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The yield of electroplating nickel in alkaline bath for fabricating Ni-63 beta source

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1. Introduction

A beta radiation source of Ni-63 with a half-life of 100.1 years is prepared using electrical deposition of radioactive Ni-63 ions on a thin non-radioactive nickel foil [1]. The electroplating process is most commonly used for Ni deposition when using 63Ni as a power source for a battery or seal source for a detector [2]. In a previous study, Ni-63 plating was fixed in a chloride bath at pH 4 [3]. The experiment results show that increasing the current density has a considerable effect on the average grain size of the deposit [3]. This process promises a fast deposition of Ni-63 to avoid a long exposure time for workers to RI. However, the diluted activity in the waste is still unsolved. However, the deposition yield of the Ni chloride bath is still unclear. Ni-63 is a pure beta emitter. The deposition yield of electroplating radioactive Ni should be conformed to identify the activity. The Ni-63 in a plating bath, after deposition, is radioactive waste. Thus, most of the Ni-63 in a bath should be coated on the substrate. Thus, the preparation of Ni-63 is an extremely expensive process.

In this study, a new base solution for plating bath, and its deposition yield were studied to decrease activity of waste, after electroplating. The newly prepared bath is consists of two base solutions,i.e., ammonium citrate (25g/l), and hydrazine hydrate (25g/l), and few drops of adjusting solution for a pH of 10. The concentration of Ni-63 is able to be significantly decreased to 0.0001 M in an alkaline bath. The deposition yields for both radioactive Ni and inactive Ni were accurately measured. In addition, the condition of electroplating radioactive Ni was optimized using a new alkaline bath.

2. Experimental Technique

The plating electrolytes are an alkaline citrate bath and an acidic chloride bath. The basic composition of the acidic bath was from 0.05 to 0.2 M of Ni, and boric acid (H_3BO_3) for buffering purposes. In the case of a citrate electrolyte, the concentration of Ni was from 0.0001 to 0.05 M. The base solution consists of hydrazine hydrate (N_2H_4-H2O), and ammonium citrate ($(NH_4)_3(C_6H_5O_7)$). The pH of the bath was adjusted to 10.0 ± 0.2 by the dropwise addition of NaOH (1N). A nickel sheet of 99.99% purity with dimensions of 10 mm×10 mm× 0.125 mm was used as a cathode (substrate) and a Pt coated Ti mesh as an anode. A Ni sheet with a high purity of 99.99 % (Aldrich) was used as the substrate. The deposition yield of Ni ions in the plating bath was measured using inductively coupled plasma (ICP, Nexion 300 D, PerkinElmer Co.) before and after electroplating. The radioactivities were estimated by comparing the plating bath measured liquid scintillation counter (LSC, Tri-Carb 2910 TR/ PerkinEmer Co.) before and after coating.

3. Results and discussion

In a previous study, we optimized an electroplating bath at pH 4. The concentration of Ni in a full chloride bath was 0.2 M. The time dependence of the deposition yields for Ni-chloride baths were measured using ICP at different Ni concentrations. Figure 1 shows deposition yields for Ni-chloride baths with a concentration of 0.05 and 0.22 M. The deposition yields were saturated at 62 % for a concentration of 0.05 M in the bath. The deposition yield was increased up to 72 %, as the concentration was increased to 0.22 M. The saturation time was quickly reached at a diluted concentration of 0.05 M. Although the concentration was increased up to 0.22 M, the deposition yield was below 74.1 %. The deposition yields show a limitation. It is well known that a full chloride bath is useful for fast deposition to fabricate a thick film [4]. Boric acid is a buffer material for stabilizing the pH. However, it was later determined that the buffering of nickel solutions below pH 5 is the result of the complexation of nickel and two boric acid molecules [4]. The time dependence of the deposition yields for $NiCl_2$ dissolved in citrate baths were measured using ICP at different concentration of Ni. Figure 1 shows the deposition yields for citrate-alkaline baths with a concentration of 0.0001 and 0.05 M. The electroplating was carried out for 8h in the same bath. The concentration of Ni in the plating bath were measured every 1 h during the deposition. Deposition yields were saturated at 91.2 % for a concentration of 0.0001 M in a bath. The deposition yield reached 90 %, although the concentration was increased up to 0.05 M. The deposition times to reach saturation were very slowly achieved for all prepared concentration of 0.0001, 0.002 and 0.05 M. The deposition times for reaching saturation were increased from 6 to 11h with a rising concentration of Ni from 0.0001 up to 0.05 M. The deposition yield was significantly increased, as the plating bath was changed from acidic chloride to alkaline citrate. Although the deposition time was increased, the radioactivity of the waste was decreased. In addition, the quantity of the total plating bath with a concentration of 0.0001 M is to be 20 ml, which is sufficient to be used as an electrode in the bath solution.

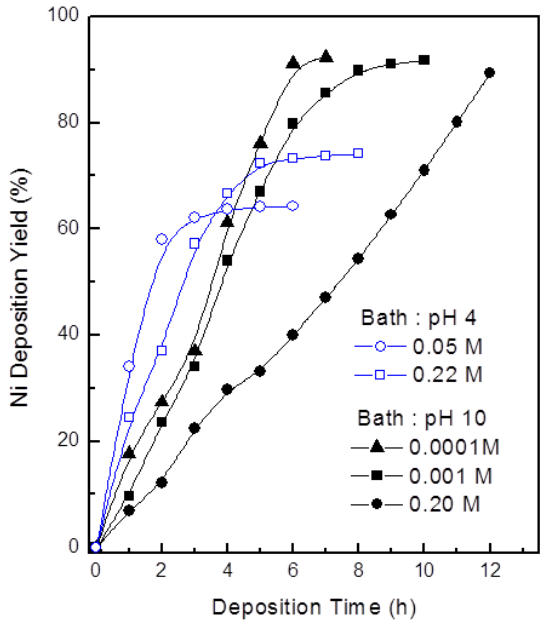

Fig. 1 Time dependent deposition yields of Ni at chloride bath (pH 4) and citrate bath (pH 10) with different concentration of Ni measured using ICP.

The time dependence of the deposition yields for Ni-63 dissolved in citrate baths was measured using LSC at a concentration of 0.0001 M. The standard solution for Ni-63 was 100.6 μCi/5ml in 0.1M HCl. The radioactivity of the electroplated 63Ni on the foil with the planar dimensions (1×1 cm2) was estimated to be about 61.38 μCi The total amount of Ni was 150 μg. The concentration of Ni in the bath was 0.0001 M. The concentration of Ni in the plating bath were measured every 0.5 h during the deposition. The deposition yields were measured to be 35.2 % after a plating time of 3 h. We also optimized the recovery process of Ni in the waste of both chloride and citrate electrolytes for the near future.

Figure 2 show SEM images of electroplated Ni on a Ni-foil. The deposition time of Ni in the chloride bath with a concentration of 0.05 M was 15 min. at 20 mA/cm^2. The Ni film prepared in a citrate bath with a concentration of 0.05 M was deposited for 45 min. at same current density. The thickness of both deposited films was almost similar, as shown in Fig 2 (a) and (b). At the same concentration of Ni, the deposition time of the chloride bath is over three times faster than that of the citrate bath. The results correspond to the time dependence of the deposition yield at the same concentration. The morphologies of the surface, such as the shape, and particle size, are almost the same, as shown in Fig 2(c) and (d).

4. Conclusions

Nickel (Ni) films were synthesized by direct current electrodeposition at a pH of 4 and 10 under a current density of 20 mA/cm2. The basic compositions of the baths were from 0.0001 to 0.05 M of Ni ions in an alkaline bath. The composition of an alkaline electrolyte was ammonium citrate (25g/l), hydrazine hydrate (25g/l), and ammonium solution (25 %) for adjusting the pH to 10. The acidic electrolyte at pH 4 was prepared using a full chloride bath. The time dependence deposition yields were measured for both acidic and alkaline electrolyte. A higher deposition yield (92 %) of the citrate electrolyte (pH 10) than 73 % of acidic electrolyte (pH4) was measured using ICP. The deposition yield of radioactive Ni-63 in a citrate bath was also same as natural Ni in alkaline electrolyte

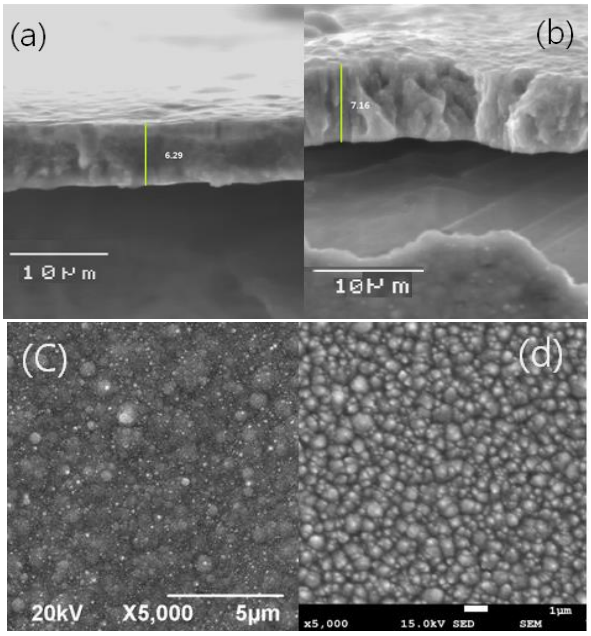

Fig. 2 SEM images of the cross section of the Ni prepared at (a) chloride bath, and (b) citrate bath. The surface layers for electroplated Ni on Ni-foil at (c) chloride bath, and (d) citrate bath.

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