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FAST FLUX
TEST FACILITY
PROJECT REPORT

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NOBLE GAS RELEASE
TO FTR COVER GAS

W. L. Bunch

March 1968

AEC RESEARCH &
DEVELOPMENT REPORT

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NOBLE GAS RELEASE TO FTR COVER GAS

W. L. Bunch

INTRODUCTION

An inert cover gas is to be provided for the FTR sodium coolant to prevent oxidation of the liquid metal and suppress inleakage of undesirable impurities. The cover gas needs to be chemically inert to the materials within the sodium system, and should not contribute significantly to the radiation problems associated with the reactor. Cost and availability of the required quantities of gas must also be considered. Gaseous fission products can enter the cover gas, either as the result of a cladding failure or if vented fuel is used in the reactor. Therefore, the influence of the fission products must be considered in the design of the cover gas system and in the selection of the cover gas. The purpose of the present study is to estimate the release of fission products to the cover gas in order to provide a basis for including this parameter in the conceptual design of the FTR cover gas system.

NUCLEAR BASES AND DATA

The noble gases (krypton and xenon) are expected to be the fission products that will be able to traverse the sodium and enter the FTR cover gas in sufficient quantities to influence the design of the cover gas system. The quantities of these gases that enter the cover gas will depend on their production rate within the fuel, their effective holdup time from formation to entrance into the cover gas, and the fraction that escapes with this effective holdup time. This study has been directed primarily at establishing the production rates and the relations that apply. Calculations have been

included for two simple cases to indicate the important roles of holdup time and release fraction.

Nuclear data for use in evaluating the release of noble gas isotopes to the cover gas system are given in Table I. The tabulated values are defined as follows:

- Y_i These values are the effective cumulative yield factors (percent) for each isotope or isomeric state considering direct yield factors together with decay chains and branching ratios, but neglecting the possible effect of subsequent neutron transmutations. These cumulative yield factors assume equilibrium conditions exist in the decay chain. Because of the short half lives of most precursors, the values would be attained after a few hours of operation. However, in the case of ^{131}Xe , several weeks of operation would be required to attain the tabulated value because of the 8.05 day half life of ^{131}I . The direct yield factors are based on values presented in LA-3383.⁽¹⁾ The effect of branching ratios through isomeric states was included by using factors tabulated by Gumprecht for use with RIBD.⁽²⁾
- P_i The effective cumulative production rate of each isotope or isomeric state has been calculated for an operating level of 400 MW. The tabulated values are based on an energy release of 215 MeV per fission event, with a resultant conversion factor of 1.16×10^{19} fissions per second at 400 MW.

(1) C. A. Anderson, Jr., "Fission Product Yields from Fast (~1 MeV) Neutron Fission of Pu-239," LA-3383, December 30, 1965.

(2) W. L. Bunch and D. R. Marr, "Fast Reactor Library for use with RIBD," BNWL-CC-868, October 17, 1966.

Table I

Nuclear Data for Noble Gas Fission Product Isotopes^(a)

	Y_i Effective % Cumulative Yield	P_i 10^{17} atoms/sec at 400 MW	τ_i Half Life, Days	λ_i^d Decay Constant Days ⁻¹	λ_i^s Decay Constant Sec ⁻¹	Decay Energy, 10^{-13} watt-sec/decay	
						E_i^β	E_i^γ
<u>Krypton</u>							
83 ^m	0.38	0.44	7.75^{-2}	8.94^0	1.035^{-4}	0	0.066
83	0.38	0.44	∞	0	0	0	0
84	0.52	0.60	∞	0	0	0	0
85 ^m	0.69	0.80	1.833^{-1}	3.78^0	4.37^{-5}	0.46	0.287
85	0.13	0.15	3.927^3	1.76^{-4}	2.04^{-9}	0.40	0.0064
86	0.90	1.04	∞	0	0	0	0
87	1.145	1.33	5.278^{-2}	1.21^1	1.40^{-4}	2.15	1.745
88	1.392	1.61	1.167^{-1}	5.94^0	6.87^{-5}	0.61	3.08
89	1.598	1.85	2.222^{-3}	3.12^2	3.61^{-3}	2.23	3.63
90	1.463	1.70	3.819^{-4}	1.814^3	2.10^{-2}	2.19	3.57
91	1.071	1.24	1.157^{-4}	5.99^3	6.93^{-2}	2.50	3.20
92	0.546	0.63	3.471^{-5}	1.995^4	2.31^{-1}	4.37	0
93	0.184	0.21	2.315^{-5}	2.99^4	3.46^{-1}	6.12	0
94	0.040	0.05	1.620^{-5}	4.275^4	4.95^{-1}	4.33	0
95	0.005	0.006	1.157^{-5}	5.99^4	6.93^{-1}	7.32	0
<u>Xenon</u>							
131 ^m	0.024	0.03	1.20^1	5.77^{-2}	6.68^{-7}	0	0.262
131	3.40	3.94	∞	0	0	0	0
132	4.55	5.28	∞	0	0	0	0
133 ^m	0.114	0.12	2.30^0	3.01^{-1}	3.48^{-6}	0	0.374
133	5.70	6.62	5.27^0	1.31^{-1}	1.51^{-6}	0.16	0.131
134	6.449	7.48	∞	0	0	0	0
135 ^m	1.85	2.14	1.111^{-2}	6.23^0	7.21^{-5}	0	0.85
135	6.843	7.94	3.833^{-1}	1.807^0	2.09^{-5}	0.496	0.418
136	6.887	8.00	∞	0	0	0	0
137	6.496	7.54	2.708^{-3}	2.56^2	2.96^{-3}	2.37	1.60
138	4.968	5.76	9.722^{-3}	7.12^1	8.25^{-4}	1.54	0.67
139	2.987	3.46	4.745^{-4}	1.46^3	1.69^{-2}	3.20	0.64
140	1.443	1.67	1.852^{-4}	3.74^3	4.33^{-2}	1.75	0
141	0.510	0.59	1.968^{-5}	3.52^4	4.07^{-1}	3.78	0
142	0.110	0.13	1.736^{-5}	3.99^4	4.62^{-1}	2.72	0

(a) Note abbreviation that 7.75⁻² = 7.75 x 10⁻²; 3.78⁰ = 3.78; 3.927³ = 3.927 x 10³; etc.

τ_i The half life of the isotope or isomeric state, in days.
These values were also taken from the RIBD library.⁽²⁾

λ_i^d The decay constant for each isotope, in reciprocal days.
The decay constant is equal to 0.693 divided by the half life. The activity (or number of radioactive nuclei) of a particular species decreases exponentially with time; i.e. as $\exp(-\lambda_i t)$, where the time t is measured in the same units as the decay constant.

λ_i^s The decay constant in reciprocal seconds, for convenience in considering the effect of short decay time.

E_i^β The average energy associated with the beta rays emitted as the nucleus decays. The average beta ray energy is significantly less than the maximum measured energy for a given species. The relationship between the maximum and the average energy is dependent on the degree of forbiddenness of the transition. The rest of the energy is presumably carried away by the neutrino and does not contribute to the local heat removal problem. Values were taken from the RIBD library developed by Gumprecht.⁽²⁾ For short half-life isotopes in which experimental data is incomplete, all of the energy is assumed to be associated with the betas and none with the gammas.

E_i^γ The average energy associated with the gamma radiation emitted as a result of the disintegration process. This energy can be associated either with a complex or simple spectrum. The total energy values were taken from the RIBD library.⁽²⁾

RELATIONS

A. Inventory in Reactor

Under the assumptions discussed above for the cumulative yield factor, the radioactive content of the core will reach an equilibrium value equal to the production rate. That is, the number of radioactive nuclei of a particular species will continue to increase until the decay rate equals the production rate. Such a condition should exist after a few weeks of operation for all of the radioactive species but ^{85}Kr . The ground state of this isotope has a half-life of 10.76 years; hence, the inventory will increase throughout the fuel irradiation. In general, the radioactive inventory of the i^{th} species, I_i^* , is given by

$$I_i^* = \frac{P_i}{\lambda_i} [1.0 - \exp(-\lambda_i T)]$$

where T is the length of time the fuel has been irradiated. After a few weeks of operation, the term in brackets is essentially equal to unity for all species but ^{85}Kr , and

$$I_i^* = P_i / \lambda_i$$

For the stable species, the inventory continues to increase throughout the irradiation and is given by

$$I_i = P_i T$$

where T must be in seconds for P_i in atoms per second.

B. Release to Cover Gas

The rate at which noble gas might be expected to enter the cover gas can be estimated by considering an effective holdup time. It is presumed the

fission products will be generated within the reactor. At the instant a noble gas nucleus is generated, it is free to begin a tortuous path to the cover gas that takes an effective length of time H . If the nucleus decays prior to reaching the cover gas, it is no longer a noble gas and its journey ends short of the cover gas. Thus, the rate at which radioactive noble gas nuclei reach the cover is

$$N_i^* = f P_i \exp(-\lambda_i H)$$

where f is the fraction of the nuclei generated that have an escape path with this effective holdup time. For the stable isotopes, the equilibrium release rate to the cover gas is

$$N_i = f P_i$$

independent of the holdup time.

C. Concentration in Cover Gas

The concentration of noble gas fission products in the cover gas can be calculated for specific systems. The simplest case is that of a single pass system in which fresh gas enters the system continuously at a flow rate F cubic centimeters per second, and leaves the system at the same rate carrying along the fission products. In practice, the gas could be filtered to remove the fission products and then released, or could be filtered, retained, and reused. For this simple system, the concentration of each isotope in the cover gas is

$$Q_i = N_i/F \text{ or } Q_i^* = N_i^*/F$$

where the Q 's are in atoms per cubic centimeter.

D. Activity of Cover Gas

The specific activity of the cover gas will be directly proportional to the concentration of radioactive nuclei:

$$G_i = \frac{Q_i^* \lambda_i}{3.7 \times 10^4} (\mu\text{Ci/ml})$$

The radiological hazard associated with this concentration can be estimated by considering (i) the gamma field associated with a specific geometry array of the contained gas, and (ii) the maximum permissible concentration that can safely be released to habitable areas, as defined by appropriate established criteria.

E. Buildup on Filter

If the cover gas is passed through a perfect filter that removes all of the noble fission products, the number of atoms trapped on the filter is given by:

$$B_i(t) = N_i t \text{ (stable nuclei, } t \text{ in seconds)}$$

$$B_i^*(t) = \frac{N_i^*}{\lambda_i} [1.0 - \exp(-\lambda_i T)] \text{ (radioactive nuclei)}$$

where N's are related to the production rate as described previously. After a few weeks of operation the term in brackets need only be considered for ^{85}Kr . In the relation for $B_i^*(t)$, the transport time in the cover gas can be included by incrementing the holdup time H to include this additional path.

F. Heat Generated by Fission Products

If it is assumed all of the radioactive fission products remain trapped in the filter, the heat generation rate in the filter can be obtained from the product of the disintegration rate and the average energy release per disintegration. The disintegration rate is $B_i^*(t) \lambda_i^S$. Essentially all of the beta energy would be deposited within the filter near the point of

disintegration. The gamma energy would be spatially distributed in a manner dependent on the gamma energy spectrum for the specific isotope and the geometric arrangement of materials in the filter. To a first approximation it can be assumed the gamma energy is also deposited at the point of disintegration, if a conservative estimate of the cooling requirements for the filter is desired. A practical filter system will probably consist of a number of separate filters in series. Each of the filters retains the gas for a finite length of time, after which it escapes to the following filter (if it has not already decayed). The amount of energy dissipated in each filter depends on the length of time the gas is retained in each. Because of half life, the contribution of each isotope must be determined separately. If each of the filters in a series system retains the gas for the same length of time t_f , then the equilibrium decay rate on the n-th filter is given by

$$D_i^* = B_i^* \lambda_i \exp[-(n-1) \lambda_i t_f] [1.0 - \exp(-\lambda_i t_f)]$$

A sufficient number of filters would need to be employed in series such that the amount of radioactive gas leaving the last filter was tolerable. This implies that essentially all of the decay energy would be deposited within the filter system.

SAMPLE RESULTS

Two sample calculations have been made to illustrate the use of the tabulated nuclear data and the relations defined. The first case is that of vented fuel, while the second is for an assumed cladding failure in non-vented fuel.

A. Vented Fuel

It was assumed that all of the fuel generating 400 MW of power would be vented to allow the noble gas to escape to the cover gas ($f = 1.0$). To eliminate the effect of short term transient effects prior to saturation of the decay chains of interest, it was assumed the reactor had operated for about 115 days or 10^7 seconds. After this length of time all of the radioactive isotopes would be effectively saturated except for ^{85}Kr , which would have attained about 2% of its saturated value. It was further assumed that a holdup time of 5 days would exist for the transfer of a noble gas nucleus from its point of formation in the core to the cover gas. A cover gas flow rate of 10^4 cubic centimeters per second was assumed. Results of the calculations for isotopes of interest are given in Table II. The contribution of the short half-lived isotopes is negligible because of the holdup time. Cover gas activity and decay heat to the filter would be due primarily to ^{133}Xe for these assumed conditions.

B. Cladding Failure

A value of 10^{-6} was assumed for f to indicate the fractional quantity of material located within the vicinity of the cladding failure and thus able to contribute. As before, it was assumed the fuel had operated for 115 days prior to the failure, and the cover flow rate was 10^4 cubic centimeters per second. An effective holdup time of 10 seconds was used. The cladding failure would initially release the existing inventory of stable gas, after which the "equilibrium" values of Table III are calculated to apply. The initial inventory for the example case would be the product of the

Table II

Vented Fuel

Assumptions: $t=115$ days, $H=5$ days, $f=1.0$, $F=10^4$ cm³/sec

	10^{17} atoms/sec to Cover Gas		10^{13} atoms/cm ³ in Cover Gas		Cover Gas Activity $\mu\text{Ci}/\text{cm}^3$	Heat Generation in Filter Watts		
	N_i^*	or N_i	Q_i^*	or Q_i	G_i	H_B	H_Y	H_{Total}
<u>Krypton</u>								
83		0.44		0.44				
84		0.60		0.60				
85	0.15		0.15		8.25^{-2}	12.00	0.19	12.19
86		1.04		1.04				
<u>Xenon</u>								
131^m	0.0225		0.0225		4.06	0	58.9	58.9
131		3.94		3.94				
132		5.28		5.28				
133^m	0.0265		0.0265		2.49^1	0	99.1	99.1
133	3.44		3.44		1.40^3	5510	4510	10,020
134		7.48		7.48				
135	0.0009		0.0009		5.09^0	4.47	3.76	8.23
136		8.00		8.00				

Note: Superscript m denotes metastable state

Table III

Cladding Failure

Assumptions: $t=115$ days, $H=10$ seconds, $f=10^{-6}$, $F=10^4$ cm³/sec

	10 ¹¹ atoms/sec to Cover Gas		Cover Gas Activity μCi/ml	Maximum Heat Generation in Filter Watts			
	*	or		N _i	H _B	H _Y	H _T
	N _i						
<u>Krypton</u>							
83 ^m	0.44		0.0123	0	.00029	0.00029	
83		0.44		0	0	0	
84		0.60		0	0	0	
85 ^m	0.80		0.00945	.0037	.0023	0.0060	
85	0.15		(8.25 ⁻⁸)	.000012	~0	0.000012	
86		1.04		0	0	0	
87	1.33		0.0503	0.0286	.0232	0.0518	
88	1.61		0.0299	0.0098	.0496	0.0594	
89	1.78		1.74	0.0397	.0646	0.1043	
90	1.38		7.82	0.0302	.0493	0.0795	
91	0.62		11.6	0.0155	.0198	0.0353	
92	0.063		3.93	0.00275	0	0.00275	
93	0.0067		0.627	0.00041	0	0.00041	
94	0.00035		0.0468	0.00002	0	0.00002	
95	0.00000		~0	~0	0	~0	
<u>Xenon</u>							
131 ^m	0.03		(5.42 ⁻⁶)	0	.00008	0.00008	
131		3.94		0	0	0	
132 ^m		5.28		0	0	0	
133	0.12		(1.13 ⁻⁴)	0	.00045	0.00045	
133	6.62		0.027	0.0106	.0087	0.0193	
134		7.48		0	0	0	
135 ^m	2.14		0.0417	0	.0182	0.0182	
135	7.94		0.0449	0.0394	.0332	0.0726	
136		8.00		0	0	0	
137	7.31		5.85	0.173	0.117	0.290	
138	5.71		1.27	0.0879	0.0382	0.1261	
139	2.92		13.4	0.0934	0.0187	0.1121	
140	1.08		12.6	0.0189	0	0.0189	
141	0.0081		0.0891	0.00031	0	0.00031	
142	0.0013		0.0162	<0.00004	0	<0.00004	

Note: Superscript m denotes metastable state

tabulated production rates and the assumed operating time of 10^7 seconds.

Thus, for example, the inventory of ^{136}Xe would be $\sim 8.0 \times 10^{18}$ atoms.

It can be seen that the short half-lived isotopes are dominant; thus, the results are sensitive to the assumed holdup time.

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