

EXTENDED ABSTRACT

Production of ^{90}Y at Hanford

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Ytrium-90 is a daughter of ^{90}Sr , a fission product with about a 30-year half-life, that decays through emission of a β particle to give stable ^{90}Zr . Among radionuclides that have been used to label antibodies for therapeutic purposes, ^{90}Y has received much attention owing to its favorable half-life of 64.1 hours and its predictable chemical properties. The procedure that we have used to produce multicurie quantities of ^{90}Y is similar to that developed at Oak Ridge National Laboratory.¹

Raw material purification

The abundance of ^{90}Sr at Hanford provides ample raw material for fabricating ^{90}Y generators from purified ^{90}Sr . For example, several tanks of solution containing ^{90}Sr , among other elements, were available to us at the start of this project. Analysis of the tank solution revealed the presence of several transition metals, ^{137}Cs and other alkali metals, and ^{90}Sr and other alkaline earths. The raw material was purified in the Shielded Analytical Laboratory of Pacific Northwest Laboratory (PNL) so that contaminants such as Fe and ^{137}Cs were not introduced into the processing area. This purification involved treatment of the tank solution with sodium hydroxide to precipitate most of the transition metals. Addition of sodium carbonate caused ^{90}Sr to precipitate and left ^{137}Cs in solution. The precipitate containing ^{90}Sr was collected by filtration, dissolved and analyzed to determine its purity. After the impurities of transition metals and ^{137}Cs were reduced to undetectable levels, the ^{90}Sr was considered suitable for use as a generator. It was then transferred to a specially designed room containing a hot cell and shielded glove box for processing.

Extraction of ^{90}Y

After a sufficient amount of daughter ^{90}Y has accumulated in the ^{90}Sr , the mixture can be

separated by solvent extraction. The extraction is carried out in the hot cell in order to provide adequate shielding. Thus, the ^{90}Sr - ^{90}Y salt mixture is dissolved in dilute acid. A solution of di-(2-ethylhexyl)phosphoric acid (HDEHP) in dodecane is used to extract the ^{90}Y away from the aqueous solution. The separation factor at this stage is about 10^{-5} parts ^{90}Sr per ^{90}Y . Since this value gives more ^{90}Sr than allowed for use in radiopharmaceutical preparations, the organic phase is scrubbed with dilute acid to reduce the ^{90}Sr level to an acceptable value. These scrub solutions are returned to the original generator (see below) in order to keep losses of ^{90}Sr to a minimum.

A problem that has arisen with prolonged use of a generator is a gradual decrease of ^{90}Y yield. We have investigated this problem as follows. A gradual increase of cloudiness of the dissolved generator is also seen over a period of weeks. In order to determine the cause of the cloudiness and whether it could be responsible for the decreased yields, a generator was filtered through a $0.2\ \mu$ filter. The solid remaining on the filter was examined under an electron microscope. The image produced revealed an amorphous solid. Electron microprobe analysis of this solid detected mainly Si, with traces of Na and Al. Therefore, it seems that the cloudiness is due to degradation of the glass vial in which the

generator is stored between runs. As expected, filtering the generator did not alleviate the problem.

Quite recently, we have been experimenting with the procedure for preparing the generator for the next run. This preparation involves adding the scrubs to the generator solution and boiling the combined generator and scrubs to dryness. Although we have been able to inhibit the decrease of yield, the reason for this success is unknown and the problem is still considered to be unsolved.

Isolation of ^{90}Y

The purified ^{90}Y is transferred from the hot cell to the shielded glove box so that the chances of accidentally introducing ^{90}Sr into the final product are minuscule. The ^{90}Y is stripped from the organic phase using strong acid. This strip solution then goes through a number of steps to remove traces of Fe that variably contaminate the product. The resulting solution of ^{90}Y is then evaporated to give a solid product that can be dissolved in the particular acid specified by the customer.

Analyses

The product solution of ^{90}Y is subjected to a number of tests to determine its purity and specific activity. The amounts of trace contaminants in the final product are determined by inductively coupled plasma atomic emission spectroscopy (ICP-AES). Since the specific activity of the final solution is usually quite high (>1 Ci/mL) and sample sizes greater than 5 mL are required, the final solution is diluted into a blank solution (usually the same acid used to prepare it) before the ICP-AES is recorded. Typical impurities that may be seen are Na, Ca, Fe, Zn, B, and Si. Several of these come from the glass vials used to contain the product. Since the glove box has a stainless steel liner and several pieces of equipment in the glove box are made of Fe alloys, special care must be taken to exclude Fe from the product. It was discovered that high levels of Zn can be introduced into the product if sterile 0.2 μ filters are not rinsed before use. Often, the product is of such high purity that the only element seen in the ICP-AES is Y.

The specific activity of the product is measured by proportional counting of highly diluted samples. The values obtained by this method are checked by measurements on a CRC-7 Radioisotope Calibrator (Capintec, USA) using correction factors that have been averaged over several measurements.

The amount of ^{90}Sr in the product is determined as follows. A sample of the product solution is added to a solution of Fe^{3+} and tracer ^{85}Sr . Aqueous NH_3 is added to precipitate $\text{Fe}(\text{OH})_3$, which carries much of the ^{90}Y with it. This precipitation is repeated three more times in order

to produce a solution of ^{90}Sr and ^{85}Sr almost free of ^{90}Y . The recovery of ^{90}Sr can be determined from the amount of tracer ^{85}Sr that is lost in the procedure. The solution from the last precipitation is filtered through a 0.2 μ filter onto a planchette in order to remove any precipitate particles. The planchette is then heated to remove NH_4NO_3 , which can interfere with the β counting. Calibrated absorbers can be used to determine whether the activity being counted is coming from ^{90}Y or ^{90}Sr since the energy of the emission from the former is much stronger than that from the latter.

Discussion

The previously published procedure to extract ^{90}Y from ^{90}Sr was improved and modified for use at Hanford.

Owing to the composition of our starting ^{90}Sr material, it was necessary to precipitate transition metals by adding hydroxide ion to the solution. Since copious amounts of salt would have been produced by neutralizing and then acidifying this basic solution, we chose a carbonate precipitation to isolate the ^{90}Sr . Since the first SrCO_3 precipitate contained detectable amounts of ^{137}Cs , the precipitation was repeated to give a material with no detectable ^{137}Cs . Thus, the $\text{Sr}(\text{NO}_3)_2$ precipitation that was used in the previous work¹ was deemed unnecessary.

We chose to do only one HDEHP extraction followed by purification of the ^{90}Y obtained. Scintillation counting of samples from the first organic extract revealed that the $^{90}\text{Sr}/^{90}\text{Y}$ ratio is typically in the 10^{-5} range. Several washes with dilute acid are sufficient to reduce this value in the final product to the 10^{-7} - 10^{-9} range, well within accepted limits. Thus, a second extraction from dilute acid, a second strip with strong acid and a second evaporation are not needed. This is a very important feature since each evaporation concentrates any impurities originating in the reagents used.

The problem of decreasing yields from the generator have previously been attributed to traces of soluble organic phosphates.¹ We have been reluctant to use aqua regia to destroy these putative contaminants in our stainless-steel-lined hot cell since this would probably create a problem with Fe contamination. Thus, we have been experimenting with various methods of preparing the generator for the next extraction. Boiling to dryness with ~ 7 M HNO_3 has apparently solved this problem for the present.

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Reference

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