The process of Ni-63 production and measurement of beta radiation of irradiated Ni-63

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1. Introduction

A betavoltaic battery is a device that converts the decay energy of beta-emitting radioisotopes into electric power. It has the following characteristics: long service lifetime, high energy density, easy small-scale fabrication, and minimum maintenance. For this reasons, betavoltaic batteries are widely used in many low-power applications such as medical applications and power sources for micro electro-mechanical system (MEMS) because they can operate effectively at extreme environments [1,2,3].

Proper selection of radioisotope is a critical factor in the design of a betavoltaic battery. The important factors affecting the performance of a betavoltaic battery are specific activity, half-life, and toxicity [4]. The Ni-63 is a pure beta emitter with a low energy spectrum of $E_{avg} = 17.4$ keV and it has significantly long half-life of 100.1 years. Therefore, it has been widely used as the power source of betavoltaic batteries. The beta spectrum of Ni-63 is below the radiation damage threshold (approximately 200 keV for Si) of semiconductors such as Si and SiC. In addition, handling of Ni-63 is easier than other beta-particles, such as H-3, Sr-90, and Pm-147 because of its low energy spectrum [5]. For this reason, Ni-63 is popular as the power source of a betavoltaic battery, whose capacity are at the range of $10^{-6} \sim 10^{-9}$ watt.

In this study, a target for irradiation was produced using 99% Ni-62 metal power concentrate. Ni-62 target of 1 g was irradiated in MARIA reactor operated in Poland for 470 hours, and estimated production of Ni-63 was calculated. Irradiated Ni-63 pellets were dissolved in HCl solution, and its beta radiation was measured by Liquid Scintillation Counter (LSC).

2. Methods and Results

2.1 Production of Ni-62 pellets



Fig. 1. The pellets of metal Ni-62.

99% Ni-62 metal powder concentrate was purchased from Isoplex in USA, and it was formed with pellets to insert into irradiation capsule. A weight of one pellet is 0.1 g, so a total of ten pellets were formed. Pellets were sealed in quartz tube as shown in Fig. 1.

2.2 Target irradiation and calculation of estimated production of Ni-63

Ni-62 pellets were irradiated in MARIA reactor operated in Poland. MARIA reactor is pool type reactor whose thermal power is 30 MWth, same as HANARO research reactor operated in KAERI. Its average neutron flux is 2.5×10^{14} n/cm²s. The neutron flux of a irradiation hole containing the Ni-62 target is 1.2×10^{14} n/cm²s, and target was irradiated for one cycle (20 days, 470 hours). The estimated production of Ni-63 irradiated from MARIA reactor was calculated as shown below:

$$N\Phi\sigma t_r(1-e^{-\lambda t}) \approx \lambda N\Phi\sigma t_r$$

where λ is the decay constant of Ni-63 (2.19 \times 10⁻¹⁰/s), N is the atomic number of Ni-63 (9.74 \times 10²¹), Φ is the neutron flux of MARIA reactor (2.5 \times 10¹⁴n/cm²s), σ is the neutron absorption cross section (14.22b), and t_r is the time of irradiation (2.42 \times 10⁶s). Therefore, estimated production of Ni-63 is calculated about 6.172 \times 10⁹ Bq (166 mCi).

2.3 Dissolution of Ni-63 and fabrication of Ni-63 foil



Fig. 2. Dissolution process of irradiated Ni-63 pellets.

Ionic solution including Ni-63 is prepared for an electroplating process. Ni metal powders of 1 g with Ni-63 were dissolved in a mixture of 18 ml HCl and 2 ml distilled water for 2 hours at 80 °C. Boric acid (H₃BO₃) of 2.1 g and saccharin of 0.4 g were added in DI water, and mixed with Ni-63 dissolved solution. 10 ml DI water and 1.5 g KOH were added, and the pH value of the plating buffer was adjusted to 2.5 with the addition of KOH. 99.99% purity of Ni sheet with dimensions of 1×1 cm² was used as the substrate. Ni-63 coatings were deposited by DC electroplating at current density of 20 mA/cm². The thickness of the Ni-63 deposition is about 4 µm as shown in Fig. 3.



Fig. 3. (a) Glove box in Bank-2 in RIPF, and (b) electroplated Ni-63 onto 99% Ni sheet at current density of 20 mA/cm².

2.4 Measurement of beta radiation using LSC

	Measurement time (min)	СРМ	DPM
Sample 1	30	12767	15736
	30	12734	15685
	30	12736	15726
Sample 2	30	13324	16497
	30	13772	17003
	30	13740	16947





Fig. 4. The quench curve of Ni-63 standard source

The beta radiation of dissolved Ni-63 is measured by Liquid Scintillation Counter (LSC, Tri-Carb 2910 TR, Perkin Elmer). Ni-63 standard source was purchased from Eckert & Ziegler Isotope Products, and its specific activity is 20.12 μ Ci/ml. Ultima gold was used as LSC cocktail. First, dissolved Ni-63 was diluted with DI water, and Ni-63 diluted solution and LSC cocktail were mixed. The CPM and DPM values of Ni-63 are shown in table 1. An average DPM values of samples are 15737 and 16975 respectively. It means that Ni-63 of 120.46 to 130.9 mCi was produced in MARIA reactor.

3. Conclusions

A target for irradiation was produced using 99% Ni-62 metal power concentrate. Ni-62 target of 1 g was irradiated in MARIA reactor operated in Poland for 470 hours at neutron flux of 2.5×10^{14} n/cm²s, and estimated production of Ni-63 was calculated. Irradiated Ni-63 pellets were dissolved in HCl solution, and Ni-63 coatings were deposited by DC electroplating at current density of 20 mA/cm². Its beta radiation was measured by Liquid Scintillation Counter (LSC). In conclusion, about 120 mCi Ni-63 was produced per 1 g of Ni-62 in MARIA reactor.

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