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Sources of Tritium

J. E. Phillips
C. E. Easterly

HEALTH AND SAFETY RESEARCH DIVISION
OAK RIDGE NATIONAL LABORATORY

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J. E. Phillips and C. E. Easterly

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ABSTRACT

A review of tritium sources is presented. The tritium production and release rates are discussed for light water reactors (LWRs), heavy water reactors (HWRs), high temperature gas cooled reactors (HTGRs), liquid metal fast breeder reactors (LMFBRs), and molten salt breeder reactors (MSBRs). In addition, release rates are discussed for tritium production facilities, fuel reprocessing plants, weapons detonations, and fusion reactors. A discussion of the chemical form of the release is included. The energy producing facilities are ranked in order of increasing tritium production and release. The ranking is: HTGRs, LWRs, LMFBRs, MSBRs, and HWRs. The majority of tritium has been released in the form of tritiated water.

1. INTRODUCTION

Much interest has centered on the radiological hazards associated with tritium since it was discovered to be a by-product of man's attempts to exploit successfully atomic energy. This interest has prevailed despite tritium's low-energy β emission (0.91 fJ average). Such a β particle will not penetrate the outer dead layer of skin; therefore, it does not pose an external radiological hazard. Tritium, however, can pose an internal radiological hazard. The annual allowable intake of tritiated water is 2.2×10^{-3} to 3.0×10^{-3} TBq for occupational exposures.

Interest in tritium is not so much due to its physical properties as to the chemical forms in which it is prevalent in the environment. Tritium is primarily found in the environment as tritiated hydrogen gas, HT, or as tritiated water, HTO. Upon exposure, tritiated water has been found to be absorbed more efficiently in the body than tritium gas by ∿4 orders of magnitude. Thus, tritiated water is the primary cause of concern over tritium releases.

Two other factors have contributed to the concern over releases of tritium. First, tritiated water is very difficult to separate from ordinary water. No truly economical, satisfactory process exists to effect such a separation, especially on a large scale. Thus, once water is contaminated with tritium, it will remain contaminated until tritium decays. A substantial period of time would be required for a significant reduction in the tritium to occur since tritium's half-life is 12.3 years. Second, water is a biologically essential molecule.

The hazard of tritium is strongly influenced by the probability of exposure. The first step in establishing the probability of exposure is to determine and quantify the sources. This paper addresses this aspect of the exposure problem as well as discussing the chemical forms of release.

2. NATURAL PRODUCTION

Tritium is produced primarily by interactions of cosmic rays with the atmosphere.[1-2] Production by cosmic ray interactions with terrestrial material is negligible in comparison.[3] Most of the atmospheric production arises from interactions of protons and neutrons with nitrogen and oxygen atoms in the atmosphere.[1,3,4] Production rates per unit volume vary with latitude and altitude. Latitude dependences are due to the earth's magnetic field shielding lower latitude regions from low-energy protons. Altitude dependences are from decreases in atmospheric density with increasing altitudes. Production rates also vary with time. These time variations can be correlated with the 11-year solar cycle during which the number of sunspots decreases and then increases. This increase and decrease in sunspot number is correlated with the solar activity. In times of high solar activity (sunspot maximum) plasma clouds are more likely to be expelled from the solar surface. As these clouds travel through interplanetary space, they can magnetically shield the earth from intergalactic cosmic radiation. This shielding decreases the cosmic radiation intensity incident on the earth's atmosphere. Particles of lower magnetic rigidity (momentum to charge ratio) are more shielded; therefore, cosmic ray spectrums are harder during sunspot maximum. Also, higher latitudes, which have lower cut-off rigidities, exhibit larger variations in cosmic ray intensities over solar cycles. A more detailed account of these variations can be found in Lal and Peters.[1-2]

Various investigators have estimated tritium production by cosmic rays. Varying degrees of corrections were made for latitude and time variations of cosmic ray intensity to obtain average global production rates. Values range from 0.14 to 0.90 tritium atoms/cm^2-s[1,3-7] with the better estimates lying between 0.20 and 0.25 tritium atoms/cm^2-s.[1-2]

Another method used to obtain average global natural tritium production rates is to measure tritium in water or rain samples and use geochemical balances to calculate production rates. An implicit assumption in this method is that all compartments in which tritium resides and exchanges are in equilibrium. This assumption allows the establishment of representative exchange rates between compartments. Another assumption is that tritium compartments are well mixed. Thus, sample concentrations are representative of concentrations in each compartment. If exchange rates are known for each compartment average global production rates can be determined.

Complications arise when this method is applied. Tritium compartments are not truly in equilibrium but exhibit fluctuation about some average state. Establishment of this state can only be determined by numerous measurements over long periods of time. Rough estimates of this state, however, are available. Furthermore, the assumption that each compartment is well mixed is erroneous. Users of the method attempt to circumvent this problem by estimating average concentrations in each compartment and determining average residence times. To do this accurately requires a knowledge of pathways for tritium movement throughout compartments. In most compartments, tritium's behavior is only partially or poorly known. In cases where the behavior is known interactions are so complicated that accurate models are unwieldy. Furthermore, data are not sufficient to allow assumptions that would simplify the model.

It is not surprising, therefore, that estimates using this method lead to a wide range of values. The range spans from 0.14 tritium atoms/cm^2-s to 2.0 tritium atoms/cm^2-s.[7-11] A major cause for the range is differing assumptions used for residence times in various compartments. The best estimate to date is 0.5 ± 0.3 tritium atoms/cm^2-s.[7]

Estimates of the average global production rate obtained from geochemical balances have often been much larger than those estimates derived from cosmic ray interactions with the atmosphere. This discrepancy led some investigators to propose that tritium can be accreted directly from solar radiation emitted during solar flares. Evidence presented in support of this position is excess tritium found in Discoverer 17 satellite material which was in orbit two days following the solar flare of November 12, 1960.[12-13] It has subsequently been suggested, however, that this tritium can be accounted for by alpha particle stripping in the satellite material.[14] Although the best estimate of the average global production rate (0.5 ± 0.3 tritium atoms/cm^2-s) determined by geochemical balances is greater than the average global production rate (0.20-0.25 tritium atoms/cm^2-s) obtained for cosmic ray interactions, the latter value falls within the error of the former. Thus, occurrence of direct solar accretion cannot be resolved at the present time.

3. LIGHT WATER REACTORS (LWRs)

3.1 Production

Tritium is produced in the fuel, control rods, burnable poisons (i.e., initial core neutron absorbers), and coolant of LWRs. Tritium production in the fuel is predominantly due to ternary fission. Tritium yields from the fissioning of several atoms of interest have been measured and are given in Table 3.1. Yields from fission of ^{235}U apparently increase as energies of neutrons initiating the fission increase. The dependence on energy, however, is not well established. In addition, thermal yields are rather uncertain. Therefore, yields from the fission of ^{235}U under reactor conditions, where a spectrum of neutron energies applies, can only be approximated. Yields of tritium from thermal fission of ^{233}U and ^{239}Pu are even less well established, and their dependence on neutron energy has not been determined. Based on current data, it is generally assumed that yields from the fission of ^{239}Pu are twice those of ^{235}U for fission by thermal neutrons. Because of these uncertainties, it is not surprising to find large variations in the predictions of tritium production in LWRs by ternary fission. Estimates range from 0.422 to 0.921 TBq/MW(e)-year.[31-36]

In addition, tritium is produced in LWRs by a number of neutron activation reactions. The more important of these are given in Table 3.2 along with their effective cross-sections. In boron carbide control rods, most of the tritium is produced by neutron activation of ^{10}B. Total production in these control rods has been estimated to be 0.074 TBq/MW(e)-year in relatively new rods increasing to 0.74 TBq/MW(e)-year in well-burned rods.[33] Increases with rod age are due to buildup of ^{7}Li in the rods. The given ranges are supported by the work of Ray, Wooten, and Barnes[34] and by Smith and Gilbert.[31] The average

Table 3.1. Ternary fission yields for tritium

Fissile nuclide	Neutron energy	Fission yield (Tritium atoms/fission)	Reference
^{235}U	Thermal	0.95×10^{-4}	15
^{235}U	Thermal	0.80×10^{-4}	16
^{235}U	Thermal	0.99×10^{-4}	17
^{235}U	Thermal	0.85×10^{-4}	18
^{235}U	Thermal	1.24×10^{-4}	19
^{235}U	Thermal	1.32×10^{-4}	20
^{235}U	Thermal reactor	1.26×10^{-4}	21
^{235}U	Thermal reactor	1.44×10^{-4}	22
^{235}U	27-112 fJ	2×10^{-4}	18
^{235}U	32-128 fJ	2.2×10^{-4}	23,24
^{233}U	Thermal	0.91×10^{-4}	25
^{233}U	Thermal	0.68×10^{-4}	26
^{233}U	Thermal reactor	1.11×10^{-4}	27
^{239}U	Thermal	1.8×10^{-4}	25
^{239}U	Thermal	1.65×10^{-4}	28
^{239}U	Thermal	1.34×10^{-4}	29
^{239}U	Thermal	1.39×10^{-4}	20
^{239}U	Thermal	1.75×10^{-4}	30

Table 3.2. Effective microscopic cross-sections for reactions producing tritium or precursors[a]

Reaction	σ_{eff} (10^{-28} m^2)
$^{2}H(n,\gamma)T$	0.000316
$^{6}Li(n,\alpha)T$	693
$^{7}Li(n,n\alpha)T$	0.0516
$^{10}B(n,\alpha)Li$	3060
$^{10}B(n,2\alpha)T$	1.27

[a] From ref. 37.

production is ∿0.35 TBq/MW(e)-year over a 15-year life.[31] Pressurized water reactors (PWRs) are presently using silver, indium, and cadmium alloy control rods in which tritium is not produced. Tritium is also produced in boron curtains which are used to control excess reactivity of initial cores of boiling water reactors (BWRs). Production is around 0.074 TBq/MW(e)-year.[33] These curtains are being replaced in newer BWRs with fuel tubes spiked with reactivity control materials. Tritium production in burnable poisons, used in PWRs, is around 0.029 TBq/MW(e)-year.[32]

In coolants, tritium is produced by activation of boron dissolved in coolants for reactor control (PWRs), activation of lithium present as an impurity or added as a lithium based cation for pH control, and from activation of deuterium. For PWRs, these mechanisms produce 0.026 to 0.032 TBq/MW(e)-year.[32] For BWRs, the rates are approximately 0.00044 TBq/MW(e)-year.[31] Thus, the production rates for typical BWRs and PWRs are: (1) BWRs, 0.78-1.3 TBq/MW(e)-year plus 0.074 TBq/MW(e) from initial core boron steel curtains, and (2) PWRs, 0.44-0.96 TBq/MW(e)-year plus 0.029 TBq/MW(e) from initial core burnable poisons.

3.2 Transport and Release

Tritium can be released at reactor sites only after entering reactor coolants. Releases of tritium from fuel elements to coolants are a function of cladding materials and core temperatures.[38] Estimates of ternary fission tritium releases from UO_2 fuels with stainless steel claddings are 80%,[32,34] supported by data from the Portable Medium Power Plant (PM-3A); 16-32% (depending on assumed ternary fission yields), supported by data from the Connecticut Yankee Reactor,[39] and 1%, supported by data from the Humboldt Bay and Big Rock Point Reactors.[31] Release estimates from rods with zircaloy claddings range from 0.013% to 1.0% of fission production tritium.[31,32,40-42]

Data relating to releases of tritium from control rods, burnable poisons, and burnable curtains are sparse. It is generally accepted that there is no evidence of releases from boron control rods or from boron-steel curtains used in BWRs; however, releases from B_2O_3 burnable poisons used in PWRs are estimated to be between 30 and 80%.[32]

In coolants, tritium atoms exchange with protium atoms in water molecules to form tritiated water molecules. As a result, most tritium releases from LWRs are HTO vapor or HTO liquid. The average proportion of total tritium releases that are released to the atmosphere as vapor or gas is larger for BWRs (40-44%) than PWRs (4-13%). These higher proportions may be due to variations in waste treatment schemes of individual reactors rather than to inherent characteristics of the two reactor types since percentage releases to the atmosphere in either BWRs or PWRs vary widely. Total releases, however, do depend on reactor type being larger in PWRs due to their use of soluble boron chemical shims and releases from burnable poisons. Measured releases in liquid and airborne effluents of some BWRs and PWRs are given in Tables 3.3 and 3.4. For PWRs, the average release in 1974 was ∿0.056 TBq/MW(e)-year while the average release for BWRs was ∿0.0041 TBq/MW(e)-year. It is also apparent from Table 3.3 that liquid releases have steadily decreased for both reactor types over the 1970 to 1977 period. In 1977, liquid releases were 0.0035 TBq/MW(e)-year for PWRs and 0.0011 TBq/MW(e)-year for BWRs. Data are insufficient to make hard conclusions for trends in atmospheric releases. Nevertheless, data for 1973 and 1974 indicate slight increases in atmospheric releases from PWRs and essentially constant atmospheric releases from BWRs.

4. HEAVY WATER REACTORS (HWRs)

4.1 Production

Tritium is produced in HWRs by the same mechanisms that produce tritium in LWRs. Contributions to the overall production, however, are vastly different for each mechanism. In heavy-water cooled, heavy-water moderated reactors, which are of popular design, neutron activation of deuterium in heavy water is the largest contributor despite a low reaction cross-section. High neutron fluxes and large amounts of deuterium in coolants and moderators account for the large production. Tritium production in coolants is not precisely known.

Table 3.3. Normalized liquid releases for light water reactors [TBq/MW(e)-year x 10^2][a]

Reactor	1970	1971	1972	1973	1974	1975[d]	1976[d]	1977[d]
BWRs								
Dresden 1	0.11[e]	0.44	1.3	1.1	1.7	0.013	0.0007	0.0048
Big Rock Pt.	4.59	1.0	0.89	1.5	0.48[b]	0.063	0.31	0.78
Humboldt Bay	0.52	0.70	1.1	3.96	2.7	1.7	2.1	
Lacrosse	4.85	14.1	16.1	16.9	11.3	15.1	7.36	15.5
Oyster Creek	0.20	0.17	0.44	0.32	0.12	0.17	0.31	0.19
Nine Mile Pt.	0.34	0.13	0.28	0.44	0.19[c]	0.30	0.019	0.028
Dresden 2,3	0.81	0.31	0.10	0.096	0.11	0.33	0.074	0.018
Millstone Pt. 1		0.11	0.21	0.063	0.22	0.67	0.17	0.030
Monticello		0.013	7.4×10^{-7}					
Quad Cities 1,2			0.041	0.078	0.14	0.24	0.20	0.10
Pilgrim 1			0.17	0.0034	0.17	0.23	0.63	0.41
Vermont Yankee				0.0036			0.017	0.0081
Peach Bottom 2,3					0.063	0.096	0.21	0.26
Brown's Ferry 1,2,3					0.024	0.17	0.041	0.063
Cooper Station					0.031	0.081	0.089	0.078
Brunswick 1,2						0.070	0.078	0.059
Duane Arnold						0.0048	0.0044	0.0024
J. A. Fitzpatrick						0.078	0.035	0.030
Edwin Hatch						0.067	0.070	0.10
Total energy prod [MW(e)-year]	1.01×10^3	1.94×10^3	3.27×10^3	4.37×10^3	5.08×10^3	6.33×10^3	8.13×10^3	9.52×10^3
Total tritium released [TBq x 10^2]	588[f]	753	1228	1219	1144	1737	1262	1025
Overall normalized release [TBq/MW(e)-year x 10^2]	0.59	0.41	0.37	0.28	0.23	0.27	0.16	0.11
PWRs								
Yankee Rowe	38.1	36	40.3	21	11	6.8	4.0	4.3
Indian Pt. 1	34	17	15		19			
San Onofre	48.5	42.6	40.0	57.4	40.0	38.5	42.2	23.6
Connecticut Yankee (Haddam Neck)	64.4	42.9	44.4	52.2	17	44.4	38.9	53.3
R. E. Ginna	1.6	1.7	1.6	2.7	3.0	2.7	3.5	1.1
Point Beach 1,2		2.5	4.1	3.2	4.1	3.8	2.9	4.1
H. B. Robinson		1.5	3.0	3.7	3.0	4.9	6.5	5.1
Palisades			3.6	2.5	3.4	0.48	0.10	0.33
Maine Yankee			0.63	1.5	2.0	1.2	1.9	0.93
Surry 1,2			0.44	2.0	1.3	1.6	3.3	1.4
Turkey Pt. 3,4				2.3	2.4	3.0	3.0	3.6
Indian Pt. 2				2.6	0.48	0.59	5.5	1.3
Ft. Calhoun				0.85	1.7	1.7	1.8	1.8
Prairie Island 1,2				1.2×10^{-4}	3.3	0.0028	0.00067	7.5
Oconee 1,2,3				0.96	2.0	7.1	5.2	4.5
Zion 1,2					1.5	3.7	2.8	2.3
Arkansas 1					1.4	3.4	2.0	1.7
Kewaunee					1.9	2.6	1.7	2.7
Three Mile Island					2.0	2.5	1.3	1.0
Calvert Cliffe 1						2.0	1.4	2.0
Cook 1						0.41	0.93	1.9
Millstone Point 2						1.3	1.9	1.6
Rancho Seco.						3.4		0.00048
Beaver Valley 1							0.44	1.1
Salem							0.085	4.6
St. Lucie							4.0	1.4
Trojan							0.52	1.6
Crystal River								1.4
Davis Besse								0.56
Total energy prod. [MW(e)-year]	1.34×10^3	2.38×10^3	2.87×10^3	5.20×10^3	7.10×10^3	1.45×10^4	1.50×10^4	2.06×10^4
Total tritium released [TBq x 10^2]	5.25×10^4	4.92×10^4	4.48×10^4	4.18×10^4	4.00×10^4	7.25×10^4	6.29×10^4	7.18×10^4
Overall normalized release [TBq/MW(e)-year x 10^2]	39.2	20.7	15.6	8.0	5.6	5.0	4.2	3.5

[a] Taken from ref. 43 and 44.

[b] Disagreement exists between ref. 43 and 44.

[c] Factor of 10 error in reference as value gives impossible estimate of efficiency.

[d] Values determined by determining efficiency using both references then using the release and energy generation data from references.

[e] Read as 0.0011 TBq/MW(e)-year.

[f] Read as 5.88 TBq.

Table 3.4. Normalized tritium release to the atmosphere for light water reactors

	Normalized release [TBq/MW(e)-year]	
	1973	1974
BWRs		
Big Rock Pt.	0.0596	0.037
Humboldt Bay	0.00148	0.0015
Lacrosse	0.0829	0.018
Oyster Creek	0.000029	0.000036
Nine Mile Pt.	0.0025	0.0016
Dresden 2,3	0.00037	0.0056
Millstone Pt.	0.00029	0.00070
Quad Cities 1,2	0.0011	0.0011
Pilgrim 1	0.0011	0.0013
Vermont Yankee	0.00018	0.00029
Peach Bottom 2,3		0.00034
Browns Ferry		0.00006
Cooper Station		0.000003
Total energy prod [MW(e)-year]	4.34×10^3	5.8×10^3
Total tritium released [TBq]	8.03	8.95
Overall normalized release [TBq/MW(e)-year]	0.0019	0.0018
PWRs		
Yankee Rowe	0.0026	0.0014
Indian Pt. 1		0.000089
San Onofre	0.0377	0.0096
Connecticut Yankee (Haddam Neck)	0.0067	0.00000085
R. E. Ginna	0.00010	0.000056
Pt. Beach 1,2	0.0014	0.0021
H. B. Robinson	0.00021	0.0035
Palisades	0.000024	0
Maine Yankee	0.00018	0.00063
Surry 1,2	0.0019	0.0033
Turkey Pt. 2,3	0.00028	0.00037
Indian Pt. 2	0.0019	0.0020
Ft. Calhoun	0.00018	0.00010
Prairie Island		0.00089
Oconee 1,2	0.0018	0.051
Zion 1,2		0.013
Arkansas 1		0.000017
Kewaunee		0.0022
Three Mile Island		0.0020
Total energy prod [MW(e)-year]	5.09×10^3	7.10×10^3
Total tritium released [TBq]	16.5	54.4
Overall normalized release [TBq/MW(e)-year]	0.0032	0.0077

[a] Taken from ref. 43.

Kouts and Long[33] give the production in coolants as 22.2 TBq/MW(e)-year. This value is not inconsistent with an estimate of 88.8 TBq/MW(e)-year for total production from activation of deuterium in HWRs.[45] Bonka[46] estimated that 5.55 TBq/MW(e)-year is produced in water by activation of ^2H; however, it is not clear whether this estimate was for coolants, moderators or both. It would appear, however, to be for coolants. Using this assumption, estimates for production in coolants by activation of deuterium range from 5.55 to 22.2 TBq/MW(e)-year, whereas production in coolants and moderators is estimated to be 88.8 TBq/MW(e)-year.

The next most important contributor to tritium production in HWRs is ternary fission. An estimate of 0.74 TBq/MW(e)-year for this mechanism is commonly given.[43,46] Modern HWRs use zircaloy-clad fuel rods; so, releases from fuel elements to coolants is certainly less than 1% and generally less than 0.1%.[31-32,40-42] These release data are for LWRs; however, operating temperatures of HWRs and LWRs are similar. Therefore, these data should be applicable to HWRs. Assuming that 1% of the ternary fission produced tritium is released to coolants, ∿0.0074 TBq/MW(e)-year of tritium is available for release from HWRs.[43] Production by other mechanism (i.e., activation of ^6Li impurities in fuels, ^{10}B impurities in fuels, and boron in absorber rods) are negligible (Table 4.1). Activation of lithium can become significant, however, when LiOH is used to control coolant water chemistry.[48] An estimation of tritium production for this situation, however, was not found.

4.2 Transport and Release

Tritium produced in or released to coolants or moderators is available for release at reactor sites. In most designs, heavy-water coolants and heavy-water moderators are separate systems. In a very popular design used extensively in Canada, coolants are contained in pressurized tubes (0.68 MPa) and reach temperatures of 290°C.[49] Moderators are contained in tubes concentric to pressurized coolant tubes and at low temperatures and pressures. These tubes are in a tank

Table 4.1. Tritium production in heavy water reactors[a, b]

Mechanism	Production [TBq/MW(e)-year]
Fission	0.74
^{6}Li in fuel (0.05 ppm)	0.030
^{10}B in fuel (0.05 ppm)	1×10^{-6}
^{2}H in water	
Coolant	5.55-22.2
Coolant and moderator	88.8
Absorber rod	0.037

[a] Taken from ref. 33, 45, and 46.

[b] Thermal efficiency of the CANDU Reactor is 29.1% (ref. 47).

of heavy-water moderator water. This tank has a helium gas cover. Coolants pass from this tank to heat exchangers where the heat is transferred to ordinary steam systems.

Because coolant and moderator systems are separate, releases from these systems must be considered separately. Most tritium released from HWRs is released from coolant systems. This is reasonable since coolants are at higher pressures and temperatures than moderators; so, leakage and diffusion from coolants can be expected to be higher. Pathways include permeation and leakage from coolant piping and diffusion through heat exchanger walls into secondary coolant systems. Tritium escapes from secondary coolant systems by pipe leakage, diffusion into condenser water, or blowdown. Releases should appear primarily as tritiated water.

Tritium releases from coolants and moderators are very dependent on leakage rates. There are two incentives to keeping leakage rates very low. The obvious incentive is to reduce releases of radioactivity. The second incentive is based on economics. Heavy water is very expensive; so, optimum release rates are lower than those for inexpensive coolants.

As mentioned above, leakages from coolants are larger than those from moderators. Generally, coolant leakages are sufficiently large that tritium concentrations in coolants reach equilibrium. On the other hand, leakages from moderators are so small that tritium concentrations in moderators continue to increase over the reactor life. Moderator concentrations can become sufficiently large to present health hazards to operating personnel. Ontario Hydro has, therefore, begun a displacement program in which portions of contaminated moderator water are removed from operating reactors to newly opening heavy-water facilities and replaced with virgin heavy water. The goal is to keep tritium concentrations in the moderator water below a guideline value of 0.074 TBq/kg.[50] Therefore, releases from moderators should increase over time until some moderator water is displaced at which time releases should fall sharply and then increase over time. This behavior assumes a constant moderator leak rate. Releases by leakage should be as tritiated water.

Another mode of tritium releases from moderators is tritium entrainment in the helium cover gas and escape through ventilation or leakage. Tritium's behavior may be dominated by the following effects:

1. Tritium formed from D_2O by neutron activation can be expelled from D_2O molecules upon formation. Expelled tritium atoms can then combine with other deuterium atoms and escape from the water as DT gas.[51]

2. Tritium formed can achieve thermodynamic isotopic equilibrium with surrounding D_2O and D_2 molecules. The equation:

   $$D_2O + DT \rightleftarrows DTO + D_2 \ ,$$

   has an equilibrium constant of approximately 1.5 at reactor temperatures. Achievement of equilibrium is slow but may be catalyzed by isotopic exchange.[51]

3. The probability of D radical formation is approximately twice the probability of T radical formation; therefore, hydrogen molecules formed are depleted in tritium.[51]

The first effect would cause tritium to be extracted from moderator water. The latter two would cause tritium to be retained in moderator water. An investigation of tritiated heavy-water chemistry indicates that the latter two possibilities are preferred so that little tritium is lost as gas through helium cover gases.[51]

No detailed modelling of tritium transport in HWRs could be found in the literature. Theoretical predictions of tritium releases from reactors are very simplified being based on observed or expected coolant releases (0.5-2.0% of the total D_2O amount/year). Using 0.5% release/year tritium releases from HWRs range from 0.1 TBq/MW(e)-year in the first year to 0.74 TBq/MW(e)-year in the tenth.[43] Approximately 80% of the tritium is released through stacks to the atmosphere.

Release data from operating reactors are scarce. Data that could be found are presented in Table 4.2. In addition, Gorman and Wong[45] have determined that the average total release was 0.89 TBq/MW(e)-year

Table 4.2. Tritium releases from heavy water reactors

Station	Year	Electrical output [MW(e)]	Atmospheric release[a]		Liquid release[a]		Normalized atmospheric release [TBq/MW(e)-year]	Normalized liquid release [TBq/MW(e)-year]
			(TBq)	(%)	(TBq)	(%)		
Pickering	1971	344	98.8	86.9	14.9	13.1	0.29	0.044
	1972	658	529	93.5	36.5	6.5	0.803	0.056
	1973	1630	1350	87.7	190	12.3	0.829	0.11
	1974	~1614	918	63.3	533	36.7	0.570	0.33[b]
	1975		752	65.5	396	34.5		
	1976		895	79.9	225	20.1		
	1977		1630	69.8	703	30.2		
Atucha	1974	109	8.03	70.6	3.34	29.4	0.074	0.031
	1975						0.14	0.1

[a] Values taken from refs. 43 and 46. Where values conflict, values from ref. 43 are taken.

[b] Value differs slightly from that of ref. 43. The value in this reference was not consistent with other data of 1974.

for the Pickering Station from 1971-1977. It is evident that releases from Pickering vary widely from year to year. This variation is largely due to variation in coolant leakage rates. Increases in tritium releases over the first few years is largely due to generating capacity increases and partly to increases in tritium concentrations in the coolant. The data are not broken down into releases from individual reactor components or into releases as elemental tritium (HT or DT) and tritiated water. For reasons given above, most tritium released should be released as tritiated water. The majority is released to the atmosphere (63-94%); the rest is released with liquid effluents (6-37%).

5. HIGH TEMPERATURE GAS COOLED REACTORS (HTGRs)

5.1 Production

Tritium is produced in the fuel, graphite moderator and matrix material, helium coolant, and boron control elements of HTGRs. One mechanism for production is ternary fissioning of the fuel. A popular fuel cycle for HTGRs is the thorium-uranium 235 cycle in which power is primarily produced from energy liberated as ^{233}U and ^{235}U fission. Tritium produced by fissioning of these isotopes can only be approximated for several reasons. Firstly, yields from fission of ^{235}U by thermal neutrons is only approximately known as shown in Table 3.1. Secondly, yields from fission of ^{233}U by thermal neutrons is even more poorly established. Thirdly, dependences of yields on initiating neutron energies is unestablished. Fourthly, neutron flux spectrums are dependent on specific reactor designs and operating conditions. Finally, fractions of fission of ^{235}U, ^{233}U, and ^{238}U per unit of power produced are dependent on relative concentrations of these isotopes within HTGRs which, in turn, is dependent on reactor designs. Productions of tritium for specified HTGR designs have been estimated. A number of these estimates are given in Table 5.1. Estimates can be converted from MW(t) to MW(e) using a thermal efficiency of 40%.[54] Thus, ternary fission is estimated to produce from ∿0.28 TBq/MW(e)-year to ∿0.61 TBq/MW(e)-year.

Table 5.1. Comparison of calculated tritium production rates for a 1000 MW(t) high temperature gas cooled reactor (TBq/year)

	Compere[a, d]		Heire[b, d]		Forsyth[c, d]		Fischer[e]	
Production mechanism	Yield	Assumptions	Yield	Assumptions	Yield	Assumptions	Yield	Assumptions
Ternary fission	114	Fission yield: 1×10^{-4} ^3H/fission	137	Fission yield: 1×10^{-14}	245	Fission yield: 1.3×10^{-4}	178	$^{235}U/Th = 1/10$ Fission yield: 1.1×10^{-4} for ^{233}U 1.3×10^{-4} for ^{233}U
Lithium-6 activation	47.4	Lithium impurity: 0.05 ppm	34.7	Lithium impurity: 0.01-0.05 ppm	29.3	Lithium-6 impurity: 0.06 ppm		
Boron-10 activation	9.7		11.3		Ignored			
Helium-3 activation	21.8	Helium-3 content: 0.2 ppm 4-6% He in core	45.5	Helium-3 content: 0.2 ppm 6% of He in core	82.9	Helium-3 content: 0.2 ppm 20% of He in core		

[a] Slow neutron flux - 6×10^{13} n/cm^2-s, fast neutron flux - 3.6×10^{13} n/cm^2-s, σ_{He-3} - 2280 barns, σ_{Li-6} - 408 barns, σ_{B-10} - 1630 barns.

[b] Neutron flux - 1×10^{14} n/cm^2-s, σ_{He-3} - 2800 barns, σ_{Li-6} - 463 barns.

[c] Neutron flux - 2.8×10^{13} n/cm^2-s, σ_{Li-6} - 5400 barns, Pu-239 fission yield - 2.3×10^{-4}, Pu-241 fission yield - 2.6×10^{-5}, low enriched fuel (7%).

[d] Taken from ref. 52.

[e] Taken from ref. 53.

Tritium is also produced by neutron activation reactions. Predominant neutron activation reactions leading to tritium formation in HTGRs are shown in Table 5.2. In addition, a chain reaction on ^{12}C may make a significant contribution to tritium inventories.[55] The production in various loci of reactors by these reactions can only be approximated for reasons similar to those given above [i.e., (1) poorly known reaction cross-sections, (2) poorly known energy dependence of reaction cross-sections, (3) poorly known isotope concentrations that vary with reactor design and time, (4) neutron flux spectra that vary with reactor design and operating conditions, and (5) coolant makeup and refueling rates that vary with reactor]. The fifth item is important since refueling and coolant makeup replenish the parent isotope. Estimates of tritium production have been made using reactor parameters that are believed to be realistic (Table 5.1). Helium, the primary coolant in HTGRs, contains small amounts of ^{3}He whose abundance depends on the source of the gas. Helium obtained from natural gas wells, which is currently being used to cool HTGRs, has ^{3}He compositions of 0.5×10^{-5} to 3×10^{-5}%.[56] Helium attained from air has higher ^{3}He contents (1.3×10^{-4}%).[56] Tritium production rates are proportional to ^{3}He concentrations. In leak tight systems, tritium production rates eventually become negligible as ^{3}He is depleted; however, helium leakage will occur. Coolant makeup helium replenishes ^{3}He so that equilibrium production rates will be obtained. Leakages may be sufficient to make production by ^{3}He activation the second most prominent source of tritium in HTGRs.

Tritium production by activation of lithium is approximately equivalent to that from activation of ^{3}He.* Lithium is primarily present in HTGRs as impurities in graphite. Lithium contents of AVR Jurlich graphites have been measured and found to vary between 0.007 ppm and 0.25 ppm.[53] Residence times of graphite in HTGRs are sufficiently

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*Some of the estimates given under lithium production include tritium from a C^{12} initiating chain reaction.

Table 5.2. Essential neutron activation reaction leading to tritium production in high temperature gas cooled reactors

	Fischer[a]		Gainey[b]	
Reaction	Energy (pJ)	Cross-section (barn)	Energy (pJ)	Cross-section (barn)
$^{3}He(n,p)^{3}H$	Thermal	5327	Thermal	5400
$^{6}Li(n,\alpha)^{3}H$	Thermal	953	Thermal	953
$^{7}Li(n,\alpha n)^{3}H$	0.5-1	90×10^{-3}	>0.029	153×10^{-3}
$^{7}Li(n,\alpha n)^{3}H$	>1	400×10^{-3}		
$^{10}B(n,\alpha)^{7}Li$	Thermal	3837	Thermal	4010
$^{10}B(n,2\alpha)^{3}H$	0.2-0.8	50×10^{-3}	>0.029	50×10^{-3}
$^{10}B(n,2\alpha)^{3}H$	>0.8	110×10^{-3}		

[a] Taken from ref. 53.

[b] Taken from ref. 52.

long for 100% of the ^{6}Li to be spent. Production rates, therefore, are initially high and decrease as ^{6}Li concentrations decrease. Total production is dependent on refueling rates.

In predictions where neutron fluences and average cross-sections have been estimated and used, tritium production by boron activation ranks fourth as a source of tritium (Table 5.1). Boron is used as a neutron absorber and is present as a burnable poison in fuel elements and absorber rods. Another estimate, therefore, has been made by using the excess reactivity of an unrodded core to estimate neutron absorption in boron. Using this method, boron production jumps from fourth to second as a source of tritium.[57]

5.2 Transport and Release

Under normal conditions, tritium produced in HTGRs must enter reactor coolants prior to escape to the environment. For fission produced tritium the path to reactor coolants is rather tortuous. In pebble-bed type reactors, escape paths include escape from fuel kernels, permeation through fuel particle coatings, permeation through graphite matrix materials in which coated fuel particles are embedded, and permeation through fuel free graphite coatings that surround graphite matrices. In prismatic type reactors, escape paths include releases from kernels, permeation through particle coatings, permeation through graphite compact matrices, and permeation through graphite sleeves into reactor coolants. Fuel kernels retain negligible amounts of tritium since tritium solubility in kernels is low and tritium diffusion coefficients through the kernel are high.[58] Negligible tritium retention in kernels has been born out in some studies of tritium distributions in irradiated coated particles.[59-60] Nevertheless, there is also some evidence that retention in kernels may be significant and play a role in tritium retention.[61] Tritium that escapes particle kernels has been found to be well retained in TRISO and BISO coatings of fissile and fertile particles. Lange and Walter[62] estimated that 2.5% of ternary fission tritium will be released for temperatures up to expected peak fuel temperatures. Burnups were typical of those in fertile particles. Other data that can be applied to fertile particles have shown retentions of 73% at 750°C and 95% at 950°C.[61] Data typical of fissile particles (i.e., TRISO UC_2 particles) indicate 34% retention at 1250°C (expected peak temperature) and 74% at 850°C.[61] Another study supports these results though particle and irradiation data are not sufficient to draw solid conclusions.[63] Larger releases at peak temperatures (1250°C) compared to lower temperatures (850°C) can be attributed in part to large temperature coefficients found for diffusion of tritium in pyrocarbon.[52]

Behavior of tritium produced in boron control rods (prismatic and pebble bed) and burnable poison spines in fuel elements is not well known. Data regarding tritium profiles in poison spines indicate that tritium is immobile[57] and not available for release. It has been suggested, based on this observation, that tritium produced in boron carbide control rods is also immobile. Some measurements, however, have indicated that retention of tritium in boron carbide control rods may not be so complete. Tritium retention in control rods apparently has a complex dependence on temperature but is independent of burnup. The retention varied from 20-80% for temperatures between 538 and 871°C.[52] Thus, tritium releases from boron control rods to coolants under reactor operating conditions could conceivably be significant. This tritium is released to reactor graphites or primary coolants. Tritium in HTGR graphites and tritium in helium coolants are governed by tritium's behavior at graphite-helium interfaces. Studies have been performed to elucidate this behavior. Kinetically, the behavior is governed by diffusion-adsorption-desorption processes. Diffusion is the long-term rate controlling mechanism. Diffusion rates are higher for fine grain graphites than for coarse grain graphites.[60] Both, however, have equivalent activation energies. Based on this mechanism and low partial pressures of tritium in HTGRs helium coolants, tritium would be expected to move from graphites to coolants. Tritium behavior in graphitic materials, however, appears to be influenced strongly by thermodynamic stability. Tritium has higher solubilities in graphites than in helium coolants.[60] Extrapolation of experimental results to equilibrium conditions indicate that the ratio of tritium concentrations in graphite to those in helium coolants is $\sim 10^3$ in unirradiated graphite and 7×10^3 in irradiated graphite.[60] Reactor graphites with oxide surfaces have been found to have higher tritium contents than those whose surface is unoxidized.[53,60] Surface oxidation increases adsorption.[53] Increased adsorption increases the diffusion rate; thereby, tritium absorption is increased. Oxidized samples, therefore, are closer to equilibrium conditions than unoxidized samples.

Graphite is not the only sink for tritium in primary coolants.[52,59-60] Other sinks include permeation and leakage through primary coolant pipings into reactor containments, permeation through steam generator walls into secondary coolants, and removal from primary coolant purge streams by coolant purification systems. Tritium behavior at these sinks is not sufficiently defined to allow detailed modelling of tritium transport in HTGRs. Nevertheless, some rough estimates have been made for tritium behavior and release.

Tritium releases from reactors are very dependent on helium purification systems. A system currently being considered for large HTGRs is shown in Fig. 5.1. It is expected to remove essentially all tritium from purge streams,[54] and to reduce tritium inventories in helium coolants by a factor of 3 (2×10^2 TBq to 0.2 TBq in the Fort Saint Vrain reactor).[35] The system includes a chiller to remove large amounts of water (including any tritiated water) followed by a molecular sieve which absorbs water that passes through the chiller. Gaseous tritium, which constitutes the bulk of tritium in primary coolants, is primarily absorbed on titanium sponge beds and will be stored permanently at the facility. Compere[55] has estimated that $\sim$0.017 TBq/MW(e)-year of tritium, averaged over six years, will be removed by HTGR purification systems and buried. Gainey[52] obtained a much higher estimate of 0.244-0.256 TBq/MW(e)-year. Goodjohn[54] has given an estimate of 0.11 TBq/MW(e)-year. The six-month accumulation of tritium as tritiated water on charcoal beds of the 330 MW(e) Fort Saint Vrain reactor is expected to be $\sim$0.11 TBq.[35]

Tritium retained on charcoal delay beds is released through the stacks when delay beds are regenerated. At the Fort Saint Vrain reactor

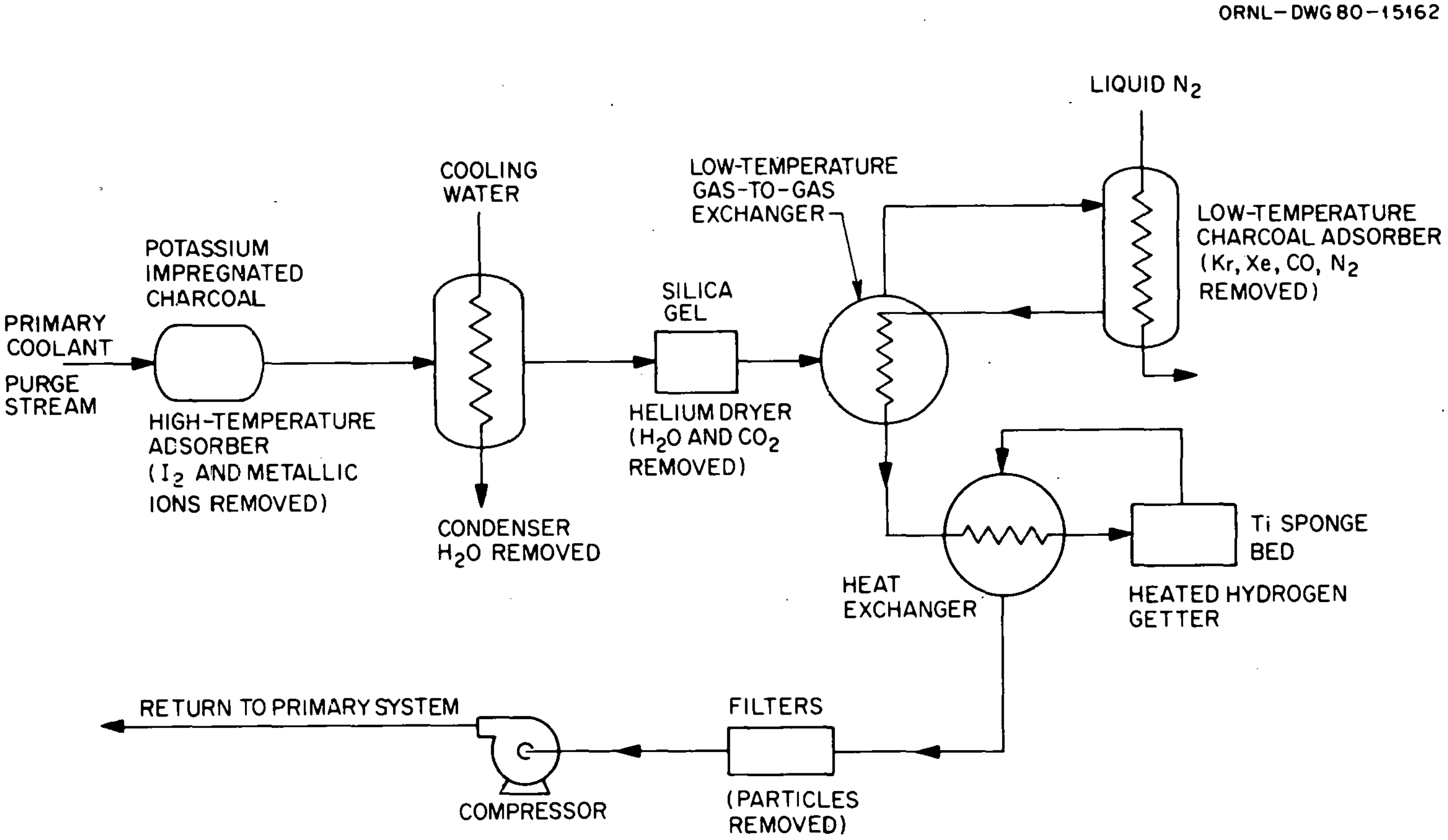

Fig. 5.1. Helium purification system proposed for high temperature gas cooled reactors.

delay beds are expected to be regenerated twice a year releasing 0.22 TBq of tritium.[35] This release corresponding to 6.7×10^{-4} TBq/MW(e)-year should be tritiated water. In addition, one turnover of primary coolant is expected to be released through the stacks. An additional 0.22 TBq of tritium[35] corresponding to another 6.7×10^{-4} TBq/MW(e)-year is released in this manner. This tritium should primarily be in the form of hydrogen molecules. Goodjohn[54] has presented somewhat lower releases from coolant leakages of 1.7×10^{-4} TBq/MW(e)-year. Therefore, a total of 8.5×10^{-4} - 1.3×10^{-3} TBq/MW(e)-year is expected to be released from stacks. Of this 22-50% is gaseous tritium, while 50-78% is tritiated water vapor.

In addition, some tritium will permeate through steam generator tubes, enter secondary coolants, and be released by blowdown, leakage or permeation into condenser water. Once tritium enters secondary coolants, it quickly exchanges with protons in water molecules forming tritiated water. It is released in this form. Though Goodjohn[54] has implied that tritium released with liquid effluents would be negligible due to its removal in the helium purification system, estimates by Compere, Fried, and Nestor[55] and by Gainey[52] indicate that this mode of release is predominant. Results of Compere, Fried, and Nestor[55] indicate releases of 0.012 TBq/MW(e) for the first year and 7.0×10^{-3} TBq/MW(e)-year averaged over six years. Their assumptions regarding tritium releases to coolants appear to be low; therefore, their estimates may be low. Gainey's results set liquid releases at 0.030 TBq/MW(e)-year for 1000 MW(e) HTGRs. Furthermore, his results indicate that releases do not increase linearly with power ratings. For example, releases are 0.024 TBq/MW(e)-year for 2000 MW(t) HTGRs, 0.035 TBq/MW(e)-year for 3000 MW(t) HTGRs and 0.037 TBq/MW(e)-year for 4000 MW(t) HTGRs.

From the presented data, the amount of tritium expected to be released from HTGRs is between $\sim 7.8 \times 10^{-3}$ and 0.031 TBq/MW(e)-year. Distributions to different effluent streams are 84-97% as liquid tritiated water, 2-8% as tritiated water vapor, and 0.6-8% as tritium gas.

6. LIQUID METAL FAST BREEDER REACTORS (LMFBRs)

6.1 Production

Tritium is primarily produced in LMFBRs by ternary fission and by neutron activation reactions with boron in B_4C control rods. A small amount is also produced by activation of lithium impurities in fuels. Tritium production in LMFBRs will be larger than that in LWRs for several reasons. First, neutron activation cross-sections for reactors producing tritium increase with neutron energy, and LMFBRs have harder neutron spectrums than LWRs. Second, there is experimental evidence that ternary fission yields increase with neutron energy. Third, larger percentages of power in LMFBRs as compared to LWRs are derived from fissioning of ^{239}Pu, which has high tritium yields compared to fissioning of ^{235}U.

Estimates of tritium production from ternary fission vary widely. A major reason is that ternary fission yields from fast fission of ^{239}Pu, the major ternary fission source, are poorly defined both in terms of thermal yields and dependences of yields on fission initiating neutron energies. The generally accepted yield for thermal fission of ^{239}Pu is 2×10^{-4} tritons/cm^2-s. Theoretical predictions have indicated that fast yields should be the same as thermal yields within ±10%.[64] Experimentally, fast fission yields as much as four times thermal fission yields have been reported for ^{235}U fission.[23-24,64] Therefore, yields from fast fission of ^{239}Pu may be larger than those from thermal fission. Tritium produced by ternary fission in LMFBRs has been estimated to be between 0.41 and 0.96 TBq/MW(e)-year depending on assumed yields.[64,67] Assumptions attributing different contributions to power from fission of ^{235}U, ^{238}U and ^{239}Pu also contribute to this range.

Other mechanisms of tritium production in LMFBRs are activation of boron in control rods and activation of lithium impurities in fuels. Extrapolating production rates by activation of boron in control rods of the FFTF and EBR-II to large power reactors may be erroneous since production rates depend upon the fuel cycle and amount of B_4C needed in cores for shim control.[65] For this reason, predictions based on existing designs are probably more appropriate. Calculated production rates based on the General Electric 1000 MW(e) follow on design give 0.3 TBq/MW(e) year from $^{10}B(n,2\alpha)^3H$ reactions and 0.093 TBq/MW(e)-year from $^7Li\ (n,n\alpha)^3H$ reactions.[65] Other processes for production in control rods are small. Production from lithium impurities in fuels have been estimated as $\sim$0.15 TBq/MW(e)-year.[65] Production from lithium impurities in coolants are small [$\sim 3.7 \times 10^{-3}$ TBq/MW(e)-year].

6.2 Transport and Release

Tritium must escape from fuel rods and control rods to be available for release from LMFBRs. Retention of tritium in fuel rods appears to be dependent on linear power levels at which fuels are irradiated but independent of burnups. Mixed oxide fuel pins irradiated at linear powers of 36-46 kW/m retained in the fuel matrix <1% of ternary fission produced tritium. Little or none was retained in trapped fission gases and stainless steel claddings.[68] Retentions in fuel matrices of 0.3 to 6.10% of ternary produced tritium have been found at ORNL.[69-72] When reported, linear power levels ranged from 33 to $\sim$46 kW/m. Retention in mixed carbide fuel matrices may be slightly lower. Observed values are <1%.[69,73-74]

The above values were obtained by dissolving irradiated fuels. Ebersole, Vroman, and Krusul[75] estimated retentions in EBR-II fuels by shutting off primary and secondary sodium coolant cold traps (which precipitate NaT) and noting rises in tritium concentrations in sodium. The results indicate that approximately 70% of ternary produced tritium was released from fuels. Fuels were metallic and had stainless steel claddings; whereas, commercial LMFBRs will have oxide or carbide fuels. Therefore, commercial LMFBR fuel pins are expected to release between 95 and 100% of tritium produced by ternary fission.

Evidence for retention in boron carbide control rods is somewhat ambiguous. It is evident that retention in control rods is a function of control rod temperatures, but different functions are indicated by the results. At EBR-II operating temperatures (550-600°C), all control rod produced tritium is retained in the B_4C matrix; however, when rods are heated to 900°C, approximately all control rod produced tritium is released within 20 hours. Most control rod produced tritium is retained at 700°C.[76] Another study indicated that 20% is retained around 900°C, 70-80% is retained between 700 and 750°C, and 40% is retained at ∿600°C.[77] Other analyses show total retention up to temperatures >800°C and 70% retention up to high temperatures (temperatures not specified).[78] Because of these discrepancies and indications that releases may be 100%, 100% release from B_4C control rods is generally assumed.

Once tritium escapes fuel elements and B_4C control rods, it becomes dissolved in primary sodium coolants. Two primary sodium coolant designs, the pool type and the loop type, are presently being considered; however, most development has focused on the pool type design. Therefore, the pool type design made for the Electric Power Research Institute (EPRI) by a team from Atomics International, Combustion Engineering, and Bechtel[79]* will be used to follow tritium through reactors. Some tritium precipitates as sodium tritide in primary coolant cold traps. Amounts trapped are very dependent on amounts of hydrogen dissolved in primary sodium coolants. This tritium is subsequently stored in adsorption bottles for burial. Tritium also becomes entrained in helium, the primary coolant cover gas. This helium and helium over spent-fuel storage pools are passed through radioactive gas purification systems which consist of duplicate, parallel lines that contain a series of two liquid nitrogen cooled charcoal delay beds. Delay beds in one of these lines are regenerated

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*A design for EPRI by a team from Westinghouse and Stone & Weber is similar except the cover gas for the primary coolant and fuel storage is argon and the tritium regenerated from the adsorber beds is monitored and released through the stack.[80]

while the other line is in operation. Tritium driven off delay beds is trapped in adsorption bottles for off-site disposition. Tritium passing through delay beds is recycled to reactor vessels. In addition, tritium can permeate primary containment walls and helium piping entering inert, nitrogen filled cells surrounding reactor vessels and piping. Of this tritium, that which does not decay is eventually released through HV plenums after passing through charcoal delay beds. Finally, tritium can permeate intermediate heat exchanger (IHX) walls and become dissolved in secondary sodium coolants.

Tritium in secondary sodium coolants behaves similarly to tritium in primary coolants. Some tritium precipitates as sodium titride in secondary coolant cold traps. As in primary systems, amounts of tritium precipitated are dependent upon amounts of hydrogen dissolved in secondary sodium coolants. Tritium also becomes entrained in argon, the secondary coolant cover gas, and is released intermittently to inert, nitrogen filled cells surrounding secondary coolants. This is eventually released through HV plenums. Similarly, tritium diffusing through containment enters inert, nitrogen filled cells and is released to HV plenums. Finally, tritium can diffuse through steam generator walls becoming entrained in steam. This tritium is released by steam leaks, blowdown, or permeation through condenser walls into condenser water.

Tritium, therefore, can be released from pool-type reactors as tritiated water in liquid or vapor phases or as tritium gas. Tritium released from primary or secondary coolant systems to inert, nitrogen filled cells should be released as tritium gas. Tritium diffusing into steam systems prior to release should be released as tritiated water. Partitioning between vapor and liquid is dependent on waste management and can vary widely.

At the present time, no commercial LMFBRs are operating in the United States. Therefore, predictions of amounts of tritium following each of the above pathways is limited to experience with experimental LMFBRs. Two operating experimental LMFBRs are the EBR-II and the FFTF. The FFTF, however, does not have steam generators. Corrosion of steam generator tubings is a predominant source of hydrogen to secondary sodium coolants, and diffusion of hydrogen through intermediate heat exchanger walls into primary sodium coolants is a major source of hydrogen in primary sodium. Since primary and secondary cold trap efficiencies are very dependent upon hydrogen concentrations in coolants, the FFTF is not a good model for commercial LMFBRs. Tritium distributions in the EBR-II have been determined.[75] Tritium distributions are very dependent on cold trap efficiencies. Under normal operating conditions, losses through steam generator walls are between 0.4 and 0.7% of tritium released to primary sodium coolants; whereas, losses to the air shield cooling are between 0.7 and 1.1% of tritium entering the primary sodium coolants.[75] Using estimated generation rates and releases to sodium coolants, releases from 1000 MW(e) LMFBRs will be 3.4×10^{-3} - 0.01 TBq/MW(e)-year as tritiated water and 6.0×10^{-3} - 0.016 TBq/MW(e)-year as tritium gas.

Renner and McPheters[81] have upgraded a tritium transport model presented by Kumar[67] and verified the model against the EBR-II. The results agreed very well with operating data for a nine day transient situation. They then applied their model to the Clinch River Breeder Reactor Plant (CRBRP) for six months operation at full power. For cases in which structural materials are free of oxide permeation barriers, ~92% of tritium released to primary coolants is retained in primary and secondary cold traps, ~5% is dissolved in primary sodium coolants and ~3% is "directly" released to the environment. Releases are ~0.026-0.089 TBq/MW(e)-year. Of these releases, 82% is released through steam generators as tritiated water and 18% is released through stacks. As indicated earlier, this should have the form of HT gas. For cases where structures have oxide diffusion barriers, the results indicate that ~94% of tritium released to primary sodium coolants is retained in primary and secondary cold traps, 5.4% is dissolved in primary sodium coolants, and ~0.02% is directly released to the environment. Releases for this case are 1.8×10^{-4} - 6.3×10^{-4} TBq/MW(e)-year. Of these releases, 85% is released through steam generators as water and 15% is released to the atmosphere as HT gas.

Technologies for reducing emissions to the degree indicated by the latter calculation through the use of oxide barriers are not currently available. Only certain oxides present very effective barriers to tritium permeation.[82] Others, apparently, reduce permeation rates only slightly. Some effective barrier oxides have been studied at less stringent laboratory conditions, but their stability under reactor conditions is not yet known.[82] Therefore, though some oxidation of piping surfaces and steam generator tubes may be expected to occur, the extent of formation and effectiveness in reducing tritium permeation is not known. It can be concluded that using present technology releases from LMFBRs will be between 0.026 and 0.089 TBq/MW(e)-year. If effective barriers can be developed, however, releases may be reduced to 1.8×10^{-4} to 6.3 to 10^{-4} TBq/MW(e)-year.

7. MOLTEN SALT BREEDER REACTORS (MSBRs)

Production rates have been estimated by Briggs[83] for a 1000 MW(t) MSBR. The results are: ternary fission 0.400 TBq/MW(e)-year; activation of ^{6}Li, and activation of ^{9}Be to ^{6}Li followed by activation of ^{6}Li to tritium, 16.2 TBq/MW(e)-year; activation of ^{7}Li, 15.7 TBq/MW(e)-year; and activation of ^{19}F, 0.11 TBq/MW(e)-year.

The distribution of tritium is expected to be rapid. In the 7.3 MW(t) Molten Salt Reactor Experiment (1964-1969), the tritium distribution was: graphite, 15%; primary off-gas, 46-50%; reactor and coolant cells, 5.5-9.4%; radiator, 5.5-9.4%, and 9.4-26% was unaccounted for.[83] It has been estimated that reactor and coolant cells will receive ∿3.3 TBq/MW(e)-year[83] while steam systems will receive ∿11.0 TBq/MW(e)-year.[84] Thus, removal systems and permeation barriers will be needed. Since these have not been developed, releases from MSBRs cannot be determined.

8. TRITIUM PRODUCTION FACILITIES

At the Savannah River Plant, tritium is produced by irradiating aluminum-lithium targets in heavy-water reactors. Tritium is then recovered in lithium-aluminum processing facilities. Tritium releases from the heavy-water moderated and cooled production reactors (primarily as DTO) have already been considered. Tritium releases from the processing facility recovering tritium from the lithium-aluminum targets are primarily as HT gas. Releases from the fuel processing area are primarily as HTO. Releases from reactors and processing facilities from 1970-1974 are presented in Table 8.1.

During normal operation most atmospheric releases are as tritiated water. In 1975, 11,300 TBq of tritium was released to the atmosphere from the reactors and processing facilities.[86] Of this 9,690 TBq (85.8%) was released as an oxide while 1,600 TBq (14.2%) was released as gas. Prior to 1975, a larger fraction of releases would appear as tritium gas since tritium releases from target processing facilities have subsequently been reduced.[86]

Approximately 1.0×10^5 TBq of tritium is buried with solid wastes. Another 1.9×10^3 TBq is stored in waste storage tanks. Small fractions of the release (<1%) result from research and development work. Two large accidental releases have also been reported in recent years. In these, 17,700 TBq (ref. 87) and 6,730 TBq (ref. 86) of tritium gas were released on May 2, 1974, and December 31, 1975, respectively. For each release it was estimated that greater than 99% was as tritium gas.

9. FUEL REPROCESSING PLANTS

Tritium released from fuel reprocessing plants originates at irradiation facilities and accompanies fuel assemblies to reprocessing plants. Inventories at discharge in LWR fuels are dependent on the fissioning isotopes, lithium impurities in fuels, chemical forms of fuels, burnups, linear power levels during irradiation, and cladding materials. Inventories at the time of reprocessing are also dependent on the cooling time. Cooling times of 150-160 days are anticipated for LWR fuels.

Fission product inventories have been estimated using the ORIGEN computer code. Based on data from the latest revision, tritium inventories of PWR fuels having burnups of 33 MWd/kg of uranium are 0.029 TBq/kg of uranium after 160 days of cooling.[88] Estimates for BWR fuels are

Table 8.1. Tritium releases from Savannah River Plant reactor and separation areas[a]

Year	Release to in-plant streams (TBq)	Release to seepage basins (TBq)	Release to atmosphere (TBq)
Reactor area[b]			
1970	881	951	8177
1971	707	640	7626
1972	1030	918	9417
1973	1510	1250	8836
1974	881	640	8603
Separation areas			
1970		1190	9209
1971		699	10,320
1972		821	19,600
1973		1180	11,540
1974		577	24,720

[a] Taken from ref. 85.

[b] Includes the Heavy Water Rework Facility.

slightly lower due to lower burnups. Inventories may range as low as 0.011 TBq/kg of uranium.[89]

Tritium is distributed in fuel matrices, gas plenums, and structural materials of fuel rod assemblies. Distributions are dependent on the burnup and linear power level during irradiation. For linear power levels presently anticipated for LWR fuels, 5-15% of the tritium is bound in zircaloy claddings, presumably as zirconium tritide.[90] Fractions of tritium in gas plenums are not accurately known. Values range from negligible amounts to 10%.[91-96]

Tritium accompanying LMFBR fuels to reprocessing plants is expected to be low despite larger productions per kilogram of fuel. Tritium production in LMFBR fuels has been estimated as 0.0396 TBq/kgHM (kilogram heavy metal) for burnups of 40 MWd/kgHM, specific powers of 0.052 MWd/kg of uranium and 30 day cooling periods.[90] A large portion (95%) of the tritium, however, diffuses from fuel assemblies during irradiation.

Several options have been considered to reduce releases of tritium to the environment. These options include recycling, head end processes which release tritium before it becomes diluted in aqueous acid solvents, and processes which enrich tritium in aqueous solvents. These have been reviewed by Schnez[92] and Brown.[97]

9.1 Commercial Facilities

No commercial fuel reprocessing plants are currently operating. The Nuclear Fuel Services Facility, West Valley, New York, operated from 1968 to 1972 processing somewhat less than 6.5×10^5 kg of uranium.[98] It closed in 1972 for modifications including enlargement of its uranium capacity from 3.0×10^5 kg/year to 7.5×10^5 kg/year but has withdrawn its application for a license due to economic considerations. General Electric has built a reprocessing plant, the Midwest Fuel Recovery Facility, at Morris, Illinois, with a uranium capacity of 3.0×10^5 kg/year.[98] It presently does not appear that the plant will operate due to some technical problems in handling radioactive powders. A third facility has been built at Barnwell, South Carolina, by Allied Chemical Corporation. The uranium capacity is 1.5×10^6 kg/year. Construction has been completed except for facilities to solidify the wastes for shipment to federal repositories and to process plutonium nitrate. Designs of these facilities hinge upon Nuclear Regulatory Commission (NRC) decisions concerning fates of these materials. The plant is awaiting license approval. Exxon has announced plans for a 1.5 x 10^6 kg/year facility near Oak Ridge, Tennessee.[98] It is awaiting a construction permit from the NRC.

The Nuclear Fuel Services Facility utilized a chop leach process to expose and dissolve LWR fuels.[98-99] Tritium in the cladding (10-16%)[90] follows the cladding to on-site burial in steel drums. Stack monitoring indicates that 1.9 x 10^{-5} TBq/MWd as HTO vapor and 6.7 x 10^{-6} TBq/MWd as HT gas is released to the atmosphere.[100] Tritium following high activity wastes (HAWs) is 5% of fuel assembly inventories.[99] Using this data and fuel inventories of 0.029 TBq/kg of uranium, with 10% of tritium inventories in fuel claddings, tritium flow rates to release routes for full capacity operation (i.e., 7.5 x 10^5 kg/year) are: 18,000 TBq/year (82.1%) to low activity waste lagoons; 167 TBq/year (0.8%) as HT gas through stacks; 459 TBq/year (2.1%) as HTO vapor through stacks; 1100 TBq/year (5.0%) to HAWs storage; and 2190 TBq/year (10.0%) buried with claddings. If HAWs are calcined in 5 years, 75% of tritium in HAWs storage will be released.[99]

The Allied Chemical Reprocessing Facility will release essentially all tritium entering the plant except for that bound in claddings and that following HAW systems. Tritium bound in claddings is expected to be 5-10% of tritium in fuel assemblies.[99] The HAWs are also expected to retain 5-10% of tritium in fuel assemblies. The HAWs are stored for 5 years before being solidified. Some tritiated water is evaporated during solidification processes. Subsequent handling of this water has not been determined. The remainder of tritium in fuel assemblies is ultimately released to the atmosphere through stacks. Assuming that 5% of tritium follows claddings and 5% is stored with HAWs, tritium releases for fuels with 0.029 TBq/kg are: 39,600 TBq/year released to the atmosphere; 2.2 x 10^3 TBq/year buried with claddings; and 2.2 x 10^3 TBq/year stored with HAWs. Of tritium released to the atmosphere through stacks, 5.3% appears as HT gas and 94.7% as HTO vapor.[99]

9.2 Government Operated Plants

Government owned and operated fuel reprocessing facilities reprocess fuels generated in military operations and tritium production. These fuels have very varied fabrication and irradiation characteristics. Facilities include the Hanford Plant operated by Atlantic Richfield, Idaho Chemical Processing Plant, and Savannah River Plant. Releases from the Savannah River Plant were discussed in Sect. 8.

The Hanford Plant utilizes a purex process in which zircaloy claddings are removed by dissolution in ammonium fluoride—ammonium nitrate solutions. Fuels are then dissolved in nitric acid solutions as in conventional purex processes. Jeppson[101] has studied the behavior of tritium at this facility during processing of zircaloy-clad fuel rods. He found that most tritium (∿68%) is in process condensates and ammonium scrubber wastes as water. This water is sent to underground storage cribs. Of the remainder, 5.1%, as water, follows high-level wastes to underground storage, 22%, as water, is released to chemical sewers eventually reaching surface ponds and 4.7% is released to stacks. Greater than 90% of tritium released to stacks is tritium gas or tritiated ammonia. The fuels have much lower tritium inventories (6.48×10^{-4} TBq/kg of uranium) than power reactor fuels; therefore, releases are less than those from commercial facilities. For full uranium capacity operation ($\sim 3.6 \times 10^{6}$ kg/year) total throughput of tritium is 2300 TBq/year based on this tritium inventory.

The Idaho Chemical Processing Plant reprocesses both aluminum-clad metallic fuels and zirconium-clad fuels.[102] Aluminum-clad fuels are dissolved in nitric acid. During this process ∿45% of tritium is released to off gases as HT gas while 55% follows acid streams as HTO to solvent extraction systems. Zirconium-clad fuels are dissolved in hydrofluric acid. During this ∿90% of tritium is released to off gas with ∿98% being HT gas. The remaining 10% follows acid as water and enters extraction systems. Tritium leaves extraction systems in two streams. Approximately 0.1% exits with products while ∿99.9% exits with high-level liquid wastes. Both streams are eventually solidified and release tritium as HTO. This HTO is released to the atmosphere or injected into wells.

10. THERMONUCLEAR REACTORS

At the present time it is impossible to quantify precisely the emissions of tritium from fusion reactor power plants because fusion reactor development is yet in the experimental stage. Exact designs for fusion power reactors, therefore, are unspecified. Further, unpredictable advances in tritium control technology will likely occur before commercial fusion reactors become available so that calculations based on present control technology will only give an estimate of tritium releases. Nevertheless, studies of tritium releases from conceptual designs of controlled thermonuclear reactors have been made. In these designs, tritium is used as a fuel and is bred in blankets containing lithium.

Most conceptual designs that have been studied for tritium emissions employ magnetically confined, toroidal shaped reaction regions. This scheme is generally considered to be most likely for power producing fusion reactors. Other studies, however, have considered reactors employing magnetic mirror confinements. This review will be limited to these two plasma confinement concepts; although, other concepts utilizing inertial confinement with laser and particle heating are being studied.

Assumptions are made for tritium release calculations when designs are not sufficient to delineate data needed for the calculations. Three methods are used to make these assumptions. In some cases, arbitrary numbers, believed to be reasonable, are placed on parameters and other values are calculated from these parameters. Another method is to specify allowable tritium release limits, develop tritium control systems that are believed to limit tritium releases to this level, and determine the economic feasibility of the design. A third method is to calculate tritium inventories and release rates using parameters specified in conceptual designs, substitute prorated values based on fission power reactor experience when conceptual designs do not give sufficient data, and make arbitrary assumptions only when these two procedures are inadequate. Release rates are very dependent on the chosen method.

Table 10.1 gives tritium inventories and tritium emissions expected for some conceptual thermonuclear reactors.[103-115] Tritium release values given in the table do not consider the entire reactor. For instance, release terms given do not include releases from fuel storage since the form of stored fuels is unspecified in these designs. Designs of the UWMAK I and PPPL tokamak are most complete, therefore, their release rates are considered to be more realistic. In other estimates either designs were too incomplete to allow identification and quantification of all release routes or the investigator was studying only one release mechanism.

Other factors also increase the uncertainty in these approximations. One of these factors is uncertainty in the dependence of tritium permeation on tritium partial pressures at very low pressures. At higher partial pressures, tritium permeation is proportional to tritium partial pressures to the half power. Studies at partial pressures less than 1 mm Hg suggest that tritium permeation may be proportional to tritium partial pressures to the first power rather than to the half power. This would greatly reduce the permeation at very low pressures which are of interest. Another factor is uncertainty in the effectiveness of oxide surfaces as permeation barriers. Oxide coatings may reduce tritium permeation by a factor of 100 relative to permeation through clean surfaces.[115]

Results given in Table 10.1 suggest that total tritium inventories in a 5000 MW(t) reactors will be between 1 and 16 kg/1000 MW(e) rated power. Significant quantities will appear in blankets and fuel processing and storage systems of the various designs. Relative amounts of tritium in these areas will depend on schemes for tritium recovery from blankets, structural materials used in blankets, fuel purification schemes, and amounts of fuel that will be stored. Variation in these parameters accounts for the wide range of estimated tritium inventories. A wide range also occurs in estimated tritium releases. Estimates of

Table 10.1. Tritium in thermonuclear reactors

Parameters	UWMAK I	BNL Blanket	UWMAK II	PPPL	ORNL	LASL θPinch	Toroidal[a]	Toroidal[b]	Toroidal[c]	Toroidal stellerator[d]	TOKAMAK	Magnetic mirror
Net Electrical Output MW(e)	1473	1605	1710	2030	518	4132			170	2000		
Net Thermal Power MW(t)	4663	5000	4712	5305	1000	12000	1000	1000	1000		1000	5000
Total Plant Inventory, KG	23.5	10.1	17.5	2.58	5.7	3.8	6.0			2.8		
Total Active Inventory, KG	13.5	0.13	4.6	0.56	0.14	2.4	1.4		0.78	0.809		
Blanket		0.014	0.4	0.01				1.68-5.28	0.46		1.335	8.0
Coolant	8.7	0.001	0.2	0.01	0.12	0.35			0.0001	0.025		0.241
Structure				0.03	0.02				0.021		0.182	
Getting System	4.5	0.007	3.9			0.30				0.03		
Fuel System		0.107		0.51		1.64			0.300	0.753		
Cryo Pumps	0.3		0.1							0.001		
Storage, KG	10.0	10.0	12.9	2.02	5.6	1.43	5.6			2.0		
Release TBq/day												
Gaseous	0.127[g]	0.263[g]	0.091[g]	0.036[g]		0.004[g]			0.075[g]		0.045[g]	
Liquid	0.128	None[h]	0.009	None[i]		0.096[j] or >1.11[l]			0.022[k] or 2.47[m]		0.091	
Major Assumptions:	Permeation α sq. root of partial pressure; ^3H to containment recovered with 90% efficiency					^3H partial pressure over lithium = 10^{-10} torr, ^3H partial pressure			^3H concentration in lithium kept at 1 ppm		^3H concentration in lithium 1.5 ppm. permeation of ∝ partial pressure of ^3H	

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[a] Taken from refs. 103 and 112.

[b] Taken from ref. 106.

[c] Taken from ref. 107.

[d] Taken from ref. 105.

[e] Taken from ref. 108.

[f] Taken from Ref. 104.

[g] Does not include estimates for releases from tritium storage systems.

[h] 81.4 TBq/year of tritiated water from power system is released through the stack.

[i] 6.8 TBq/year of tritiated water from power system is recovered and assumed stored as a liquid.

[j] Cu barrier in steam generator.

[k] Stainless steel steam generator.

[l] No Cu barrier.

[m] Tungston clad stainless steel steam generator.

gaseous releases vary from 1.0×10^{-3} TBq/d to ∿0.16 TBq/d for 1000 MW(e) reactors operating at full power. Liquid release estimates range from ∿0 when tritium is recovered from water systems to slightly less than 5.3 TBq/d for 1000 MW(e) reactors operating at full power. The wide range is caused by different management techniques and use of permeation barriers.

Accidental releases are also difficult to generalize due to the large variation in reactor designs that may occur. Probabilities of accidental releases of tritium from reactor areas are very small since reactor cells must rupture simultaneously with components within the cells. Draley et al.[109] have suggested that the maximum credible accident for CTRs is a liquid metal fire releasing the blanket inventory of 1.7×10^5 TBq of tritium as HTO or HT. In other types of blankets (FLIBE for instance) releases may have another chemical form. Accidental releases should be rare, if at all, and should make only minor contributions to releases from CTRs.[109]

11. NUCLEAR EXPLOSIVES

Tritium is produced in pure fission detonations by ternary fissioning (direct production) and by neutron activation reactions (indirect production). Direct yields are estimated to be ∿25 TBq/Mton equivalent explosion[116-117] by assuming that one tritium atom is produced in 10,000 fissions and 1 Mton explosion requires 1.4×10^{26} fissions. Indirect yields depend on detonation environments. Terpilak[118] has estimated that indirect yields are ∿7.2×10^3 TBq/Mton. Thus, total yields derive almost entirely from neutron activation reactions.

For fusion detonations, Miskel[116-117] has reported that tritium yields lie within 2.5×10^5 and 1.8×10^6 TBq/Mton with an average of 7.4×10^5 TBq/Mton. Other predictions are: 2.5×10^5 TBq/Mton,[119] 1.9×10^5 - 7.4×10^5 TBq/Mton,[120] 1.5×10^5TBq/Mton (fusion yield) plus 5.6×10^4 TBq/Mton (neutron activation),[121] and 4.1×10^5 TBq/Mton.[11] The latter estimate resulted from widespread water sampling following the 1953 Castle tests.

In atmospheric detonations most of the tritium is oxidized at the time of detonation and is eventually removed by precipitation.[122] Tritium entering the troposphere is rapidly removed by precipitation and ultimately appears in ocean and inland waters.[123] Tritium entering the stratosphere is gradually introduced into the troposphere and then removed by precipitation. Fractions of tritium reaching the stratosphere are dependent on heights, latitudes, and yields of explosions. Typical values are 20% for ground surface detonations and 80% for water surface detonations.[122]

Distribution of tritium between HTO and HT following underground detonations is dependent upon availability of iron and water in detonation environments.[116] For most underground detonations a reasonable, upper limit for tritium in the elemental form is 1% of that produced.[116] Release studies of 17 vented radioactivity tests occurring at a rate of ∿60 kton/year indicate that at most, 4.4×10^{-3} - 444 TBq/year of tritium is released to the atmosphere.[116-117] Even the larger values are small in comparison to the earth's surface inventory from natural production.[117]

Several estimates of weapons detonation contribution to the earth's tritium inventory have been made. In the latest UNSCEAR report,[43] Michel's estimate[124] for earth surface tritium in 1970 (1.1×10^8 TBq) was corrected for decay to obtain a total injection of 1.7×10^8 TBq by weapons detonation before 1970. Another estimate in UNSCEAR resulted in 1.3×10^8 TBq injected before 1970. Earlier estimates were 3.0×10^8 TBq and 6.3×10^7 TBq. It is evident that weapons detonations are the major contributor to the current earth's surface inventory of tritium.

12. CONCLUSIONS

Tritium production and release are summarized in Table 12.1. The ranking of electrical generating facilities according to increasing tritium production is: HTGRs, LWRs, LMFBRs, MSBRs, and HWRs. Overlaps in the range of production estimates occur for HTGRs and LWRs as well as for LWRs and LMFBRs; therefore, their ranking can be argued. For these facilities tritium is only a by-product. Fusion devices have not been included in the table because production estimates are speculative. In addition, fusion devices differ from other energy producing facilities in that tritium is produced as fuel. Net production rates, therefore, correspond to tritium loss rates.

Tritium produced in electrical generating facilities may be released from reactors and from fuel reprocessing facilities. Releases from reactors are greatest for HWRs being 890 TBq/year for 1000 MW(e) facilities operating at full power. These releases are much greater than average releases of 2.4 TBq/year derived for 1000 MW(e) LWR facilities operating at full capacity. Release rates projected for HTGRs and LMFBRs are similar to those for LWRs. They are 2.2 TBq/year and 2.0 TBq/year, respectively, for 1000 MW(e) facilities operating at full capacity. Currently, there are no commercial fuel reprocessing facilities operating in the United States. The technology, however, does exist for reprocessing of LWR fuels. One commercial facility has operated and another is currently awaiting an operating license. Most of the tritium released from closed LWR fuel cycles are released at fuel reprocessing facilities. Expected releases based on current estimates of tritium inventories in spent LWR fuel elements correspond to 930 TBq per 1000 MW(e)-year of energy generated. Reliable release estimates for reprocessing of fuels from HWRs, HTGRs, LMFBRs, and MSBRs were not found. It can be stated, however, that releases of tritium during reprocessing of LMFBR fuels should be low since 95% of the tritium produced in LMFBR fuels escapes to coolants during operation.

Tritium is also produced commercially at the Savannah River Plant in South Carolina. The average release rate from 1970 to 1974 was

Table 12.1. Tritium production rates and releases to the earth's surface[a]

Source	Production			Release		
	Range	Value used	Rate	Range	Value used	Rate
Natural production	0.2-0.5 ^3H atoms/cm^2-s	0.25 ^3H atoms/cm^2-s	7.2×10^4 TBq/year			7.2×10^4 TBq/year
LWRs	0.44-1.3 TBq/MW(e)-year	1.0 TBq/MW(e)-year	1.0×10^3 TBq/1000 MW(e)-year	0.0019-0.038 TBq/MW(e)-year	0.024 TBq/MW(e)-year	24 TBq/1000 MW(e)-year
HWRs	∼89.5 TBq/MW(e)-year	89 TBq/MW(e)-year	8.9×10^4 TBq/1000 MW(e)-year	0.89 TBq/MW(e)-year	0.89 TBq/MW(e)-year	890 TBq/1000 MW(e)-year
HTGR	0.48-0.89 TBq/MW(e)-year	0.74 TBq/MW(e)-year	7.4×10^2 TBq/1000 MW(e)-year	0.0078-0.031 TBq/MW(e)-year	0.022 TBq/MW(e)-year	22 TBq/1000 MW(e)-year
LMFBR	0.93-1.5 TBq/MW(e)-year	1.3 TBq/MW(e)-year	1.3×10^3 TBq/1000 MW(e)-year	0.010-0.026 TBq/MW(e)-year	0.020 TBq/MW(e)-year	20 TBq/1000 MW(e)-year
TPF Savannah River				2.0×10^4 - 3.2×10^4 TBq/year	2.6×10^4 TBq/year	2.6×10^4 TBq/year
Detonations[b]				6.3×10^7 - 3.0×10^8 TBq	1.7×10^8 TBq	1.7×10^8 TBq
Fuel reprocessing[c]	0	0	0	0.089-0.93 TBq/MW(e)-year	0.89 TBq/MW(e)-year	890 TBq/1000 MW(e)-year

[a] Most current available release figures have been used in an attempt to reflect state of the art.

[b] Values are the release to the earth's surface before 1970. Atmospheric weapon test bans have limited subsequent production.

[c] Do not produce tritium, but release some of the tritium produced in irradiation facilities. Put on a per MW(e) basis by assuming 0.029 TBq/kg of uranium ^3H inventory, 33 MW(e)/kg of uranium burnup, and 365 d/year.

26,000 TBq/year. Tritium has also been introduced into the atmosphere before 1963 by atmospheric weapon tests. The atmospheric test ban has subsequently reduced the frequency of atmospheric tests. As a consequence, the tritium inventory in the hydrosphere from weapons detonations is decreasing.

Natural tritium production has been included for the sake of comparison. Of the reactors used for electrical generation, only HWRs and MSBRs produce tritium at a rate that is comparable to the natural production rate. Production rates for 1000 MW(e) facilities operating at full capacity are 124% and 45%, respectively, of the natural production rate. Other reactor types produce tritium at rates that are two orders of magnitude lower than the natural production rate. The tritium release rate from the Savannah River Plant is significant being 36% of the natural production rate. Tritium introduced into the environment by atmospheric weapons detonations before 1963 is $\sim$130 times the steady state inventory of 1.3×10^6 TBq that can be obtained from the global natural production rate of 0.25 tritium atoms/cm^2-s.

An attempt has also been made to specify the chemical form of tritium released from other than natural sources. The majority of tritium was found to be released as tritiated water. Thus, assumptions made in any analysis of impact associated with tritium release that any tritium released has the form of tritiated water are not unreasonable.

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