet For Overall Waste Disposal.

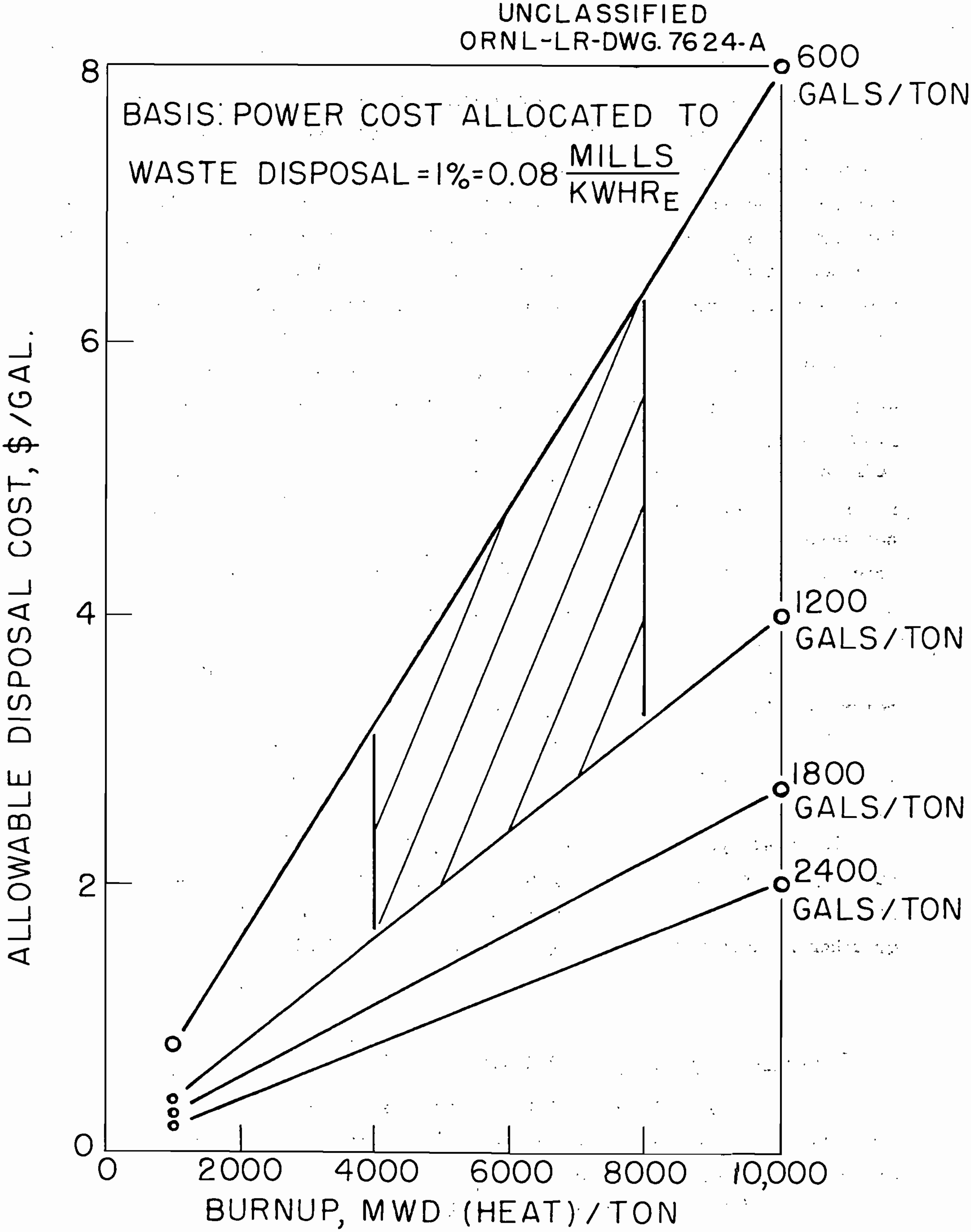


Fig. 2. Allowable Waste Disposal Cost as a Function of Burnup and Process Waste Volume.

per ton of natural uranium (or equivalent) processed. At 4000 Mwd/t burnup and a waste production of 800 gallons per ton of uranium, an 8 mills/kwh economy could support an approximate waste cost of $2.50 per gallon of high level wastes. We suggest that this number be used to roughly measure the economic advisability of steps suggested for waste disposal, recognizing that cost of all of the steps shown in Figure 1 must be covered assuming that no supplementary income is obtained from the irradiation potential in the wastes.

Both the activity level of radioactive wastes and their physical form affect costs of processing, packaging, shipping, storage and ultimate disposal. Activity levels of liquid wastes now produced as solvent extraction raffinates from irradiated fuel processing can be as high as 1000 curies per gallon (proposed power reactor fuels may be higher) to a few millicuries per gallon. Radioactive solids can be pure or almost pure fission product concentrates (example: Carrier-free Cs^{137} with specific activity of approximately 500 curies per gram) or very slightly contaminated solids for laboratory tracer level studies.

The type of radiation also contributes to the cost. High energy alpha emitters with long biological half-lives, such as plutonium, require special care which will increase the costs of handling. Alpha and beta emitters can be handled without heavy shields, but gamma emitters require shielding supplementary to the container itself.

The costs for quite a variety of operations for all types of contaminated solid or liquid wastes have been collected from a large number of sources. Frequently a specification of activity level, plant capacity, or other factors pertinent to the cost was not available.

10.2 Costs of Evaporation of Radioactive Wastes(4)

Costs of evaporation of radioactive wastes are summarized in Table I, annotated along with activity levels of feeds, capacity of units involved and other pertinent information.

We should point out that high level wastes release sufficient fission product heat to self-concentrate in the storage vessels. Since reflux condensers are provided for most high level waste tanks, this self-concentration,

Amortization:
10 years for building
5 " " equipment
300 days per year

Table 1

SUMMARY OF SELECTED RADIOACTIVE WASTE EVAPORATION COSTS

Unit	Estimate Date or Year Built	Approximate Activity Level	Nominal Capacity	Installation Cost $	Operating Costs $/gal	Approximate Total $/gallon cost Including Amortization	Reference
Oak Ridge National Laboratory	1949	10^6-10^8 d/min/ml 2 years or older	300 gph as condensate	45,000 building 45,000 equipment	0.054	0.06	ORNL - 1513
Idaho Chemical Plant	1952	10^8-10^9 d/min/ml 120 days cooled	350 gph as condensate	450,000 equipment 300,600 building	0.149	0.179	by W. G. Stockdale
Estimate by Mound Lab for High Level Solvent Extraction Raffinates	1952	a. Highly salted full level feed	a. 100 gph (as feed)	a. 200,000 building 200,000 equipment	a. @ 20:1 = 0.169	0.253	Mound Lab MLM-672(1)
		b. " "	b. 1000 gph (as feed)	b. 800,000 building 800,000 equipment	b. @ 20:1 = 0.037	0.069	" " " "
Westinghouse Atomic Power Division Low Level Concentration		Very low	1,600,000 gal/yr$^+$ as condensate	43,000 building 71,100 equipment	0.023	0.035	NYO - 7830(3)
Brookhaven Low Level Evaporator	1952	Low level	367,000 gal$^+$ per year as feed	92,900 building 204,400 equipment	@ 130:1 = 0.033	0.17	Same(3)
Knolls Atomic Power Laboratory Low Level Wastes	1950	Low level	4,500 gal/day$^+$ as feed	324,000 building 570,000 equipment	@ 200:1 = 0.025	0.156	Same(3)

$^+$Actual processing rate rather than nominal capacity taking advantage of the "free" heat source, can be accomplished with essentially no extra costs. It has been conservatively estimated that neutralized Purex type wastes can be concentrated by factors of 4:1 to 6:1. Condensate can be sent to further purification if necessary. In practice it is dumped to the ground into low level "cribs" under controlled and closely monitored conditions. A brief description of each of the evaporators follows.

10.21 Oak Ridge National Laboratory Liquid Waste Evaporator System

Capacity: Design, 300 gallons per hour

General Description:

The waste evaporator consists of a shielded pot evaporator with feed tank, an entrainment, separator, condensers, and a condensate tank housed in a one cell concrete-concrete block structure. Feeds vary from 10^6 to 10^9 disintegrations/min/ml.

References: ORNL-393, "Design and Initial Operation of the Radiochemical Waste Evaporator".

ORNL-1513, ORNL Radiochemical Waste Evaporator Performance Evaluation - December 1949 through December 1950.

10.22 Idaho Chemical Processing Plant Low Level Waste Evaporator System

Capacity: Design, 350 gallons per hour

General Description:

The evaporator for concentrating dilute radioactive stream is a thermal cycling type with the thermal-leg or steam chest external. The evaporator is equipped with pneumatically operated density and liquid level indicator recorders and temperature indicators for both the liquid and vapor. The evaporator pressure is controllable between atmospheric and 22 inches of Hg vacuum. Suitable entrainment separator, condenser, and condensate receivers are installed.

References: IDO-14334, "Experience of Handling Low Level Active Liquid Wastes at the Idaho Chemical Processing Plant".

ORNL-1792, "A Cost Analysis of the Idaho Chemical Processing Plant".

10.23 Mound Laboratory Estimate of Cost for High Level Waste Evaporation at 100 gph Hot Feed Capacity

Capacity: 720,000* gallons salted feed/year

Amortization of building: 10 year period

Amortization of equipment: 5 year period

(BASIS: 100 gph - 24 hours/day - 300 days/year)

10.24 Mound Laboratory Estimate of High Level Waste Evaporation at 1000 gph Hot Feed Capacity

Amortization of building: 10 year period

Amortization of equipment: 5 year period

(BASIS: 1000 gph - 24 hours/day - 300 days/year)

7,200,000 gals salted feed per year

10.3 Waste Tank Costs(4)

Data on costs of tankage for the storage of high and low level wastes is available. Table 2 summarizes waste tank costs; details of waste tank system costs follow:

10.31 Site A. High Level Waste Storage Tank Costs

Construction Period - 1952-54

Tank Capacity: 600,000 gallons per tank

General Construction:

Flat roof, carbon steel, 75-foot-diameter tank, encased in concrete. Roof supported by eight 2'-0" O.D. seamless pipe columns filled with concrete. Earth cover 9'-0". Ground water to top of concrete.

Cost:

Figures listed in table are based on material, labor and distribution (wage increases, administration charges, overhead, etc.).

Number of tanks	8	8	8
Coöling Coils	4 Yes, 4 No	No	Yes
Vent. Condenser and Filter	4 Yes, 4 No	Yes	Yes

Table 2

CAPITAL COSTS OF RADIOACTIVE WASTE STORAGE TANKS

Installation at	Capacity gallons	Year Constructed	Material of Construction	Internal Cooling Provided	Reflux Condensers	Total Cost $	$ Cost per gallon capacity	Comments
Site A High Level Waste Tanks	600,000	1952-1954	Carbon steel	Yes	Yes	Avg. 1,080,000	1.80	Can handle neutralized wastes only
Site B High Level Waste Tanks	1,000,000	1954-1955	Carbon steel-Concrete	No	Yes	Avg. 403,000	0.40	Can handle neutralized wastes only
Site B High Level Waste Tank Estimates	1,000,000	Bids 1954	Carbon steel-Concrete	No	Yes	172,000	0.17	
Hanford Hot Semi Works Tank	30,000	1951-1952	Stainless steel liner	No		83,090	2.77	
I CPP High Level Waste Tanks	318,000	1951-1952	Stainless steel-Concrete	No	Yes	546,415	1.77	Can store acid raffinates
" " "	300,000	1954-1955	Stainless steel-Concrete	Yes	Yes	869,390	2.80	" " " "
I CPP High Level Special Interein Tanks	30,700	1954-1955	316 Stainless Steel	Yes	Yes	255,000	8.20	Interein storage of acid fluoride and sulfate wastes
Hope Pit for Intermediate Levels	5,000,000	1954	Asphalt lining in earth pit	No	No	700,000	0.14	Not in use; still being studied

10.32 Site B. Waste Storage Tank Costs

Construction Period - 1954-55

Tank Capacities: 1,000,000 gallons each - Total 6,000,000 gallons

General Description:

Tank farm consisting of six buried storage tanks of prestressed, reinforced concrete with 3/8-in. carbon steel liners with appurtenant control structures, connecting lines and pipe encasements. Control structures include diversion box, reinforced concrete ventilation building, and a control house of insulated metal siding on a conrcete foundation. Also included are a pump pit, waste cribs, and sampler pits. Chain link fence surrounds the tank farm and woven wire fence encloses the waste cribs. Tanks are of domed roof design, 75 feet i.d. by 34 feet high at the wall. Capacity 1,000,000 gallons each.

10.33 Site B. Waste Storage Tank Costs

Bids Received 1954

Tank Capacity: 1,000,000 gallons each

General Description: Bid on Work:

Work consists of 15 underground steel-lined reinforced concrete tanks having a gross capacity of approximately one million gallons each. These tanks are arranged in three rows of five tanks each. Tank bottoms and walls are lined on the interior with 3/8" steel plate; tank dome interior is not lined. The bottom of the base slabs average 50' below the natural ground level, and the domes of the tanks have an average of 8' of earth coverage. The tanks are a nominal 75' in diameter and are spaced on approximately 100' centers.

In addition to the above tanks, work inlcudes a diversion box with catch tank, concrete encasement with stainless steel tubing, etc.

Alternate bids which called for extending the 3/8" steel place liner to include the dome of the tanks were 12T higher, or a difference of approximately $300,000 in total project cost. The tanks would have the same effective capacity, wall height would be approximately 4-1/2' less, all other features of the work remain the same

10.34 Hanford Waste Storage Hot Semi-Works

Construction Period - 1951-52

Tank Capacity: 30,000 gallons

General Description:

A 30,000-gallon underground storage tank 20 feet i.d. by 14 feet 3 inches high. One-foot thick cone wall and roof. Lining 1/4-inch stainless steel.

10.35 Idaho Chemical Processing Plant Waste Storage

Construction Period - 1951-52

Tank Capacity: 318,000 gallons each

General Description:

Tank farm consists of two 318,000-gallon 347 stainless steel storage tanks, each 50 feet in diameter and 32 feet tall with umbrella roofs capable of supporting themselves without beams. The tanks are housed in two octagonal concrete enclosures designed to support 8 feet of earth cover.

Both tanks are equipped with liquid level and density recorders, multiple thermocouples, and pressure-vacuum relief valves. One tank is provided with two reflux condensers.

Reference: ORNL-1687

10.36 Idaho Chemical Processing Plant Waste Storage

Additions to Original Construction

Construction Period - 1954-55

Tank Capacity: 300,000 gallons each

General Description:

Addition to the tank farm consisted of three (3) stainless steel 300,000 gallon tanks (two equipped with cooling coils) erected on concrete pad, with precast concrete enclosures. Instrument control house is included in contract. The following features are included:

(a) Tie-ins to existing first and second cycle systems to permit by-passing the installed tanks (WM-180, WM-181) to fill the new tanks.

(b) Piping arrangements to permit adding future tanks with minimum of personnel exposure to radiation.

(c) Overflow between first cycle waste tanks (those equipped with cooling coils).

(d) Access means for portable pumping device in case of tank failure.

(e) Liquid level, temperature, and pressure recording instrumentation.

(f) Jets in tank enclosures.

(g) Vent system, with relief valves, to stack.

(h) Nine feet of earth cover.

The first cycle tanks have the following additional features:

(a) Cooling coils with 100% spare capacity and suitable valving for testing and removal of the system in case of failure.

(b) Recirculated water system consisting of heat interchangers surge tanks, and circulating pumps.

(c) Vent condensers for each tank.

(d) Suitable instrumentation.

Reference: IDO-24011

10.37 Idaho Chemical Processing Plant Waste Storage Costs

Construction Period - 1954-55

Tank Capacity: 30,700 gallons

General Description:

Four (4) temporary underground 316 stainless steel waste storage tanks (30,700 gallon capacity each), including cooling coils and four (4) condensers, set on concrete drainage pad. Auxiliaries consist of waste storage control house, instruments and instrument control panel, facilities for monitoring and sampling, process and utility piping, electrical equipment, and waste lines (750 feet) between process building and tank farm. Earth cover approximately 24 feet. (Concrete cradles approximately 35 feet below grade.) No concrete enclosures. Expected tank life (from corrosion) five years.

Reference: IDO-24011

10.4 Cost of Collection and Disposal of Low Level Liquid and Solid Wastes - and Some Notes on Current Practice

In 1955 A. B. Joseph, Johns Hopkins University, made a survey(5) of a number of atomic energy sites to determine practices being followed and the approximate costs of handling low level wastes in solid and liquid form. He divided the low level wastes into two very general catagories:

1) Wastes which require limited personnel exposure or shielding because of a gamma radiation rate of 2 roentgen per hour in air a short distance from their surface. This level can be achieved with a very few curies of mixed fission products. For example, the number of curies of mixed fission products, aged about one year, (which emit gamma energies of about 0.7 Mev) in a cubic foot of material that would give a dose rate of 2 r/hr at a distance of one foot in air is as follows:

Specific Gravity	Typical Material	No. Curies
1.0	Water	1.89
1.2	Compressed Wastes	1.97
2.5	Concrete	3.42

2) Wastes which give a dose rate of 0.05 r/hr at a distance of one foot in air. This dose rate can be reached at a distance of one foot in air from a one foot cube containing the following number of curies:

Specific Gravity	Typical Material	No. Curies
1.0	Water	0.047
1.2	Compressed Wastes	0.049
2.5	Concrete	0.085

We should point out that there is a great difference between the level of these wastes and those which are produced as raffinates from high level radiochemical reprocessing. Most of these wastes result from laboratory investigations and are sink drains, laundry wastes; possibly rinses or equipment which contained traces of radioactivity; solids such as kleenex, contaminated containers, etc.

Although the cost accumulations are not highly accurate nor the individual units discussed analysed on a common basis of comparison, the information summarized by Joseph represents an approximation of what present low level waste handling and disposal may cost. The summaries also indicate current practice for this type of waste at various AEC sites.

The following is taken from the summary of Joseph's report.

There is no established nationwide AEC policy concerning the disposal of radioactive wastes other than having those wastes which are returned to nature be below certain allowable limits of activity. The practice of releasing waste to the environment varies among all the installations. Some installations release curies of activity at their sites every year. Others release little or no activity at their sites, transferring their waste materials elsewhere for release instead. In general, those installations which have large areas of land release low level liquids and bury radioactive solids within their site boundaries. Necessarily they keep a continuous check to determine if any activity escapes and to measure effects of the released activity on the environment. Those installations which are relatively small in size package and transport the bulk of their waste radioactivity to the larger sites for disposition or to the sea coast for dumping into the oceans.

Waste Collection

The objective of waste collection is to gather the wastes into one or more places so that they may be: a) treated, b) packaged or c) policed and released in a safe and economical manner. Collection practices are rather uniform throughout the country. All of the installations take extra precautions with hazardous wastes, whether they be alpha emitters or beta-gamma emitters. Low level wastes which are relatively non-hazardous are handled with less restraint. Low level liquids are collected in regular sewer systems and solids are collected by crews using motorized carriers.

Liquid Wastes: Most liquid wastes are collected on site in sewers; some are handled in integral containers. Sewer systems vary in size, extent and method of construction. Wastes flowing in them are under more or less continual scrutiny as they pass through monitoring points. Usually monitoring points are located at the point of origin of the wastes, the point of treatment and the point of discharge to the environment. Collection in sewers is not a fool-proof method. Sewers have been known to leak causing a great deal of consternation about uncontrolled release of radioactivity to the environment. Sewers which carry high level wastes have built-in safeguards, such as a sewer within a sewer, which gives more positive control over the wastes. Monitoring points, built-in safeguards and other special facilities and controls make the collection of liquid wastes more expensive than the municipality counterpart of sewer collection.

Solid Wastes: Almost all solid wastes are accumulated in laboratories and working areas in regular G.I. (galvanized iron) cans, cardboard cartons, kitchen style garbage cans and 30 gallon and 55 gallon steel drums. The cans usually have paper or polyethylene liners. There is no reason, as far as could be ascertained, for the wide variation in accumulation methods other than differences in opinions. The simplest procedure is that of collecting the wastes in the containers which are shipped. The next simplest appears to be the can method using 32 gallon G.I. cans lined with polyethylene bags. This method is low in cost, especially at those places purchasing the bags at a low unit cost.

Another problem of collection is that of transportation, i.e., physically moving the wastes from the point of origin to a central or convenient location for further treatment. This handling cost varies with the level of activity of the waste, the type of container used, the distance between point of origin and point of treatment and the kind of equipment used to make the transfer. The more spread out the installation, the more time spent on the vehicle and vice versa.

Refinements of the central storage area also directly affect the cost of collection (as considered in this report). Provisionsfor decay storage, usually underground, again follow different philosophies. Safety at low cost is desirable, but safety nevertheless. In this phase, philosophies of safety are measurable in relative dollars and cents because they are directly reflected in construction costs.

Waste Treatment

The primary reason for treating radioactive wastes is to reduce the volume in which the radioactivity is contained. It is less difficult to monitor and package small volumes than large volumes. The cost of reducing the volume must offset the cost of disposing the untreated volume minus the treated volume, otherwise, there is no economic basis for treatment. Each installation has made a study of its problems. The methods in use reflect the results of those studies.

Liquid Wastes: Liquid wastes in the atomic industry are treated by any of three methods: evaporation, co-precipitation with chemical coagulants and ion exchange. Each method has its special merits and special applications. Large volumes of more or less homogeneous liquids are effectively handled by co-precipitation. Heterogeneous wastes are more easily concentrated by evaporation processes. Ion exchange is used mostly for selective removal of certain isotopes.

Solid Wastes: Solid wastes are reduced in volume by one of two methods, namely, combustion or compression. Combustion gives the greater volume reduction but it also introduces the further problem of treating combustion gases which carry off some of the contaminating isotopes. Presently only one installation, WAPD at Bettis Field, uses incineration. There, the level of contamination of the wastes is rather low and does not present a very difficult gas problem. Actual results with the Bettis Field incinerator have not yet been made known.

Several installations compress their trash materials in baling machines to reduce the volume of wastes they must transport. Since common carrier charges are based on weight, the principal reason for baling is to reduce the packaging costs. An analysis of the economy of solid waste volume reduction is presented in Appendix 2.

Packaging for Disposal

Packaging for on-site burial is minimal. The wastes are just sufficiently restrained for expeditions and safe handling in collection and transfer to burial. Considerable preparation and packaging are involved in shipments off-site. The ultimate repository, i.e., on land or in the sea, determines the characteristics of the packages. Those packages of waste which are disposed of into the sea are made heavy with concrete to ensure sinking. Waste packages which are to be buried on land do not have the added concrete and consequently involve less shipment weight per volume of waste. Packages shipped off-site for both land and sea disposal are made tight so there will be no spillage in transit.

There are no regulations, per se, which specify packaging materials or methods of constructing packages. Interstate Commerce Commission regulations state that a package containing radioactive materials should be "tight" allowing no leakage from a package. ICC regulations do specify a limit of tolerable emitted radiation. These are based on the possible fogging of X-ray film which may be in transit. AEC shipments of radioactive materials are exempted from ICC regulations if a courier accompanies the shipments. All those installations utilizing common carriers have reported that their shipments do comply with the ICC regulations.

Two kinds of packages predominate in waste shipments: steel drums and wooden boxes. Other packaging materials include fiber drums, fabricated steel boxes and poured concrete boxes. Steel drums and the concrete boxes contain most of the wastes that go to sea. All the other packaging materials together with steel drums contain shipments destined for land burial. Drums usually contain slurries and loose bulky materials and the other kind of packages contain trash and miscellaneous items of waste.

Waste Transportation

The mode of transportation to any disposal site is determined by convenience and economy. AEC contractor owned trucks are used in some cases but mostly the shipments are by the common carriers, both railroad and truck. Most of the wastes are packaged according to ICC specifications and these are transported without the accompaniment of a courier. A radiation surveyor does accompany those shipments whose radiation levels are above ICC tolerances; a few such shipments are made from Brookhaven, Berkeley and Livermore. To limit possible contamination and to facilitate decontamination most of the installations cover the floor and walls of the conveying vehicle with a protective layer of paper.

The wastes are loaded aboard the conveying vehicle by AEC contractor personnel and with one or two exceptions they are also unloaded by AEC contractor personnel. The exceptions are those cases in which the Navy unloads the wastes at dockside. The routes traveled by the waste carriers are most direct; public highways by the trucks and regular freight routes by the railroads. So far as is known all shipments have been made without any loss of life, limb or time for all individuals involved.

Disposition of Wastes on Land

Most of the waste materials that are shipped to land destinations are buried in the earth. (Some of the wastes sent to the Lake Ontario Storage Area are still stored above ground.) These wastes include low and intermediate level solids and liquids. The intermediate level liquid wastes are the residues of evaporation or concentration processes in the form of sludges and are more or less solid in physical form. Usually wastes that are buried are buried in their shipping containers. Radioactive wastes are buried by a method similar to a sanitary landfill operation. A trench or hole is first excavated in a geologically suitable area. The packages of wastes, those shipped cross-country and those collected locally, are dumped into the excavated hole and covered with dirt. Some installations backfill by alternating layers of dirt and wastes and others use only one cover layer of dirt on top. The excavations range from 10 to 20 feet in depth. Dirt cover is proportional to radioactivity, ranging from a minimum of 3 ft. for low level wastes to 6 or 8 ft. for high level wastes.

Decontaminated liquids and low level radioactive liquids are released to the environment at the installations. Some places release to nearby surface watercourses and others to underground strata by seepage from surface lagoons, pits, cribs or by discharge to reverse wells. All the installations have established certain tolerance levels for the release of radioactive material to the environment. Area monitoring programs at all of the installations releasing materials are pursued to determine the effects, if any, on the environment.

None of the installations release high level liquid wastes to the environment. These particularly hazardous wastes are confined within specially constructed, underground storage tanks.

Sea Disposal

On the West Coast, the San Francisco Navy Shipyard (Hunters Point) is the home port of the YGN-73, a modified dump scow whose mission is to carry out and dump radioactive wastes at sea. The Naval Radiological Defense Laboratory at Hunters Point co-ordinates truck shipments from the Berkeley and Livermore sites of the University of California Radiation Laboratory. Wastes from these sites and from the USNRDL constitute the bulk of the matter taken out to sea and dumped in a designated area about 20 miles off San Francisco where the water is at least 500 fathoms (3,000 ft.) deep.

On the East Coast the Navy operates a disposal service for defective ammunition. An LST puts in at selected ports along the Atlantic Coast to pick up unwanted ammunition and haul it out to sea. Co-operating with the AEC the Navy also accepts packaged radioactive wastes and disposes of it along with the defunct ammunition. The designated dumping areas in the Atlantic are at the edge of the continental shelf about 100 miles from the mainland where the water is at least 1000 fathoms deep.

Both in the East and in the West, most of the waste material is packaged in 55 gallon drums. Unique to the West Coast operation are some large (6' x 6' x 12' max) concrete disposal units which contain contaminated isolation (glove) boxes and other bulky items.

Conclusions Regarding Liquid Wastes Disposal Costs

On the basis of available data it is difficult to make equitable comparisons of the costs of radioactive liquid waste disposal among the installations surveyed. Table 3 summarizes the principal items which make up these costs. To make a comparison one has to calculate a unit cost -in the case of liquids say, cost per gallon. The column headed "Collected after Monitoring" includes all of the wastes in some cases and only part of the waste in others. As a divisor in determining unit costs it is not equitable because it vaires with the different kinds of wastes. The largest item of collection cost, namely the amortization cost of the collection systems is lacking. In most cases it was not available. In others many years of installation records would have had to be scrutinized and summarized. The other possible index of "total volume", the column "Effluent Discharged" also has an element of unreliability. Unit costs computed from either one of these indices could be made lower by diluting the waste stream with water, possibly storm water, and thus increasing volume.

The costs of liquid waste treatment and waste concentrate disposition operations are summarized in Table 4. The reader is cautioned against comparing these costs. The basis for each item is not the same.

Conclusions Regarding Solid Waste Disposal Costs

Tables 5 and 6 summarize, by installations, the costs of the various phases of handling low level and high level wastes. For preparing these

Installation	Collected after Monitoring	Transferred in Containers	Radioisotope Concentration Treatment				Effluent Discharged		Stored in Underground Tanks
			Evaporated	Chemically Precipitated	Ion Exchanged	Concentrate Produced	To Surface	To Subsurface (1)	
Argonne Nat. Lab.	5,732 (2)	176	66	25	1.9		46,400	500	none
Bettis Field (WAPD)	1,679 (3)	79	1,600	none	none	3.96	137,000	none	none
Brookhaven Nat. L.	3,267 (4)	12	367	none	none	3.67	120,000	none	none
Fernald (FMPC)				none	none		94,000	none	none
Knolls At. Pow. Lab.	1,130	5	1,125	none	none	4.9	126,000	none	42
Los Alamos	13,900	none	none	13,900	none	33.2	13,900	none	none
NRTS (CPP) (5)	671	none	252	none	none	3.56	none	671	3.56
Oak Ridge (ORNL)	1,000	none	none	none	none	none	158,000	1,000	none
Rocky Flats	4,800	none	none	1,570	none	45.5	40,000	786	none

(1) Discharged either to a surface holding pond or directly to subsurface.
(2) Does not include high level wastes collected in "pots".
(3) Does not include wastes dumped to sewer system.
(4) The quantity monitored was between 367,000 and 120,000,000 gallons.
(5) The quantities listed are for a 6 month period. They would not be true for a 12 month period if doubled because waste production varied.

Table 3 Volumes of Liquid Wastes Handled (by stages) During Fiscal Year 1955
(Units in 1000's gallons)

Installation	Wastes Treated (1000 gal)	Gross Cost of Concentration	Concentration Cost Per Gallon	Operating Cost(1)	Operating Cost Per Gallon	Concentrate Produced (gallons)	Concentrate Packaging Cost	Concentrate Packaging Cost Per Gallon	Concentrate Shipping Cost(2)	Cost of Underground Storage Facilities(3)	Summary Cost = Concentrating Packaging Shipping &/or Storing Costs	Unit Summary Cost (per gal.)
NO TREATMENT GIVEN												
Oak Ridge (ORNL)	1,000	N.A.	N.A.	N.A.	N.A.	N.A.	N.A.	N.A.	N.A.	N.A.	$ 85,400	$ 0.085
BY EVAPORATION												
Argonne Nat. Lab.	66	$	$	$	$		$	$	$	$	$	$
Bettis Field (WAPD)	1,600	56,060	0.04	37,510	0.023	3,960	3,110	0.79	3,150	N.A.	62,320	0.039
Brookhaven Nat. L.	367	62,400	0.17	12,230	0.033	3,670	770	0.21	830	N.A.	64,000	0.174
Knolls At. Pow. Lab.	1,125	60,900	0.05	8,500	0.025	4,900	860	0.36(4)	610(5)	2,400	64,770	0.057
NRTS (CPP)(6)	252					3,565	N.A.	N.A.	N.A.	8,200		
BY EVAPORATION AND CALCINING												
Fernald (FMPC)		718,141		328,831			N.A.	N.A.	N.A.	(7)		
BY CHEMICAL PRECIPITATION AND VACUUM FILTRATION OF SLUDGE												
Argonne Nat. Lab.	25											
Los Alamos	13,900	136,840	0.01	75,630	0.005	33,200	nil(8)	nil(8)	1,870(9)	N.A.	138,710	0.010
Rocky Flats	1,570	126,120	0.08	66,870	0.043	43,000	4,140	0.10	5,940	N.A.	136,200	0.087

N.A. signifies "not applicable".

(1) Total treatment cost minus fixed cost.

(2) Estimated as a proportion of the gross weight or volume shipped.

(3) Construction unit cost of storage facilities multiplied by annual input. Does not include operational and maintenance costs.

(4) Only 2,400 gallons packaged.

(5) Theoretical cost of shipping 2,400 gallons to ORNL. (21,800 lbs at $2.81/cwt.)

(6) The quantities listed are for a 6 month period. They would not be true for a 12 month period if doubled because waste production varied.

(7) Construction unit cost is $1.61 per cu. ft.

(8) Used drums are obtained from other operations on the site.

(9) The cost of hauling and burying the sludge.

Table 4 Costs of Liquid Waste Treatment and Waste Concentrate Disposition Operations
Fiscal Year 1955

Installation	Waste Collection			Waste Concentration					Waste Packaging			Shipping and Terminal Handling Data					Overhead	Total Known Cost	Total Cost per cu. ft. Collected
	Volume (cu. ft.)	Cost	Unit Cost	Method	Volume in cu ft. Before	Volume in cu ft. After	Cost	Unit Cost(1)	Volume (cu. ft.)	Cost	Unit Cost	Destination	Overland Distance (miles)	Volume (cu. ft.)	Cost	Unit Cost			
Argonne Nat. Lab.	14,290	$ 18,530	$ 1.30	baling	10,000	2,580	$2,580	$ 614	6,660	$11,330	$1.70	Oak Ridge	530	6,660	$3,460	$0.52	$12,230	$48,270	$ 3.38
Bettis Field (WAPD)	18,000	3,760	0.21	incin'n	13,200	360	4,510	123	710	2,530	3.56	Earle, N. J.	370	710	3,440	4.84	3,320	17,560	0.97
Brookhaven Nat L.	16,140	6,520	0.40	baling	2,000	640	1,190	380	745	2,650	3.55	Floyd Bennett Field	70	745	1,030	1.38	6,740	18,130	1.12
Knolls At. Pow. Lab.	36,880	7,910	0.21	baling	20,000	3,300	5,170	861	20,950	17,840	0.85	Lake Ontario St. Area Oak Ridge	300 850	6,980 13,970	2,735 5,470	0.39 0.39	13,320	53,445	1.45
Los Alamos	62,400	32,030	0.51	none	N. A.	N. A.	N. A.	N. A.	N. A	N. A.	N. A.	Site burial	-	62,400	8,970	0.14	14,960	55,960	0.90
NRTS, Idaho	27,000	6,280	0.23	none	N. A.	N. A.	N. A.	N. A	N. A.	N. A.	N. A	Site burial	-	27,230	9,900	0.36	4,620	20,800	0.77
Oak Ridge	116,400(2)	38,440	0.33	none	N. A.	N. A	N. A.	N. A.	N. A.	N. A.	N. A.	Site burial	-	116,400	25,830	0.22	28,900	93,170	0.80
Rocky Flats	N. A.	N. A.	N. A.	none	N. A.	N. A.	N. A.	N. A.	33,900	15,380(3)	0.45	NRTS, Idaho	700	33,900	35,390	1.04	2,700	53,470	1.58
U. of Cal. Rad. Lab Berkeley	6,200	3,610	0.58	none	N. A.	N. A.	N. A.	N. A.	6,180	11,330	1.83	San Francisco	20	6,180	4,850	0.77	11,290	31,070	5.01
Livermore	4,260	3,520	0.83	none	N. A.	N. A.	N. A.	N. A.	4,160	8,640	2.08	San Francisco	50	4,160	3,550	0.86	8,910	24,620	5.78

N. A. signifies "not applicable"

(1) Concentration unit cost measured in dollars per unit of concentration.

(2) For 12 months based on 6 months of record.

(3) Materials only.

Table 5 Costs of Very Low Level Solid Wastes (0.05 r/hr) Disposal Practices

Fiscal Year 1955

Installation	Collection (and Storage)			Packaging			Shipping and Terminal Handling Data				Capital Investment Depreciat'n Charged (1)	Overhead	Total Known Cost (3)	Total Cost per cu. ft. Collected
	Volume (cu. ft.)	Cost	Unit Cost	Volume (cu. ft.)	Cost	Unit Cost	Destination	Volume (cu. ft.)	Cost	Unit Cost				
Argonne Nat. Lab.	391	$10,440(2)	$ 26.70	N.A.	N.A.	N.A.	Site storage	N.A.	N.A.	N.A.	$ 10,260	$ 100	$10,540	$ 27
Bettis Field (WAPD)	26.8	900	33.60	26.8	$23,930	$ 893	Earle, N.J.	26.8	$10,600	$392	none(4)	3,510	38,940	1,450
Brookhaven Nat. L.	462	3,600(2)	7.80	462	4,200	9	Floyd Bennett Field	462	1,620	3.52	1,420	4,870	14,290	30
Knolls At Pow. Lab.	5.6	2,900(2)	518.00	N.A.	N.A	N.A.	Site storage	N.A.	N.A.	N.A.	1,950	300	3,200	572
NRTS, Idaho	230	600	2.61	N.A.	N.A.	N.A.	Site burial	230	80	0.36	none(4)	375	1,055	4.58

N.A. signifies "not applicable".

(1) Cost of maintaining and guarding high level wastes in storage represents an additional cost not included.

(2) Includes amortization of construction cost of storage facilities.

(3) Sum of collection, packaging, shipping and overhead costs.

(4) Some of the depreciation charged to low level wastes practices could be charged here.

Table 6 Costs of Low Level Solid Wastes (2 r/hr) Practices

Fiscal Year 1955

tables an attempt was made to ascertain the total cost of disposal per cubic foot of waste. In some cases this was not possible because all the costs are not known. Cost data for those installations disposing of wastes to the oceans lack one important item, namely the true charges for the ship and crew which took the material out to sea. This service was rendered as any inter-governmental agency service. Another unknown cost item is that cost for removing high level wastes from storage or for maintaining and guarding the high level waste storage facilities in the years of storage. Even without these realistic cost items one can draw some conclusions regarding the costs of disposing of solid radioactive wastes.

10.5 Costs of Drum Drying Low Level Radioactive Wastes(6)

Drum drying of radioactive waste was tested at KAPL over a period of time between 1949 and 1952 with the following results and costs.

"Drying was discontinued because of the numerous troubles encountered with the dryers, the lack of any appreciable reduction in volume of the concentrated evaporator slurry by drum drying, and the additional expense.

The cost of drying in addition to the cost of evaporation based on processing 3,000,000 gallons of raw waste a year is estimated at 4 cents per gallon of raw waste. The drying rate obtained was 50 pounds of solids per hour with the larger of the two dryers, or 1.6 pounds per hour per square foot of drying surface. The design rate for this dryer was 75 pounds of solids per hour." Gamma activity of the waste processed ran as high as 1.7×10^3 - $1.7 \times 10^7 \mu$c per gallon.

TABLE 7

71,000 LBS OF SOLIDS PER YEAR

	Estimated Costs	Depreciation	Annual Costs
Building	$158,000	3%	$ 5,740
Equipment, Installed	221,680	20%	44,340
Operating Costs	--	--	74,940
TOTAL			$125,020

Cost per pound of waste dried is $1.76 per lb. of dried solids.
Cost per gallon of raw waste is $0.04 per gallon.

Note:

Storage costs for slurry	$1.18/gallon (Drum costs)
Storage costs for dried powder	$1.43/gallon (Drum costs)

REFERENCES

1. MLM-672, "Reactor Waste Processing Cost Analysis", Monsanto Chemical Company, Mound Laboratory, March, 1952.
2. H. R. Zeitlin, "Economic Requirements for Radioactive Waste Disposal in a Nuclear Power Economy", ORNL CF 55-6-152.
3. Arnold B. Joseph, "Radioactive Waste Disposal Practices in the Atomic Energy Industry, A Survey of Costs", NYO-7830, Dec. 31, 1955, The Johns Hopkins University.
4. F. L. Culler, J. O. Blomeke, W. G. Stockdale, "Unit Costs and Economic Relationships for Certain Radioactive Waste Disposal Steps", ORNL CF 57-5-25, pp. 4-11, May 10, 1957.
5. Arnold B. Joseph, op. cit.
6. D. M. Lewis, "Drum Drying Radioactive Waste at the Knolls Atomic Power Laboratory", KAPL-720, April 8, 1952.

REFERENCES

1. National Academy of Science - National Research Council, "The Biological Effects of Atomic Radiation, A Report to the Public", 1956.

2. Ibid., Summary Reports, 1956.

3. Ibid., pp. 28-29.

4. H. F. Reichard et al., "Homogeneous Reactor Fuel Reprocessing Quarterly Progress Report", KLX-10034, April 1 - June 30, 1956, (also in previous or later Vitro Progress Reports on Homogeneous Reactor); additional methods in HW-20803 and HW-21103.

5. W. L. Carter, "Suggested Design of a Unit for the Removal of Iodine and Bromine from Arco Dissolver Off-Gas System, ORNL CF 51-3-27.

6. D. W. Moeller, "Theoretical Evaluation of Induced Radioactivities in Reactor Cooling Water", TID-7517 (Pt 1b), pp. 597-635, Unclassified.

7. K. Z. Morgan, "Maximum Permissible Internal Dose of Radionuclides: Recent Changes in Values", Nuc. Sci. and Engr. 1, No. 6, (Dec. 1956) pp. 477-500.

8. K. Z. Morgan, "Handbook of Physics, Health Physics", to be issued by National Research Council.

9. Federal Register (10 CFR, Part 20), "Standards for Protection Against Radiation" (proposed and still pending), July 15, 1955.

10. Reference 2 pp. 21-22

11. AEC Division of Reactor Development, Stack Gas Problem Working Group, "Fourth Atomic Energy Commission Air Cleaning Conference Held at Argonne National Laboratory, November 1955", TID-7513, (Part I unclassified, Part II confidential), June 1956.

12. J. B. H. Kuper et al., "Special Reactor Hazards Study", prepared at Brookhaven National Laboratory, December, 1956 (to be released).

13. E. D. Arnold, unpublished calculations and estimates, Oak Ridge National Laboratory.

14. P. L. Robertson, W. G. Stockdale, "A Cost Analysis of the Idaho Chemical Processing Plant", ORNL-1792 (Jan. 3, 1955), unclassified.

15. J. A. Lane, "Determining Nuclear Fuel Requirements for Large-Scale Industrial Power", Nucleonics 12(10), 65 (1954).

16. Report of the Panel on the Impact of the Peaceful Uses of Atomic Energy to the Joint Committee on Atomic Energy, U.S. Government Printing Office, Jan., 1956.

17. "Major Activities in the Atomic Energy Program", January - June 1955 and July - December 1955, U.S. Government Printing Office.

18. "Jane's Fighting Ships", McGraw-Hill Book Co., Inc., New York, 1953-1954 ed.

19. H. R. Zeitlin, E. D. Arnold, J. W. Ullmann, "Economics of Waste Disposal", Nucleonics, 15(1) 58-62 (1957). (formerly ORNL-CF-55-6-152, ORNL CF-55-10-101, and ORNL-CF-56-1-162).

20. J. O. Blomeke, Mary F. Todd, "U^{235} Fission Product Production as a Function of Thermal Neutron Flux, Irradiation Time and Decay Time", ORNL-2127 (in press).

21. E. D. Arnold, "Effect of Recycle of Uranium Through Reactor and Gaseous Diffusion Plant on Buildup of Important Transmutation Products in Irradiated Power Reactor Fuels", ORNL-2104 (August 21, 1956).

22. J. O. Blomeke, "The Buildup of Heavy Isotopes During Thermal Neutron Irradiation of Uranium Reactor Fuels", ORNL-2126 (Dec. 20, 1956).

23. E. D. Arnold, Oak Ridge National Laboratory, private communication, 1957.

24. Calculated and prepared by E. D. Arnold from ORNL-1982 by E. D. Arnold and A. T. Gresky, "Relative Biological Hazards of Radiation Expected in Homogeneous Reactors", (Nov. 15, 1955) (confidential).

25. K. Z. Morgan, M. R. Ford, "Developments in Internal Dose Determinations", Nucleonics, 12(6): 32-39 (1954) (revision of paper presented at the American Industrial Hygiene Conference, Los Angeles, April 23, 1953).

26. W. A. Rodger, "Waste Handling Methods and Costs in Solvent Extraction Processes", IDO-14363 (Part I) (April, 1956) (confidential).

27. K. Z. Morgan, Oak Ridge National Laboratory, private communication.

28. Prepared by E. D. Arnold, A. T. Gresky, ORNL, July 1956.

29. E. D. Arnold, A. T. Gresky, J.O. Blomeke, W. DeLaguna, E. D. Struxness, "Compilation and Analysis of Waste Disposal Information", ORNL-CF-57-2-20, February 11, 1957.

30. E. D. Arnold, ORNL, private communication, February 22, 1957.

31. A. T. Gresky, ORNL, private communication, 1957.

32. J. R. Dietrich, "Experimental Investigation of the Self-Limitation of Power During REactivity Transients in a Sub-cooled Water-Moderated Reactor - Borax-1 Experiments, 1954", ANL-5323.

33. W. B. Lewis, "The Accident to the NRX Reactor on Dec. 12, 1952", Reactor Sci. Technol. 3(4), 9-19 (1953) (TID-2011).

34. "EBR-1 Core: After Meltdown", Nucleonics 15 (1) 84,(1957).

35. J. W. Ullmann, "Effect of Plant Size on Costs of Processing Heterogeneous Power Reactor Fuel", ORNL-2020 (Dec. 18, 1955).

36. National Committee on Radiation Protection, Handbook 52, "Maximum Permissible Amounts of Radioisotopes in the Human Body and Maximum Permissible Concentrations in Air and Water", National Bureau of Standards, Washington, D. C.

37. Stuart McLain, R. O. Brittan, "Safety Features of Nuclear Reactors", Problems in Nuclear Engineering, Vol. I, pp. 1-10, Pergamon Press, 1957.

38. R. J. Creagan, "Safety Aspects of Water Cooled and Moderated Reactors", Problems in Nuclear Engineering, Vol. I, pp. 11-14, Pergamon Press, 1957.

39. W. J. McCarthy, Jr., F. C. McMath, "Safeguard Features of a Fast Breeder Reactor Power Plant", Problems in Nuclear Engineering, Vol. I, pp. 15-21, Pergamon Press, 1957.

40. C. R. McCullough, M. M. Mills, E. Teller, "The Safety of Nuclear Reactors", International Conference on the Peaceful Uses of Atomic Energy, 1955.

SUPPLEMENTARY BIBLIOGRAPHY

Introduction

1. "Radiological Health Handbook", compiled and edited by Simon Kinsman, Radiological Health Training Section, Sanitary Eng. Center, Ohio, October, 1955.

2. K. Z. Morgan, W. S. Snyder, M. R. Ford, "Maximum Permissible Concentrations of Radioisotopes in Air and Water for Short Period Exposure", P. 79.

3. H. M. Parker, "Radiation Exposure from Environmental Hazards, P. 279.

4. L. P. Hatch, "Ultimate Disposal of Radioactive Wastes", BNL-1345, Jan., 1953.

5. E. Glueckauf, "Long-term Aspect of Fission Product Disposal", p.3.

6. Abel Wolman, A. E. Gorman, "The Management and Disposal of Radioactive Waste", p. 9.

7. L. P. Hatch, "Ultimate Disposal of Radioactive Wastes", American Scientist, Vol. 41, No. 3, pp. 410-421, July, 1953.

8. A. B. Joseph, J. M. Morgan, "Radioactive Wastes in the Atomic Energy Industry", Mar. 31, 1955, AEC-Cont. No. AT(30-1) 1477. Also "Report of Meeting on Subterranean Disposal of Reactor Wastes", Nov. 15, 1954. "Report of Meeting on Ocean Disposal of Reactor Wastes", Aug. 5, 1954.

9. A. B. Joseph, "Radioactive Waste Disposal Practices in the Atomic Energy Industry, NYO-7830, Dec. 31, 1955.

10. Abel Wolman, A. E. Gorman, J. A. Lieberman, "Disposal of Radioactive Wastes in the U. S. Atomic Energy Program", WASH-408, May 17, 1956.

11. "Sanitary Engineering Aspects of the Atomic Energy Industry. A Seminar sponsored by the AEC and the Public Health Service. Held at the Robert A. Taft Eng. Center, Cincinnati, Ohio, Dec. 6-9, 1955". Technical Information Service Extension, Oak Ridge, Tenn. Parts Ia, Ib, unclassified. Part II, confidential. TID-7517.

12. P. F. Brown, "Minutes of ICPP Waste Disposal Conference", IDO-10030, Idaho Falls, Idaho, Aug. 22-23, 1955.

13. P. F. Brown, "Minutes of ICPP Waste Disposal Conference", IDO-10030, Idaho Falls, Idaho, Aug. 22-23, 1955, Confidential.

14. "The Reactor Handbook", Vol. 4, Fuel Processing, Technical Information Service, USAEC, Oak Ridge, Tenn, May, 1953, Secret.

15. "Uranium Recovery Technical Manual", HW-19140, Secret.

16. "Redox Technical Manual", HW-18700, Secret.

17. Du Pont Savannah River Technical Manual, DPSTM-200, Secret.

18. T. C. Runion, "Tributyl Phosphate as an Alternate Solvent for Extraction of "25" from Spent MTR Fuel Assemblies", ORNL-801, Sept. 26, 1950.

19. F. L. Culler, "The Nature and Magnitude of Radioactive Wastes as Influenced by Types of Reactors and Fuel Processing - Present and Prospective", ORNL CF 56-5-2.

20. "Laboratory Demonstration of Redox: Feed, Head-end Treatment, Ru Volatilization and MnO_2 Scavenging, July 15, 1951", HW-22076.

21. R. E. Tomlinson, "Purex Process Evaluation", HW-22888, Dec. 10, 1951.

22. R. E. Tomlinson, F. W. Woodfield, "Purex Chemical Flowsheet No. 2", HW-27277, Feb. 25, 1953.

23. C. A. Rohrmann, "Process Modifications 202-S Bldg.", HW-27968, May 5, 1953.

24. R. H. Beaton, "Redox Plant Alternative Flow Schemes", HW-28424, July 2, 1953.

25. E. J. Fuller, R. B. Lemon, R. H. Perkins, C. M. Slansky, "The Application of TBP to the "25" Process", IDO-14051, July 25, 1952.

26. E. J. Fuller, R. B. Lemon, "Revised Feed Preparation and Solvent Extraction Flowsheets for the Idaho Chemical Processing Plant", IDO-14250, May 11, 1953.

27. "Feasibility of the Conversion of ICPP to TBP - Amsco Solvent for Extraction", IDO-15002, Engineering Department, Atomic Energy Division, Jan. 15, 1953.

28. "Fuel Recovery Process for the SIR Mark A", KAPL-933, July 17, 1953.

29. "Decontamination of UO_2SO_4 Solutions (57-C)", KLX-1617, Project Summary, July 31, 1953.

30. "The Dissolution of Zr and Corrosion of SS in H_2SO_4 and HNO_3-HF Mixtures", LRL-78.

31. E. L. Zebroski, "The Purex Process at KAPL", TID-2009, Vol. 13, No. 2, June, 1953.

32. F. L. Culler, "Design of the Idaho Chemical Processing Plant", TID-2011, Vol. 3, No. 4, Dec., 1953.

33. D. Duffey, "Review of the Processing Development Program for Reactors", TID-5133, Jan. 15, 1953.

34. F. R. Bruce, "Solvent Extraction Chemistry of the Fission Products", p. 100.

International Conference on the Peaceful Uses of Atomic Energy, Vol. 7, Nuclear Chemistry and Effects of Radiation, United Nations Publications, N. Y., 1956.

35. J. R. Clark, "Radioactive Wastes at the Savannah River Plant", p. 40: In Report of meeting on ocean disposal of reactor wastes held at Woods Hole, Mass., Aug. 5-6, 1954; Atomic Energy Commission, NYO, Waste Disposal, p. 79 (mimeo), May 15, 1955.

Chemical Processes Associated with Waste Disposal

36. I. R. Higgins, "Alkaline Method for Treatment of High Radiation Level Aluminum Wastes", ORNL CF 57-1-139, Jan. 17, 1957.

37. D. L. Barney, et al, "Investigation of Methods for the Removal of Fission Products from Coating-Removal Solution", KAPL-1460, Jan. 30, 1956.

38. R. S. Pressly, "Preparation of Fission Rare-Earth Isotopes", ORNL-2252, May 7, 1957.

39. H. L. Krieger, B. Kahn, C. P. Straub, "Removal of Fission Products from Reactor Wastes", ORNL-2297, May 3, 1957.

40. A. S. Wilson, "Ruthenium Behavior in Nitric Acid Distillation", HW-45620, Sept. 1, 1956.

Chemical Processes Associated with Waste Disposal (cont'd)

41. E. A. Coppinger, R. E. Tomlinson, Chem. Eng. Prog. 52, 417, (1956).

42. A. T. Gresky, "The Preparation of Ru^{106} Tracer from Waste Metal Solution", ORNL-275, July 21, 1949.

43. R. E. Blanco et al, "Separation of Fission Products from Aluminum Waste Solutions by Ion Exchange", ORNL-301, June 26, 1953. (Revised)

44. A. T. Gresky, "The Recovery of Cs^{137} from ORNL Radiochemical Waste", ORNL-742, Jan. 8, 1951.

45. P. O. Schallert, "Production Separations of Fission-Product Groups for the Radioisotope Program", ORNL-1144, 1952.

46. "Laboratory Development of the MTR - RaLa Process for the Production of Ba^{140}, ORNL-1148.

47. R. H. Burns, E. Glueckauf, "Disposal of Industrial Atomic Waste Products", in Disposal of Waste Materials, Soc. Chem. Industry, 1957.

48. R. S. Pressly, A. F. Rupp, "Purification of Fission Product Rare Earths by Ion Exchange", ORNL-1313, 1953.

49. J. M. Chandler, D. O. Darby, "Terminal Report for the ORNL Pilot Plant Investigations of the Purex Process", ORNL-1519, Feb. 24, 1954.

50. R. A. Charpie, "A Chemical Reprocessing Plant for a Nuclear Power Economy", ORNL-1638, Project Hope.

51. H. L. Krieger, B. Kahn, C. P. Straub, "Removal of Fission Products from Acid Aluminum Nitrate Solutions by Co-precipitation Methods", ORNL-1966, Sept. 28, 1955.

52. E. D. Arnold, A. T. Gresky, "Relative Biological Hazards of Radiation Expected in Homogeneous Reactors TBR and HPR, ORNL-1982, Nov. 15, 1955.

53. I. R. Higgins, R. G. Wymer, "Diban-Ion Exchange Waste Disposal Scheme I", ORNL-1984, 1955.

54. R. E. Burns, "Decontamination of Metal Recovery Process Wastes", HW-28408, June 22, 1953.

55. R. J. Sloat, "Copper Ferrocyanide Scavenging of TBP Wastes", HW-28715, July 14, 1953.

56. "Decontamination of Bismuth Phosphate Process First Cycle Waste Supernatants", HW-29308, Sept. 10, 1953.

57. "Progress Report on Fission Products Utilization, II", BNL-161, Brookhaven National Laboratory, Jan. 1, 1952.

58. F. V. Caccavo et al, "Proposals for Concentrating Savannah River High Level Wastes", BNL-211, Part 2, April 1, 1953.

59. "Progress Report on Waste Processing Development Project; the Concentration of Purex Waste", BNL-266, Dec. 1, 1953.

60. B. Manowitz et al, "Progress Report on Waste Processing Development Project", BNL-293, May 1, 1954.

61. F. Hittman, B. Manowitz, "Progress Report on Waste Processing Development Project - Description of Calciner Pilot Plant", BNL-323, Dec. 1, 1954.

62. B. Manowitz, F. Hittman, "Proposal for Waste Processing at Idaho Chemical Processing Plant", BNL-329, Feb. 1, 1955.

63. "Recovery of Aluminum Nitrate from Redox Aqueous Waste Streams", KAPL-213, July 29, 1949

64. "Concentration of Wastes from Saltless Processes", KAPL-427, Dec. 4, 1950.

65. "Recovery of Aluminum Nitrate by Crystallization", KAPL-474, Feb. 24, 1951.

66. "An Investigation of the $KMnO_4$-MnO_2 Head-End Procedure for the Removal of Ruthenium and Niobium, Zirconium", KAPL-795, March 5, 1953.

67. T. Aponyi et al, "Pilot Plant Studies of the Ferrous Sulfide Process for the Decontamination of Purex Type Reactor Wastes", MLM-835, Final report, April 17, 1953.

68. G. Yasui, J. Schilb, J. Settle, "Distillation and Recovery of Nitric Acid According to Flow Sheet No. 1", ANL-4720, Summary report - July, August, September, 1951.

69. "The Processing of Radioactive Liquid Wastes", SRO-1, May 1, 1952.

70. E. Glueckauf, V. T. Healy, "Chemical Processing of Fission Product Solutions", p. 634.

International Conference on the Peaceful Uses of Atomic Energy, Vol. 9, Reactor Technology And Chemical Processing, United Nations Publications, N. Y., 1956.

71. A. T. Gresky, R. P. Wischow, Proposal for "25" or Thorex Waste Disposal: "Selective Recovery of Fission Products in Relation to the Long-Range Aspects of Chemical Waste Economics", ORNL CF 55-11-97.

72. R. E. Tomlinson, "The Isolation and Packaging of Fission Products at Hanford", HW-43835, June 27, 1956.

73. B. Manowitz, R. H. Betton, R. V. Horrigan, "Entrainment in Evaporators",, Chemical Engineering Progress, Vol. 51, No. 7, July, 1955.

Transportation of Active Wastes

74. H. R. Zeitlin, J. W. Ullmann, "Radioactive Waste Economics: Optimum Storage Time Prior to Shipping to Disposal Site", ORNL CF 55-10-101.

75. H. Blatz, "Transportation of Large Quantities of Radioactive Materials", P. 86.

Ultimate Disposal and Present Disposal Practices

76. "The Underground Disposal of Liquid Wastes at the Hanford Works", HW-17088, Washington, Feb. 1, 1950.

77. W. W. Koenig, K. L. Sanborn, "Corrosion of Redox Waste Storage Tank Construction Materials", HW-18595, August 21, 1950.

78. J. W. Healy, "Release of Radioactive Wastes to Ground", HW-28121, May 20, 1953.

79. M. J. Rutherford, "Waste Disposal Design Criteria - Purex Facility", HW-28320.

80. H. V. Cluckey, "TBP Waste Disposal Project for Cribbing Scavenged RAW", HW-30652, Jan. 27, 1954.

81. F. N. Browder et al, "Idaho Chemical Processing Plant, Liquid Waste Operating Manual", IDO-14094-ACCO, Vol. III, July 15, 1952.

82. A. L. Biladean, "Radioactive Waste Removal in a Trickling Filter Sewage Plant", IDO-24010, May, 1953.

83. B. Manowitz et al, "Progress Report on Fission Product Utilization III - Proposals for Concentrating Savannah River High Level Wastes", BNL-191, Aug. 1, 1952.

84. L. P. Hatch, J. J. Martin, W. S. Ginell, "Ultimate Disposal of Radioactive Wastes", BNL-1781, Feb., 1954.

85. W. S. Ginell, "Ultimate Radioactive Fission Waste Disposal", BNL-Summary Report No. 1, Nuclear Engineering Department, July 1, 1952.

86. "Liquid Waste Disposal Research", MLM-614, June 4, 1951 to Sept. 3, 1951, Mound Laboratory.

87. "Decontamination of Wastes", - Final Report, MLM-662, March 1, 1952, Mound Laboratory,

88. "Fixation of Mixed Fission Product Activity by Soils of the Savannah River Project", MLM-665, Feb. 25, 1952.

89. D. C. Hampson, E. H. Hykan, W. A. Rodger, "Basic Operational Report of the Argonne Active Waste Incinerator", ANL-5067, Feb. 6, 1953.

90. J. V. Natale et al, "Solidification of High Activity Aqueous Wastes", ANL-5103, Chem. Eng. Division, Summary Report, April, May, June, 1953.

91. James E. Evans, "Disposal of Active Wastes at Sea", DP-5.

92. I. C. Russell, "Water Resources of the Snake River Plains of Idaho", USGS-199, 1902.

93. Stearns, Crandall, Lynn and Steward, "Geology and Ground-Water Resources of the Snake River Plain in Idaho", USGS-774, 1938.

94. R. C. Newcomb, J. R. Strand, "Geology and Ground-Water Characteristics of the Hanford Reservation", USGS-WP8.

95. G. E. Siple, "Progress Report on Ground Water Investigations in South Carolina", Bulletin No. 15, 1946.

96. C. B. Amphlett, "Fixation of Radioactive Effluent", Nuclear Engineering, p. 119, June, 1956.

97. R. J. Morton, E. G. Struxness, "Ground Disposal of Radioactive Wastes", American Journal of Public Health, Vol. 46, Feb., 1956.

98. "National Petroleum Council Report of the Committee on Underground Storage for Petroleum", April 22, 1952.

99. F. N. Browder, "Liquid Waste Disposal at Oak Ridge National Laboratory", Industrial Engineering Chemistry, Vol. 43:7, p. 1502, July, 1951.

100. B. Manowitz, L. P. Hatch, "Processes for High Level Waste Disposal", Chemical Engineering Progress, Vol. 50:12, p. 144, 1954.

101. C. D. Watson, J. C. Bresee, J. S. Watson, "Gamma Radiation Damage Studies of Organic Protective Coatings and Gaskets", ORNL-2174, pp. 18-22, Oct. 29, 1956.

102. E. G. Struxness et al, "Status Report Ground Disposal of High-Level Radioactive Wastes", ORNL CF 55-1-188, Jan. 1, 1955.

103. S. H. Jury, "Heat Transfer in Waste Basins", ORNL CF 55-8-76, Aug. 11, 1955.

104. N. D. Groves, M. C. Fraser, W. L. Walker, "Field Corrosion Tests in Redox and Purex Underground Waste Storage Tanks", HW-37642, June 28, 1955.

105. "Recommendations for Waste Disposal of Phosphorus-32 and Iodine-131 for Medical Users", NBS Handbook 49, Nov. 2, 1951.

106. "Recommendations for the Disposal of Carbon-14 Wastes", NBS Handbook 53, Oct. 26, 1953.

107. C. A. Mawson, "Report on Waste Disposal System at the Chalk River Plant Of Atomic Energy of Canada Limited", CRB-658, July, 1956.

108. C. A. Mawson, "Waste Disposal into the Ground", P. 12.

109. J. C. Geyer, L. C. McMurray, A. P. Talboys, H. W. Brown, "Low-Level Radioactive Waste Disposal", P. 312.

110. C. R. Anderson, C. A. Rohrman, "The Design and Operation of High-Level Waste Storage Facilities", P. 552.

111. F. N. Browder, "Idaho Chemical Processing Plant Liquid Waste Facilities: Design Report", ORNL-1687, Aug. 6, 1954.

Ultimate Disposal and Present Disposal Practices (cont'd)

112. "Underground Storage for Petroleum", a report of The National Petroleum Council, Washington 6, D. C., March 7, 1957.

113. "Radioactive Waste Disposal in the Ocean", Handbook 58, National Bureau of Standards, Aug. 25, 1954.

Economic Considerations

114. F. L. Culler, "Preliminary Draft - Economics of Waste Disposal", ORNL CF 56-5-160.

115. "Preliminary Estimates of Costs for Processing Power Reactor Fuels in the Expanded Metal Recovery Facility", Letter, C. E. Center (UCNC) to S. R. Sapirie (AEC), July 27, 1956.

116. "Reactor Fuel Processing Costs: Papers Presented at the Idaho Symposium", IDO-14363 (3 parts), Jan 18-20, 1956.

117. "Economics of Concentrating Savannah River High Level Wastes and Off Site Disposal", BNL-197.

118. "Industrial Applications of Gross Fission Products", KLX-1394, Vitro Laboratories report, Jan. 1 - July 31, 1953.

119. "Economic Evaluation of Potential Methods for Concentrating Reactor Wastes for Storage-Study of Idaho Chemical Processing Plant Wastes", KLX-1723, Vitro Laboratories report, June 15, 1954.

120. "Economic Evaluation of Potential Methods for Concentrating Reactor Wastes for Storage-Study of Purex Waste from the Savannah River Project", KLX-1729, Vitro Laboratories report, Sept. 30, 1954.

121. "Economic Evaluation of Potential Methods for Concentrating Reactor Wastes for Storage-Study of Hanford Works Purex- and Redox-Wastes", KLX-1734.

122. The Reactor Handbook, KLX-54-24-N, Vol. 4, Fuel Recovery, Appendix A, Economics of Fuel Recovery, March 2, 1953.

123. "Reactor Waste Processing Cost Analysis", MLM-672, Mound Laboratory, March 5, 1952.

124. A. C. Herrington et al, "Permanent Methods of Radioactive Waste Disposal, Economic Evaluation", K-1005, M.I.T. Practice School, Oak Ridge, March 11, 1953.

125. E. C. Dybdal, "Engineering and Economic Evaluations of Projects", Chemical Engineering Progress, Vol. 46, 58, Feb., 1950.

126. A. C. Harrington, R. G. Shaver, C. W. Serenson, "Permanent Methods of Radioactive Waste Disposal - An Economic Evaluation", K-1005, Waste Disposal, p. 50, March, 11.

127. F. L. Culler, J. O. Blomeke, W. G. Stockdale, "Unit Costs and Economic Relationships for Certain Radioactive Waste Disposal Steps", ORNL CF 57-5-25, May, 1957.

Miscellaneous

128. R. Revelle, T. R. Folsom, E. D. Goldberg, J. D. Isaacs, "Nuclear Science and Oceanography", P. 277.

129. W. C. Hanson, H. A. Kornberg, "Radioactivity in Terrestial Animals Near an Atomic Energy Site", P. 281.

130. R. H. Simon, J. A. Consiglio, "Design and Operation of a Pilot Plant to Product Kilocurie Fission Product Sources", P. 317.

131. J. H. Rediske, F. P. Hungate, "The Abosrption of Fission Products by Plants", P. 278.

132. D. R. R. Fair, A. S. McLean, "The Radioactive Survey of the Area Surrounding an Atomic Energy Factory", P. 771.

133. R. F. Foster, J. J. Davis, "The Accumulation of Radioactive Substances in Aquatic Forms", P. 280.

International Conference on the Peaceful Uses of Atomic Energy, United Nations Publications, N. Y., 1956.

[redacted]

APPENDIX I

Description of Present Reactor - Chemical Processing - Waste Complex

Characterization of the nature of radioactive wastes starts with the nuclear reactor, in which the following variables have a decided effect on the radioactive wastes produced. The first set of variables, relating to the nuclear properties of the fissionable and fertile material, and the manner in which the reactor is designed to operate, shall be labeled nuclear variables.

Nuclear Variables

1. The fissionable material used
    - A. Uranium-235, highly enriched. Used for reactors for research (high neutron flux), mobile power, core elements of two-region power producers
    - B. Natural uranium - containing 0.71% U^{235} initially. After start of irradiation, which is supported by U^{235} fission, Pu^{239} is produced, which in turn fissions at increasing rate depending upon concentration of Pu produced by capture of neutrons in fertile nucleus U^{238}. Natural uranium is now used in production reactors for Pu, and can be used in stationary power reactors as a combined fuel and fertile material.
    - C. Partially enriched uranium - containing from 1% to high enrichment (~90%) U^{235}. This type of feed is being studied for many stationary power reactors; there is a high probability of producing more Pu^{239} than fissionable material consumed.
    - D. Plutonium-239. This second fissionable isotope has potentially the same applications as highly-enriched U^{235} reactors. It is being considered for fuel cores for "fast" stationary power reactors operating on a breeding cycle using natural or depleted uranium as the fertile material.
    - E. Uranium-233. This third fissionable material is produced by neutron capture in thorium-232. Major probable use may be in "thermal" reactors operating with a fully enriched U^{233} core and a breeding cycle using Th^{232} as the fertile material.

(Comment: The products of fission of U^{235}, Pu^{239}, and U^{233} are approximately the same for thermal neutron (2200 m/sec) fission. The fission product spectrum changes with the energy of the neutrons causing fission. Complete data on changes in fission product spectrum with fission neutron energy level are not availabe for all three fissionable materials. In this first report, all fission product yield data have been calculated from U^{235} thermal fission values, a safe generalization. The latest compilation of U^{235} thermal fission values, a safe generalization. The latest compilation of U^{235} fission yield data for thermal fission is given in works by Blomeke(1), Glendenin and Steinberg(2).)

2. The fertile material used

   A. U^{238} - in natural, depleted (from gaseous diffusion cascades or other reactor cycles) or partially enriched. Natural uranium is used as both fuel and fertile material for Pu production; for "fast" reactors operating with a plutonium breeding cycle in which natural or depleted uranium will be the blanket material.

   B. Thorium-232. The most probable use will be for thermal reactor breeding cycles; however, it can be used as a fertile blanket for intermediate and fast reactor breeding cycles. As in the case of U^{238} fertile production of Pu in a reactor, U^{233} produced from Th^{232} fissions at a rate lower than its total production rate during irradiation.

3. The "specific power" of the reactor system, or the number of fissions per unit weight of fissionable material, or the energy release per unit weight of fissionable material per unit of time.

For each specific power of the fissioning system, a different total quantity of fission products is produced and the fission product spectrum differs for finite periods of radiation, as shown by the following equations (using the nomenclature of Glasstone(3)):

$$\frac{dA}{dT} = -\lambda_A A - \sigma_A \phi A + \gamma_A \Sigma_f \phi \quad \text{(assuming no chemical removal)}$$

where

$\frac{dA}{dT}$ = net rate of increase of fission product A with time

λ_A = radioactive decay constant of A, $\lambda_A = \frac{0.693}{T_{1/2A}}$

A = number of nuclei/cm^3 of element A at any instant

σ_A = neutron capture cross section

ϕ = neutron flux of reactor

γ_A = fission yield of A, expressed as a fraction

$\Sigma_f \phi$ = number of fissions/(cm^3)(sec)

For periods of operation at constant power in which $\Sigma_f \phi$ is constant, an equilibrium for each fission product is reached where the rate of formation of A equals its rate of disappearance, or $\frac{dA}{dT} = 0$; then, defining A_o as equilibrium concentration of A,

$$A_o = \frac{\gamma_A \Sigma_f \phi}{\lambda_A + \sigma_A \phi} = \frac{\gamma_A \Sigma_f \phi}{\lambda_A^*}, \text{ where } \lambda_{A*} \equiv \lambda_A + \sigma_A \phi$$

Thus, the equilibrium amount of A is seen to depend upon the neutron flux or the specific power of the reactor.

4. Since many fission products have long effective half-lives and small neutron capture cross sections, as illustrated by the examples in Table No. 1, their concentration in the reactor may not reach equilibrium before the fuel element is removed. Thus, the time of irradiation, assuming a constant neutron flux or, more exactly the total of ϕ T product, determines the production of a fission product of known fission yield and capture cross section, assuming that the nucleus is the first element in the decay chain. This time dependence is expressed by the following equation:

$$\frac{dA}{dT} + \lambda_{A*} A = \gamma_A \Sigma_f \phi;$$

if $\Sigma_f \phi$ is constant, this can be integrated to

$$A(T) = \frac{\gamma_A \Sigma_f \phi}{\lambda_A^*} \left(1 - e^{-\lambda_A^* T} \right) + A(0)e^{-\lambda_A^* T}$$

where A(T) and A(0) are concentration of A at times T and zero.

The determination of other than primary yield fission products in a nulti-membered decay chain $A \longrightarrow B \longrightarrow C \longrightarrow D$ is treated in many published sources.(4)(5)

[redacted]

TABLE 1

Properties of Long-lived Fission Products

FP	Yield,%	Half-Life	Barns Thermal Capture Cross Section
Sm^{151}	0.5	73 y	7000
Cs^{137}	5.9	33 y	< 2
Sr^{90}	5.9	28 y	~ 1
Kr^{85}	0.3	10.27 y	>15
Pm^{147}	2.6	2.6 y	~ 60
Nb^{93m}	2.1	4.2 y	-

5. The energy of neutrons that cause the fission event, or enter in other reactions with nuclei in the reactor neutron field affect the nature and hazard of wastes. As previously pointed out, the fission product spectrum changes slightly with neutron energy; compilations in this report are based on thermal fission.

Even in a reactor in which almost all neutrons have been slowed down to thermal energies (2200 m/sec) by moderators such as heavy water, water, carbon, beryllium, or aluminum, a significant "fast" neutron flux may exist, significant in that neutrons of sufficient energy to produce appreciable quantities of products of reactions of the type (n, 2n) may exist. The significance of (n, 2n) parasitic reactions is illustrated by the production of U^{232} by the neutrons of energy in excess of 6.37 Mev on Th^{232}. The build-up of heavy element chains by (n, γ) and other reactions and the effects of products of these on the hazards in reactor fuel recycles and wastes are discussed in [redacted] 5.0.

6. The reactor "burn-up" is the degree to which the available fissionable material is used is usually expressed as atom per cent or weight per cent consumed, or more generally, in energy per unit weight of irradiated fuel. The per cent of utilization of available fission element in any reactor per pass is determined by many factors, the most important of which are:

A. Radiation damage to fuel elements - particularly important in metal fuel reactors.

B. Corrosion - particularly important in circulating fuel reactors

C. Build-up of fission product poisons, primarily from such nuclei as xenon-135; samarium-149 and -151; gadolinium-155 and -157, cadmium-113; iodine-131; europium-151, -153, -155; and others. Several reports have been prepared on fission product poisons and their effects on reactor design, two of which are referenced.(6)(7)

D. Depletion of fissionable material inventory in a reactor of specific design.

E. Growth of parasitic heavy elements by complex (n,γ) or $(n, 2n)$ capture chains

The per cent burn-up in a reactor determines the frequency of chemical processing and hence, the volume of fission product wastes produced per unit of energy from nuclear fission. The total quantity of fission products produced per unit of energy obviously is independent of the number of times that chemical processing occurs. However, the growth of certain parasitic heavy elements that may provide hazards to fuel element recycle and to waste disposal is dependent upon the number of recycles and the nature of the chemical process.

Type of Reactor

The type of reactor has a definite effect on the nature of hazards from fuel element recycle and waste fission products. Classifying reactors is a somewhat inexact procedure, but for this report we have chosen to discuss two broad categories: heterogeneous and homogeneous. It should be pointed out that other characterizations are possible, such as by neutron velocity, type of fuel, enrichment of fuel, et cetera.

1. Heterogeneous Reactors

Heterogeneous reactors usually are fueled by metallic fuel elements. The fuel elements can contain natural, partially enriched, or highly enriched uranium. In most cases the fissionable or fertile material is contained and even alloyed with a metal of low neutron capture cross section that imparts properties of corrosion resistance, temperature resistance, dimensional stability or other desirable characteristics. Since most fissionable or fertile material is contained in a protective metal cladding to prevent loss of fission products to coolant or moderator and to prevent corrosion, all fission products and neutron-produced heavy elements remain with the irradiated material in a heterogeneous reactor. Thus, the removal of fission product poisons and the recovery of new fissionable material can be accomplished only by removal of fuel from the reactor, followed by chemical reprocessing.

The hazard potential from accidental release of fission products in a clad metallic fuel or fertile element is slight; if a rupture occurs in a single element, only a small portion of the total fission product activity accumulated in the reactor would be released at any time. The hazard evaluation for a solid fuel element reactor can be assisted by analysis of data from the reports on the accident with the NRX reactor at Chalk River(8) and the Borax experiment.(9) The hazards involved in handling, transporting, and storing metal fuel elements are those resulting from the high contained fission product activity: (1) fission product heat removal; (2) biological shielding from gamma rays; and (3) ingestion or inhalation hazard of fission products released by an improbable fuel element rupture in transit. For most power reactor fuel elements, residual fission product heat will be sufficient to require cooling for several weeks to several months to prevent large temperature rises under near adiabetic conditions during transport and storage. The heat due to decay of fission products can be estimated from empirical equations accurate to a factor of about 2 for decay periods of ten seconds to several months(10)(11) as given in Appendix I.

The heterogeneous reactors have dominated the nuclear picture from 1942 until the present time. Reactors used for production of Pu are thermal machines, water of heavy water moderated. Early experimental reactors used natural uranium as fuel; later machines such as the MTR use highly enriched U^{235}--aluminum alloy clad in aluminum. The Experimental Breeder Reactor is a "fast" reactor, cooled with a liquid metal; it uses highly enriched uranium metal canned in stainless steel. Naval and submarine reactors employ zirconium-clad highly enriched uranium-zirconium alloy.

Many of the proposed stationary power reactors are of the heterogeneous type. Most are designed to breed more fissionable material than they consume, and thus require processing to recover new fissionable material and probably to recycle partially depleted core fuel. In order to extract the energy released by the fission process, to produce electricity with reasonable thermal efficiencies, high temperature metals are employed in power producer fuel elements. Such materials, new to reprocessing technology, are zirconium and stainless steel and variations. Cladding and alloying elements, together with possible bonding agents, brazing or welding materials have a controlling influence on the chemical process and ultimately on the volume of fission product wastes.

For example, an MTR fuel element contains roughly 200 grams of uranium along with about 4400 grams of aluminum, 15-17 grams of silica in brazing flux and other lesser impurities. To recover uranium it is necessary to dissolve the entire fuel element in nitric acid (catalyzed with mercury in a concentration equivalent to about 2 per cent of the Al weight). Thus, the volume of chemical plant feed is determined by aluminum, and inactive salts of aluminum control the volume of the aqueous fission product wastes. The uranium concentration is less than two grams per liter.

This same condition is true for most of the proposed power reactor fuels: liquid wastes will contain large concentrations of inactive salts.

Most of the reactor types now being considered for large scale installation for research, power demonstration, and large scale power production are of the heterogeneous type. A listing of types, taken from a recent declassified publication(12) includes the following:

A. Pressurized Water - all reactors are thermal, use highly enriched to slightly enriched uranium as fuel, have rod or plate fuel elements using Al, Zr, Zircalloy-2 and stainless steel as fuel diluent and cladding material. Examples of this type of reactor:

   a. Materials Testing Reactor(13) - Al and enriched U, no blanket. This reactor design typifies all others of this class and has provided the basic reactor type for most of the reactor proposals.

   b. Submarine Thermal Reactor - Zirconium-clad enriched uranium fuel, no blanket; for mobile power. An MTR type reactor using zirconium; typifies many other reactor designs, such as the Pressurized Water Reactor(14) for stationary power.

   c. Engineering Test Reactor(15) - A giant MTR using Al and enriched uranium. For engineering research.

   d. Army Package Power Reactor(16) - A stainless steel MTR, with notable difference in that UO_2 is used in the fuel and then clad with stainless steel to form the fuel element. No blanket. For small power station use, particularly for outlying and remote areas where power cost is not restrictive.

B. Boiling Water Reactors(17) - All, so far, are experimental, of the MTR type, sponsored by Argonne National Laboratory. Fuel elements are enriched uranium and aluminum with no blankets. The Nuclear Power Group has proposed a zirconium-uranium boiling water reactor.

C. Swimming Pool Reactors(18) - Research reactors of MTR type, of which the ORNL Bulk Shielding Reactor was the first in the family, the Geneva Conference Reactor a famous daughter, and the Oak Ridge Research Reactor is the maturing and powerful research-oriented senior member of the clan. All are aluminum-enriched uranium fuel assemblies of the MTR plate type, some with UO_2 rather than uranium metal as the fissionable material.

D. Heavy Water Cooled Reactors(19) - Most are fueled with uranium metal, and are used for research or plutonium production and do not breed. Most notable examples:

  a. Savannah River Reactors
  b. NRX and NRU Canadian Reactors
  c. CP-3 and CP-3' (enriched U) - first of this type, built at Argonne National Laboratory
  d. Most foreign research reactors - British, French, Swedish, Norwegian, Russian, Canadian

E. Graphite Moderated Reactors(20)(21) - The classic type first built in Chicago in 1942, with many production and research descendants. Uranium metal fuel is canned in aluminum or zirconium. Power reactors may be the single region-type plutonium producers.

F. Sodium Graphite Reactors(22)(23) - pioneered by North American Aviation. The Sodium Reactor Experiment which is scheduled for completion in 1956-1957 will be the first of this type. The fuel is slightly enriched uranium metal clad in zirconium or stainless steel, with a bonding agent of sodium metal. Coolant will be sodium. Its long-term use will be for power and plutonium production.

G. Liquid Metal Cooled Reactors - built for fissionable material consumption only. The first reactor of this type was Clementine, a mercury cooled, plutonium fueled, fast reactor built at Los Alamos. The classic reactor design for power may be the Submarine Intermediate (intermediate neutron energies) Reactor built by General Electric at Knolls Atomic Power Laboratory. The fuel is enriched uranium; no blanket is included.

H. Fast Breeder Reactors(24)(25)(26)(27) - the classic type is the Experimental Breeder Reactor, with highly enriched U^{235} core, clad in stainless steel, a natural uranium blanket, and a sodium-potassium (NaK) alloy coolant. The fast breeder cycle may utilize plutonium as fuel and depleted or natural uranium as blanket. Coolants will always be some material other than a hydrogeneous one, probably liquid metals or fused salts. Cores of fuel elements will vary greatly. Many reactor concepts of this nature have been proposed, such as that of the Atomic Power Development Associates, EBR-2, and the British Fast Power Breeder. Thorium can be used as a feast breeder blanket. Reactors will be used for power and fissionable material production. The Liquid Metal Fuel Reactor concept, pioneered by Brookhaven National Laboratory(28), is a graphite moderated homogeneous U^{233} breeder.

TABLE 2

Classification of Heterogeneous Fuel Elements Based on Reprocessing Principles

Classification	Principle
1. Enriched (90% U^{235}) UO_2-S.S. sinter	1. Requires criticality control 2. No Pu recovery 3. Not directly HNO_3 soluble 4. High inert content
2. Enriched (90% U^{235}) U and Zr	1. Requires criticality control 2. No Pu recovery 3. Not directly HNO_3 soluble 4. High inert content
3. Enriched (90% U^{235}) U-Al alloy, Pu alloy, Pu, UO_2 or UC_2	1. Requires criticality control 2. No Pu recovery (enriched U case) 3. Soluble in HNO_3 catalyzed by Hg or HF 4. High inert content (~96% Al for U-Al alloy)
4. Enriched (20% U^{235}) U and Zr	1. Requires criticality control 2. Requires Pu recovery 3. Not directly soluble in HNO_3 4. Low inert content (~10% Zr)
5. Natural or slightly enriched (~ 2% U^{235}) U and Zr	1. Essentially no criticality control 2. Requires Pu recovery 3. Not directly soluble in HNO_3 4. Low inert content (<15% Zr)
6. Natural or slightly enriched (~ 2% U^{235}) U metal, oxide, or Mo alloy	1. Essentially no criticality control 2. Requires Pu recovery 3. Soluble in HNO_3 4. Low inert content (<15% Mo)
7. Thorium	1. Essentially no criticality control 2. Requires U^{233} recovery 3. Soluble in HNO_3 catalyzed by HF 4. No inert content

From the standpoint of chemical reprocessing and from the production of radioactive wastes resulting from the recycle of fuel and blanket material, the array of possible fuel types is formidable and will require process development of a highly diverse nature. R. E. Blanco has summarized possible fuels from heterogeneous systems in seven categories(29) as shown in Table No. 2.

2. <u>Aqueous Homogeneous Reactors</u>

The homogeneous reactor is one in which the fuel, coolant and moderator (if any) are combined in a single phase, usually a fluid. The two primary types that are emerging are the aqueous homogeneous and the liquid metal fuel reactor. A third type, employing fused salts as the fuel carrier has promise. The types which we shall consider in this discussion are the aqueous homogeneous and the liquid metal fuel reactor.

The Aqueous Homogeneous Reactor combines fuel and moderator (usually heavy water) in a fluid that can be used as the primary heat transfer medium. The first circulating fuel model was built and operated by Oak Ridge National Laboratory. Many combinations of fuel and blanket arrangements are possible, such as:

A. Two-region Machines - circulated fuel and blanket

- a. Core - uranium salt dissolved in D_2O or a slurry of UO_3, usually fully enriched uranium-235, or for a power breeder cycle, U^{233}. Core tank probably zirconium.
- b. Possible blankets
  - 1'. Thorium oxide slurry in D_2O
  - 2'. Possible thorium salt soluble in aqueous medium that is stable chemically to reactor conditions and does not have prohibitive parasitic neutron capture
  - 3'. Natural or depleted uranium in D_2O solution. Pu will be produced but breeding not possible. A uranium oxide or other insoluble salt slurry is possible.

B. Single-region Machines - circulating fuel

- a. Partially enriched uranium salt dissolved in heavy water Plutonium is produced by excess neutron capture in U^{238}
- b. Slurries of uranium oxide or other water insoluble salts
- c. Reactor built to consume enriched U^{233}, U^{235} or Pu without any fissionable material recovery for production of high neutron flux or for mobile power application. It should be noted that to increase neutron flux levels appreciably above those attainable in the MTR - (2-3 x 10^{14} n/cm^2/sec) an aqueous homogeneous reactor may be necessary for two reasons:

1'. The burn-up rate and consequently the fuel replacement schedule of fissionable material (1% of core per day in the MTR) would require prohibitive material handling and very short reactor cycles. For example, at a flux of 2×10^{15} neutrons/(cm^2)(sec), the fissionable material consumption would be approximately 10%/day. For a core of fixed geometry the operating cycle would be from two to ten days maximum.

2'. For systems other than the aqueous homogeneous, the maximum concentration of U^{235} may be too low to achieve flux levels of the order of 10^{15} to 10^{16}. Dilution and parasitic neutron capture by materials other than water will prevent the attainment of high neutron flux levels.

Thus, the ideal experimental and engineering development reactor, i.e., one in which it is possible to obtain much higher neutron fluxes than will be utilized in reactors built to produce power, could be of the aqueous homogeneous type.

The aqueous homogeneous reactor possesses a high negative temperature coefficient of reactivity and remains stable even with large additions of reactivity in a short period of time. Its fission rate is self-regulating depending upon the power demand. A more complete picture of homogeneous reactors can be obtained from numberous reports published in the declassified literature.(30)(31)(32)

The hazard from sudden release of fission products from a homogeneous reactor is greater than for heterogeneous reactors with solid clad fuel elements unless the entire element vaporizes. Fission products, fissionable and fertile material, and the parasitic neutron capture products exist in a very mobile form, either in solution or as a suspension in a liquid under pressure. In the event of a reactor failure, or even a leak of reactor fuel, the hazard is greatest in the immediate vicinity of the reactor. To minimize the hazard of an aqueous homogeneous, or for any circulating fuel, the reactor and its associated chemical plant, are contained in a sealed vessel built to withstand the energy release of a reactor break. The long-term hazard from a release of reactor fluids from a homogeneous reactor would not be greater than those from a heterogeneous one. In fact, since homogeneous reactors can be operated with a continuous chemical cycle that removes biologically dangerous isotopes from the reactor, potentially the hazard from the ultimate development of the homogeneous reactor can be less than that of the heterogeneous case.(33)(34

Processing of homogeneous reactors is simplified somewhat in that it is possible to remove fission product and corrosion product poisons continuously from the reactor circuit without having to process the uranium fuel. However, a small bleed-off of fuel must occur to maintain the enrichment level of the reactor, assuming a maximum concentration limit on the fule solution. Fortunately, in the aqueous homogeneous case, the allowable concentration range for fuel can be from a few grams per liter to several hundred grams per liter for UO_2SO_4 in heavy water. In the homogeneous case of a thermal breeder with a thorium oxide blanket, thorium oxide must be removed and processed by solvent extraction to separate U^{233} and Pa^{233} from thorium. In the case of the plutonium producer, plutonium and high cross-section fission products can be removed from the reactor continuously. In either case, final separation of products must be performed by solvent extraction (or some other satisfactory technique). However, the quantity of fuel and blanket material to be processed, (and as a consequence the waste volumes) may be less than for most heterogeneous cases, since the fissionable and fertile material are present in relatively pure form uncontaminated by diluents, and since the achievable burn-up fraction in the homogeneous case can always be high.

The aqueous homogeneous reactor produces and releases gaseous fission products during its operation, unlike the heterogeneous case. A list of radioactive gaseous fission products from U^{235} thermal fission would include the elements listed in Table No. 1, Section 3.0 of main report.

3. Liquid Metal Homogeneous Reactors(35)

The liquid metal fuel reactor is similar in concept to the aqueous homogeneous reactor except that a relatively low melting metal of low neutron capture cross section is substituted for D_2O. Bismuth with a melting point of 271°C and a neutron capture cross section for 2200 m/sec neutrons of 0.030 barns will dissolve about one-half atom per cent uranium at 500°C. This is sufficient solubility for an enriched uranium to support a chain reaction. With bismuth the two-region type of reactor can be built economically; because of solubility restrictions a single-region reaction for plutonium production is not feasible. The reaction being studied at Brookhaven National Laboratory is a two-region machine consisting of a graphite moderated core of highly enriched uranium (U^{233} in the long range power picture) dissolved in bismuth with a blanket of some form of thorium, although natural or depleted uranium could be used. Several blanket systems are possible:

(1) Thorium oxide slurry in D_2O

(2) Suspension of thorium bismuthide (Th_3Bi_5) in liquid bismuth

The processing of core fuel will be similar to the aqueous homogeneous case in that high cross-section fission products will be removed continuously. With the uranium-bismuth system this will be accomplished by treating a portion of the circulating core solution with a fused fluoride or chloride salt of sodium, potassium, magnesium, calcium or zirconium.

As in the aqueous homogeneous case the fission products, fissionable and fertile material, and the parasitic capture products of the heavy elements will be present in a mobile form, but not under high pressure. However, the liquid metal system possesses a high chemical reaction potential on exposure to oxygen or moisture. The fission gases must be vented from the reactor circuit, offering the same problem as described for the aqueous homogeneous case. Fission products will be removed in a fused salt mixture along with some of the valuable fuel, which may require processing to recover, processing such as the addition of magnesium metal to the fused salt to reduce uranium for return to the bismuth phase. The small percentage of fuel with the fission products in the fused salt can be purified by solvent extraction. Recovery of new fissionable material from the blanket probably will have to be accomplished by solvent extraction.

The liquid metal homogeneous reactor using bismuth-209 as the carrier presents an additional hazard which results from the following parasitic neutron capture:

$$\mathrm{Bi}^{209}\,(n,\gamma)\ \xrightarrow{0.030\mathrm{b}^{*}}\ \mathrm{Bi}^{210}\ \xrightarrow[5.0\mathrm{d}]{\beta^{-}}\ \mathrm{Po}^{210}\ \xrightarrow[138.3\mathrm{d}]{\alpha}\ \mathrm{Pb}^{206}$$

Polonium-210, with an alpha emission of 5.29 Mev, is retained in the body by the spleen (soluble) or lungs (insoluble) and is particularly hazardous because of the high ionizing potential of its radiation.

Other liquid metal reactor systems are possible, using either thermal or fast neutrons for fission. For example, a slurry of some salt or uranium in sodium or sodium potassium alloy; a solution of uranium in lead or mercury.

The Chemical Processing Cycle

A large chemical complex is required to supply fuel to reactors and to recover from them partially depleted and new fissionable material. The functioning of this complex can be affected at many points by changes in allowable radiation exposures to operating personnel. In this chemical-metallurgical complex, exposure potentials comparable or greater than those provided by a single reactor are possible. The recycle complex for a nuclear power economy involves the transportation, storage and processing of the radioactive output of all reactors. The inventory of radioactivity in a chemical plant, because of the probable economics of reprocessing, represents an integration of hazard from long-lived fission products produced by many reactors. Stored wastes by virtue of accumulation and degree of dispersability may represent the greatest potential long-term hazard to the general population.

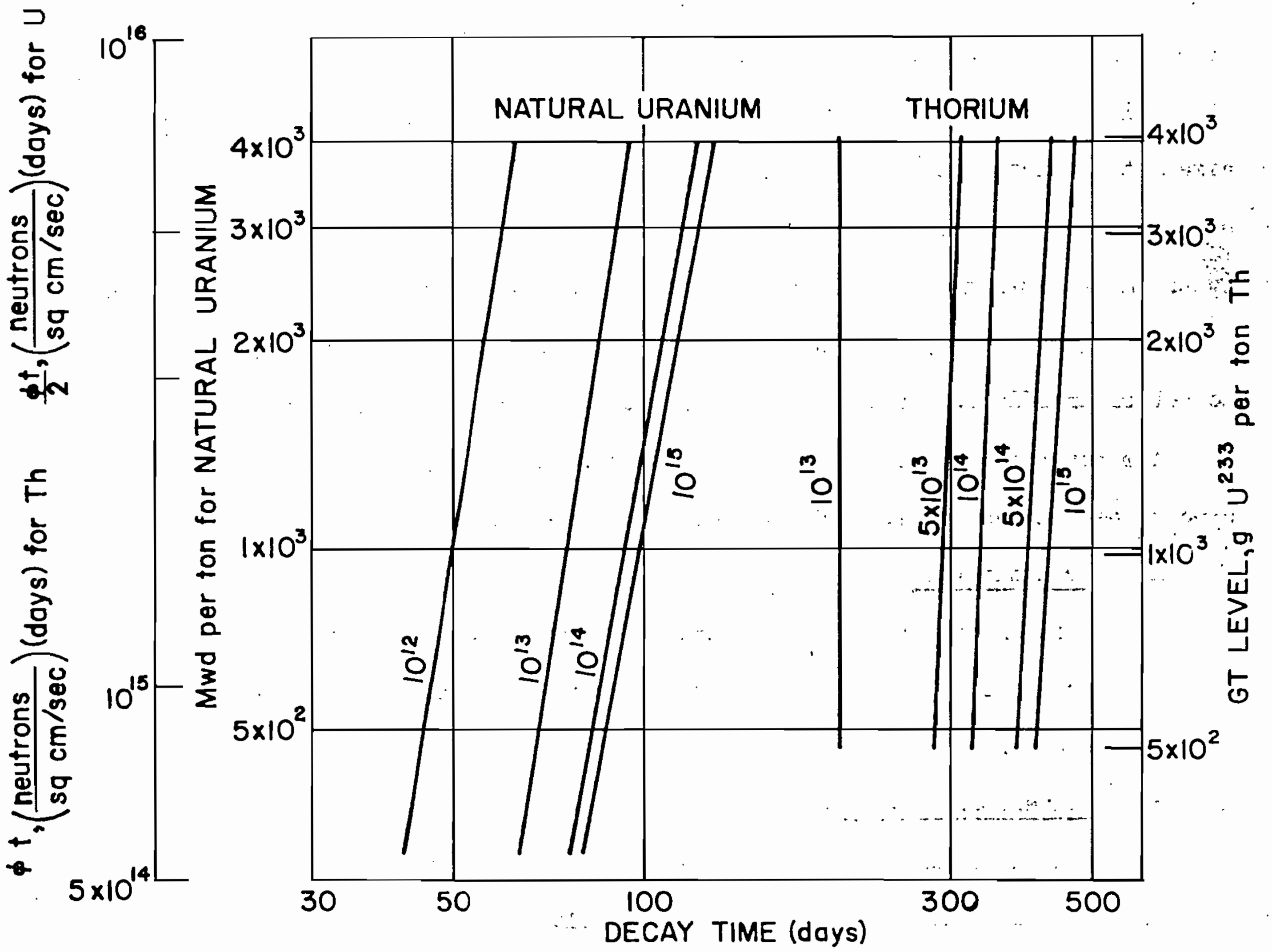
Chemical Processes for Fission Product Removal and Separation of Fissionable and Fertile Material

Following a suitable cooling period, as determined for thermal fission in Figure 1, all reactor fuel and blanket materials are ready for chemical processing to separate fissionable and fertile material from fission products and from each other. Two types of processes can be considered: (1) where fissionable and fertile materials are removed from fission products, and (2) where fission products are removed from fissionable material. Various degrees of separation can be considered for the long-term development. However, to allow for direct (or better, unshielded) handling of fissionable and fertile materials, complete removal of fission products is required. With the development of methods of remotely accomplish-

---

*Total cross section of 0.030b includes capture to form ~10^6 year alpha emitting Bi-210m.

UNCLASSIFIED
ORNL-LR-Dwg6736-A



U^{235} Enrichment (%)	Additional Decay Time Required (days)
2	10
5	19
10	26
15	30
20	33
30	37
40	40
50	42
100	49

Fig. 1 Time Required for U^{237} from Uranium and Th^{234} from Thorium to Decay.

ing all other phases of the recycle after chemical separation, partial decontamination as possible. As far as the reactor is concerned, only fission products of high capture cross section need be removed. To recover new fissionable material produced by neutron capture in blankets or fertile regions of a reactor, either complete or partial decontamination may ultimately be used, depending upon the economics of the particular reactor cycle.

All chemical processes developed to the pilot plant stage to date have been aimed at complete decontamination. All are based upon selective organic solvent extraction of uranium, plutonium, and thorium from aqueous nitrate systems (with the exception of the first process for separation of Pu, the bismuth phosphate precipitation process). Figure No. 2(37) gives the decontamination factors required to return irradiated fissionable and fertile material to natural background.

The reprocessing of reactor fuel elements may be accomplished by a variety of process techniques. A list of some of the better known possibilities follows:

A. Precipitation

Bismuth phosphate(39) for the recovery of Pu from uranium and fission products. Precipitated fission product sulphates from aqueous homogeneous reactors.

B. Solvent Extraction

Add the following possible reagents as extractants:

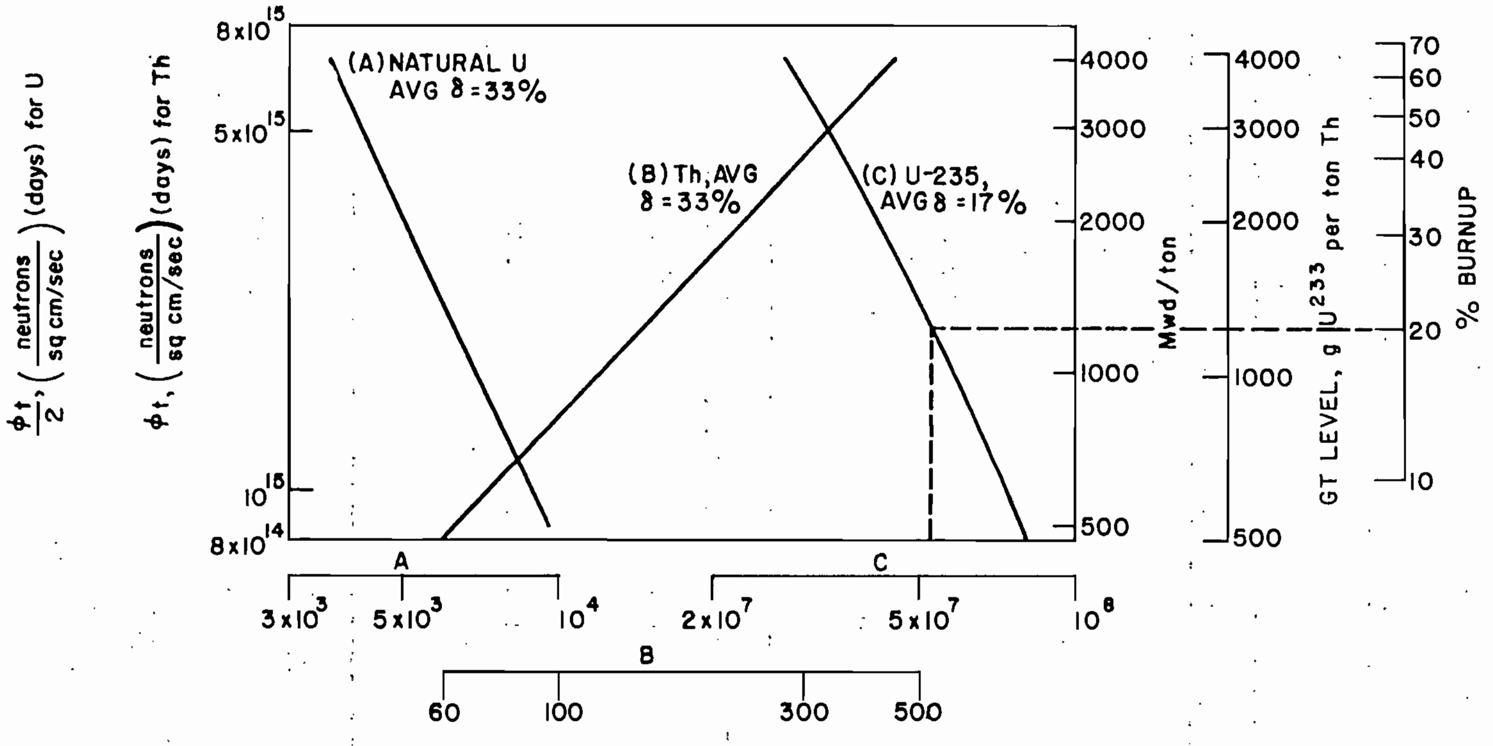
a. Triglycol Dichloride (Trigly)

b. Dibutyl Carbitol (Butex)

c. Ethers, such diethyl, dibutyl, diisoprophyl, and cellosolve

The solvent extraction processes have been described in numerous papers.(39) Since the basic principles of the separations by solvent extraction are so well reported and since the technique is well established, we will not describe any of the processes. We are primarily interested in the chemical and radioactive nature of the wastes from these processes. Table No. 3

UNCLASSIFIED
ORNL-LR-Dwg 6723



$$A = \frac{DF}{Mwd/t}\left(\frac{1.5\times10^{13}}{\phi}\right)^{1/2}$$

$$B = \frac{DF}{GT}\left(\frac{1.5\times10^{13}}{\phi} \text{ or } \frac{\phi}{1.5\times10^{13}}\right)^{1/2}$$

$$\phi < 1.5\times10^{13}, \quad \phi > 1.5\times10^{13}$$

$$C = \frac{DF}{\%B.U.}\left(\frac{1.5\times10^{13}}{\phi_0}\right)^{1/2}$$

Decay Time (days)	Avg. Decay[a] Factor, from 10 days
10	1.0
30	1.7
60	3.0
100	5.2
150	8.3
200	12.5
300	20

[a] Based on 100 days Irradiation at 3×10^{13}

Fig. 2. Required Decontamination Factors for Irradiated Natural Uranium, Enriched Uranium, and Thorium. Based on background activity of 10^9 disintegrations/min/kg; decay time, 10 days. Consider the following example: Assume that U-235 was irradiated to 20% burnup at flux of 10^{14}; the decontamination factor $= 20\times5.3\times10^7\times(6.67)^{1/2} = 2.74\times10^9$ after 10 days decay and 3.3×10^8 after 100 days. (ϕ_0 = initial flux; flux increases with burnup in order to maintain constant power)

[redacted]

Table 3

Processes for Chemical Separation of Fissionable and Fertile Materials

Process	Organic Solvent	Aqueous Solvent	Irradiated Metal Feed	Salting Agent
For Separation and Decontamination of U and Pu				
Redox	Hexone	HNO_3	Natural U, Al can	$Al(NO_3)_3$
Metal Recovery	12.5% TBP in hydrocarbon	HNO_3	Caustic precipitated U, fission products	HNO_3
Purex	30% TBP in hydrocarbon	HNO_3	Natural U, Al can	HNO_3
TTA chelation	0.25 M TTA in hexone	HNO_3	Natural U, Al can	$Al(NO_3)_3$
For Separation and Decontamination of Enriched U and Al				
25	Hexone	HNO_3, $Hg(NO_3)_2$ catalyst	U-Al alloy	$Al(NO_3)_3$
25, TBP	5% TBP in hydrocarbon	HNO_3, $Hg(NO_3)_3$ catalyst	U-Al alloy	HNO_3 - $Al(NO_3)_3$
For Separation of U-233, Pa, and Th				
Interim-23, hexone	Hexone	HNO_3, F^- catalyst	Th, Al can	$Al(NO_3)_3$, HNO_3
Interim-23, TBP	1.5% TBP in hydrocarbon	HNO_3, F^- + Hg^{++} catalyst	Th, Al can	$Al(NO_3)_3$, HNO_3
Thorex	42.5% TBP in hydrocarbon	HNO_3, F^- + Hg^{++} catalyst	Th, Al can	$Al(NO_3)_3$, HNO_3

gives a list of the solvent extraction processes, along with their main features.

For materials that are initially insoluble in nitric acid such as stainless steel and zirconium, dissolution methods in other mineral acids have been developed. After dissolution, the stainless or zirconium fuels can be converted to a nitrate aquoues system by the addition of $Al(NO_3)_3$ and HNO_3 and solvent extracted by a modified Purex process. Stainless steel elements can be dissolved in concentrated sulfuric acid or aqua regia; zirconium and zirconium-uranium alloy can be dissolved in concentrated hydrofluoric acid.

C. Volatilization of Chemical Compounds

a. Fluoride volatility(40-43) for removal of UF_6 as a gas. Wastes probably produced as a fused fluoride salt waste. Fluorinating agents could be F_2, BrF_3, ClF_3, BrF_5, and other interhalogens. May be useful only for enriched uranium fuels.

b. Chloride volatility, from which wastes would appear in fused salt chlorides

D. Ion Exchange

a. Possibly applicable to separation of U and Pu or Th and U^{233}. Wastes from primary recovery of fissionable and fertile material will be similar to those produced from solvent extraction. Most useful for recovery and final purification of solvent extraction plant products. Further development of inorganic ion exchange materials and permeable ion exchange membranes may increase the applicability of ion exchange.

b. Can be used to remove fission products from process solutions or wastes.

E. High Temperature Process for Partial Decontamination from Fission Products

a. Distillation(44-46) of Pu and high cross-section fission products from

[redacted]

molten uranium. This type of process must be followed by shielded transport, fabrication, reactor loading.

b. Slagging of molten uranium-plutonium(47)(48)(49) with uranium oxide to remove high cross-section fission products and volatile components. This, too, must be followed by remote metallurgy, fuel element handling and reactor loading. Fission product wastes will appear in vapor phase and in oxide slag.

c. Molten salt(50)(51) or metal extraction(52) for high cross-section fission product removal and possible partial separation of uranium and plutonium. The same general comments as for slagging are applicable. This type of process may be used for primary separation of fuel and blanket of liquid metal fuel reactor.(52)

d. Fused salt electrolysis in which a partially decontaminated product will result.(53)

e. Modified de Boer process(54) for uranium recovery.

Most of the process in the above list, with the exception of solvent extraction and bismuth phosphate precipitation, are still in rather early stages of development. Insufficient information has been developed on the nature of liquid or solid wastes from these processes to provide a basis for discussion in this paper although further study will define their nature.

[redacted]

REFERENCES

(1) Blomeke, J. O., Nuclear Properties of U^{235} Fission Products, ORNL 1793, October 21, 1955, Unclassified.

(2) Glendenin, L. E., and Steinberg, E. P., Survey of Radiochemical Studies of the Fission Process, Paper 614, International Conference on the Peaceful Uses of Atomic Energy, August, 1955.

(3) Glasstone, Samuel, Principles of Nuclear Reactor Engineering, pp. 121-126, D. Van Nostrand, Inc., (1955).

(4) Glasstone, Samuel, op. cit.

(5) AECD-3645, The Reactor Handbook, Physics, Vol. I, March, 1955.

(6) Gresky, A. T., Arnold, E. D., Poisoning of the Core of the Two-Region Homogeneous Thermal Breeder: Study No. 1, ORNL CF 53-12-165, Secret; and Study No. 2, ORNL CF 54-2-208, Secret; and Weinberg, A. M., Survey of Fuel Cycles and Reactor Types, Paper 862, International Conference on the Peaceful Uses of Atomic Energy, (1955).

(7) Lock, C. J. L., Fission Product Formation in a Homogeneous Power Reactor, AERE C/R 1715, June, 1955.

(8) Gilbert, S. W., Decontamination of the Canadian Reactor, Chemical Engineering Progress 50:267 (1954).

(9) Dietrich, J. R., Experimental Determinations of the Self-Regulation and Safety of Operating Water Moderated Reactors, Paper 481, International Conference on the Peaceful Uses of Atomic Energy, (1955).

(10) Way, K., and Wigner, E. P., Rate of Decay of Fission Products, Paper 43, NNES Div. IV, Vol. 9, McGraw-Hill (1951)

(11) Untermeyer, S., and Weills, J. T., Heat Generation in Irradiated Uranium, AECD-3454 (1952).

(12) Raytheon Manufacturing Company, Nuclear Reactor Data, November 15, 1955; brochure available from Raytheon, Waltham, Massachusetts.

(13) Doan, R. L., Huffman, J. R., Materials Testing Reactor Experimental Program and Reactor Operation, Paper 485, International Conference on the Peaceful Uses of Atomic Energy, (1955).

(14) Rengel, J. C., Pressurized Water Reactor, Atomic Industrial Forum Report No. 7, September, 1955.

(15) Zartman, I. F., AEC Engineering Test Reactor, Atomic Industrial Forum Report No. 7, September, 1955.

(16) Kaschau, K. A., The Army Package Power Reactor, Paper 30, Nuclear Engineering and Science Congress, Cleveland, Ohio, December, 1955. May be obtained from AIChE, 25 West Forty-fifth Street, New York 36, New York.

(17) Dietrich, J. R., et al, Design and Operating Experience of a Prototype Boiling Water Power Reactor, Paper 851, International Conference on the Peaceful Uses of Atomic Energy.(1955).

(18) Gill, J. P., A High Performance Research Reactor, Nuclear Engineering, Part II, Symposium Series of AIChE, No. 12, Vol. 50, June, 1954.

(19) Stevens, J. E. S., Canadian Chemical Processing, 40, 80 (1956).

(20) Carson, A. B., A Graphite Moderated Nuclear Power Plant Design, Paper 492, International Conference on the Peaceful Uses of Atomic Energy (1955)

(21) Hinton, C., The Graphite Moderated, Gas-Cooled Pile, Paper 406, International Conference on the Peaceful Uses of Atomic Energy (1955)

(22) Starr, C., A Sodium Graphite Reactor, 75,000 Electrical Kilowatt Power Plant Paper 493, International Conference on the Peaceful Uses of Atomic Energy (1955)

(23) Parkins, W. E., The Sodium Reactor Experiment, Paper 499, International Conference on the Peaceful Uses of Atomic Energy (1955)

[redacted]

(24) Zinn, W. H., Review of Fast Power Reactors, Paper 814, International Conference on the Peaceful Uses of Atomic Energy (1955).

(25) Kendall, J. W., and Fry, T. M., The Downreay Fast Reactor Project, Paper 405, International Conference on the Peaceful Uses of Atomic Energy (1955).

(26) Barnes, A. H., The Engineering Design of EBR-II, Paper 501, International Conference on the Peaceful Uses of Atomic Energy (1955).

(27) Lichtenberger, H. C. et al, Operating Experience and Experimental Results Obtained from a NaK Cooled Fast Reactor, Paper 813, International Conference on the Peaceful Uses of Atomic Energy, (1955).

(28) Miles, F. T., and Williams, C., Liquid Metal Fuel Reactor, Paper 494, International Conference on the Peaceful Uses of Atomic Energy, (1955).

(29) Blanco, R. E., Processing Power Reactor Fuels, to be presented at meeting the American Nuclear Society, Chicago, June, 1956.

(30) Briggs, R. B., and Swartout, J. A., Aqueous Homogeneous Power Reactors, Paper 496, International Conference on the Peaceful Uses of Atomic Energy (1955).

(31) Beall, S. E., and Swartout, J. A., The Homogeneous Reactor Test, Paper 498, International Conference on the Peaceful Uses of Atomic Energy (1955).

(32) Beall, S. E., and Winters, C. E., The Homogeneous Reactor Experiment, ORNL CF 54-2-128, Unclassified.

(33) Arnold, E. D., and Gresky, A. T., Relative Biological Hazards Associated with Nuclear Reactors, Presented, at meeting of American Nuclear Society June 27-29, 1955.

(34) Arnold, E. D., Gresky, A. T., Relative Biological Hazards of Radiations Expected in Homogeneous Reactors, Thermal Breeder and Homogeneous Plutonium Producer, ORNL 1982, Confidential, November 15, 1955.

(35) Miles, F. T., and Williams, C., Liquid-Metal-Fuel Reactor Systems for Power, Nuclear Engineering, Part I, Chemical Engineering Progress Symposium Series, AIChE, Vol. 50, No. 11 (1954).

(36) Culler, F. L., Reprocessing Reactor Fuel and Blanket Materials by Solvent Extraction, Paper 822, International Conference on the Peaceful Uses of Atomic Energy (1955).

(37) Lawroski, S., Survey of Separation Processes, Paper 832, International Conference on the Peaceful Uses of Atomic Energy (1955).

(38) A selected list of papers presented at the International Conference on the Peaceful Uses of Atomic Energy in Geneva, Switzerland, August, 1955 follows:

Author	Title	Paper No.
Culler, F. L.	Reprocessing of Reactor Fuel and Blanket Materials by Solvent Extraction	P-822
Flanary, J. R.	Solvent Extraction of Uranium and Plutonium from Fission Products (Purex Process)	P-539
Gresky, A. T.	Solvent Extraction of U^{233} and Thorium Fission Products	P-540
Bruce, F. R. and Culler, F. L.	The Processing of Uranium-Aluminum Fuel Elements	P-541
Bruce, F. R.	The Solvent Extraction Chemistry of Fission Products	P-719
Goldshmidt, B.	Solvent Extraction of Plutonium from Uranium Irradiated in Atomic Piles	P-349

Author	Title	Paper No.
Fletcher, J. M.	Chemical Principles of Separation of Fission Products from Uranium and Plutonium by Solvent Extraction	P-413
Fletcher, J. M., and Martin, F. S.	The Chemistry of Ruthenium	P-437
McKay, H. A. C.	TBP as an Extracting Agent for the Nitrates of the Actinide Elements	P-441
Hyde, E. K.	Radiochemical Separations Methods for the Actinide Elements	P-728

(39) Lawroski, S., op. cit.

(40) Hyman, H. H., et al, Decontamination of Irradiated Reactor Fuels by Fractional Distillation Processes Using Uranium Hexafluoride, Paper 546, International Conference on the Peaceful Uses of Atomic Energy (1955).

(41) Malm, J. G., Weinstock, B., The Properties of Plutonium Hexafluoride, Paper 733, International Conference on the Peaceful Uses of Atomic Energy (1955).

(42) Cathers, G. I., and Leuze, R. E., A Volitilization Process for Uranium Recovery, Paper 278, Presented at Nuclear Engineering and Science Congress, December 16, 1955. (Reprint available from AIChE, 25 West Forty-fifth Street, New York, N. Y.)

(43) Lawroski, S., op. cit.

(44) Motta, E. E., High Temperature Fuel Processing Methods, Paper 542, International Conference on the Peaceful Uses of Atomic Energy (1955).

(45) Phipps, T. E., et al, Vapor Pressure of Liquid Plutonium, Paper 735, International Conference on the Peaceful Uses of Atomic Energy(1955).

(46) Feder, H. M., Pyrometallurgical Processing of Nuclear Materials, Paper 544, International Conference on the Peaceful Uses of Atomic Energy (1955).

(47) Motta, E. E., op. cit.

(48) Foltz, J. R., Rosen, F., and Gardner, W., *Pyroprocessing of Nuclear Reactor Fuels*, Paper 145, Presented at Nuclear Science and Engineering Congress, December, 1955. (Reprint available from AIChE, 25 West Forty-Fifth Street, New York 26, N. Y.)

(49) Motta, E. E., op.cit.

(50) Lawroski, S., op. cit.

(51) Voigt, A. F., *The Purification of Uranium Reactor Fuel by Liquid Metal Extraction*, Paper 545, International Conference on the Peaceful Uses of Atomic Energy, (1955).

(52) Dwyer, O. E., *High Temperature Processing Systems for Liquid-Metal Fuels and Breeder Blankets*, Paper 550, International Conference on the Peaceful Uses of Atomic Energy, (1955).

(53) Niedrach, L. W., and Glamm, A. C., *Electrorefining as a Method for Removing Fission Products from Uranium Fuels*, Paper 200, Presented at Nuclear Engineering and Science Congress, December, 1955. (Reprint available from AIChE, 25 West Forty-Fifth Street, New York 36, N. Y.).

(54) Lawroski, S., op. cit.

General Reference on Chemical Processing

Bruce, F. R., Fletcher, J. M., Hyman, H. H., Katz, J. J., Process Chemistry, Progress in Nuclear Energy, Series III., Pergamon Press and McGraw-Hill, 1956

[redacted]

APPENDIX II

REACTOR EXCURSIONS

1.0 Borax Incident(32)

Those who had witnessed the Borax excursion (power reached 13-20 x 10^9 watts in a reactor which normally operated in the 1-10 MW range) were asked to compare it with the dynamite explosion. Shots of 1, 2, 3 and 4 sticks were made, each stick being about 1/2 pound of 40% dynamite. Opinions varied considerably. Those who had been inside the control trailer for both kinds of explosion had the opinion that the Borax excursion sounded like 2 sticks of dynamite, but not as sharp as the dynamite. Most of the observers who had been in the open thought that about 3, but not more than 4, sticks was a proper comparison, but they felt that the Borax excursion was a sharper explosion.

The damage done to the equipment perhaps gives a more quantitative idea of the explosive effect. The breaking of the reactor tank was the most striking manifestation of explosive force.

The fact that the reactor tank failed was not surprising, since it was constructed of 1/2-inch carbon steel for a design pressure of 125 psi. It was not expected to withstand pressures greatly in excess of 1,000 psi. The manner in which it fragmented, without regard for flanges or other reinforcements, was, however, striking.

It was estimated by examining the lines of parting, that the breaking process had occupied a time of about 1/2 to 1 millisecond, and that the pressure had therefore been maintained at a high level for at least that length of time.

Calculations indicated that explosive charges in the range 6 to 17 pounds of TNT would produce comparable damage to the reactor and shield tanks. The equivalent, energy-wise, of the 135 Mw-sec of nuclear energy released in the excursion is about 70 pounds of TNT.

During ~~or~~ immediately following the excursion an instantaneous dosage rate in excess of 400 mr/hr was indicated on survey meters at the distance of approximately 1/2 mile from the reactor. Within approximately 30 seconds the rate had decreased to 25 mr/hr and continued to decrease rapidly. Within less than 5 minutes the readings had decreased to less than 1.0 mr/hr.

These intensities, which were undoubtedly from pure gamma radiation, seem consistent with the energy release of the excursion. At the time of the excursion the air temperature was 53.8°F, and the barometric pressure was 25.285 inches. Taking an effective air attenuation length of 270 meters for delayed fission-product gammas under these conditions, one estimates that, if no other shielding than air were present around the total mass of fission products from an excursion of 135 Mw-sec, the radiation intensity at 1/2 mile would be about 110 mr/hr after 30 seconds. Actually, something like half the fuel remained in the shield tank and pump pit. Of the remainder, it seems reasonable to assume that its radiation might be attenuated by a factor of 2 or 3 by local obstructions.

Beta-gamma film packets located at 1500 feet from the reactor, roughly downwind but outside the path of fall-out, recorded exposures of 50 mr/hr. These exposures were probably primarily from gammas originating in the material which stayed in the reactor vicinity. The total integrated exposure from complete gamma decay of all fission products of a 135 Mw-sec excursion would be about 140 mr if only air shielding were present.

The air-dispersed material from the reactor was blown in a direction about 35 degrees west of south. Mobile monitoring teams crossing the trajectory of the material at 8:35 A.M. (15 minutes after excursion) at a distance of about 0.8 mi from the reactor, recorded a maximum reading of 5 mr/hr with open-window survey meter, 3 feet above ground. A similar reading at 8:45 A.M., about 2.3 miles from the reactor, gave 2.0 mr/hr. The maximum readings at these same distances, at 9:15 - 9:20 A.M., after the air-borne material had definitely passed over, were 6 mr/hr, with open-window meter 1 inch above ground. By 10:30 the following morning these intensities had decayed at 0.05 mr/hr. The intensities were maintained at roughly the maximum level over a path width of about 500 feet, and fell to about 1/10 the maximum along the edges of a path about 1500 feet wide.

For comparison with the foregoing observations, an estimate has been made of the total activity of the fuel left in the immediate vicinity of the reactor, by extrapolation of later surveys in the reactor area. This estimate indicates that the fuel, if spread uniformly over an area of 5×10^8 square feet, would give a reading of 6 mr/hr on an open-window meter 1 inch above ground, 1 hour after the excursion.

The integrated radiation intensities of fuel in the various locations on this date are given in the Table below.

Description of Fragments		Area Integral of Intensity on 8/19/54, $(cm^2)(r)/hr$
General distribution of fragments outside fence (Figure 38)		24×10^4
General distribution of fragments inside fence		38×10^4
Fragments remaining in shield tank		4×10^4
Fragments remaining in pump pit		33×10^4
Large pieces of fuel element		10×10^4
Fragments mixed with other miscellaneous debris		4×10^4
	TOTAL	113×10^4

2.0 Early Observations and Deductions of 1952 NRX Incident(33)

The activity discharged by the air through the stack behaved like fission products from a very short irradiation and is attributed to the excape of volatile and gaseous fission products from the uranium with ruptured sheathing together with most of the fission products from the melting, fracture, and rapid oxidation of the uranium of the air-cooled rod of previously unirradiated uranium.

The best estimate which it has been possible to make is that the total fissions involved would be 10^{18}, and, assuming the power surge was 4000 megawatt-sec, if all the activity were supposed to come from the air-cooled rod, it would require the excape of the products from 30 kg of natural uranium at the center of the rod. Much less than this is likely to have been involved because there would have been a considerable excape of volatile and gaseous fission products from other ruptures.

The estimate is that of Drs. W. G. Cross and S. A. Kushneriuk based on the exposure of 350 mr on a film worn by an electrician up a pole adjacent to the reactor stack at the time.

It was not considered safe to stop the flow of water to the basement since the condition of the uranium was not known. It was feared that, since some of the metal had been so highly irradiated (about 3000 Mwd/ton), it would heat itself up, oxidize rapidly, and might even catch fire if not cooled. The flow of water was cut back as low as considered sufficient to reach all the uranium. This flow was about 70 gal/min. It was not discharged to the river but was pumped from the basement to a storage tank. The total water collected amounted to about k,000,000 gal and contained about 10,000 curies of long-lived fission products. This water was successfully disposed of by pumping it through a 1 1/4-mile pipeline to a trench system in a disposal ground where it was allowed to seep away. A check was kept on activity in water draining from this area, but no detectable activity was found even in the creek draining the area to a small lake.

3.0 EBR Incident(34)

On November 29, 1955, at AEC's reactor testing station near Arco, Idaho, the world's first fast breeder reactor, EBR-I, was undergoing the last series of experiments scheduled at that time. Object of these difficult tests was to measure transient temperature coefficients, by measureing changes in reactivity of the reactor as the temperature of the fuel elements was increased. The reactor was placed on a short positive period and the fuel temperature permitted to rise to 500-600°C. To obtain the temperature coefficient of the fuel only, the liquid NaK coolant flow had to be shut off - so that the machine was actually operating not as a reactor at all but rather as a critical assembly. (The core pot was filled with NaK, but it was static.) On the last test in the series of deliberate power surges, the scientist in charge, watching special fast-acting neutron and temperature recorders and realizing a runaway was imminent, gave verbal instructions to the operating technician for immediate shutdown. The technician misunderstood and pressed the button activating the normal motor-driven shut-off rods. The scientist reached over and pushed the scram button. The interval, a delay of at most two seconds, was enough to permit power to overshoot to a level where the fuel rods melted down, some uranium alloying with core steel.

One possibly encouraging thing that remains to be verified has to do with the whitish encrustation on the reflector elements and on the surface of the melted mass. This is due to oxides of sodium and potassium from the NaK coolant. The core underwent a significant decrease in density due to boiling and volatilization of the NaK, and thereby became less reactive, from the nuclear point of view. It has yet to be established whether this took place an instant before or an instant after the meltdown. If the former, it would mean this phenomenon was operating as an added safety factor.

APPENDIX III

CRITICALITY HAZARD IN REPROCESSING OF NUCLEAR FUELS

by

J. W. Ullmann
Chemical Technology Division
Oak Ridge National Laboratory

INTRODUCTION

Origin of the Problem

Fissionable materials uranium-233, uranium-235 or plutonium-239, may have to be recovered from bred or partially spent fuels to permit economic operation of nuclear reactors. The recovery process consists of chemical or metallurgical separation of the fissionable material from undesirable contaminants and any chemical, metallurgical and mechanical steps necessary to restore the fuel to its original form. Precautions must be taken in the design and operation of reprocessing plants against the creation of a critical assemble.

Definitions

An assembly is said to be critical if the number of neutrons produced in each generation by fission equals the total number of neutrons absorbed and lost by leakage. Such a system if self-sustaining and can be operated at various power levels. If fewer neutrons are produced by fission than are absorbed and lost, the system is designated subcritical; if more neutrons are produced than absorbed and lost, the system is supercritical.

If a source of neutrons is directed at a subcritical assembly the steady state neutron flux will exceed that due to the source alone. The closer an assembly is to criticality the greater will be this flux multiplication. Criticality measurements can therefore be made by extrapolation of data from subcritical systems to zero reciprocal multiplication.(1)

When a system attains criticality without requiring the delayed neutrons which are produced by post-fission decay it is said to be prompt critical. If the delayed neutrons are needed to reach criticality the system is called delayed critical.

System Parameters

The factors affecting the criticality of a system are:

(1) The mass of fissionable material which determined the potential number of fissions.

[redacted]

(2) The specific fissionable isotope which determines the number of neutrons produced per fission and the ratio of fission to non-fission absorptions.

(3) The degree of moderation since the probability of effecting fission depends on neutron velocity. The probability is greater for slow neutrons than fast neutrons.

(4) The size and shape of the system which determine the extent of neutron leakage.

(5) The degree of poisoning since neutron absorption by non-fissionable poison atoms competes with the fission reaction.

(6) The degree of homogeneity of the system since the presence of voids will increase the mass required for criticality.

(7) The degree of reflection since reflection of neutrons back into the system reduced the effect of leakage.

(8) The presence of fertile material which can fission with fast neutrons.

(9) The size, shape and spacing of lattice elements in a heterogeneous system will influence leakage and the degree of interaction between elements.

The types of systems which can be encountered in a processing plant are:

(1) Slow neutron systems which result from the fissionable materials in water or organic solvents.

(2) Fast neutron systems which result from the fissionable material handled as oxide or metal.

(3) Intermediate systems which may result from aqueous slurries or hydrated solid salts of fissionable material.

Since the slow neutron system leads to the smallest critical mass, it is potentially the most dangerous, the most of the following remarks concern solutions of fissionable material in a moderating solvent.

METHODS OF CONTROL

Mass Limitation

Criticality may be avoided if the minimum mass of fissionable material capable of sustaining a chain reaction is never assembled in any single location, provided care is taken to prevent interaction with surrounding materials. Table No. 1 gives published safe upper mass limits for reflected and unreflected solutions with optimum geometry and moderation and no poison present.

TABLE 1

EXTIMATED MINIMUM CRITICAL MASSES (GRAMS)

	U^{233}	U^{235}	Pu^{239}
Unreflected	*1150(2)	*2130(2)	1244(3)
Water reflected	588(4)	800(4)	510(4)

*Lowest reported mass, not necessarily minimum.

Criticality control by mass limitation requires strict accounting by analysis and material balance to determine at all times the quantity of fissionable material present in a piece of equipment or in storage. Mass limitation inherently requires batch operation with holdup of batches for analysis before each step. It is therefore usually more costly than control methods amenable to continuous processing and is certain only to the extent permitted by the available techniques of analysis and mensuration.

Geometry Control

A sufficiently high rate of neutron leakage from a system averts criticality. Table No. 2 shows the minimum demensions of spheres, infinitely long cylinders and infinite area slabs required to sustain criticality of solutions.

TABLE 2

ESTIMATED MINIMUM CRITICAL DIMENSIONS (CENTIMETERS)

	U^{233}	U^{235}	Pu^{239}
Bare Sphere, Diameter	-	-	33.8(3)
Water-reflected Sphere, Diameter	18.1(4)	23.0(4)	20.5(3)
Bare Infinite Cylinder, Diameter	-	-	16.0(3)
Water-reflected Infinite Cylinder Diameter	-	-	11.2(3)
Bare Infinite Slab, Thickness	-	-	17.6(3)
Water-reflected Infinite Slab Thickness	-	-	4.0(3)

[redacted]

It should be noted that the mass contained in a geometry controlled system can exceed the [redacted] critical mass of Table No. 1 without achievement of criticality and [redacted] the minimum masses and minimum volumes occur at different concentrations of fisionable material.

Geometry control is adaptable to safe continuous operation, but as can be seen, the equipment size is necessarily small and parallel lines may be required. Adequate spacing of individually safe units must be allowed to prevent criticality of an assembly of units by interaction.

Concentration Control

Change in concentration of a fissionable material in a water or hydrocarbon solvent will alter the atomic ratio of hydrogen to fissionable isotope. Both the degree of moderation and the degree of posioning are affected by this ratio. The combined effect is illustrated in Figure 1. At low hydrogen concentrations per fissionable atom, the critical mass is large since there is only a small amount of moderation. As the hydrogen concentration increases, the mass decreases to a minimum. With further increase in hydrogen concentration, poisoning increases the critical mass until a region of infinite critical mass is reached. This is the limit for concentration control. Any solutions more dilute in fissionable isotope than corresponding to this hydrogen to fissionable isotope ratio are safe. Table No. 3 lists the maximum safe concentrations for aqueous solutions in optimum geometry.

TABLE 3

ESTIMATED MINIMUM CRITICAL AQUEOUS CONCENTRATIONS(5)

	H/Fissionable Isotope	Grams/Liter
U^{233}	2330	10.9
U^{235}	2220	11.6
Pu^{239}	3600	7.3

UNCLASSIFIED
ORNL-LR-DWG 12928

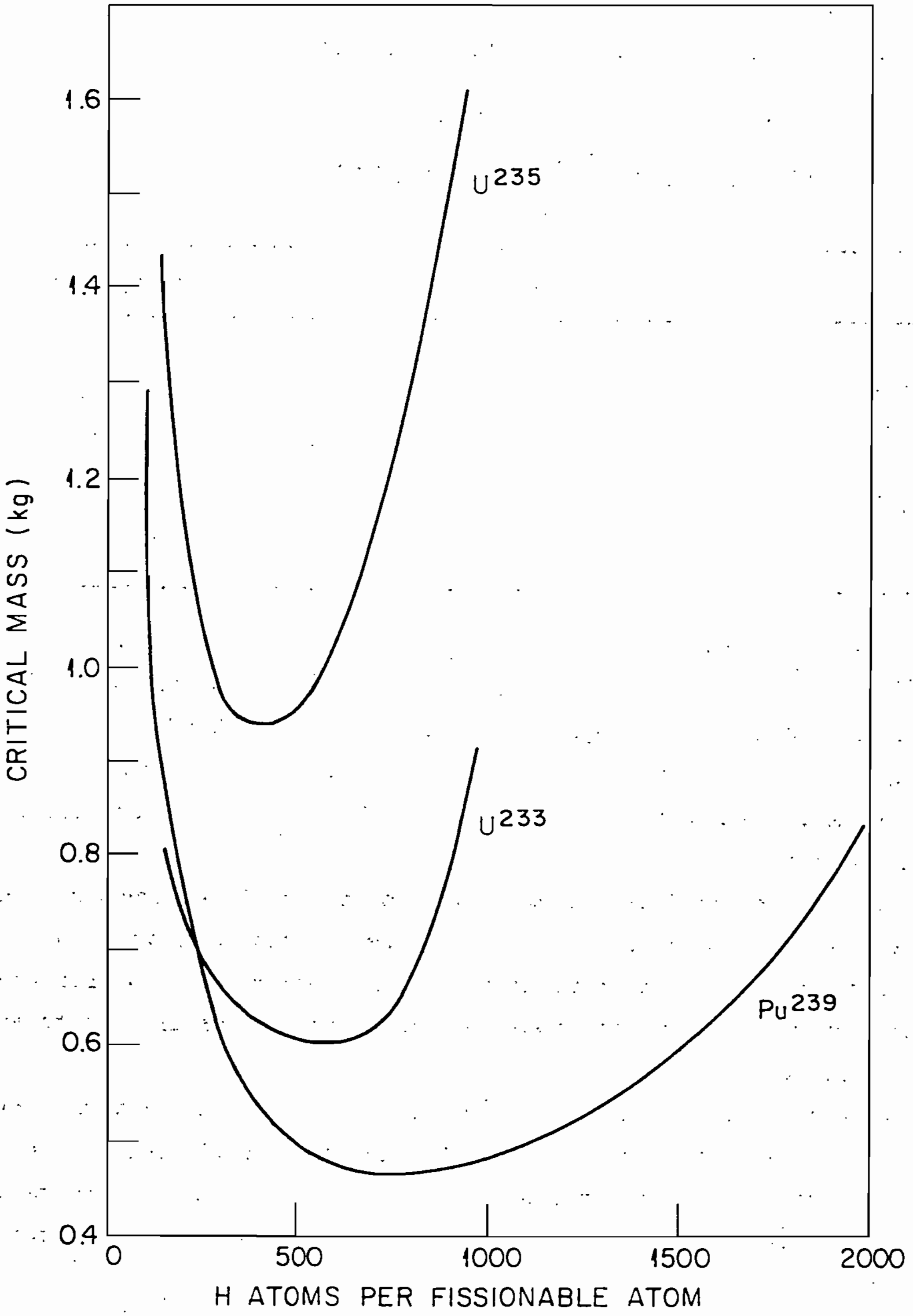


Fig. 1. Effect of Moderation on Critical Mass of Water Reflected Aqueous Solutions in Spherical Vessels. (Data of McKay and Nicholls).

[redacted]

In the absence of any moderation, at a hydrogen to fissionable atom ratio of zero, essentially only fast fission can occur. Table No. 4 gives values obtained with spherical assemblies of 50 to 90% U^{235} metal reflected with metallic natural uranium at various effective densities produced by voids.

TABLE 4

CRITICAL MASS U^{235} METAL SPHERES(6)

Core	Density (Grams/cm^3)	Critical Mass (Kg U^{235})
~90% U^{235}	18.7	16
~90% U^{235}	13.1	25
~90% U^{235}	9.3	37
~65% U^{235}	18.7	21
~50% U^{235}	18.7	27

The data of the above table are correlated by assuming critical mass propostional to U^{235} enrichment to the -0.7 power and to core density to the -1.2 power.

The effect of slight hydrogen moderation is illustrated by the critical mass of a highly enriched uranium hydride assembly reflected by natural uranium. The average empirical formula for the hydride was $UH_{2.97}C_{1.11}O_{0.25}$ and the critical mass was 12.1 Kg U^{235}.(6)

Concentration control is effective in processing dilute aqueous solutions if there is assurance that the fissionable isotope concentration will not exceed the values of Table No. 3, and precipitation will not occur. Instruments are therefore used to monitor stream concentrations and flows, and alarms are set to sound in advance of a critical condition.

In the handling of metallic fissionable material, concentration control permits the assembly of masses far in excess of the minimum of Table No. 1. The spacing of metal pieces, possibility of water being present and similar factors require detailed study of each individual situation to determine the most desirable method of safe handling.

Combined Mass, Geometry and Concentration Control

The maximum safe values of mass, size and concentration of Tables Nos. 1, 2, and 3 are each tabulated separately for optimum values of the other two parameters. It is possible to relax two of the three restrictions if the third is well on the safe side of the optimum value. For example, the diameter of a cylinder may be increased beyond the limit of Table No. 2 if the concentration can be guaranteed to be at all times different from that corresponding to optimum moderation. It is also possible to exceed the limits of Table No. 2 if the units are, in a criticality sense, far short of infinite cylinders or slabs. The basis for such relaxations from the limiting values is illustrated by Figure 2, a plot of critical cylinder heights as a function of diameter for two hydrogen to U^{235} atom ratios.

Poison Control

If it is compatible with the process, a poison can be deliberately introduced into the equipment or process streams to prevent criticality. Cadmium foil surrounding a vessel can essentially negate the effect of a water reflector, and the insertion of poison rods into the interor of vessels has been proposed. The latter method requires certainty that the poison is always present in the proper geometry.

The poison effect of the nitrate ion and the presence of the 240 isotope of plutonium is illustrated by the estimate of 510 grams of Pu^{239} as the minimum critical mass compared to 690 grams found by experiments with $Pu(NO_3)_4$.(4) Fission products present in irradiated fuel add poison to the system to an extent dependent on the irradiation history of the material. Fertile isotopes also exert a poisoning effect. For example, the minimum critical mass of 4.9 per cent enriched U^{235} is reported as somewhat less than 2 kilograms compared to 0.8 kologram for ~90 per cent enrichment.(4) Because of U^{238} poisoning, there exists a minimum U^{235} enrichment below which uranium cannot be made critical with light water moderation.

UNCLASSIFIED
ORNL-LR-DWG 12929

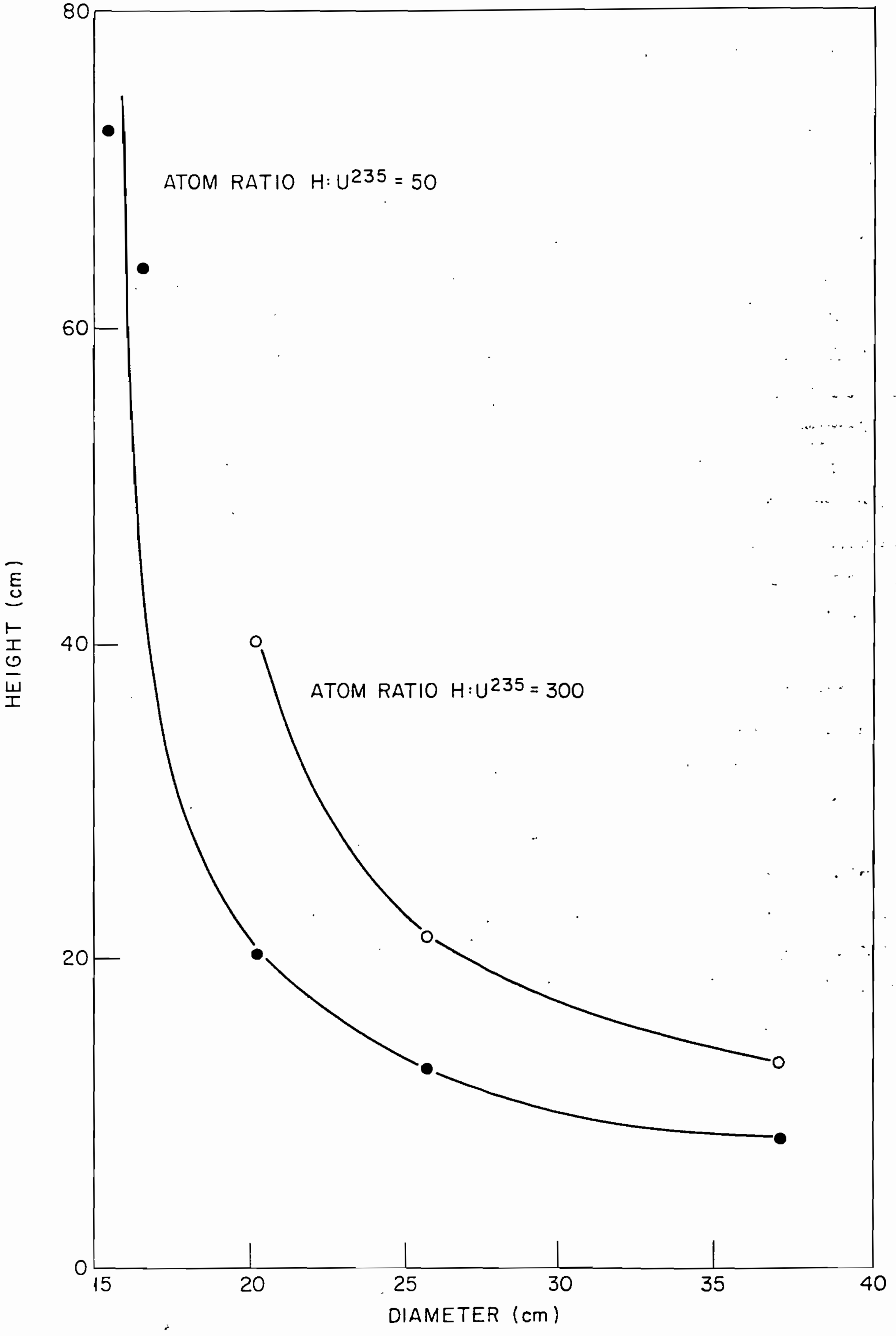


Fig. 2. Critical Dimensions of Water Reflected Cylinders Containing Aqueous U^{235} Solutions. (Data of Callihan, Morfitt and Thomas).

SAFETY PHILOSOPHY

The aim of criticality control in processing is to prevent a nuclear incident from occurring. Since it is almost impossible, and certainly uneconomical, to design against all possible operational errors or sabotage, the consequences of an accident are of interest.

In a process plant it is most unlikely that fissionable material can be assembled fast enough to cause a nuclear detonation. Since nuclear explosion requires assembly in less than a millisecond, a solution brought to supercriticality by precipitation or overfilling will probably disperse itself by boiling.(3) The consequences of such an incident would, however, lead to serious radiation and toxic hazards and damage to the equipment.

The present practice is to design for routine safe operation with substantial safety factors, to check the mode of operation with expert opinion or experiment, to monitor the plant to prevent accumulation by leakage or deposition, and to provide instrumental interlocks and alarms such that at least two independent mistakes are necessary to create a dangerous situation.

REFERENCES

(1) Horton, C. C., and McCullen, J. K., Paper P-428, International Conference on the Peaceful Uses of Atomic Energy, Geneva, Switzerland (1955).

(2) Thomas, J. T., Fox, J. K., and Callihan, D., Nuclear Science and Engineering 1: 20 (1956).

(3) McKay, H. A. C., and Nicholls, C. M., Paper P-454, International Conference on the Peaceful Uses of Atomic Energy, Geneva, Switzeralnd (1955).

(4) Callihan, A. D., Morfitt, J. W., and Thomas, J. T., Paper P-834, International Conference on the Peaceful Uses of Atomic Energy, Geneva, Switzerland, (1955).

(5) Thomas, J. T., ORNL Internal Memo CF 55-11-104 (1955).

(6) White, R. H., Nuclear Science and Engineering, 1: 53 (1956).