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REACTION PRODUCTS IN HIGH nvt IRRADIATED BERYLLIUM

John E. Evans

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UNITED STATES ATOMIC ENERGY COMMISSION

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A B S T R A C T

Calculated estimates of the amounts of reactor poisons and other interesting reaction products expected to exist in beryllium that has been irradiated in the MTR (nvt of 2×10^{22} thermal neutrons/cm², 5×10^{21} fast neutrons/cm²) are listed in the table below. The values show that these reaction products (except B¹⁰) can be experimentally measured.

It is proposed that a piece of beryllium from a burned-out MTR shim rod be utilized by the MTR Technical Branch for experimental measurements of these products.

Table I. Reaction Product Concentrations per cm³ of Be.

B ¹⁰ (yearly vol. effect)*	$\sim 3 \times 10^{12}$ atoms	$\sim 10^{-5}$ wt ppm
B ¹⁰ (0.004" surface effect)	$\sim 10^{11}$	$\sim 10^{-6}$ " "
Be ¹⁰	2.2×10^{19}	180 " "
Li ⁶	$> 1.2 \times 10^{18}$	> 10 " "
He ⁴	3×10^{19}	0.8 cm ³ NTP
He ³ (10/1/56)	$> 1.7 \times 10^{17}$	> 0.003 " "
H ³	$> 4.8 \times 10^{18}$	> 0.1 " "

* starting 2/27/56

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I. INTRODUCTION

Changes detrimental to reactor operation are produced in beryllium subjected to high neutron and gamma fluxes. The reactivity losses caused thereby are not serious over a period of a few years at MTR fluxes but may become serious in higher flux reactors. The production of Li^6 , and its subsequent conversion to H^3 , which in turn decays to He^3 was suggested as a possible explanation for a loss of reactivity experienced in the early operating period of the MTR.⁽¹⁾ Morris⁽²⁾ and McMurtry and Webster⁽³⁾ calculated the loss expected from this source.

Structural damage has not been evidenced in MTR Be, but there exists the possibility that production of He^4 in large amounts could cause pressure damage to fabricated pieces.

It is proposed that beryllium from an MTR shim rod recently removed (after 31,000 mwd of operation) and cut into small sections be analyzed to determine quantitatively the reaction products contained. Some of the important results that can be obtained from these analyses are:

1. A check of calculated values for poison buildup in Be at MTR flux levels: this would show whether the various processes are understood and would demonstrate whether or not the processes now being considered are sufficient and can be used with confidence in

estimating Be life in future reactors with fluxes of 10^{16} or more.

2. An estimate of the amount of the produced tritium, if any, that diffuses out into the cooling water. Such diffusion would decrease the reactor poison effect but would be a health hazard.
3. An estimate of the fast nvt level at which structural damage caused by pressure buildup of He and H^3 could be a factor. Possibly this pressure buildup can be correlated with changes in the metallurgical properties of irradiated Be.
4. A measurement of the amount of Be^{10} produced which would serve to check whether or not Be^{10} has a high absorption cross section, and it would be a preliminary step toward the production and separation of appreciable amounts of this isotope. Detailed studies of its nuclear properties could show whether or not small amounts of this isotope are unduly harmful to the moderating and reflecting properties of Be.

There are a number of reasons for carrying out this program here at the

MTR:

1. It builds on earlier work done at the MTR by R. H. Lewis and T. O. Passell on activation and chemical analyses of the impurities in reactor-grade Be and by Morris, McMurtry and Evans on the buildup of reactor poisons in Be.
2. A study of the metallurgical properties of irradiated Be is now in progress at the MTR and this program would mesh well with it.

3. This program would provide a basis for future work; as samples irradiated to higher nvt become available, experimental techniques developed and the data accumulated in its course will become more valuable.
4. It will be necessary to carry out these measurements here in order to be sure that we get, by doing the proper analyses, all the information that is available and pertinent to an adequate study of irradiated Be.
5. It will serve as a useful confirmation to analyses obtained in other laboratories (which are to receive portions of the present sample) which in turn will provide a cross check on the correctness of our techniques.

THE MAIN REACTIONS, YIELD ESTIMATES, AND SUGGESTED METHODS OF ANALYSIS

The main products which can be measured with a reasonable amount of laboratory effort and without the procurement of elaborate special equipment are the concentrations of Be^{10} , Li^6 , He^4 , He^3 and H^3 which are produced in the body of the Be pieces.

Surface effects, produced by reactions with the proton recoils from a thin layer of cooling water next to the Be surfaces, are smaller effects and will be mentioned only briefly in this report. When accurate proton cross sections, as a function of energy, become available for these reactions which take place near the surface, a more accurate calculation of their product yields will be justified.

The more important nuclear reactions which take place are the following:

For a thermal nvt of 2×10^{22} we get 180 ppm of Be^{10} or 2.2×10^{19} atoms $\text{Be}^{10}/\text{cm}^3$. Be^{10} has a half-life of 2.7×10^6 years and emits a β particle with a maximum energy of 0.55 Mev. No gamma rays are observed. From this calculation we expect 100 disintegrations/sec per mg of Be^9 . Thus the Be^{10} content can be analyzed by scintillation counter techniques provided there is no appreciable interference from the activation products of impurities in the Be. It has been assumed in this calculation that Be^{10} has a low absorption cross section (since Be^{10} is a neutron rich even-even nucleus) and that a negligible number of Be^{10} atoms produced will be destroyed by the neutron or gamma fluxes present. The lowest threshold gamma reaction (γ_{th}) has a threshold of 6.81 Mev.

The average cross section for this reaction for fission neutrons is given as 10 mb by Hughes, Pile Neutron Research, Addison-Wesley, Table 4-1, p. 100, 1953. However, recent work at ORNL by Campbell and Stelson⁽¹⁾ gives an average value of more than 30 mb for this cross section.

For a fast nvt of 5×10^{21} neutrons/ cm^2 and the 10 mb cross section, I calculate that $60 \mu\text{gm}$ of He^6 which goes in 0.8 sec to Li^6 will be produced for each cm^3 of Be. However, the burnup

half life of Li^6 (see Fig. 1) in a thermal flux of 2×10^{14} is 90 days. the burnup half life is about 1/10 of the 1000 days of reactor operation in which the Be piece was in the reactor. Thus the Li^6 content expected is twice the amount produced in one burnup half life or $12 \mu\text{gm per cm}^3$ of Be (10 ppm by weight). If 30 mb is a better cross section value than the 10 mb used then we could expect $36 \mu\text{gm of Li}^6 \text{ per cm}^3$ of Be. It seems to be worthwhile to try to analyze chemically for the Li^6 . Another possibility, if the chemical analysis fails, is to use fission counter techniques in the MTR thermal column and count the alpha particles and tritons from the disintegrations of Li^6 by thermal neutrons.

This reaction will convert 4/5 of the Li^6 produced into nearly equal amounts of H^3 and He^4 or $24 \mu\text{gm}$ of tritium and $32 \mu\text{gm}$ of helium per cm^3 of Be^9 . This assumes 10 mb for the production of Li^6 so amounts up to three times the above amounts may be produced. Since tritium weighs $270 \mu\text{gm/cm}^3$ and helium weighs $180 \mu\text{gm/cm}^3$ at NTP we expect to find at least 0.1 cm^3 of tritium and 0.15 cm^3 of helium per cm^3 of Be dissolved. By reducing the oxygen from the gas and water of the solution a mixture of hydrogen and helium should remain. This can be separated into the two components by means of a palladium leak. The hydrogen can be introduced into a counter

and its tritium content determined. The helium content can be determined volumetrically. Other reactions which add to the helium content are discussed below.

the burnup half life of He^3 ($\sigma_{n,p} = 5400$ barns) in a thermal flux of 2×10^{14} is 15 days. Thus the equilibrium concentration of He^3 is that produced in two half lives or 30 days is 2.2×10^{16} atoms per cm^3 of Be. Also, since the material has been out of the reactor (it was removed 2/27/56) the H^3 has been decaying into He^3 at the rate of 2.2×10^{16} atoms/month. Thus the expected concentration 10/1/56 will be 1.7×10^{17} He^3 atoms per cm^3 of Be or 0.003 cm^3 NTP. By placing the helium gas collected in a proportional counter, the protons from the n,p reaction on He^3 can be counted to give a quantitative analysis of the He^3 .

The average value of the cross section for this reaction is about 0.8 mb for gamma rays with energy >1.67 Mev. The number of gamma rays having this energy should be of the same order of magnitude as the fast neutron flux or 5×10^{21} . This yields 10^{18} helium atoms or about 0.03 cm^3 NTP helium per cm^3 of Be.

This reaction cross section has been measured to be 300 ± 100 mb for Ra-Be neutrons. Thus if we use 100 mb for an average cross section for neutrons > 1.67 Mev and a fast nvt of 5×10^{21} we expect to produce by this reaction 2.3×10^{19} helium atoms or 0.6 cm^3 NTP helium per cm^3 of Be.

The following four reactions are induced by the proton recoils from a thin layer of cooling water next to the Be surfaces.

└→ Probably $p + 2\text{He}^4 + 0.18 \text{ Mev.}$

Protons having energies > 300 kev are effective in the first three reactions but the last requires protons with energies greater than 2 Mev. In a 0.004 inch layer on the Be surface about 10^{12} B^{10} atoms/ cm^3 would be produced by reaction G. Since the burnup half life of B^{10} in a flux of 2×10^{14} is 21 days about four percent of the B^{10} produced or $\sim 10^{11}$ atoms of $\text{B}^{10}/\text{cm}^3$ would still be present in the surface layer of Be. About 10^{12} atoms $\text{B}^{10}/\text{cm}^3$ will also be produced each year as a volume effect by the decay of the Be^{10} atoms. Reaction H will produce of the order of 10^{15} Li^6 atoms/ cm^3 . The yields of reactions I and J are expected to be of the same order of magnitude as reaction H. Thus the last four reactions are not expected to make an appreciable contribution to the poison or to the pressure effects in the Be.

CONCLUSIONS

The amounts of Be^{10} , Li^6 , He^4 , H^3 , and He^3 in the pieces of MTR irradiated Be are sufficient to be measured by available methods of analysis. The amount of B^{10} in the surface layer is about 10^{-5} ppm and is not detectable by chemical, neutron, or mass spectrometer analysis. The amounts of Li^6 and He^4 produced by proton recoils in a 0.004" surface layer is of the order one percent of the concentrations produced throughout the volume by neutron reactions.

REFERENCES

1. Evans, EV-1-52A, May 19, 1952.
2. Morris, IDO-16024, July 1, 1952.
3. McMurry and Webster, IDO-16017, June 13, 1952.
4. Campbell and Stelson, ORNL-1975, 1956.

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FIG. I

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