SILVER-111 BETA-RAY SOURCES

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Multi-millicurie sources of beta emitting materials have found many uses during the past decade. Biological systems have most often been subjected to these radiations (1-5), although investigators have also used the sources with chemical (6), and physical (7,8) systems. Investigators report using sources of phosphorous-Bakelite (9,10), Sr^{90}-Y^{90} (5,11), Ru^{109}-Rh^{109} (4,12), Tl^{204}(13), and Pd^{109} (8).

Development in this laboratory (14,15) of a new, rapid, high decontamination method for the separation of silver by isotopic exchange makes possible the routine production of large beta-ray sources of Ag^{111} from neutron bombarded palladium to supplement this list of sources.

Source Preparation:

Palladium metal, when irradiated in a nuclear reactor, forms 13.6-hr Pd^{109}, 26-min Pd^{111}, and a small amount of 17-day Pd^{103}. The Pd^{111} decays to 7.6-day Ag^{111}. In processing, these Ag^{111} atoms must be separated from grams of palladium--an element which is quite similar chemically to silver. Previous methods of separation proved tedious and not readily adaptable to high level processing.

The separation of silver by isotopic exchange, however, proves to be a very specific type of reaction which suffers little interference from macro amounts of palladium. Silver iodide is the compound used, since it suffers less interference from palladium than the silver chloride described previously. A "silver iodide" electrode is prepared and the electrode immersed in the radioactive solution to allow the process of isotopic exchange to occur and thus separate the radioactive silver from impurities. The silver iodide is then dissolved from the electrode and silver electrolysed from this solution as the metal on a platinum electrode of desired shape to form the source. For this final step, the silver may be plated on a variety of basic metals such as copper, brass, steel, nickel, and many ferrous and non-ferrous alloys, as well as platinum. Instructions for such electroplating processes may be found in a book on electroplating by Gray (16).

To prepare the electrode, a platinum gauze about 1-1/4" high and 3/8" in diameter is coated with metallic silver by plating from 10 ml of solution

containing 10 mg of silver as the nitrate in 3 M NaCN and 0.5 M NaOH. The plating process requires about 4 minutes at 3 volts and results in the deposition of about 5 mg of silver. The electrode is washed with water for 30 seconds and immersed in 10 ml of a solution 0.05 M in HI and 0.05 M in $NaHSO_3$. The current is then reversed, the silver surface used as the anode, and electrolysed at 1-1/2 volts for about 5 minutes changing the silver to silver iodide. The finished electrode is washed and can be stored in 1-1 HNO_3.

To separate radioactive silver from other activities (except halides) the "silver iodide electrode" is immersed for 5 minutes at room temperature in about 10 ml of an acid solution containing the active material. The solution may be stirred magnetically. The electrode is then removed and washed with a stream of 1-1 HNO_3 for one minute, the thorough wash being essential when high decontaminations are required. To form the Ag^{111} source, the electrode from above is again immersed in the alkaline cyanide bath and the silver iodide coat dissolved and the silver plated as metal on a source holder of platinum or other metal.

Sources prepared in this way give an adherent coat of metallic silver. Contamination by rub-off may be lessened by plating a thin coat of inactive silver over the active silver. When silver is used as the support for the silver halide surface, much poorer deposits are obtained. It is also possible to use the silver iodide electrode directly and eliminate an extra step in the procedure. The silver iodide tends to be somewhat chalky and brittle, however, and would require a heavier coating to prevent it from contaminating materials it touches than does the metal source.

These sources can be prepared in any of a number of shapes and forms from a point to a line or a cube or a gauze, or an irregularly shaped object merely by changing the shape of the cathode in the final plating step.

Yields of the radioactive silver vary with the type of electrode used in the exchange step. For the platinum gauze used in these experiments, greater than 98% yield was obtained in 10 minutes at room temperature. Increasing the temperature decreases the time required to obtain this yield. Smaller amounts of inactive silver may be used on the gauze if a source of very high specific activity is required. When submilligram amounts of silver are used, near quantitative yields are not obtained because of chalking of the silver iodide.

Concentrated solutions of palladium inhibit this exchange process only at concentrations greater than 0.5 M. Similarly, other foreign ions have an inhibiting effect at concentrations of about 2 M and above. Results of a number of experiments on yields, decontaminations and interferences using this

procedure will be reported elsewhere.

Characteristics of the Source:

The only silver isotopes present in a freshly bombarded sample of pure palladium would be the short-lived isomers of 44-sec Ag^{107} and 39-sec Ag^{109} in addition to the 7.6-day Ag^{111}. Unfortunately, "high-purity" palladium often contains some silver metal impurity [see typical analysis (8)] which would give small amounts of 270-day Ag^{110} by neutron capture. A typical impurity of 0.002% silver would give only about 0.002% as much activity of this long-lived isotope as of the 7.6-day activity in a one week's bombardment, and would be negligible. Thus the primary silver activity present in these sources will be Ag^{111}.

A source of pure Ag^{111} would weigh only 6.5×10^{-9} gm/mc, but since several mg of silver carrier are required to utilize this exchange method, the silver will not be carrier-free. The Ag^{111} emits several beta rays of about 1 Mev and a small percentage (10%) of gamma rays of about 0.3 Mev. The surface density of silver metal is about 26 $mg/cm^2/mil$. Thus, the "half-thickness" for the silver beta rays is about 2-1/4 mils of silver. (This term "half-thickness" is useful for our discussion, although it admittedly is not very exact.) Since in this procedure 10 mg or less of silver carrier is used, at least half of all the beta rays emitted will not be absorbed in most source arrangements. Hence, much less activity is lost by absorption in these silver sources than in the P^{32} plaques.

The most active source made to date in this laboratory has been several millicuries, but the same procedure is suitable for many curies with the same amount of carrier. The graph in Fig. 1 shows the amount of Ag^{111} per gram of Pd which may be obtained in a 1-week irradiation at various reactors. Reactors such as the NRX at Chalk River and the MTR in Idaho can accept up to 50 or more grams of palladium in their high flux position. Thus, multicurie sources could be made by this exchange procedure. Regardless of the activity level of the Ag^{111}, the source can still be kept in an easily handled form.

Comparison of Sources:

Characteristics of a number of kinds of beta-ray sources are listed in Table I. Sr^{90} suffers the major disadvantage of difficulty of mounting and of high personnel hazard. P^{32} has been used extensively in Bakelite plaques for many types of irradiations--especially in the biological and medical field, but is inherently bulky and prone to contamination.

Sources of Ag^{111} compare favorably with the other sources given in the table. The 7.5-day half life is some disadvantage, but the ease of preparation should offset this. In its favor is the possibility of preparing multicurie

sources in any of a multitude of shapes which best fit the material or object being irradiated. Since silver is relatively non-toxic (20) in small concentrations, it does not present the problems of Sr^{90}. The gamma ray associated with the silver beta ray is of low energy and low abundance and should have a negligible effect on the handling problems.

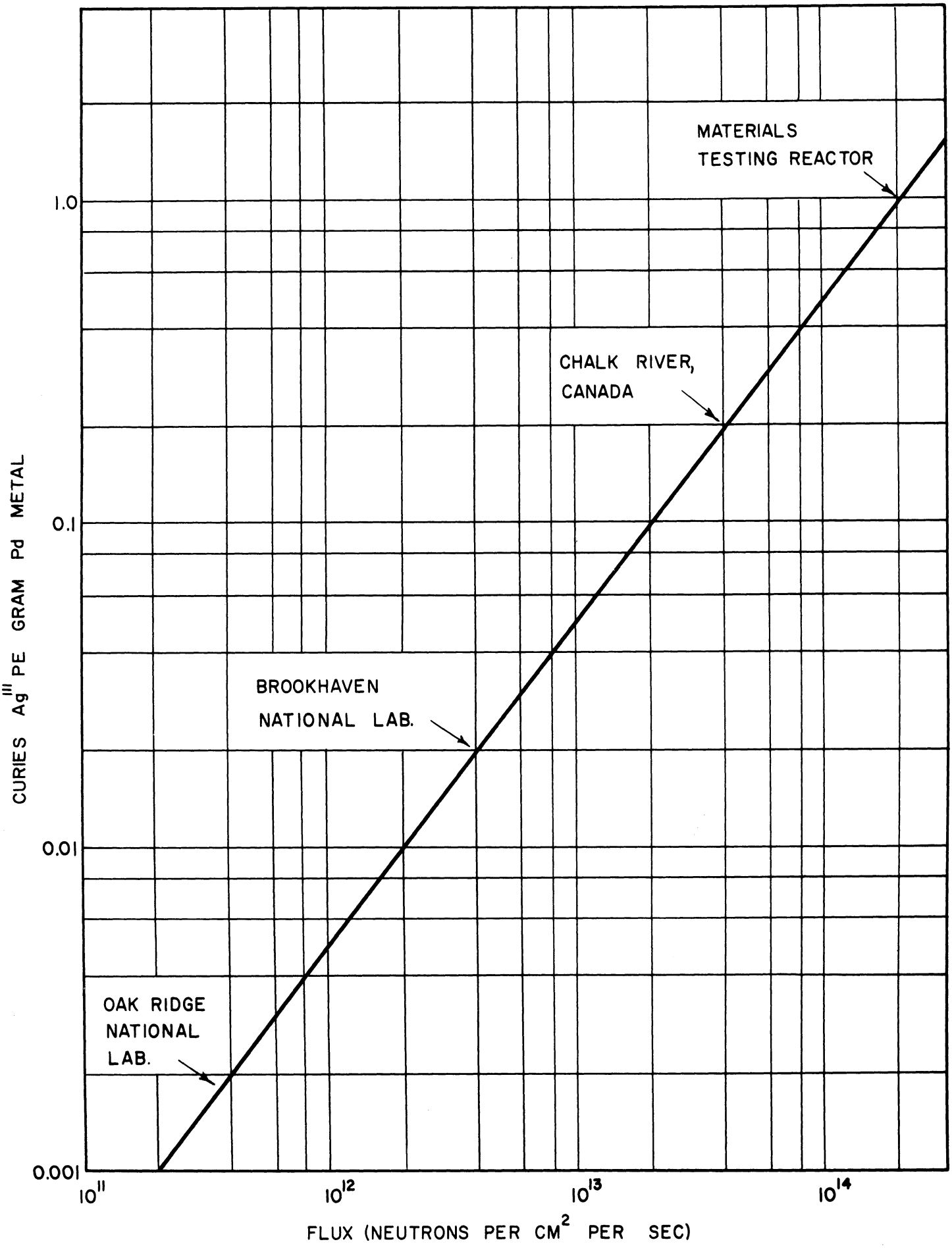
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TABLE I. Comparison of beta-ray sources.

	Half-Life (17)	Radiations (Mev)(17) β^-	γ	Form	Specific Activity used or available (curies/gm)	Personal hazard(18,19)	Combustible?	How obtained
Pd^{109}-Ag^{109m}	13.6 hr	Pd 0.961 Ag e^-	none 0.087	Metal foil	X-10 - MTR 0.2 - 100	low	no	Irradiation of Pd metal in neutron flux
Ag^{111}	7.60day	1.04(91%) 0.80 (1%) 0.70 (8%)	0.340(8%) 0.243(1%)	Ag metal electroplated on metal cathode. Thin layer of inactive Ag gives protection.	1 - 1000	low	no	Formed carrier-free from 26-min Pd^{111} in palladium irradiated with neutrons
P^{32}	14.30day	1.701	none	Red phosphorus mixed with Bakelite	X-10 - 0.0058	low	yes	Irradiation of plaque in neutron flux
Ru^{106}-Rh^{106}	1.0 yr	Ru 0.0392 Rh 3.53(68%) 3.1 (11%) 2.44(12%) 2.0 (3%) others (6%)	none 0.513(23%) 0.624(12%) others(3%)	Ru metal electroplated on Cu, Pt, Au or Ni cathode and covered with thin layer of silver for protection	~20	Intermediate	no	Fission products
Tl^{204}	4.1 yr	0.765(98%) EC (2%)	none	Thallium salt or metal	0.02	low	no	Irradiation of thallium in neutron flux
Sr^{90} - Y^{90}	19.9 yr	Sr 0.61 Y 2.18	none none	$SrCl_2$ incorporated in vials or sealed in plastic or metal with thin window	~200 (carrier free)	high	no--but easily ruptured	Fission products

Fig. 1. Ag^{111} activity attainable in one week's bombardment at different neutron flux levels.



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