Preparation of Ni-63 source for the fabrication of betavoltaic battery

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1. Introduction

Ni-63 is pure beta-emitter with a low energy spectrum and significantly long half-life of 100.1 years and thus is widely used as the power source of betavoltaic batteries. The beta spectrum of Ni-63 is below the radiation damage threshold of semiconductor, for this reason, it is suitable for the power source of a betavoltaic battery to be within the nano- to microwatt range [1-2].

A beta radiation source, Ni-63, is prepared by electrical deposition of radioactive Ni-63 ions on a thin non-radioactive nickel foil. Ni-63 plating is similar to other electroplating processes that employ soluble metal anodes. Specifically, it requires the passage of direct current (DC) between two electrodes that are immersed in a conductive, aqueous solution of nickel salts. The flow of a DC causes one of the electrodes to dissolve and the other electrode to become covered with nickel [3]. To establish the coating conditions for Ni-63, Niplating solution is prepared by dissolving metal particles, and the deposition conditions have been optimized by studying the influence of the current density.

Also, Ni-63 is prepared by the electrophoretic deposition (EPD) is widely used for shaping, and coating for preparing uniform ceramic films with various thickness and assembling charged particles on electrodes with various morphologies [4]. Since the discovery of repairing pit by using EPD method, well dispersed metal nanoparticles in suspension are applied to self-repairing pit on the metal surface [5]. Electrophoretic deposition consists of two processes; the movement of the charged powder particles in suspension under the applied electric field between the working electrode and counter electrode, and then, electrophoresis by deposition of particles on the working electrode. These proposed models can also be applied for radioactive Ni-63 electroplating.

2. Methods and Results

2.1 Ni electroplating

Ni coatings were deposited by DC electroplating at current densities of 1, 5, 10, and 20 mA/cm². The basic composition of the bath was 0.2 M Ni and 25 g/l of boric acid (H₃BO₃). Ni metal powders were dissolved in a mixture of HCl and distilled water. The pH of the bath was adjusted to 4.0 ± 0.2). A nickel sheet of 99.99 % purity with dimensions of $10 \times 20 \times 0.125$ mm³ was used as a cathode and a Pt-coated Ti mesh with dimensions of $25 \times 135 \times 1$ mm³ as an anode. A Ni

sheet with a high purity of 99.99 % (Aldrich) was used as the substrate. The deposition time was adjusted to achieve an average thickness of 3 µm based on Faraday's law [6]. The microstructure of the coatings was studied by scanning electron microscopy (SEM) and X-ray diffraction (XRD). XRD investigations were carried out using a Philips X'Pert-Pro instrument operated at 40 kV and 30 mA with CuK α radiation ($\lambda =$ 1.5418 Å). The average particle size of the nanocrystalline nickel coatings was calculated from XRD patterns using the modified Scherrer relationship expressed in [7].

Figures 1 shows the results of SEM for Ni-coated Ni sheets at current densities of 1, 5, 10, and 20 mA/cm². The average particle size decreased with an increasing current density up to 10 mA/cm². The shape of the particles changed from needle-like to spherical as the current density was increased.



Fig. 1. SEM images for the Ni coated Ni sheet at a current density of (a) 1, (b) 5, (c) 10, and (d) 20 mA/cm². The basic composition of the bath consists of Ni (0.2 M), and H_3BO_3 .

Figure 1(a-c) shows SEM images observed for electrodeposited Ni on Ni sheets at current densities of 1, 5, and 10 mA/cm², respectively. The minimum particle size was observed at a current density of 10 mA/cm². The particle size became larger as the current density was increased beyond 15 mA/cm². Figure 1(d) presents SEM images for electrodeposited Ni on Ni sheets at current densities of 15 mA/cm². The particles on the Ni sheet had a spherical shape, as shown in Figure 1(d). The electroplating of Ni was not possible at and above a current density of 30 mA/cm² because salts in the solution were also coated on the Ni substrate.

2.2 Ni electrophoretic deposition

Ni nanoparticles were synthesized by a pulsed wire evaporation (PWE) method [7]. It was observed that the average particle size of the Ni nanoparticles about 70 nm. Hypermer KD-2 (HKD-2, Uniqema UK) was used as a dispersant. To prepare the Ni-dispersed solution, HKD-2 ($1 \sim 13$ wt%) was first dissolved into the ethanol, and then Ni nanoparticles added into a dispersant containing ethanol solution. The zeta potential of Ni nanoparticles in surfactant dispersed ethanol was measured from -32 to -17 mV in values by using a Nano-ZS zeta sizer (Malvern, UK).

A two-electrode electrochemical cell was employed for the electrophoretic deposition. The scratched specimens of Ni plate and a Pt coated mesh were used as a working electrode and counter electrode, respectively. The Ni-dispersed solution was used as the electrolyte. The area of the specimen exposed to the electrolyte amounted to 0.25 cm². Constant electric fields of 180 V cm⁻¹ were applied to the specimens using a DAP-2002R dc power supply, and the electrophoretic current value during electrophoretic deposition was measured using a multimeter.



Fig. 2. SEM images of Ni particles deposited on the sulface of a scratched plate subjected to a constant electric field, E, of 180 V cm⁻¹.

SEM images of Ni particles deposited on the surface of a scratched plate subjected to a constant electric field, E, of 180 V cm⁻¹ for 0, 300, and 600 s are shown in Figure 2. Figure 2(a-b) shows a time-dependent EPD sequence for 0.005 wt% Ni. As EPD time was increased, the size of the pits decreased substantially because of agglomeration of Ni nanoparticles deposited at the scratched plate. Figures 2(c-d) show the amount of Ni nanoparticles had no effect on the time-dependent EPD sequence. The SEM images show that as deposition time increased from 5 to 10 min aggregation of the particles became more remarkable, and cracks and holes appeared. The results of this study show that the EPD can be used to manufacture Ni films directly from Ni particles without melting. To obtain the optimum surface and deposition thickness, the effect of voltage, deposition time, and sintering after EPD should be optimized.

3. Conclusions

Nanocrystalline Ni coatings were synthesized by DC electrodeposition at a current density from 1 to 20 mA/cm² and pH 4. The results showed that the average grain size decreased to 25 nm as the saccharin concentration increased to 2 g/l, and increasing the current density had a considerable effect on the average particle size of the deposits.

In order to deposit Ni nanoparticles, the EPD was executed by using two electrode cells in the suspension which was produced in organic solvents using HKD-2 as a dispersant. The coating of Ni particles on substrate of Ni plate was fabricated in suspension at 7 wt% HKD-2 or more. It was found that the EPD process was important not only zeta potential of the suspension but also conductivity.

Acknowledgement

This work is supported by the National Strategic R&D Program for Industrial Technology(10043868), funded by the Ministry of Trade, Industry and Energy(MOTIE).

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