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Polonium Purification

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ABSTRACT

Three processes for the purification of ^{210}Po from irradiated bismuth targets are described. Safety equipment includes shielded hotcells for the initial separation from other activation products, gloveboxes for handling the volatile and highly toxic materials, and provisions for ventilation. All chemical separations must be performed under vacuum or in inerted systems. Two of the processes require large amounts of electricity; the third requires vessels made from exotic materials.

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POLONIUM PURIFICATION

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POLONIUM PURIFICATION

I. Introduction

Polonium (Po) was discovered in 1898 by Pierre and Marie Curie (1). It was named in honor of the homeland of Marie Curie. Madam Curie and A. Derbiern processed 2 tons of uranium ore and produced 2 mg of a concentrate that contained 100 μg of Po. The Po was further purified by Marie Curie using vacuum distillation. One of the most important isotopes of Po, ^{210}Po , is in the natural decay chain of $^{238}\text{Uranium}$ (^{238}U). It is the decay product of $^{210}\text{Bismuth}$ (^{210}Bi), see Fig. 1. In the early part of this century, ^{210}Po was important as a source of alpha particles and, when mixed with Be, neutrons. Both of these particles were used as early probes in the development of our knowledge of the atom and its nucleus (2).

With the advent of nuclear reactors, the laborious process of separating ^{210}Po from natural sources became obsolete. Today, the production method of choice is neutron irradiation of purified, natural Bi, which is 100% ^{209}Bi . The production sequence is shown below (3):

The production cross section for ^{210}Po is ~ 15 mb (3). Thus, for irradiations in a thermal neutron flux of 1×10^{14} n/cm²-s the ^{210}Po activity will be ~ 110 Ci/kg of Bi at 200 days of irradiation (3).

Figure 1. Production and Decay of ^{210}Po

U 92	U^{238} 4.51×10^9 years		U^{234} 2.48×10^5 years				
Po 91	α	Po^{234} 1.17 minutes β	Po^{234} 6.75 hours β	β (99.85%) I.T. (0.15%) α			
Th 90	Th^{234} 24.1 days		Th^{230} 8.0×10^4 years				
Ac 89			α				
Ra 88			Ra^{226} 1600 years				
Fr 87			α				
Rn 86			Rn^{222} 3.824 days				
At 85			α	At^{218} ~2 seconds			
Po 84			Po^{218} 3.05 minutes	β (0.02%) α	Po^{214} 1.62×10^{-4} second	Po^{210} 138.4 days	
Bi 83			α (99.98%) β	Bi^{214} 19.8 minutes α (0.02%) β	β (99.98%) α	Bi^{210} 5.01 days β	α
Pb 82			Pb^{214} 26.8 minutes α	Pb^{210} 21 years β	α 1.3×10^{-4}	Pb^{206} (stable)	
Tl 81			Tl^{210} 1.3 minutes β	α (1.7 x $10^{-6}\%$) β	Tl^{206} 4.19 minutes β		
Hg 80				Hg^{206} 8.5 minutes β			

^{210}Po decays 100% by alpha emission, 99+% through a 5.305 MeV alpha and 0.001% through a 4.502 MeV alpha (4). There is a 803 KeV gamma-ray associated with 0.0011% of the decays (4). Due to its short half life, a gram of ^{210}Po emits 1.7×10^{14} alphas per second (4504 Ci). By combining ^{210}Po and low atomic number elements, neutrons are produced from an (α, n) reaction. Typically, ^{210}Po is alloyed with Beryllium (Be) and for every 10^6 alpha disintegrations, ~65 neutrons are generated (5). A 1 curie ^{210}Po source, therefore, could produce up to 2.4×10^6 neutrons per second.

The heat generated by 1 g of ^{210}Po is ~140 watts (6). Thus, two major uses of this isotope are neutron generators and high specific, short-lived thermoelectric batteries. Manufacturers of isotopic neutron generators have switched to americium/beryllium and plutonium/beryllium sources. However, some nuclear reactor start up sources are of the ^{210}Po /beryllium type.

II. Safety

Polonium tends to accumulate in the spleen and kidneys (7). The maximum permissible safe level for the intake of ^{210}Po is 6.8×10^{-12} g (7). Because ^{210}Po has both high specific activity and high vapor pressure, it is particularly hazardous to handle. Any work with an open source of ^{210}Po , above ~mCi amounts, should be performed in a glove-box or some other contained system.

III. ^{210}Po Recovery and Purification

The original separation of ^{210}Po from irradiated Bi must be performed in a hot cell (8) because of the radioactivity from activation of the Bi cladding material and impurities in the Bi. Typical cladding materials are aluminum and stainless steel. Aluminum would have the least activation products; however its melting point and mechanical strength are much lower than that of stainless steel. Common impurities found in bismuth that must be kept to a minimum are: arsenic (As), scandium (Sc), tellurium (Te), antimony (Sb), silver (Ag), iron (Fe) and lead (Pb). Most of these are relatively volatile, and therefore difficult to remove by distillation, a common technique used for the purification of ^{210}Po .

The distillation process requires that the irradiated Bi target be placed in a leak-tight stainless steel crucible, inerted with helium, and heated to 600-650°C. The outlet of the heated crucible must have a ^{210}Po condensation device that is enclosed in a glove box. To increase the efficiency of ^{210}Po recovery, helium is passed through the molten Bi. A properly designed and operated fractional distillation system can recover >95% of the ^{210}Po with high purity (9). The Bi target, at this point, can be cast as a target for further irradiation.

The ^{210}Po product from the distillation unit will need further chemical purification; however, this step will have separated it from the large mass of Bi and most of the gamma-ray emitting nuclides. Therefore, further purification is of a relatively small scale and can be performed inside a glove box.

There are several potential methods to perform the final purification. One of the most straight forward techniques is to dissolve the ^{210}Po in hydrochloric acid (HCl) and electrochemically redeposit the ^{210}Po onto a noble metal such as gold or platinum, or permit its spontaneous deposition on silver (10). The ^{210}Po is mostly metallic at this point. If it is desired to make high purity metal, then the ^{210}Po is redissolved in HCl and precipitated as the sulfide. This precipitate is dried and placed in a inerted chamber. By heating, the polonium sulfide decomposes to the metal and vaporizes. The metallic vapor is usually collected on a platinum foil which is kept under helium. The ^{210}Po can be dispensed as needed by evaporation from the platinum foil. The above purification process is probably the least complex, however, the energy costs to melt the bismuth targets are rather high.

Another purification scheme employs dissolution of the Bi target followed by spontaneous deposition on a much smaller amount of particulate Bi. This process is repeated several times to increase the purity and concentration of the ^{210}Po . This is the process used at Mound Laboratory in its later years (11).

The Bi targets are decontaminated and dissolved in HNO_3/HCl (aqua regia). The HNO_3 is decomposed by adding formic acid. The solution is passed through small column of granulated Bi. The ^{210}Po spontaneously plates on the Bi. The granulated Bi is cooled in order to eliminate localized boiling as the ^{210}Po is concentrated. The complete procedure is repeated, using smaller Bi columns, until the ^{210}Po is concentrated and purified to the desired level. The final purification is by electrodeposition onto gold or platinum (12). In order to make the ^{210}Po metal, the last step in the above procedure can be

used. A disadvantage of this technique is that the Bi target is dissolved and must be reduced to the metal for reirradiation.

If a new ^{210}Po chemical purification system were to be built, the pyrochemical extraction process, developed at Pacific Northwest National Laboratory, should be considered (13). This process has not been time tested. However, a pilot plant which was operated for ~6 months and processed 16,700 Ci of ^{210}Po from 222 kg of Bi. The overall extraction efficiency was >96%; and the ^{210}Po was extremely pure.

The process consists of contacting molten Bi with molten sodium hydroxide (NaOH) in the ratio of 1 part Bi to 0.044 parts NaOH. It takes five successive extraction to recover 96% of the ^{210}Po . The extraction is performed at 450°C. The melting and pyrochemical extractions are performed under inert gas. The equipment developed to perform these extractions is fabricated from Inconel 600 and is described in reference 13.

After separating the NaOH from the Bi, the NaOH, containing the ^{210}Po , is cooled and dissolved in 7 M HNO_3 . The ^{210}Po is precipitated with manganese dioxide (MnO_2) and filtered from the solution. The MnO_2 is redissolved in HNO_3 containing hydrogen peroxide. Finally, the ^{210}Po is electrodeposited from this solution onto platinum gauze. In the pilot plant tests, the ^{210}Po separation stopped at this point.

The ^{210}Po can be reduced to the metal as described above, or by another, slightly more difficult procedure, yielding higher purity ^{210}Po metal. In this procedure the ^{210}Po is vaporized at 500-800°C, with a heated, oxygen getter in the chamber to sorb the oxygen. Further down stream, the ^{210}Po is condensed on platinum. This material will be essentially free of oxygen.

Metallic ^{210}Po , usually on some supporting material (Pt), must stay either in a vacuum or in an inerted container. It is best to keep the ^{210}Po at ambient temperature to minimize potential oxidation. It would be prudent to put an oxygen getter in the storage device, if the material is going to be stored for extended periods.

IV. Summary

Three different ^{210}Po purification processes are described. All of the processes will require a shielded hot cell to perform preliminary decanning of the irradiated Bi targets, as well as the initial chemical purification. Once the ^{210}Po has been purified from the bulk of the Bi and other gamma-ray emitting radionuclides, the remainder of the purification will need various glove boxes. Both hot cells and glove boxes need HEPA filtered air exhaust systems. In addition, this exhaust system will need some type of device to sorb the ^{210}Po vapors that will be present.

All of the purification processes require that the chemical separation be performed under vacuum or in an inerted system. Two of the purification processes require large amounts of electricity for melting the Bi targets, while the third needs exotic metal vessels and fairly large amounts of HNO_3 and HCl .

Once the preliminary separation is completed, the final purification and concentration will probably consist of spontaneous deposition on to silver or electrodeposition onto platinum. If it is required, the ^{210}Po can be more completely reduced to the metal by converting it to the sulfide and decomposing this compound with heat in a vacuum or in an inert gas.

In most respects, the types of equipment that would be used for processing ^{210}Po are similar to that used in many industries. The principal difference is that this processing needs to be performed in shielded and carefully ventilated rooms.

Identification of ^{210}Po above background in a locale is not an unambiguous indicator that ^{210}Po is being manufactured. If there is ore smelting, such as recovering fertilizer from phosphate rock, or burning of coal containing uranium, there will probably be high background levels of ^{210}Po in the vicinity.

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