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Preparation of the electroplated Ni and Co films for applying betavoltaic battery and Mössbauer source

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1. Introduction

Ni-63 and Co-57 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 aconductive, aqueous solution of nickel salts. The flow of a DC causes one of the electrodes (the anode) to dissolve and the other electrode (the cathode) to become covered with nickel. The nickel in the solution is present in the form of divalent positively charged ions (Ni^{2+}, and Co^{2+}). When the current flows, the positive ions react with two electrons (2e-) and are converted into metallic nickel ($Ni^0$and Co^0) at the cathode surface. The reverse occurs at the anode where metallic nickel is dissolved to form divalent positively charged ions that enter the solution. The nickel ions discharged at the cathode are replenished by those formed at the anode [1-2]. In this study, a Co-and Ni- plating solution is prepared using two different baths. One is the acid-based buffer (pH 3-4) containing boric acid, sodium chloride, and saccharin. The rest is the base-based buffer (pH 10) containing hydrazine hydrate and ammonium citrate. The optimization of the electroplating parameters for the deposition of Co on plate was considered as indispensable. A betavoltaic battery was fabricated using Ni-63 attached on a P-N junction semiconductor, and the I-V characteristics were measured using a probe station. The thickness-dependent self-shielding effect of the radioisotope layer was investigated [3] Also, the aim of this work was determination of the optimal parameters for thermal diffusion of the electroplated Co into the Rh structure.

2. Experimental Technique

The composition of alkaline electrolyte was ammonium citrate (25g/l), hydrazine hydrate (25g/l), and ammonia solution (25%) for adjusting the pH to 10. The acidic electrolyte contained 0.1 M HCl and 0.1M NaCl in such proportions so as to assure the pH 3. The volumes of the electrolytes were 10 ml. Charged Co, and Ni ions are formed by chloride. The concentration of Co in both the alkaline electrolyte and the acidic bath used in these experiments were 10^{-2} mg/ml. A platinum foil was used as the anode. The deposited Co diffused almost completely into a rhodium matrix without substantial loss at an annealing temperature from 1,100 to 1,200℃ for various times under the vacuum or inert gas of Ar. The deposition yield was measured by the concentration of Co and Ni in the bath before and after deposition using inductively coupled plasma-atomic emission spectroscopy (ICP-AES). Scanning electron microscope (SEM) was investigated to identify the Co diffused Rh layer. The IV characteristics of a Ni-coated semiconductor were investigated using a Probe Station of the Precision Source/Measure Unit, B2911A.

3. Results and discussion

, the relationship between the current density and plating time was determined in order to realize equal electric quantities for each condition. The rate of deposition for Co-57 on Rh foil with thickness of 4.5 μm as a function of pH was measured. 1. Definitely, lower values have been obtained for a chloride electrolyte (pH 3) than for a citrate electrolyte (pH 10). The rate of deposition shows almost a linear function of deposition time at and below 4h. So, the deposition time below 4 h was adjusted to achieve an average thickness based on faraday's low for the electroplating of Co. If the deposition time is increased, the rate of deposition is saturated [4]. In this investigation, the current density for electroplating Ni prepared by dissolving metal powder in an acidic solution was lower than that used to prepare Ni by dissolving Ni-chloride of a commercial product. However, other conditions for electroplating, such as pH, the concentration of grain refiner, and the deposition time, were similar to those used in conjunction with the commercial plating solution. The prototype for electroplating radioactive Ni-63 has been established. The electroplating was carried out by two-step processes such as preparation of ionic solution including Ni-63, and coating processes on the substrate. To evaluate the P-N junction prepared by ETRI, the electron beam induced current technique has been employed to experimentally simulate the beta emission of ^{63}Ni and to estimate the total device current [3]. From the e-beam illumination test, we confirmed the good operation of PN absorber. A Ni-plating solution is prepared by dissolving metal particles, and the deposition conditions have been optimized by studying the influence of the current density in a previous study. In addition, we prepared electroplated ^{63}Ni with the specific activity of 0.45 mCi for the characteristics of a nuclear battery. The proposed prototype for the synthesis can be applied to the electroplating radioactive ^{63}Ni. An accurate measurement for the specific radioactivity electroplated ^{63}Ni will be carried out in a future study. The range of the beta particles and the location of their deposited energy are deep within the silicon substrate. We attached prepared beta source on the P-N junction using vacuum. The I-V curves of both dark and deposited ^{63}Ni show almost the same values. The difference between the pre-deposition (dark) and deposited Ni-63 can be obtained through a magnification of the I-V curve. The difference of the short circuit current between the pre-deposition and post deposition of Ni-63 on foil with a thickness of about 3 μm was found to be 5.03 nA. This value of a single cell operated at the nominal specific radioactivity, 0.45 mCi was approximately same comparing with previous measured a single cell with seed layer on the P-N junction at 2.5 mCi. XRD for the deposited Co on a Rh foil under the plating condition of pH at 10 and 3. XRD patterns were observed showing that the crystal structure of the Co deposits is pure hcp (hexagonal close packing). For initial annealing attempts, Co electroplated Rh foil with thickness of 4.5 m were used, and a relatively low vacuum 10-2 hPa under inert Ar gas atmosphere was maintained during the diffusion process. Also, the annealing process was performed at higher vacuum 10^{-5} hPa in quartz and ceramic tubes at different temperatures. Fig. 1 shows SEM images for surface morphology and cross-section for Co electrodeposited Rh foil. The Co particles on the Rh plate had a spherical shape. The particles are diffused into Rh matrix after annealing. The mapping images of elemental energy dispersive spectrum (EDS) for the cross section of Co coated Rh foil. The Co particles are observed on the surface of Rh plate, only.

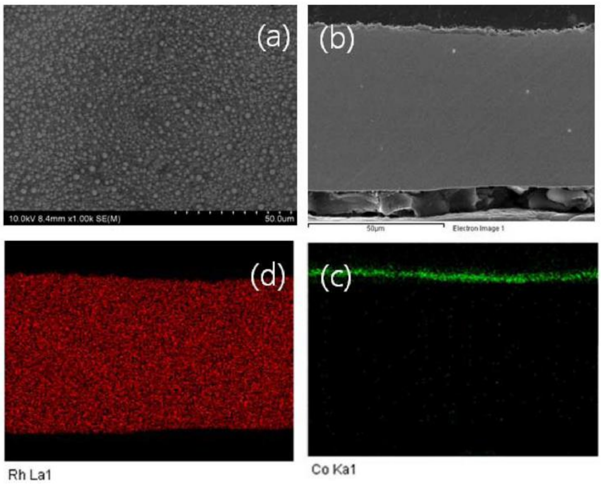

Fig. 1 SEM images for (a) surface of Co layer on Rh foil, (b) cross section images for Co coated Rh foil. Mapping of EDS for (c) Co layer, and (d) Rh layer in SEM image of (b).

We are able to establish the prototype for electroplating radioactive 57Co and diffusion condition. For Mössbauer investigations the electroplated and annealed foil is mounted in to Ti capsule sealed with epoxy or brazed Be window [4]. All Mössbauer measurements were carried out on a pure iron foil as absorber. The proposed condition in this study will be applied to prepare radioactive career free 57Co/Rh Mössbauer source at near future.

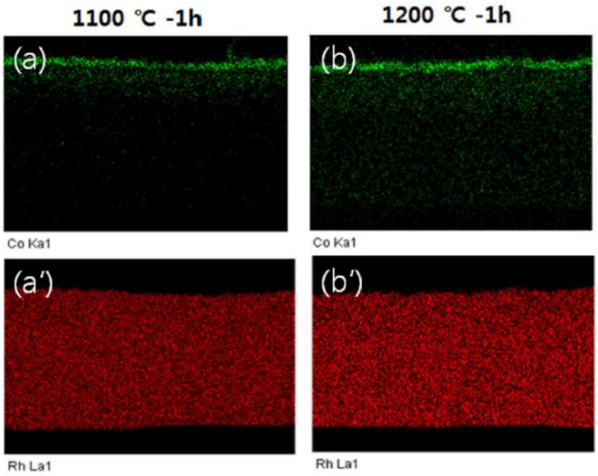

Fig. 2 Mapping of EDS for Co layer annealed at (a) 1100 ℃, and 1200 ℃ (b), and Rh layer annealed at (a') 1100 ℃, and (b') 1200 ℃ for 1h.

4. Conclusions

To establish preparation of betavoltaic battery, and Mössbauer source, natural Ni and Co were electroplated on the Rh plate. Both the acid-based buffer (pH 3) and the alkarine-based buffer (pH 10) are used for plating bath. The deposition yield of the alkarine electrolyte is relatively higher than those of the acidic bath. optimum conditions for the homogeneous diffusion is determined at annealing temperatures of 1100℃ for 3h under the high vacuum atmosphere (10^{-5} hPa). The proposed condition in this study should be applied to prepare 57Co/Rh Mössbauer source at near future..

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