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ATOMIC ENERGY COMMISSION

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A REVIEW OF THE POSSIBILITY OF REACTOR PRODUCTION OF ISOTOPES CURRENTLY PRODUCED BY CYCLOTRON BOMBARDMENT

A report by the Manager, Oak Ridge Operations Office

THE PROBLEM

1. To determine the possibility of uranium nuclear reactor production of the radioisotopes now being made by cyclotron bombardment.

BACKGROUND

2. "Program for Production and Distribution of Cyclotron-produced Isotopes," AEC 195, was approved by the Commission on March 23, 1949. Part "b" of the Commission's action on AEC 195 was as follows:

> "Requested that the staff work out and report procedures to insure that wherever possible, as the Commission's reactor program develops, isotopes distributed by the Commission are reactor-produced."

3. Staff paper, "The Procedure Used by the Isotopes Division to Insure Distribution of Reactor-Produced Rather than Cyclotron-Produced Radioisotopes Wherever Possible," dispatched February 2, 1950, indicated that periodic studies are conducted by the staff of the Isotopes Division, Oak Ridge, to determine possibilities of production of cyclotron radioisotopes in the uranium reactor. This is the first of a series of

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papers complying with this commitment. Others will be prepared as improved new reactors become available for radioisotope production.

DISCUSSION

4. The twenty-five radioisotopes approved for possible cyclotron production (as listed in Appendix D of AEC 195) are: Beryllium 7, Fluorine 18, Vanadium 49, Manganese 52, Manganese 54, Nickel 57, Xenon 127, Lead 203, Astatine 211, Sodium 22, Iron 55 free of Iron 59, Iron 59 free of Iron 55, Arsenic 73, Arsenic 74, 34-hour Krypton, Cobalt 56, Cobalt 57, Cobalt 58, Silver 106, Iodine 126, Iodine 130, Rhenium 184, Gold 196, Thallium 202, and Bismuth 206.

5. To date, eleven of twenty-five radioisotopes listed above have not been reported as producible by neutron-induced reactions. These are: Beryllium 7, Vanadium 49, Manganese 52, Manganese 54 Iron 55 free of Iron 59, Cobalt 56, Cobalt 57, Arsenic 73, Xenon 127, Bismuth 206 and Astatine 211.

6. Those radioisotopes which have been experimentally produced by neutron irradiation through a (neutron, gamma), (neutron, 2 neutron), (neutron, proton) or (neutron, alpha) reaction, together with the reaction energies at which they were produced and known cross sections are presented in tabular form below.

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Isotope	Reaction	Absorption Cross Section	Energy of Bombarding Neutrons
F 18	F 19(n,2n)		90 Mev
Ni 57	Ni 58(n,2n)		12.6-16 Mev
As 74	As 75(n,2n)		Fast
Ag 106	Ag 107(n,2n)		20 Mev
I 126	I 127 (n,2n)		Fast
I 130	Cs 133(n,α)		Fast
Re 184	Re 185(n,2n)		Not given
Au 196	Au 197(n,2n)		Fast
Tl 202	Tl 203(n,2n)		Fast
Pb203	Pb 204(n,2n)		Not given
Kr 79	Kr 78(n,γ)	0.27 b	Thermal
Fe 59	Co 59(n,p,)	92 mb	1
		0.04 mb	Fission (about 1 M
Co 58	Ni 58(n,p)	0.7 mb	Fast
Na 22	Na 23(n,2n)	0.006 mb	Fast ~1 Mev

7. The 34-hour isotope of krypton has recently been assigned a mass number of 79. Krypton 79 has been produced in the pile and the isotopic cross section of Kr 78 found to be 0.27 barn. Krypton 78 has a natural abundance of .342%. Calculations show that if we assume a flux of 2 x 10^{13} thermal neutrons (given as the highest flux of the Hanford pile in HW14135), saturation activity would be about 3.6 mc per gram of krypton (Appendix "A"). At S.T.P. this amounts to 13.3 microcuries per milliliter. Other radioisotopes of krypton would also be present. The most objectionable of these would be the 4.5 hour Krypton 85. The activity of Kr 85 under the above conditions would amount to 212 mc per gram of krypton or 786 microcuries per milliliter. In order to use the Kr 79 at least one of its half-lives would be wasted in allowing the decay of Kr 85. One or more half-lives would be used in packaging and shipping. In order to procure a useable amount, 10 mc for example, 40 mc of Kr 79 would need

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to be present at removal from the pile. This would represent a volume of 4 liters of target material. Therefore, the volume of gas needed to produce a usable amount of Kr 79 would make its manufacture very difficult in the present facilities available.

8. The possibility of production of carrier-free Iron 59 in the pile has been rather extensively investigated and a recommendation prepared in MonC-142, the conclusion of which is:

> "In general, it can be concluded that production of radiochemically pure Fe 59 by pile irradiation does not compare favorably with its production by fast neutrons from the cyclotron. In the latter case a yield of 10 microcuries per hour can be achieved with a beam of 300 microamperes of 14 Mev deuterons on a Be target and 40 grams of cobalt directly behind the beryllium. The Co 60 activity is approximately a factor of 10 greater than the iron activity under these conditions making separation a simple operation."

This conclusion was drawn from the fact that the ratio of cobalt activity to iron activity produced under neutron bombardment is about 1,000 to 1. This would make a separation of cobalt and iron difficult, impracticable and expensive from the quantitative standpoint unless extremely large amounts of cobalt were to be produced.

9. Cobalt 58 has been produced by the Nickel 58 (n,p) reaction. Production and separation of any quantity of Cobalt 58 in this manner

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would be difficult, since Nickel 59 is also formed from the (n,γ) reaction on Ni 58. The cross section for thermal neutrons of the (n,γ) reaction is 4.5 barns as compared to 0.7 millibarn for the (n,p) reaction.

10. Sodium 22 has been produced in the pile and the cross section for its production studied extensively. However, because of the extremely small neutron absorption cross section for a (n,2n) reaction, 0.006 millibarn, the yield is very small. Calculations show that if one mole of sodium were bombarded at the highest appropriate flux available for this reaction (5×10^{12} as given in HW14155 for the maximum flux of 0.5 to 1.5 Mev neutrons in the Hanford pile) for one month, only 9 microcuries of Na 22 would result. This would mean a specific activity of about 0.4 microcurie per gram of total sodium. At saturation, the specific activity of Na 22 would be only about 21 microcuries per gram and would be unsuitable for biological use, its principal application. The calculations of yield may be found in Appendix "B".

CONCLUSIONS

11. The uranium nuclear reactors presently available for isotope production cannot economically make, in proper form, any of the cyclotron-produced radioisotopes approved for distribution under the Atomic Energy Commission's cyclotron program.

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LIST OF APPENDICES

APPENDIX "A"

SATURATION ACTIVITY IN MILLICURIES OF KRYPTON 79 PER GRAM OF KRYPTON ASSUMING FLUX OF 2×10^{13} THERMAL NEUTRONS

APPENDIX "B"

PRODUCTION OF SODIUM 22 FROM 1 MOLE Na 23 AFTER ONE MONTH BOMBARDMENT ASSUMING A MAXIMUM FLUX OF 5×10^{12}

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APPENDIX "A"

SATURATION ACTIVITY IN MILLICURIES OF KRYPTON 79 PER GRAM OF KRYPTON ASSUMING A FLUX OF 2 x 10^{13} THERMAL NEUTRONS

$$Mc = \frac{N_o \sigma \phi (1 - e^{-\lambda t})}{3.7 \times 10^7}$$

Mc = Millicuries

N_o = Number of target atoms

σ = Cross section

ϕ = Flux

λ = Decay constant

t = Time (hours)

$$N_o = \frac{6.02 \times 10^{23}}{84} \times 0.00342$$

$$\sigma = 0.27 \times 10^{-24}$$

$$\phi = 2 \times 10^{13}$$

$1 - e^{-\lambda t} = 1$ (saturation is desired)

$$Mc = \frac{6.02 \times 10^{23} \times 3.42 \times 10^{-3} \times 2.7 \times 10^{-25} \times 2 \times 10^{13}}{8.4 \times 10 \times 3.7 \times 10^{7}}$$

$$= \frac{6.02 \times 3.42 \times 2.7 \times 2 \times 10^{8}}{8.4 \times 3.7 \times 10^{8}}$$

$$= \frac{111.19}{31.08} = 3.578 \text{ mc/gram}$$

The density of Krypton at S.T.P. (0°C 760 mm Hg) is 3.708 grams per liter.

$$1 \text{ gram} = \frac{1000}{3.708} = 269.7 \text{ milliliters/gram krypton}$$

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Saturation activity of 1 gram of Krypton 85 (4.5 hour) assuming a flux of 2×10^{13}

$$Mc = \frac{N_o \sigma \phi (1 - e^{-\lambda t})}{3.7 \times 10^7}$$

$$N_o = \frac{6.02 \times 10^{23}}{84} \times .5702$$

$$\sigma = 0.096 \times 10^{-24}$$

$$\phi = 2 \times 10^{13}$$

$$Mc = \frac{6.02 \times 10^{23} \times 5.7 \times 10^{-1} \times 9.6 \times 10^{-26} \times 2 \times 10^{13}}{8.4 \times 10 \times 3.7 \times 10^7}$$

$$= \frac{6.02 \times 5.7 \times 9.6 \times 2 \times 10^9}{8.4 \times 3.7 \times 10^8}$$

$$Mc = \frac{658.8}{31.08} \times 10 = 211.9$$

211.9 mc per gram of Kr at saturation

$$\frac{\frac{211.9}{1000}}{3.708} = \frac{211.9 \times 3.708}{1000} = 795.72 \text{ microcuries per milliliter}$$

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APPENDIX "B"

PRODUCTION OF SODIUM 22 FROM 1 MOLE Na 23 AFTER ONE MONTH BOMBARDMENT ASSUMING A MAXIMUM FLUX OF 5 x 10^{12}

$$Mc = \frac{N_o \sigma \phi (1 - e^{-\lambda t})}{3.7 \times 10^7}$$

N_o = No. of target atoms

σ = Cross section

ϕ = Flux

t = Time (months)

λ = Decay constant

$N_o = 6.02 \times 10^{23}$

σ = .006 mb = 6×10^{-30} barn

$\phi = 5 \times 10^{12}$

$\lambda = \frac{.693}{36} = 0.0193 \quad t = 1$

$$\text{Mc of Na 22} = \frac{6.02 \times 10^{23} \times 6 \times 10^{-30} \times 5 \times 10^{12} (1 - e^{-0.0193})}{3.7 \times 10^7}$$

$$= \frac{6.02 \times 6 \times 5 \times 10^5 (1 - .98088)}{3.7 \times 10^7}$$

$$= \frac{1.8 \times 10^7}{3.7 \times 10^7} \times .01912 = 0.00929 \text{ mc or } 9.29\ \mu c$$

Specific activity in $\frac{c}{gm}$ = 0.404 μc/g

Saturation activity would be 21.1 μc/gram.

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