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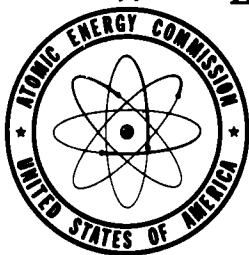
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**THEORETICAL STUDY OF FISSION PRODUCT  
GASEOUS ACTIVITY FROM A HOMOGENEOUS  
REACTOR**

By  
J. R. Donaldson

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Livermore Research Laboratory  
California Research and Development Company  
Livermore, California



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**ABSTRACT**

Buildup of the gaseous radioisotopes which are expected to be found in the atmosphere above a solution-fueled reactor has been calculated as a function of operating time. Krypton, iodine, xenon, and bromine have been considered. Direct fission and daughter yields are included in the summations of activities. Decay upon shutdown following various periods of equilibrium operation are tabulated.

THEORETICAL STUDY OF FISSION PRODUCT  
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HOMOGENEOUS REACTOR

1. INTRODUCTION

This report is concerned with the radioactivity of the gaseous fission products from the operation of a homogeneous reactor. The activity is calculated for each significant isotope found as a gas in the reactor atmosphere. The maximum permissible concentrations in populated areas are also listed. These values are applicable especially to reactors of the water boiler-type. Several such reactors (1)(2)(3)(4) are operated in this country, and they have proved useful tools in the study of reactors and neutron physics. They are the simplest and safest reactors operated to date. More of them will undoubtedly be built as research tools in the near future by universities, industrial concerns, and laboratories of the Atomic Energy Commission. For any such reactor a study of the exhaust gas activity and hazard will be of considerable interest and importance.

2. CALCULATION OF ACTIVITY

It is desired to know the activity of each component of the fission gases as a function of the time of buildup and decay. The activity of each isotope for which data are available is calculated using standard equations for fission product buildup and decay together with the known fission product decay schemes (5), independent fission yields, and decay constants (6). A second source (7) of decay constants has been used if values given in the primary reference are approximate.

2.1 Buildup

The equations for fission product buildup are easily derived, and well known. They give in each case expressions of the form

$$N_i = FG_i$$

where  $N_i$  = number of atoms of isotope  $i$   
 $F$  = number of fissions per minute in the reactor  
 $G_i$  = a function depending on the isotope  $i$ .

Converting the number of atoms to curies and the number of fissions per minute to power in the reactor, the number of curies present is

$$C_i = \frac{\lambda_i N_i}{3.7 \times 10^{10} \times 60} = \frac{\lambda_i W \cdot 3.1 \times 10^{10} \times 60 G_i}{3.7 \times 10^{10} \times 60} = 0.837 W \lambda_i G_i$$

where  $\underline{W}$  = operating power of the boiler in watts

$\lambda_i$  = decay constant ( $\text{min}^{-1}$ ) of isotope  $i$

$3.7 \times 10^{10} \text{dps} = 1 \text{ curie}$

$3.1 \times 10^{10} \text{ fissions per second} = 1 \text{ watt}$

Assume operation at a constant power of 2000 watts. The number of curies of any isotope is

$$\begin{aligned} C_i &= 0.837 \times 2 \times 10^3 \times \lambda_i G_i \\ &= 1.675 \times 10^3 \lambda_i G_i \end{aligned}$$

Now assume a fission product chain such that

Isotope 1  $\longrightarrow$  Isotope 2  $\longrightarrow$  Isotope 3  $\longrightarrow$  . . .

and let  $P_i$  be the direct fission yield of isotope  $i$  in number of atoms per fission. Then the equations for the activity of the isotope in curies are:

$$C_1 = 1.675(10^3) P_1 (1 - e^{-\lambda_1 t})$$

$$C_2 = 1.675(10^3) \lambda_2 \left\{ \frac{P_1 + P_2}{\lambda_2} (1 - e^{-\lambda_2 t}) - \frac{P_1}{\lambda_2 - \lambda_1} (e^{-\lambda_1 t} - e^{-\lambda_2 t}) \right\}$$

$$C_3 = 1.675(10^3) \lambda_3 \{ K - L e^{-\lambda_1 t} + M e^{-\lambda_2 t} - (K - L + M) e^{-\lambda_3 t} \}$$

where  $K = \frac{P_1 + P_2 + P_3}{\lambda_3}$

$$L = \frac{P_1 \lambda_2}{(\lambda_1 - \lambda_2)(\lambda_1 - \lambda_3)}$$

$$M = \frac{P_1 \lambda_2}{(\lambda_2 - \lambda_1)(\lambda_3 - \lambda_2)} - \frac{P_1 + P_2}{\lambda_3 - \lambda_2}$$

$$C_4 = 1.675(10^3) \lambda_3 \lambda_4 \{ R + S e^{-\lambda_1 t} - Q e^{-\lambda_2 t} + U e^{-\lambda_3 t} - W e^{-\lambda_4 t} \}$$

where  $R = \frac{\lambda_3 K + P_4}{\lambda_3 \lambda_4}$

$$S = \frac{L}{\lambda_1 - \lambda_4}$$

$$Q = \frac{M}{\lambda_2 - \lambda_4}$$

$$U = \frac{K - L + M}{\lambda_3 - \lambda_4}$$

$$W = R + S - Q + U$$

## 2.2 Decay

Similarly, the equations for fission product decay are:

$$C_1 = C_1^0 e^{-\lambda_1 t} (*)$$

$$C_2 = \frac{\lambda_2}{\lambda_2 - \lambda_1} C_1^0 (e^{-\lambda_1 t} - e^{-\lambda_2 t}) + C_2^0 e^{-\lambda_2 t}$$

$$C_3 = A e^{-\lambda_1 t} + B e^{-\lambda_2 t} + (C_3^0 - A - B) e^{-\lambda_3 t}$$

$$\text{where } A = \frac{\lambda_2 \lambda_3 C_1^0}{(\lambda_2 - \lambda_1)(\lambda_3 - \lambda_1)}$$

$$B = \frac{\lambda_3 C_2^0}{\lambda_3 - \lambda_2} - \frac{\lambda_2 \lambda_3 C_1^0}{(\lambda_2 - \lambda_1)(\lambda_3 - \lambda_2)}$$

$$C_4 = E e^{-\lambda_1 t} + H e^{-\lambda_2 t} + J e^{-\lambda_3 t} + (C_4^0 - E - H - J) e^{-\lambda_4 t}$$

$$\text{where } E = \frac{\lambda_4 A}{\lambda_4 - \lambda_1}$$

$$H = \frac{\lambda_4 B}{\lambda_4 - \lambda_2}$$

$$J = \frac{\lambda_4 (C_3^0 - A - B)}{\lambda_4 - \lambda_3}$$

## 3. DECAY SCHEMES

The isotopes of interest are those formed in fission which are found as radioactive gases. We must, therefore, consider isotopes of bromine, krypton, iodine and xenon, plus any others which are parents of these isotopes and appear in the equations for buildup and decay.

A complete table of decay schemes calculated is given below. The table is somewhat simplified, as several isotopes are neglected entirely. Isotopes with half lives of the order of 10 years or longer were neglected, as it was assumed that the fission gases would be flushed out at much shorter intervals than this and the buildup would be negligible. Isotopes with half lives of the order of one second were also neglected, as it was assumed their decay would be essentially complete before the gas could escape from the solution in appreciable yield. In three cases, no yields were available, and the effect was obviously small in any case. These were also neglected.

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\* $C_n^0$  denotes the number of curies of isotope n present when the reactor is shutdown.

TABLE A  
DECAY SCHEMES

Br-82  $\longrightarrow$  stable  
 Se-83m  $\xrightarrow{90\%}$  Br-83  $\longrightarrow$  Kr-83m  $\longrightarrow$  stable  
 Se-83m  $\xrightarrow{10\%}$  Se-83  $\longrightarrow$  Br-83  $\longrightarrow$  Kr-83m  $\longrightarrow$  stable  
 Se-84  $\longrightarrow$  Br-84  $\longrightarrow$  stable  
 Br-85  $\longrightarrow$  Kr-85m  $\longrightarrow$  stable  
 Br-87  $\longrightarrow$  Kr-87  $\longrightarrow$   
 Br-88  $\longrightarrow$  Kr-88  $\longrightarrow$   
 Kr-89  $\longrightarrow$   
 Kr-90  $\longrightarrow$   
 Kr-91  $\longrightarrow$   
 Kr-92  $\longrightarrow$   
 Kr-93  $\longrightarrow$   
 Kr-94  $\longrightarrow$

Te-131m  $\longrightarrow$  Te-131  $\longrightarrow$  I-131  $\xrightarrow{1\%}$  Xe-131m<sub>2</sub>  $\longrightarrow$  stable  
 Te-131m  $\longrightarrow$  Te-131  $\longrightarrow$  I-131  $\xrightarrow{99\%}$  stable  
 Sb-132  $\longrightarrow$  Te-132  $\longrightarrow$  I-132  $\longrightarrow$  stable  
 Sb-133  $\longrightarrow$  Te-133  $\longrightarrow$  I-133  $\longrightarrow$  Xe-133  $\longrightarrow$   
 Sb-134  $\longrightarrow$  Te-134  $\longrightarrow$  I-134  $\longrightarrow$  stable  
 Te-135  $\longrightarrow$  I-135  $\xrightarrow{30\%}$  Xe-135m  $\longrightarrow$  Xe-135  $\longrightarrow$   
 Te-135  $\longrightarrow$  I-135  $\xrightarrow{70\%}$  Xe-135  $\longrightarrow$   
 I-136  $\longrightarrow$  stable  
 I-137  $\xrightarrow{94\%}$  Xe-137  $\longrightarrow$   
 I-137  $\xrightarrow{6\%}$  stable Xe-136  
 I-138  $\longrightarrow$  Xe-138  
 I-139  $\longrightarrow$  Xe-139  
 Xe-140  $\longrightarrow$   
 Xe-141  $\longrightarrow$

4. SUMMARY OF ASSUMPTIONS

(1) The reactor is operated at a equilibrium power of 2000 watts. The activity, of course, is directly proportional to the power.

(2) The time after formation for the gases to escape the reactor is long compared with 1 second.

(3) The accumulated gas is flushed out so frequently that isotopes with half lives of the order of 10 years or longer can be neglected.

(4) Neutron "burnout" does not exist.

5. MAXIMUM PERMISSIBLE BREATHING CONCENTRATIONS

The maximum permissible breathing concentrations of a number of the gaseous radioisotopes have been listed by the National Council for Radiation Protection (1951). These are included with others in an effort to make this report more useful in the calculation of potential radiation hazards from radioactive gases generated by homogeneous reactors.

TABLE B  
MAXIMUM PERMISSIBLE BREATHING CONCENTRATIONS

<u>Isotope</u>	<u>MPC(<math>\mu\text{c}/\text{cm}^3</math>)</u>	<u>Reference</u>
Br-82	$7 \times 10^{-7}$	(8)
Br-83	$5 \times 10^{-6}$	(8)
Br-84	$2 \times 10^{-6}$	(8)
Kr-85	$2 \times 10^{-6}$	(8)
Kr-85m	$2 \times 10^{-6}$	(8)
Kr-87	$6 \times 10^{-7}$	(8)
Kr-88	$4 \times 10^{-6}$	(8)
I-131	$3 \times 10^{-9}$	(9)
I-132	$8 \times 10^{-8}$	(9)
I-133	$9 \times 10^{-9}$	(9)
I-134	$2 \times 10^{-6}$	(8)
I-135	$3 \times 10^{-8}$	(9)
I-136	$2 \times 10^{-5}$	(8)
Xe-133	$4 \times 10^{-6}$	(9)
Xe-135	$2 \times 10^{-6}$	(9)

6. DISCUSSION AND CONCLUSIONS

The calculations were carried out with the aid of an IBM Card Programmed Calculator. A number of checks were made by hand to confirm the accuracy of the machine. The results have been found trustworthy except in a few instances where two large numbers must be subtracted to find the activity whose numerical value is small. This was the case with some of the third and fourth members of a chain, for short periods of buildup. Since the calculation is imperfect, only where the activity is low, the inaccuracies have very little effect on the results.

Appendix A contains a sample calculation. The gross activities of the four gaseous elements involved and their totals as a function of time are presented graphically in Fig. 1. A complete tabulation of the calculation results are included as Appendix B. Buildup times of 0.1 to  $10^5$  minutes in decade steps were considered. Decay values were calculated for similar time increments.



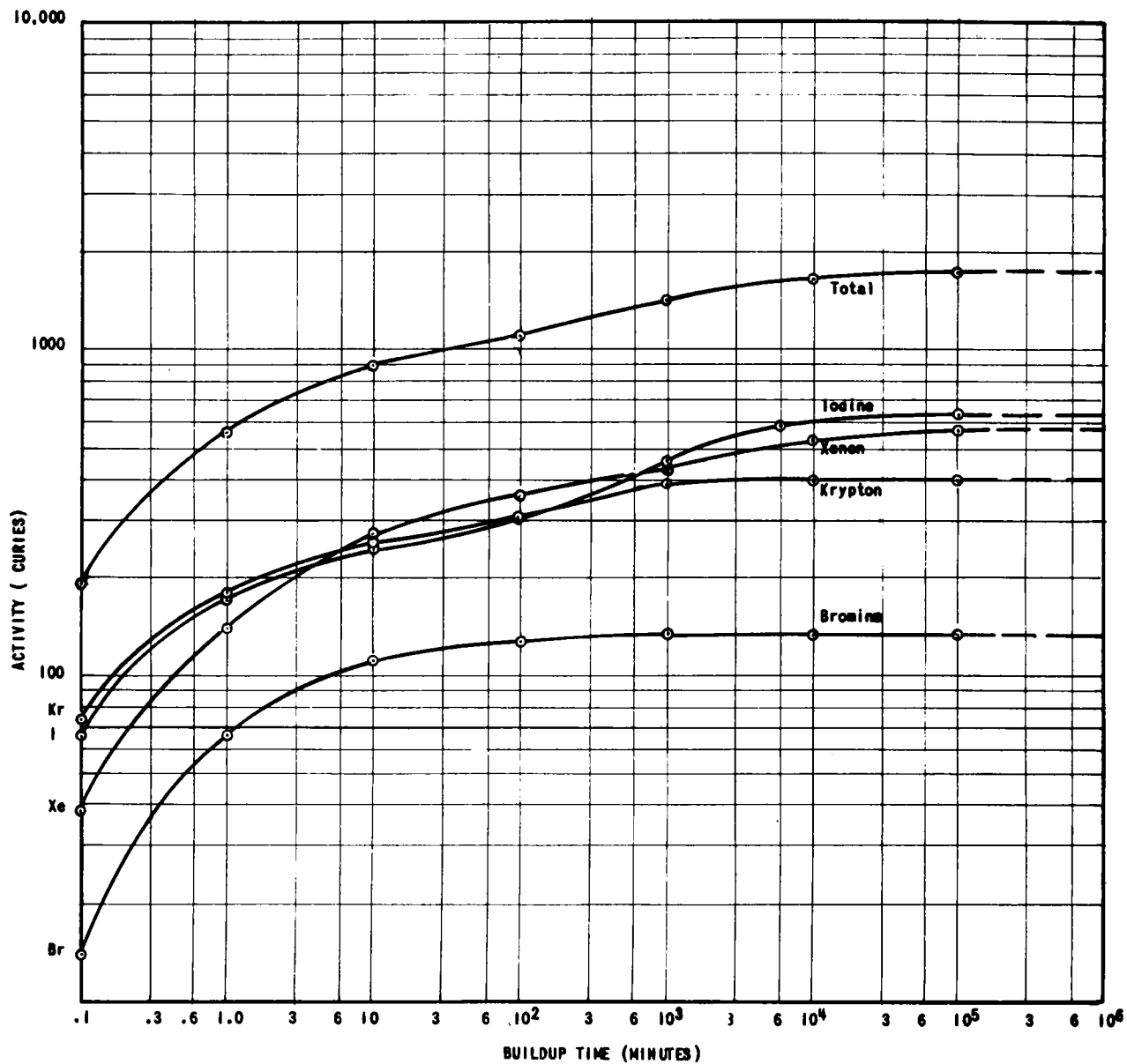


FIG. 1 - BUILDUP OF GASEOUS ACTIVITY, 2000 Watts

## 7. ACKNOWLEDGEMENTS

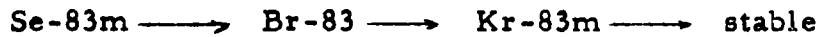
The author wishes to thank Mr. J. W. Flora for many helpful discussions of the calculations, and Mr. N. C. Ostrander and Miss Peggy Sweeney and the other members of the Computer Group for their fine cooperation in making the calculations.

## 8. REFERENCES

- (1) The Los Alamos Homogeneous Reactor, Supo Model, LA-1301, 2-7-52, Unclassified.
- (2) Low Power Water Boiler Reactor Neutron Source, NAA-SR-Memo 784, Unclassified.
- (3) Further Design Features of the Nuclear Reactor at North Caroline State College, NCSC-46, 1-52.
- (4) Description and Startup of a Water Boiler Reactor, LRL-136, Unclassified
- (5) National Nuclear Energy Series, Radiochemical Studies: The Fission Products, Book 3, Coryell and Sugarman, McGraw-Hill Publishing Co., 1951.
- (6) ADC-65, Hunter and Ballou, Unclassified.
- (7) Hollander, Perlman, Seaborg, Reviews of Modern Physics, 25, 469 (1953)
- (8) Private Communication, R. C. Thorburn.
- (9) National Council for Radiation Protection (1951).
- (10) Composition and Decontamination of Radioactive Gas Mixtures, Nucleonics, Vol. 12, No. 5 (1954).

9A. APPENDIX A

Calculate the activity of Kr-83m after 1,000 minutes of operation at 2000 watts and 1,000 minutes of decay from the decay chain



<u>Isotope</u>	<u>P<sub>i</sub>(per fission)</u>	<u>λ<sub>i</sub>(min<sup>-1</sup>)</u>
1- Se-83m	0.90(28.6) x 10 <sup>-4</sup>	0.620
2- Br-83	0	4.81(10 <sup>-3</sup> )
3- Kr-83m	0	6.13(10 <sup>-3</sup> )

Actually, the total independent yield of Se-83m is 28.6 x 10<sup>-4</sup> atoms per fission, but only 90 per cent of this decays through the chain of interest. The yield for this calculation, therefore, is taken as P<sub>i</sub> = 0.90 x 28.6 x 10<sup>-4</sup>.

I. Buildup

$$\text{Kr}^{83\text{m}}: C_3 = 1.65 \times 10^3 \lambda_3 \{K - L e^{-\lambda_1 t} + M e^{-\lambda_2 t} - (K - L + M) e^{-\lambda_3 t}\}$$

$$\text{where } K = \frac{P_1 + P_2 + P_3}{\lambda_3} = \frac{0.90(28.6)10^{-4} + 0 + 0}{6.13 \times 10^{-3}} = 0.420$$

$$L = \frac{P_1 \lambda_2}{(\lambda_1 - \lambda_2)(\lambda_1 - \lambda_3)} = \frac{0.90(28.6)10^{-4}}{(0.620 - 4.81 \times 10^{-3})(0.620 - 6.13 \times 10^{-3})}$$

$$= 6.83 \times 10^{-3}$$

$$M = \frac{P_1 \lambda_2}{(\lambda_2 - \lambda_1)(\lambda_3 - \lambda_2)} - \frac{P_1 + P_2}{\lambda_3 - \lambda_2}$$

$$= \frac{0.90(28.6)10^{-4} \cdot 4.81(10^{-3})}{[4.81(10^{-3}) - 0.620][6.13(10^{-3}) - 4.81(10^{-3})]} - \frac{0.90(28.6)10^{-4} + 0}{(6.13 - 4.81)10^{-3}}$$

$$= -1.97$$

$$K - L + M = -1.55$$

$$\begin{aligned} \therefore C_3 &= 1.675(10^3)(6.13)10^{-3} \{0.420 - 6.83 \times 10^{-3} e^{-0.620t} - \\ & 1.97e^{-4.81(10^{-3})t} + 1.55e^{-6.13(10^{-3})t}\} \\ &= 10.26\{0.420 - 6.83 \times 10^{-3} e^{-620} - 1.97(8.13)10^{-3} + 1.55(2.19)10^{-3}\} \end{aligned}$$

After 1000 min.  $C_3 = 4.18$  curies.

Similarly, the other required buildups ( $Se^{83m}$  and  $Br^{83}$ ) are found to be 4.31 and 4.19 curies respectively.

## II. Decay

$$C_3 = Ae^{-\lambda_1 t} + Be^{-\lambda_2 t} + (C_3^0 - A - B)e^{-\lambda_3 t}$$

$$\text{where } A = \frac{\lambda_2 \lambda_3 C_1^0}{(\lambda_2 - \lambda_1)(\lambda_3 - \lambda_1)} = \frac{4.81(10^{-3})6.13(10^{-3})4.31}{(-0.615)(-0.614)} = 3.37 \times 10^{-4}$$

$$\begin{aligned} B &= \frac{\lambda_3 C_2^0}{\lambda_3 - \lambda_2} - \frac{\lambda_2 \lambda_3 C_1^0}{(\lambda_2 - \lambda_1)(\lambda_3 - \lambda_2)} \\ &= \frac{6.13(10^{-3})(4.19)}{1.32 \times 10^{-3}} - \frac{4.81(10^{-3})6.13(10^{-3})4.31}{(-0.615)(1.32)10^{-3}} \\ &= 1.94 \times 10 + 1.57 \times 10^{-1} = 19.6 \end{aligned}$$

$$\begin{aligned} \text{and } C_3 &= 3.44 \times 10^{-4} e^{-620} + 19.6 e^{-4.81} + (4.18 - 3.44 \times 10^{-4} - 19.6) e^{-6.13} \\ &= 19.6(8.13)10^{-3} - 15.4(2.19)10^{-3} = 0.125 \text{ curies} \end{aligned}$$







