FISSION PRODUCT CONTROL IN HTGR PLANTSH. J. de Nordwall,[†] W. E. Bell^{*}

ABSTRACT

Fission product control in a typical 2000 MW(t) HTGR plant is discussed in terms of (1) radiation dose guidelines as established by the AEC, (2) fission product pathways to the environment, (3) fission product distributions under normal and selected accident conditions, and (4) the physical models and basic data used to describe fission product transport. Six key fission product nuclides, namely ^3H , ^{88}Kr , ^{90}Sr , $^{127\text{m}}\text{Te}$, ^{131}I , and ^{137}Cs are taken to represent chemically and radiologically all significantly mobile fission products.

Results of release calculations for the six nuclides show substantial margins (or safety factors) between acceptable (guideline) and predicted releases to the environment. For normal conditions, the smallest apparent margin, a safety factor of 22, is for ^{88}Kr . For accident conditions, the smallest apparent safety factor is 330, which is for ^{90}Sr in the design depressurization accident.

1. INTRODUCTION

The goals of fission product control are minimization of radiation dose to man, optimization of equipment associated with the handling of fission products and description of the radiological consequences of accidents in which the normal barriers between the fission products and the environment are breached.

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Standards for acceptable radiation doses to man aim at controlling the maximum dose commitment made by any individual and by the population as a whole. The current objective is to limit the maximum dose commitment rate from nuclear power to the whole body or any organ of any off-site individual to about 5 mrem/yr, which is a small fraction of the natural background of 100-250 mrem/yr.¹

Total population exposure from a given plant is currently determined by integrating (dose x population) over a 50-mile radius.² Integral dose rates of 10-30 man-rem per plant per year are being projected for large reactors. This population dose does not include any allowance for doses received during unanticipated events such as accidents or the repair of major components.

Once in a lifetime maximum individual dose guidelines for use in accident analysis are established by regulation³ as 25 rem to the whole body and 300 rem to the thyroid. A bone dose guideline of < 150 rem is being used in relation to the potential release of ⁹⁰Sr during a fast depressurization. No population dose guide for use in accident analysis has been defined numerically.

Practical and economically acceptable goals for fission product control in large HTGR systems are currently being established. Following some quantification of these overall goals in terms of specific plant release goals the paths by which fission products can reach the environment and the status of methods for assessing fission product transport within an HTGR will be discussed.

While the emphasis in this paper will be on demonstrating that specific release goals can currently be met, it must be emphasized that if probable radiation cost in man-rem is ultimately to be assessed, best-value calculations and error assessments rather than the conservative estimates accepted for licensing will ultimately be required.

2. PLANT RELEASE GOALS

The selection of a small set of nuclides to characterize the behavior of radiologically important fission products in an HTGR has been described by Flowers.⁴ Tritium has been added to the list.

The fission product isotopes selected are ³H, ^{127m}Te, ¹³¹I, ⁸⁸Kr, ¹³⁷Cs, and ⁹⁰Sr. Chemically this group represents all significantly mobile fission product elements. Satisfactory control of these six key fission products in an HTGR system means satisfactory control for all fission products. The list has been restricted to one isotope per element for the sake of brevity.

Restrictions may be imposed for reasons other than radiological toxicity, for example, the release of stable and long-lived tellurium

could be limited by its reaction with nickel alloys. This effect has not been observed to date in an HTGR. Plant maintenance considerations^{4,5} may also lead to restrictions on the release of γ -emitters such as $^{140}\text{Ba-La}$ and ^{137}Cs from the core.

The radiological consequences of release of the key nuclides at ground level on a hypothetical site are compared in Tables 1 and 2 under normal and accident conditions in terms of mrem/yr received at the exclusion area boundary (EAB)* per curie/year released under normal conditions, and rem/Ci released in 2 hr during an accident.

3. FISSION PRODUCT PATHWAYS TO THE ENVIRONMENT

The barriers and sinks provided in the HTGR to prevent fission products from being transported to the environment from the fuel kernels where they are born are shown in Fig. 1. The barriers and sinks within the PCRV liner determine the radionuclide inventory in the primary circuit; this inventory is important because it is most readily available for release during accidents involving breaches of the primary containment. Because of the sluggishness with which heat and fission products are transported in the core, it is unlikely that significant release of fission products from the core will occur during currently analyzed accidents. Components outside the PCRV liner attenuate and/or contain radionuclides that leak from the PCRV.

The first and most important fission product barrier is the fuel particle coating, whose effectiveness depends upon material properties, service temperature, and fission product half-life and atomic number. Under normal operating temperature, SiC coatings provide an absolute barrier for all fission products; pyrocarbon coatings provide a selective barrier, which is impermeable to gases and iodine and more or less permeable to metals. In escaping from the fuel rod, which is the next larger core structural unit, a fission product must pass through the fuel rod matrix and across a helium-filled gap between the fuel rod and the fuel element structural graphite. Both the matrix and graphite contain large surface areas upon which less volatile fission products can adsorb, but the effectiveness of the matrix as a diffusion barrier (or delay) is minimized by the homogeneous distribution of fuel within it and its higher operating temperature. The structural graphite does however provide an effective diffusion delay for metallic fission products, again depending on half-life, atomic number and temperature. Structural graphite has little effect on gas transport on account of its high permeability.

Within the primary circuit, the purification plant and the walls of the system serve as sinks for fission products, their effectiveness

*Federal regulations define an exclusion area within which access is controlled by the reactor operator.³

Table 1. Dose Commitment Rate per Unit Release
Under Normal Conditions

Release Rate = 1 Ci/yr at ground level

$X/Q = 1 \times 10^{-5} \text{ sec/m}^3$

Concentration at EAB^a = $3.175 \times 10^{-13} \text{ Ci/m}^3$

Isotope	EAB Dose Commitment Rate for Most Limiting Organ ^{b,c}		Limiting Path, Organ
	$\left(\frac{\text{mrem/yr}}{\text{Ci/yr released}} \right)$	$\left(\frac{\text{Ci/yr}}{\text{mrem/yr}} \right)$	
³ H	0.00026	3900	Inhalation, W. Body
^{127m} Te	83	0.012	Ingestion, Kidney
¹³¹ I	380	0.0026	Ingestion, Thyroid, Child ^d
⁸⁸ Kr	0.0051	200	Submersion, W. Body
¹³⁷ Cs	89	0.011	Ground shine, W. Body
⁹⁰ Sr	750	0.0010	Ingestion, Bone

^aEAB = Exclusion Area Boundary.

^bOrgan dose = Σ dose via submersion, ground radiation, inhalation, and ingestion.

^cDose factors from Ref. 6. Ingestion path dose factors are divided by 3 since no one is assumed to grow more than 1/3 of his food.

^dDose factors from Ref. 7. Ingestion path dose factor is adjusted.

Table 2. EAE Dose Commitment per Unit Release During an Accident

Activity Released to Atmosphere = 1 Ci^a

$$\chi/Q = 1 \times 10^{-3} \text{ sec/m}^3$$

Isotope	Dose Commitment ^a (rem/Ci released)			Ci to Exceed Limiting ^b Dose	Limiting Organ, Path
	W. Body	Thyroid	Bone		
³ H	0.00038	0.00038	0.00038	66,000	W. Body, Inhalation
^{127m} Te	0.00057	0.019 ^c	0.0043		Kidney, Limit undefined
¹³¹ I	0.0010	0.51	0.0010	590	Thyroid, Inhalation
⁸⁸ Kr	0.00050	0.00050	0.00050	50,000	W. Body, Submersion
¹³⁷ Cs	0.011	0.011	0.021	2,300	W. Body, Inhalation
⁹⁰ Sr	0.45	0.45	6.8	22	Bone, Inhalation

^aActivity released and inhaled over a 2-hr period; breathing rate, 30 m³/day.

^bThis is not a working limit.

^cKidney pathway.

HTGR SYSTEM COMPONENTS CONTROLLING FISSION PRODUCT
TRANSPORT TO ENVIRONMENT

Fig. 1. HTGR System Components Controlling Fission Product Transport to Environment.

depending on fission product half-life, volatility, and chemical reactivity. The coolant circuit surfaces can become a source of fission products during certain accidents, as indicated by the double arrows in Fig. 1.

Pathways for fission products to reach the environment under normal operating conditions are (1) leakage of primary coolant through the PCRV liner [$\tau(5,9)$ and $\tau(5,10)$] and (2) leakage of primary coolant into the reheater sections of the steam generators [$\tau(5,8)$]. (The steam pressure in the reheaters is lower than the helium coolant pressure.) Accidents involving large increases in both these leakage paths are analyzed in HTGR safety reviews. The concrete wall of the PCRV and the water in the cooler parts of the secondary coolant system function as sinks for elemental iodine and less volatile metallic fission products passing through them.

Fission products reaching the containment are controlled by (1) the containment air cleanup system, (2) deposition on containment surfaces, (3) filtration of air leaving the containment, or (4) decay. During accidents involving release of primary coolant into the secondary containment, ventilation is stopped, isolation valves are closed, and leakage from the secondary containment [$\tau(10,16)$] becomes the dominant path to the atmosphere.

Fission products reaching the secondary coolant (water side of the steam generator) can reach the environment either as liquids, or gas via the steam air ejector [$\tau(14,16)$]. Tritium is unique in that it can reach the environment via diffusion through the steam generator tube walls as well as via the pathways open to the noble gases. It therefore appears in both liquid and gaseous effluents.

4. CURRENT FISSION PRODUCT DISTRIBUTION ESTIMATES FOR NORMAL AND ACCIDENT SITUATIONS

This section summarizes and discusses the results of fission product release and distribution calculations made for a typical 2000 MW(t) HTGR using the plant parameters taken from the Preliminary Safety Analysis Report⁸ (Table 3). Fission product distributions under normal conditions and following three accidents involving breach of the primary containment are considered. The physical models used to represent fission product transport and the parameter values used in these models are discussed later in Section 5.

Table 4 summarizes the expected fission product distribution under normal conditions at the end of reactor life. Inspection of Table 4 permits some important conclusions to be drawn:

1. The inventory of fission products closest to the environment is well controlled by the graphite core and the particle coatings. (See also ref. 9.)
2. Only small fractions of the radiologically toxic elements, iodine and strontium, remain gas-borne in the primary circuit, and hence are able to be transported by gas leaks through the PCRV liner.

**Table 3. Plant Parameters Used in Fission Product
Transport Calculations**

Primary Coolant Purification Rate	5.3/day
PCRVR Leak Rate	0.01%/day
Reheater Leak Rate	0.001%/day
Containment - Atm Purification Rate During Accident	1 vol/hr ^a
- Recirculation Filter Efficiency	
I, Cs, Sr	90%
Organic Iodide	30%
Containment Leak Rate During Accident (first 24 hr)	0.5%/day

^aPurification starts 1 hr after accident.

Table 4. Predicted Normal Fission Product Distributions in a 2000-MW(t) HTGR^a

Isotope	Equilibrium Core Inventory (Ci)	Primary Circuit Inventory After 40 yr ^b		Rate of Gaseous Release to Environment (Ci/yr)	EAB Concentration (Ci/m ³)
		Ci gas-borne	Ci deposited		
³ H	1.7 x 10 ⁴	1.9	0	48	1.5 x 10 ⁻¹¹
^{127m} Te	1.5 x 10 ⁶	5.6 x 10 ⁻⁴	6.8 x 10 ²	1.6 x 10 ⁻⁷	5.0 x 10 ⁻²⁰
¹³¹ I	6.0 x 10 ⁷	8.5 x 10 ⁻²	8.6 x 10 ³	2.5 x 10 ⁻⁵	7.7 x 10 ⁻¹⁸
⁸⁸ Kr	1.0 x 10 ⁸	2.3 x 10 ³	0	45	1.4 x 10 ⁻¹¹
¹³⁷ Cs	4.1 x 10 ⁶	1.7 x 10 ⁻⁴	1.1 x 10 ⁴	6.2 x 10 ⁻⁸	2.0 x 10 ⁻²⁰
⁹⁰ Sr	4.3 x 10 ⁶	4.1 x 10 ⁻⁵	2.7 x 10 ³	1.5 x 10 ⁻⁸	4.8 x 10 ⁻²¹

^aχ/Q = 10⁻⁵ sec/m³ as in Table 1.

3. There are considerable margins between the reference release limits (Ci/yr/mrem/yr) listed in Table 1 and the releases to the environment (Ci/yr) in Table 4.
4. While the projected release of cesium to the environment is clearly acceptable, the presence of ~ 11,000 Ci of ^{137}Cs in the primary circuit must be considered in terms of the contribution it could make to the total man-rem dose delivered by an HTGR if major maintenance such as removal of a steam generator were necessary.

Since ^{85}Kr can be separated and removed for offsite disposal, it was not considered in assessing the radiological impact of the plant.

Table 5 summarizes the 2-hr activity release to the environment following a design basis depressurization accident involving a penetration failure with 100 in.² flow area. In this accident, the containment is isolated by closing of isolation valves, and activity release to the environment occurs by leakage from the containment.

Table 6 summarizes the activity release to the environment resulting from a single steam tube rupture in the reheat section of a steam generator. In this accident, activity monitors detect radioactivity in a reheat line, the defective reheater is isolated, and the loop is shut down. Release to the atmosphere is via the steam air ejector.

The third accident involves steam leakage into the primary coolant system with concurrent failure of the moisture monitor. It is assumed that the steam leak is not isolated quickly and the leaking loop is not dumped automatically. As the leak continues, the pressure relief valve opens, once due to the initial pressure buildup, and again as a result of steam-carbon reactions. Some fission products are released as a result of reaction of steam with core graphite and failed fuel particles. The radiological consequences of this accident are less severe than for the design basis depressurization accident.

Let us now consider the estimates of releases during the first 2 hr of these accidents in conjunction with the plant release limits in Table 2. Again substantial margins appear to exist; in fact some margins are larger in the accident case because the release limits are higher and the containment is isolated except for an assumed leak rate. This is true in spite of the additional safety factor of 10 applied to the primary circuit inventory for use in accident analyses.

Some of these margins are large enough to permit design changes without significant effect on potential radiological impact. For example, if one were not limited by requirements relating to access to the primary circuit one could use only pyrocarbon coated fuels. One also has considerable tolerance (on the same basis) toward undetected releases of metallic fission products from partially or completely blocked coolant channels. Alternately one does not need to be so concerned about the

Table 5. Estimated Activity Release (2-hr) to the Environment Following a Design Basis Depressurization Accident

Isotope	Release to Containment (Ci)	Containment Inventory after 2 hr		Two-hr Release to Atmosphere (Ci)
		Circulating (Ci)	In Cleanup System Filter (Ci)	
^3H	19	19	0	0.008
$^{127\text{m}}\text{Te}$	74	33	41	0.026
^{131}I	950	420	530	0.33
^{88}Kr	2.3×10^4	1.3×10^4	0	7.4
^{137}Cs	59	24	35	0.020
^{90}Sr	190	76	110	0.067

Normal primary circuit inventory multiplied by 10 for use ⁱⁿ accident analysis.

**Table 6. Estimated Activity Release to Environment
After a Reheater Tube Accident**

Isotope	Release to Environment (Ci)
^3H	0.037
$^{127\text{m}}\text{Te}$	1.1×10^{-6}
^{131}I	1.6×10^{-4}
^{88}Kr	44
^{137}Cs	3.2×10^{-7}
^{90}Sr	7.9×10^{-8}

fraction of a deposited fission product that could re-enter the gas phase during an accident [$\tau(6,5)$]. In the accidents listed in Tables 5 and 6, 100% of the primary circuit inventory of iodine, cesium, and strontium could be released to the isolated containment without exceeding dose limits.

However since the ^{88}Kr margin is relatively small, increases in core temperature in response to pressure for higher thermal efficiency can only be made if accompanied by corresponding improvements in fuel quality and failure rates.

It is also apparent from scaling the doses per curie entering the containment, that the containment is capable of holding substantial fractions of the core inventories of the more volatile nuclides such as iodine, krypton, tritium, and cesium.

In view of the reliance placed upon the containment, one should ask what the consequences of failure to close the isolation valve would be. Using current re-entrainment factors [$\tau(6,5)$], depressurization without containment would lead to dose guidelines being exceeded if the "accident (X10)" primary circuit inventory were used; if the expected (normal) inventory were used, one would not expect to exceed the limits.

Using information similar to that reviewed in Section 5, the radiological impact of a 2000 MW(t) HTGR may be calculated for all fission products, not just the six reference nuclides. Taking into account all fission products does not significantly alter the conclusion that there are substantial margins between acceptable and predicted releases, particularly under accident conditions.

5. CURRENT BASIS OF FISSION PRODUCT DISTRIBUTION ESTIMATES

Assessment of the radiological consequences of HTGR operation and accidents revolves around a description of the amounts and mobilities of fission products in the primary coolant circuit. This section will indicate how these quantities are currently estimated for the reference elements. For brevity, individual steps in the fission product path will be identified by the τ s in Fig. 1. For example $\tau(M,5,6)$ means the transport of isotope or element M from a capacitance (5) to capacitance (6).

5.1 Noble Gases

It is now well established that the coatings of HTGR fuel particles are, for practical purposes, impermeable to krypton and xenon; therefore $\tau(\text{Kr } 2,3)$ and $\tau(\text{Xe } 2,3)$ are zero. Gas release is thus a sign of coating failure or contamination of the outer coatings by uranium or thorium and is represented by $\tau(1,3)$ in Fig. 1. For newly manufactured particles $\tau(1,3)$ is the sum of contributions from imperfect, damaged and contaminated

particles and is determined experimentally by measuring the steady state release (R/B)* of ^{85}mKr on a sample of production fuel rods. Fort St. Vrain specifications required R/B (^{85}mKr) to be $< 3 \times 10^{-5}$ at 1100°C , a value which was achieved.

The percentage of failed fuel is expected to be $< 1\%$ at full burnup and fast neutron exposure for both fuel types (BISO fertile and TRISO fissile). Mathematical models are used to determine the core-average fuel failure-fraction, which is currently 0.27%. The release of krypton and xenon from failed fuel has been measured. A recent study¹⁰ indicated a ^{85}mKr R/B value of 5×10^{-3} at 1100°C for failed particles.

Experiments have demonstrated repeatedly that the steady-state release of Kr and Xe from fuel kernels varies with $\lambda^{-1/2}$ and $e^{-E/RT}$, where λ is the radioactive decay constant and E is an activation energy characteristic of the release process. At low temperatures ($< 500^\circ\text{C}$), diffusional release tends to become zero and fission recoil controls release.¹⁰ At normal fuel temperatures (700 to 1200°C), E is ~ 23 kcal/mole, which is a relatively low value, suggesting a diffusion process more rapid than bulk diffusion. At temperatures above 1300°C , E is higher, suggesting some contribution from bulk diffusion. The same activation energy applies to both intact and failed particles in fuel rods. On the basis of experimental results, xenon release is taken to be $\sim 1/2$ that of a krypton isotope of the same half-life. Values of R/B and E are less well defined for fuel at very high burnup, and E may continue to increase above 1200°C .

In calculating circulating gas inventories, a factor of 4 increase is explicitly applied to the reference fission gas release (R/B) data for particles failing in service. This increase allows for the potential increase in gas release following hydrolysis or oxidation of the kernel by impurities in the coolant or following carburization by interaction of oxide kernels with carbon. The reference R/B data for as-manufactured rods are reduced by a factor of 2.5 to account for the actual fuel temperature distribution, which yields an average temperature of 880°C . The contribution from particles with failed coatings is not similarly reduced since the potential for particle coating failure is greater at higher temperature and the average temperature for fuel failed in service may therefore differ from that of all fuel. The R/B value for ^{85}Kr , and other long-lived gases, is assumed to be 1.0 for failed fuel particles. In-pile tests of prototype fuel for large HTGRs indicate that the release parameters used in the above procedure are well-grounded. Fort St. Vrain is expected to finally validate this analysis.

Given rates of release from the fuel, the primary circuit inventory corresponding to an equilibrium core is readily calculated since krypton

* $R/B = \frac{\text{release rate}}{\text{birth rate}}$ at steady state.

and xenon do not
or the walls.
3.

interact significantly with the core structural graphite
plant purification and leakage rates are given in Table

Noble gas
environment
ment are delayed
toxic short-
accidents gas
for an assumed

leaving the PCRV via route $\tau(5,8)$ are assumed to reach the
at delay; gases passing through the ventilated contain-
an average of 120 minutes, long enough for the more
gases to decay to filterable solid daughters. During
entering the secondary containment are retained, except
leakage rate from the containment.

5.2 Iodine and Tellurium

The trans-
the noble gas
deposit on an

behavior of iodine and tellurium differs from that of
that iodine sorbs to some extent on graphite and can
react with steel, concrete and other surfaces.^{11,12,13}

It has been
about the same
half-life.¹⁴
may be used
conservative.

found that iodine isotopes are released from fuels at
the same or more slowly than xenon isotopes of comparable
therefore the equations and parameters used for xenon
iodine. A similar assumption for tellurium is even more

Iodine and
through fuel
fully understood
for iodine and
experiments
graphite would
taken.

tellurium have been observed to be delayed in passing
out sleeves below 900°C¹⁵ although the reasons are not
Published adsorption¹⁶ and diffusion coefficients¹⁷
white are too small to explain the delay. Adsorption
very low concentrations where metallic impurities in the
some significant may permit credit for delay to be

The gas
primary circuit
tion rate of
weakly adsorbed
the circuit.

the concentrations of iodine and iodine compounds in the
are determined by the release rate of iodine, the deposi-
tion rate, the purification rate, and the rate of formation of
volatile compounds such as CH_3I in the cooler parts of

The rate
circuit involves
the Peach Bottom
The capacity
the primary
of iodine and
exist yet.
reduces the
of the Dragoon
of their low
in service;
as regards
purged core

deposition currently used in calculating the primary
in Table 4 is 40% per pass, based on observations in
reactor,¹⁹ and the General Atomic in-pile loop (GAIL).²⁰
the available surface and the distribution of iodine in
it cannot be calculated accurately since a complete set
of isotherms for all the materials present does not
as at ORNL indicate that oxidation of a metal surface
of the surface for iodine.²¹ The observed capacity
of exchangers could be unrepresentative, partly because
of temperature (200°C) and partly because of their short time
of the Peach Bottom steam generators would be realistic
of iodine, but the minute amounts of iodine released by a
that the iodine loading in the steam generators is trivial

compared with the iodine loading in the steam generators of a large HTGR after 40 years.^{18,22} Examination of the behavior of the AVR steam generator could assist in resolving this uncertainty.

Currently, the capacity of the primary coolant system for iodine is assumed to be sufficient for the average deposition rate to remain constant over reactor life. However, even if this were not so, exchange between long-lived and stable iodines on the steam generator and ¹³¹I in the coolant could reduce the partial pressure of ¹³¹I substantially at the end of life.²³

During accidents, the opposite processes of evaporation and the re-entrainment of surface dust deposits must be considered. Evaporation of iodine in the absence of steam is slower than the assumed rate of dust re-entrainment which is 1% under design basis depressurization conditions. The re-entrainment value of 1% is based on experiments²⁴ which showed that < 0.5% of the iodine could be blown off with particulates. Steam is assumed to have no effect on the partition of iodine between surfaces and the coolant. Account is taken of fission product release resulting from hydrolysis of exposed fuel and oxidation of graphite. Factors relating to the adsorption and desorption of iodine are still being investigated.

Since the seals of the PCRV penetrations are purged with clean helium, all iodine leaving the PCRV under normal conditions is assumed to either pass through concrete [$\tau(5,9)$] or the turbine [$\tau(5,8)$]. The decontamination factors applied in these regions for all iodine species are: passage through PCRV wall, 100; condensation of steam, 2000; and evaporation at cooling-tower temperature, 100. Iodine leaving the containment in ventilation air is not assumed to be further attenuated except for decay in the containment. Some iodine will undoubtedly deposit on surfaces in the containment, and a further fraction may be converted to an organic form.²⁵ During an accident, containment air is assumed to be purified with 90% efficiency for (iodine) and 30% for organic species.

5.3 Metallic Fission Products

Cesium and strontium interact strongly with graphite and metallic surfaces at all HTGR temperatures and are able to form compounds with impurities in the coolant.

Cesium and strontium do not permeate silicon carbide at normal temperatures. TRISO particles having cracked SiC coatings and intact pyrolytic carbon coatings will behave like BISO coatings.

The release of cesium from BISO coated particles is however a complex process not readily described in terms of diffusion theory.²⁶ Laboratory experiments show that if the concentrations of fission products are not too high (corresponding to < 10% burnup in uranium kernels), thorium kernels

can retain up to 90% of cesium even at 1600°C.²⁷ In-pile confirmation of this behavior is being sought.

In the meantime, diffusion through the coatings is assumed to be rate-determining. The diffusion coefficients used represent a synthesis of the work of groups at ORNL^{9,26} and GAC.¹⁰ D_{Cs} in pyrocarbon is strongly dependent on temperature and coating structure; the effects of fast neutron exposure lie within experimental uncertainty.

In calculating the inventories reported in Table 4 end-of-life cesium release was set at 1% for all TRISO fuel equal to the failed fuel fraction, and 10% for BISO particles.

For strontium, a similar situation obtains for ThO_2 kernels and release has been conservatively set at 50%.

The fluxes of cesium and strontium from fuel rod to graphite are influenced by sorption on the adjacent fuel rod matrix and graphite materials. At equilibrium, this may be described by a partition coefficient or sorption ratio (ϕ)* which may be computed from vapor pressure versus composition curves for the matrix and graphite involved. Measurements indicate that matrix-type materials, compared to fuel element graphite, will sorb up to 50 times more cesium and strontium on a volume basis.²³ Like the vapor pressures, the partition coefficients are structure and irradiation sensitive. However, the temperature-sensitivity of the vapor pressure curves is not reflected fully in ϕ since the heats of adsorption are not strongly temperature dependent.

Some consideration has been given to the possibility that equilibrium across the gap between fuel rod and graphite may not be reached as a result of the very low pressure (and hence mass transfer coefficient) so use of an equilibrium based ϕ may be unduly conservative.²⁹ Data from operating fuel elements are needed. In calculating the primary circuit inventories in Table 4, ϕ_{Cs} and ϕ_{Sr} were taken to be 50 and 20, respectively, on a volume basis.

The ratios of $\tau(4,5)$ to $\tau(3,4)$ for cesium and strontium are determined by the vapor pressures of the metals at the graphite-helium interface and their rate of transport through the graphite. Though this graphite is cooler than the fuel coating and ~ 100 times thicker, it is considerably more porous. Those species able to move as vapors will therefore move much faster than in pyrocarbon, particularly if there is a permeation flow of helium in the same direction;^{30,31} those species whose transport involves movement ^{inv} on internal surfaces or within crystallites will be

* $\phi = \frac{\text{conc. of metal in fuel matrix}}{\text{conc. of metal in graphite}}$

more effectively delayed though D is known to increase at higher concentrations. Helium flow and graphite structure sensitivity are greater for cesium than for strontium. Transport of the two metals through graphite as a result of helium flow through the graphite is currently assumed to be unimportant in comparison with condensed phase transport on the basis that the diffusion coefficients used are adequately conservative.

Metal vapor pressures over graphite generally exhibit Freundlich-type adsorption behavior except at low concentration. Vapor pressure varies logarithmically with concentration as well as temperature. At low concentrations, a transition from Freundlich sorption behavior to Langmuir behavior occurs.³² The latter is characterized by a constant heat of adsorption and a linear dependence of vapor pressure on concentration. The vapor pressure data currently used are represented by the Freundlich-type equations:

$$\ln P_{Sr} = [8.65 - \frac{36,000}{T}] + [\frac{45,900}{T} - 0.57] \ln C_{Sr}$$

and

$$\ln P_{Cs} = [28.4 - \frac{56,000}{T}] + [\frac{31,700}{T} - 19.0] \ln C_{Cs}$$

where C is in $\mu\text{moles/m}^2$, P is in atm, and T is $^{\circ}\text{K}$. The average specific surface area of graphite is about $1 \text{ m}^2/\text{g}$; that for matrix graphite is much higher. Transition from Freundlich to Langmuir behavior has been observed in the case of the cesium-graphite system at approximately $0.2 \mu\text{mole Cs/m}^2$ C. For other metal-graphite systems, Langmuir behavior is assumed to begin at the lowest experimentally determined vapor pressure. Theoretically computed corrections are applied to allow for the presence of other elements that may block adsorption sites.³³

Transport through the graphite is described by applying Fick's law as if graphite were a homogeneous isotropic solid. Release from the fuel and ϕ constitute the inner boundary conditions; evaporation through a gaseous boundary layer controls the flux at the outer boundary. The reference diffusion coefficients used are

$$D_{Cs} = 0.368 e^{-39,000/RT}$$

$$D_{Sr} = 299 e^{-60,000/RT}$$

where D is in cm^2/sec .^{8,34}

These diffusion coefficients lead one to predict that only small fractions of the strontium released from the fuel reach the coolant; for cesium one has to rely more heavily upon retention by the fuel. For the accidents considered in the PSAR, the diffusion coefficients are too small to permit any significant quantities of metallic fission products from being released from the core, even if fuel were to fail.³⁵

It is recognized that the present representation of transport through the structural graphite may oversimplify a very complex process. Refinement is being sought experimentally and through experiments in the Peach Bottom HTGR.

In the primary circuit, strontium is expected to adhere upon impact to surfaces $< 750^{\circ}\text{C}$ with unit efficiency, based upon experimental data obtained in loops.³⁶ A somewhat lower deposition efficiency is found for cesium above 600°C .³⁷ Surface capacity for metallic fission products depends upon the presence of oxide films and dusty deposits; however in this instance, as distinct from the situation with iodine, oxidation is beneficial and there are presently no reservations about capacity.³⁸

There is, however, uncertainty about the behavior of deposited metals during depressurization accidents, since it is difficult to model re-entrainment of small fractions of deposited dust under reactor conditions. On the other hand, because of the large margins between acceptable and predicted releases, it is not necessary to assume low re-entrainment fractions during a depressurization in which the secondary containment system works. It may be sufficient to point out observations that suggest that concern about re-entrainment is being overemphasized. Steep axial gradients of surface activity have been maintained for several million coolant cycles in loop and reactor systems in which strontium and cesium concentrations exceeded those of their precursors.³⁷ Even in the Peach Bottom reactor, where dust from the steam generator has been shown to play a part in cesium transport in the gas duct further downstream, the redistribution of cesium on a per pass basis was trivial.¹⁰ Cesium and strontium are also expected to transfer, at least partially, from carbon dust to surface oxide films³⁹ or even to dissolve in base metals at high enough temperatures.⁴⁰ Finally, prolonged cooking is expected to make dust, as distinct from fission products, adhere more effectively to surfaces. Further information on re-entrainment is expected from the planned Peach Bottom HTGR post-power generation program and parallel laboratory investigations.

The ratio (metal in coolant)/(metal on surfaces) for cesium and strontium nuclides has been found to be very much smaller than for iodine in loops and reactors.^{18,37} Therefore, the value of 40% plateout per pass used in calculating the primary circuit circulating activity appears to be quite conservative for cesium and strontium.

Cesium and strontium are assumed to be removed from helium leaking through the PCRV wall with the same efficiency as iodine.

5.4 Tritium

The principal sources of tritium in HTGR systems are ternary fission and neutron activation of (1) He-3 present in the helium coolant, (2) Li-6 and Li-7 present as impurities in the core and reflector materials, and (3) B-10 in the control rods and reflector materials. Other tritium-producing reactions are relatively unimportant. The tritium production rate in a typical 2000 MW(t) HTGR is about 12,000 Ci/yr.

The release of tritium from the fuel elements, control rods, and burnable poison rods is currently treated by assuming that the tritium from failed fuel particles, Li-6 in the graphite, and B-10 in the control material is all released into the helium coolant. Tritium behavior has been recently reviewed.⁴¹

Some work has been done by GAC⁴² on the retention of tritium by fuel kernels and coated particles. The results of this work generally indicate that tritium is retained in intact coated particles at normal operating temperatures and that the retention decreases rapidly with increasing temperature. Silicon carbide coatings appear to be more retentive of tritium than are pyrocarbon coatings.

Tritium produced from He-3 can also be retained by core materials (either by exchange or sorption), but the capacity of the graphite is unknown.⁴³ Appreciable retention of tritium born in control materials is also indicated. Confirmation is required.

Tritium is currently assumed to diffuse through steam generator walls at a rate of 48 Ci/year, based upon scaling of observations made at Peach Bottom. Tritium entering the secondary coolant system will be divided between the gas and liquid waste systems, somewhat at the operator's option. Gaseous release would ultimately be as water vapor containing HTO and a very small fraction as HT.

During an accident, tritium in the primary and secondary coolant systems could be released, depending on where a rupture occurred. No tritium from the core is assumed to be lost at that time. Condensation of steam in the containment vessel reduces the gas-borne ³H concentration.

6. CONCLUSIONS

1. The inventory of fission products in the primary circuit is well controlled by the graphite core and particle coatings.

2. The 2000 MW(t) HTGR has substantial margins between calculated fission product releases and guideline releases established by the AEC. For normal conditions, the smallest margin, a safety factor of 22, is for ⁸⁸Kr. The predicted ⁸⁸Kr release is 45 Ci and the guideline release is 1000 Ci (based on a whole-body dose at the site boundary and the AEC guideline of 5 mrem/yr). For accident conditions, the smallest safety factor is 330, which occurs for ⁹⁰Sr in the design basis depressurization accident. The predicted ⁹⁰Sr release in this accident is 0.067 Ci and the guideline release is 22 Ci (based on a bone dose guideline of 150 rem).

3. The mathematical models and parameters used in computing the primary circuit inventory, which can be regarded as the source of fission products during accidents and normal operation are currently chosen to yield conservative margins. However, in view of the complexity of some of the phenomena involved, verification of margins by surveillance and additional R&D are clearly desirable.

4. The pressures of HTGR development are in the direction of higher helium outlet and core temperatures and therefore tend to decrease available margins. If the apparently high safety margins of HTGR are to be maintained in advanced systems, not only improved fuels but more precisely defined fission product transfer functions will be needed.

5. While the projected release of ^{137}Cs is acceptable from a safety standpoint, the presence of this and other condensable gamma-emitting isotopes in the primary circuit must be considered from the standpoint of plant maintenance.

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