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Cyclotron Targets: Preparation and Radiochemical Separation. II. Krypton

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Radioactive krypton ($T=34h$, e^+, γ) has become a very useful gas for tracer experiments. This isotope can be made by $Br^{79,81}(d, 2n)Kr^{79,81}$. Preparation and separation of this gas have been a routine matter in this laboratory for some time and a brief description of the methods used is of interest. Two different methods will be described. Both are equally good as far as the separation of the krypton is concerned. One method involves very simple manipulation, but requires an apparatus made of monel metal, fused quartz, and Pyrex glass; the other uses only readily obtainable and inexpensive apparatus, but slightly more complicated manipulation.

FUSION METHOD

SODIUM bromide was chosen for bombardment because of its high bromine content and relatively small tendency to be hygroscopic.

The Target

U.S.P. grade NaBr is used as the target material, and the amount used is of the order of 40 grams. The crystals are spread in a thin layer ($\frac{1}{16}$ inch thick) over the surface of a water-cooled copper target plate, which has the surface scored by parallel grooves similar to a coarse file to hold the powder in place. This target plate fits into a cylindrical target chamber.

The target chamber is mounted at the end of a divergent spout attached to the cyclotron[1] through which the emergent beam passes. Between the spout and the target chamber is a dural foil vacuum window to transmit the beam. The energy loss in the 0.001-inch foil is approximately 0.5 Mev and the average distance traversed in the helium before striking the target reduces the energy by another 0.1 to 0.2 Mev. Thus the effective energy of the 14.0-Mev deuteron beam (measured) is about 13.3 Mev.

The target chamber is evacuated and then filled to near atmospheric pressure with helium gas during the bombardment. The helium serves as a cooling agent for the target. The emergent deuteron beam has dimensions of approximately 6 inches by $\frac{3}{4}$ inch high at the location of the target chamber. The target plate is 6 by $5\frac{1}{2}$ inches and is arranged so that it can be tilted into a nearly horizontal position, so the $\frac{3}{4}$-inch beam is spread over the $5\frac{1}{2}$-inch width.

Separation of the Target

After the NaBr has been bombarded, it contains three main radioactive elements: sodium, bromine, and krypton.[2] The radioactive bromine and krypton are to be found as gases trapped in the crystal lattice of the NaBr. Tests of the activity of the helium from the cyclotron target chamber show that practically none of the radioactive gases is lost from the crystals. The separation of the target, therefore, involves breaking up the crystal lattice of the salt to release the gases. If the sodium bromide crystals are placed in a monel metal tube and melted, the released gas can be shown to contain radioactive krypton free from measurable amounts of radioactive bromine.

Since the number of radioactive bromine atoms is so much smaller than the number of inactive bromine atoms in the NaBr, and equilibrium exists, the number of radioactive bromine atoms free from the NaBr is completely negligible. The fact that exchange is responsible for the separation of the radioactive krypton from the radioactive bromine can be demonstrated in the following manner. Radioactive bromine gas is placed in contact with inert NaBr crystals for ten or fifteen minutes. The bromine gas is then pumped off, and the NaBr crystals are baked and pumped for twelve hours. At the end of this time, if the NaBr is tested for activity, it will be

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[1] M. S. Livingston, J. App. Phys. **15**, 2, 128 (1944).

[2] A. H. Snell, Phys. Rev. **52**, 1007 (1937); E. P. Clancy, Phys. Rev. **58**, 88 (1940); E. C. Creutz, L. A. Delsasso, R. B. Sutton, M. G. White, and W. H. Barkas, Phys. Rev. **58**, 481 (1940).

found that the activity which was present in the bromine gas is now to be found in the NaBr crystals.

The apparatus for the separation of the krypton therefore consists merely of a vessel in which the NaBr may be melted, fitted with inlet and outlet tubes so that the gas may be swept from the vessel. NaBr melts at 755°C, and, therefore, a container must be used which will stand this temperature. Quartz would stand the temperature and would be convenient, since we can see when the NaBr is melted. Unfortunately, however, quartz is attacked by the molten NaBr and shatters after a few separations. Monel metal has been found to stand up as a fusion vessel for the crystals. A hot-rolled monel metal rod 10 inches long and $1\frac{1}{4}$ inches in diameter is drilled out to form a tube closed at one end. The open end is turned to a standard taper and ground to fit the male part of a Pyrex glass ground joint. A few turns of $\frac{1}{4}$-inch copper tubing are soft-soldered onto the upper end of the tube as cooling coils for the grind. A sketch of the apparatus is shown in Fig. 1. The bombarded crystals are placed in the bottom of the tube and the apparatus is evacuated with a mechanical vacuum pump. If the krypton gas is to be swept into an evacuated flask this may be connected to the outlet of the apparatus and both the flask and the monel metal apparatus evacuated together. The metal tube is tipped to an angle of about thirty degrees to the horizontal. The bottom end containing the crystals is heated in a large soft oxygen flame to a bright red heat and kept there for about five minutes. Water running through the cooling coils keeps the grease in the ground joint from getting hot. After the NaBr is melted under a vacuum it is allowed to cool, and the released krypton gas is swept into an evacuated bulb by letting any convenient sweeping gas in through the inlet going to the bottom of the metal vessel.

SOLUTION METHOD

Sodium bromide was chosen for bombardment for this separation method also, not only because of its high bromine content and relatively small tendency to be hygroscopic, but also on account of its high solubility.

Preparation of the Target

Although the crystalline reagent salt is soluble, its rate of solution is slow. The salt is fused in a nickel crucible and the melt poured into a porcelain dish. When cool it is ground in a mortar, and a fine powder is obtained which dissolves quickly. Because the total activity of the target is high, this high rate of solution is necessary to reduce the time of handling. A slight tendency to cake has been observed with this fused powder. This is minimized by preparing small quantities (40 g), drying the final powder at 150°C for several hours, and bottling and sealing while warm.

Separation of the Target

The apparatus used for this method of separation is shown in Fig. 2. The bombarded NaBr

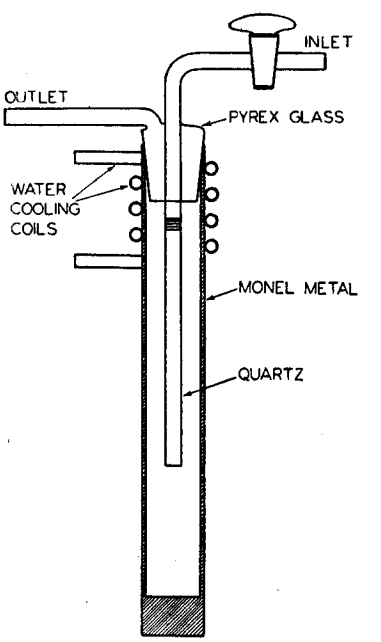

FIG. 1. Apparatus for the fusion method of separation.

is placed in the part of the apparatus marked A through the ground joint at the top. The stopcock between parts A and B is closed. The part of the apparatus marked B (volume of 70 cc) is filled two-thirds full with a solution of $0.01M$ in NaOH. The whole apparatus is then evacuated to the vapor pressure of the solution with a mechanical vacuum pump. After the apparatus

is pumped down, all the stopcocks are closed. Opening the stopcock between the parts of the apparatus *A* and *B* will allow the target material to fall into the solution. If the powdered NaBr has taken on a little water, the material may not

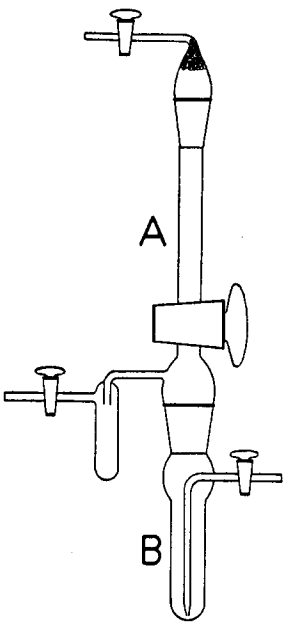

FIG. 2. Apparatus for the solution method of separation.

fall down easily, in that case either tapping or letting in a small amount of gas through the stopcock at the top will drive the powder down into the solution.

Although NaBr is quite soluble in water, shaking the mixture will greatly increase the rate of solution of the crystals. Since the whole apparatus is at reduced pressure, there is no danger that the ground joints will come apart and the whole apparatus can be taken up and shaken vigorously until the crystals are all dissolved.

Dissolving the sodium bromide removes the gases from the crystals by destroying them. The radioactive bromine is held in solution by the exchange phenomenon discussed in the fusion separation method. The purpose of the NaOH is to insure the absence of HBr in the gaseous phase above the liquid. The only remaining step is to get the krypton out of the solution. This can be done mechanically by sweeping gas through the solution. If the krypton is to be collected in a bulb, the evacuated bulb can be connected by rubber tubing to the stopcock to the left of part *B* in Fig. 2. If air is used as a sweeping gas, the stopcock to the left of part *B* is first opened and then the stopcock to the right is opened slowly. As the air bubbles through the solution, it will carry along the krypton gas. If gas other than air is used, so that it comes from a high pressure tank, a manometer must be included in the system so that the apparatus will not be filled to greater than atmospheric pressure.

To collect the gas in a very small volume, the krypton can be swept into an evacuated bulb with carbon dioxide. This gas can then be absorbed with fused sodium hydroxide and the krypton forced into a capillary side tube by displacement with mercury.

Yield

Rough measurements of the yield of the 34-hour krypton isotope have been made. Using the preparation and either separation method just described the gas was swept into an evacuated bulb. The krypton intensity measurements were made by placing the bulb containing the radioactive krypton three meters from a platinum screen cathode Geiger-Müller counter. The counter was covered by 1.1 mm of lead. All activity data were obtained by comparison with calibrated radium standards.

TABLE I.

Mg Ra γ-ray equiv. / μ amp. hr.	Mev
0.03	12
0.06	13
0.13	14

Table I gives the measured thick target yields for the 34-hour krypton isotope; the number of milligrams radium gamma-ray equivalents per microampere hour beam is given for three corresponding values of the measured beam current energy.