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Fabrication of ^{58}Co positron sources

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Abstract

A technique for producing ^{58}Co positron sources for use in slow positron beams has been developed. The method has been successfully tested at the μCi and mCi level. Scaling up of the technique is under way to consistently produce 1–2 Ci sources from 60 g of irradiated nickel.

1. Introduction

In the last 15 years, physics and materials research with slow positron beams have become quite active [1]. The need for more intense sources of positrons has concomitantly grown with experimental requirements for higher positron beam currents and beam brightness. For typical laboratory-scale beams the beta-decaying isotopes of ^{22}Na (0.1 Ci, $T_{1/2} = 2.6$ yr, $f_{\beta^+} = 0.90$) and ^{58}Co (0.5 Ci, $T_{1/2} = 71$ d, $f_{\beta^+} = 0.15$) are the most common commercially available sources. The high cost, inconsistent quality, and the need for even more intense sources has prompted consideration of alternative isotopes ^{64}Cu ($T_{1/2} = 12.7$ h) [2–4], ^{126}I ($T_{1/2} = 13.02$ d) [5], ^{48}V ($T_{1/2} = 16$ d) [6], ^{68}Ga ($T_{1/2} = 67.6$ min) [7], ^{79}Kr ($T_{1/2} = 35$ hr) [8,9], and further study of ^{22}Na [10], and ^{58}Co [11]. With the ability to cheaply produce ^{58}Co sources of 10 Ci or more at the University of Michigan Ford Nuclear Reactor (FNR), ^{58}Co becomes the most attractive isotope for source fabrication. The potential for producing very intense sources (over 10^3 Ci) of ^{58}Co at other reactors [12,13], including the proposed Advanced Neutron Source [14], lends motivation as well.

Using the general procedure for producing ^{58}Co sources described by Sherief and Grass [15], an investigation into producing sources at FNR has been conducted. The goal of this investigation is to develop a technique to produce thin metal electrodeposits of ^{58}Co on copper to use as a source. It was decided that the deposits should be no thicker than one exponential attenuation range of positrons in cobalt, 22 mg/cm^2, [16] so as to attenuate no more than 20% of the positrons emitted from the source. The procedure that suits our available facilities consists of four steps. First, nickel is irradiated in FNR. The nickel is then electrodissolved in HCl acid. Next, anion exchange chromatography is used to extract the cobalt. The purified cobalt is finally electrodeposited and encapsulated for use as a source. These four steps are discussed in the next section.

2. Experimental technique

2.1. Irradiation

Irradiation of a nickel target with fast neutrons produces ^{58}Co via the ^{58}Ni(n, p)^{58}Co reaction. Since natural nickel is 68% ^{58}Ni it is not necessary to use an isotopically pure target. However, if the reactor flux has thermal or epi-thermal neutrons, ^{58}Co is converted into ^{59}Co via the ^{58}Co(n, γ)^{59}Co reaction. This is the so called "burn-up" of ^{58}Co. The activity, A, of ^{58}Co produced per gram of natural nickel is given by the equation:

$$A = A'_{\mathrm{sat}}\left[1 - \exp(-\lambda_{\mathrm{tot}} t)\right], \tag{1}$$

where $\lambda_{\mathrm{tot}} = \lambda_{\mathrm{n}} + \lambda_{\mathrm{b}}$ and $A'_{\mathrm{sat}} = A_{\mathrm{sat}}\,(\lambda_{\mathrm{n}}/\lambda_{\mathrm{tot}})$, λ_{n} is the natural decay rate and λ_{b} is the reactor flux-dependent burn-up decay rate. A_{sat} is the saturation activity without burn-up and is given by:

$$A_{\mathrm{sat}} = n\sigma\Phi, \tag{2}$$

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where n is the number of ^{58}Ni atoms per gram of natural nickel, σ is the ^{58}Ni(n, p)^{58}Co cross section and Φ is the fast neutron flux.

As can be seen from Eq. (1) after some long irradiation time, t, compared to $1/\lambda_{tot}$ the cobalt activity saturates. At this point the production of ^{58}Co is balanced by its disappearance through natural beta decay and burn-up. The burn-up process continues to produce ^{59}Co which decreases the specific activity of ^{58}Co. Thus the irradiation time should be optimized, typically several times $1/\lambda_{tot}$. It may also be possible to selectively reduce burn-up by cladding the nickel sample in a thermal neutron absorber, e.g. cadmium or boron. We have not pursued cladding since an earlier study [10] suggested that there was little improvement to be obtained in our particular reactor.

In order to determine the parameters in Eq. (1) and thus the requisite irradiation time, a study of ^{58}Co production in the FNR was conducted using two separate techniques. The first method involves the irradiation of a nickel wire, a cobalt–aluminum neutron flux wire, and an iron wire for eight hours in FNR. An estimate of the thermal, epi-thermal, and fast neutron fluxes can then be found from:

$$^{58}\mathrm{Ni(n, p)}^{58}\mathrm{Co}, \tag{3}$$

$$^{58}\mathrm{Fe(n, \gamma)}^{59}\mathrm{Fe}, \tag{4}$$

$$^{59}\mathrm{Co(n, \gamma)}^{60}\mathrm{Co}, \tag{5}$$

where the fast neutron flux can be found from reaction (3) and the thermal and epi-thermal fluxes can be found from reactions (4) and (5). Using these fluxes, a prediction of the saturation activity, A'_{sat}, and the burn-up rate, λ_b, in Eq. (1) can be made. At our current site, we calculate $A'_{sat} = 47 \pm 12$ mCi/g and $\lambda_{tot} = 6.7 \pm 1.6 \times 10^{-4}$ h^{-1} (and therefore, $\lambda_b = 2.6 \pm 1.6 \times 10^{-4}$ h^{-1}). The contribution to λ_b from the meta-stable state of cobalt, ^{58m}Co ($T_{1/2} = 9.2$ h), was considered and found to be negligible at the 3% level. This method can be used to give an estimate of production rates in any site after an irradiation of about 24 hours. The ability to predict ^{58}Co production is especially useful if the reactor core has been re-configured or a new site is used.

In the second technique for measuring ^{58}Co production a series of equal mass nickel wires are irradiated for increasing periods of time. After cooling for several days, the ^{58}Co activity is measured by Ge γ-ray spectroscopy and plotted as a function of irradiation time (Fig. 1). The data are then fitted to Eq. (1). With this direct technique we determine $A'_{sat} = 34 \pm 5$ mCi/g and $\lambda_{tot} = 9.1 \pm 1.1 \times 10^{-4}$ h^{-1} (and therefore, $\lambda_b = 5.1 \pm 1.1 \times 10^{-4}$ h^{-1}). The agreement between the results is reasonable, given the uncertainties involved with each technique. With an exponential production time ($t = 1/\lambda_{tot}$) of approximately 1100 hours (45 days) we decided to use 60 to 70 days as an irradiation time. With this irradiation time, it is necessary to use 60 g of nickel in order to produce 1.5 Ci of activity.

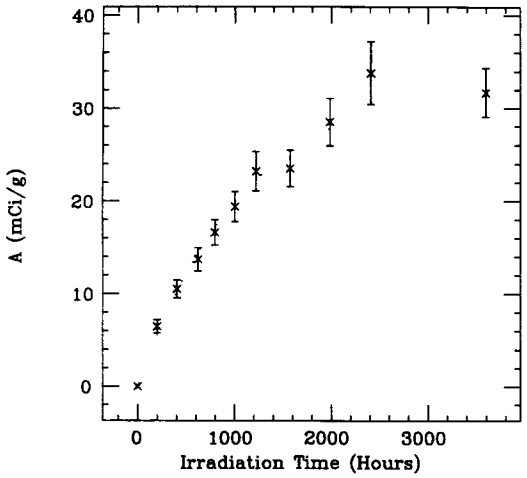

Fig. 1. The production curve of ^{58}Co is found by irradiating a series of equal mass nickel wires in FNR for increasing periods of time and plotting the measured ^{58}Co activity vs. the irradiation time. The irradiation time is defined as reactor full-power hours corresponding to about 10 days out of every 14 day reactor cycle. The parameters $\lambda_{tot} = 9.1 \pm 1.1 \times 10^{-4}$ h^{-1} and $A'_{sat} = 34 \pm 5$ mCi/g are found by fitting to Eq. (1). The burn-up is found to be $\lambda_b = 5.1 \pm 1.1 \times 10^{-4}$ h^{-1}.

After the nickel is irradiated, it is transferred to the electrodissolution apparatus.

2.2. Dissolution

Use of anion exchange chromatography requires that the nickel and cobalt be in solution and that the cobalt, which is naturally cationic, be in an anionic form. HCl acid may be used to dissolve the nickel as well as make the cobalt anionic by a complexing reaction (e.g.):

$$\mathrm{Co^{2+} + 4Cl^- \Leftrightarrow (CoCl_4)^{2-}}. \tag{6}$$

Common HCl dissolution techniques include the use of room temperature HCl acid, heated HCl acid [10], and a heated mixture of HCl and HNO_3 acids [15]. However, on larger ($\gtrsim 30$ g) samples of solid nickel the first method requires too much time, and the latter methods, while faster, outgas considerable amounts of corrosive acid vapor which is destructive to a hot cell environment. The last method also leaves a HNO_3 residue which damages the anion exchange resin. To accommodate these restrictions an alternate technique using electrodissolution was pursued.

Electrodissolution of the nickel is accomplished by using a graphite electrolytic cell immersed in acid. The irradiated nickel plate is put into the cup cathode of the cell which is parallel to a plate anode. The nickel and cell are then placed into 1800 ml of concentrated (12 M) HCl acid. A current of a few amperes is run through the cell. To prevent a reduction in the dissolution rate due to $NiCl_2$

precipitation on the nickel surface, the acid is vigorously agitated. Heat generated during processing will cause some HCl to outgas reducing the molarity of the acid below 12M. We found that at about 6M cobalt began to deposit onto the anode of the cell. In order to prevent cobalt deposition, the acid bath is cooled externally, preventing the acid from falling below 7M. Dissolution rates for this technique are 0.5–0.75 g/h and there is no appreciable cobalt deposition on the anode.

After 3–4 days the 60 g nickel sample is dissolved and the resulting solution is prepared for separation by adjusting the HCl molarity to 9M. At this point the volume of the 9M solution is typically about 2.5 l.

2.3. Separation

Standard anion exchange chromatography is used to extract the cobalt from the nickel solution. Two columns of Dowex 1X-8 resin, with volumes of 147 ml and 19.2 ml, are used consecutively to insure a high level of purity of the eluted cobalt. The resin will pass ions and retain anions. Because most metals complex at different molarity of HCl [17], material can be selectively "stripped" from the column by washing it with decreasing molarity of acid.

The first (large) column is prepared by conditioning it with 9M HCl. The nickel solution is then pumped through the column where the ionic nickel will pass through while the anionic cobalt complex will adhere to the resin. Cobalt is isolated from other impurities present at the same or higher level by stripping. First, manganese and magnesium are removed with 6M HCl. The cobalt is then removed with 3M acid. The column is finally washed with water to remove any remaining material. This procedure is then repeated with the second, smaller, column using the cobalt wash. All of the solutions are pumped through the columns at 7 ml/min. The cobalt is removed in 60–80 ml from the first column and 30–40 ml from the second column. Starting with about 3 ppm (by weight) cobalt in the 60 g nickel plate, the net efficiency of the two-column extraction of cobalt is $\geq 95\%$. When complete, the elution from the second column is transferred to the deposition system.

2.4. Electrodeposition

The final step of the procedure is to electrodeposit the separated cobalt in solution onto a cathode. The deposition cell consists of a pyrex test tube, a helical platinum anode and a cathode which is a 3 mm diameter rod. The cathode is prepared by polishing the end and coating the sides with a commercial masking paint to restrict deposition to the end. Although the cathode could be much smaller (< 1 mm diameter) we chose 3 mm to facilitate electrodeposition and because that is the diameter of our present ^{22}Na sources. Several materials were tested for use as a cathode and copper was found to produce the most adherent plates.

Previously, an electrolyte was combined with the eluted solution from the column in the cell and plating begun [10]. However, it was found that low concentrations of cobalt in the solution yielded black, non-adherent plates. For a 1–2 Ci source there is approximately 0.05 mg of ^{58}Co and 0.1–0.2 mg of inactive cobalt in the solution. To increase the cobalt concentration 0.5 mg of inactive cobalt in solution is added to the elution of the second column. The total mass of cobalt, 0.7 to 0.8 mg, is still a factor of three below the one exponential attenuation range of ^{58}Co positrons. The resulting solution is then taken to dryness. The cobalt is then picked up with 0.5 ml of electrolyte, which is described in Ref. [15]. The cobalt concentration of the solution is now approximately 1.5 mg/ml. The plating cell is run at 1–2 V and currents of a few mA. In order to prevent the formation of bubbles on the cathode surface, which hinders deposition, the solution is agitated with a magnetic stirrer. It was found that about 90% of the material plates out in 24 h.

After the deposition is complete, the cathode is rinsed and then soaked in acetone to remove the masking paint. The surface of the cathode is wiped to remove any loose activity. The cathode is then encapsulated into a stainless steel holder which has a 3 mg/cm^2 nickel window directly over the source deposit. This is strictly a precautionary measure to seal the source and should attenuate less than 15% of the positrons.

3. Results

This procedure has been used to make many μCi and several mCi level sources. The activity was closely monitored after each step in order to determine the efficiency of the process. A reproducible efficiency of 85–95% for producing the μCi sources has been achieved. The plates were consistently shiny and adhesive. No activity at the 1% level was lost when the surface of these sources were wiped. The production of the mCi level sources was conducted so as to mimic the production of a 1–2 Ci source in all aspects except total activity. This was accomplished by irradiating a 60 g piece of nickel for only 15 h and then processing as per our prescription. The mCi sources had net efficiencies starting at 50% and increasing to 60%. The difference in efficiencies between the μCi and mCi levels was mainly caused by poor control of impurities, with most of the impurities introduced after the chromatography step via improperly deionized water used in processing. These efficiencies with mCi sources should improve to that of the μCi sources. A 1.5 mCi source was encapsulated and tested in a well-characterized positron beam. Using a commercial ^{22}Na source for calibration, a rate three times greater than expected was found. The conclusion is that the ^{58}Co source was successfully thin while the ^{22}Na source was thick enough to self-absorb $\frac{2}{3}$ of its positrons.

4. Conclusions

These tests, with an immediate goal of routinely producing 1–2 Ci ^{58}Co sources, have become consistent at the μCi level, and are improving at the mCi level. Development of an apparatus that can duplicate this procedure in the remote operated, shielded environment of a hot-cell is underway. This apparatus should be able to regularly produce reliable high level sources. While a 1–2 Ci source is useful for many applications, some experiments require a higher level of activity. Allowing for a maximum source thickness of one exponential attenuation range for ^{58}Co positrons, we could theoretically produce a 60 Ci source of pure ^{58}Co on a 3 mm diameter cathode. However, given limitations in removing processing impurities, burn-up of ^{58}Co into ^{59}Co, and radiation safety considerations our longer term goal is to irradiate 10 Ci of ^{58}Co in FNR and deposit it on a 5 mm diameter active area using this technique.

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