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Thick tellurium electrodeposition on nickel-coated copper substrate for ^{124}I production

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Abstract

Tellurium electrodeposition on a nickel-coated copper substrate was investigated for production of iodine-124. The electrodeposition experiments were carried out by the alkali plating baths. The optimum conditions of the electrodeposition of tellurium were as follows: 6 g l^{-1} tellurium, pH = 10, DC current density of ca. 8.55 mA cm^{-2} and room temperature.

Keywords: Thick electrodeposition; Tellurium target; ^{124}I; Production; TeO_2

1. Introduction

Due to its favorable nuclear properties, the 4.18 d radionuclide ^{124}I (22.0% β^+, $E_{\beta^+} = 2.13$ MeV, 78% E.C., $E_\gamma = 603$ keV) has great potential for application in nuclear medicine. The main research areas are the functional imaging of cell proliferation in brain tumors using [^{124}I]iododeoxyuridine (Blasberg et al., 1996), the imaging of immunoreactions in tumors using ^{124}I-labelled monoclonal antibodies (Wilson et al., 1991; Daghighian et al., 1993), the in vivo imaging of ^{124}I-labelled tyrosine derivatives (Langen et al., 1990) as well as the classical imaging of thyroid diseases with [^{124}I]iodide (Frey et al., 1986; Flower et al., 1990); all of those were demonstrated by positron emission tomography.

Targets with isotopically enriched tellurium dioxide (TeO_2) are often used in production of ^{121}I (Helus et al., 1979), ^{123}I (Van den Bosch et al., 1977; Michael et al., 1981; Oberdorfer et al., 1981; Zaidi et al., 1983; Beyer et al., 1988; Scholten et al., 1989) and ^{124}I (Lambrecht et al., 1989; Firouzbakht, 1994; Weinreich and Knust, 1996; Knust et al., 1990) for medical purposes. Usually, the production process is performed according to the requirements formulated by Van den Bosch et al. (1977). This includes irradiation of solid TeO_2 deposited on a platinum support and followed by volatilization of radioiodine after heating of the target to its melting point in a stream of oxygen for some minutes. Electroplated targets with enriched Te are used in ^{123,124}I production by Van den Winkel, (2004).

^{124}I is routinely produced via the nuclear reaction ^{124}Te(d,n)^{124}I in a cyclotron by irradiation of isotopically enriched ^{124}Te, which is used either in metallic form (Lambrecht et al., 1988; Firouzbakht et al., 1994) or as TeO_2 (Firouzbakht et al., 1994; Weinreich and Knust, 1996; Guenther et al., 1998). In the cited references, the energy of the deuterons is given as 15 and 14 MeV, respectively.

Accelerator production of ^{124}I is largely achieved via nuclear reactions ^{124}Te(p,n)^{124}I and ^{124}Te(d,2n)^{124}I which is well suited for medium to low-energy cyclotrons. The NRCAM (Agricultural, Medical and Industrial Research School) employs a Cyclone-30 while CCHEN, a Cyclone18/9 (both accelerators from IBA, Belgium). Solid targetry systems on these accelerators are made up of pure

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copper backings onto which target materials are electrodeposited.

2. Experimental

The production of ^{124}I is mainly achieved via nuclear reactions $^{124}Te(p,n)^{124}I$ or $^{124}Te(d,2n)^{124}I$ which is well suited to low-energy cyclotrons. To take full benefit of the excitation function and to avoid the formation of the radionuclide impurities, the proton entrance energy should be 14 MeV (Bastian et al., 2001; Scholten et al., 1995). The physical thickness of the tellurium layer is chosen in such way that for a given beam/target angle geometry the particle exit energy should be 8 MeV. According to SRIM code, the thickness has to be 470 μm for 90° geometry. To minimize the thickness of the tellurium layer (and hence lowering the cost price per target), a 6° geometry is preferred, In which case a 47 μm layer is recommended.

Tellurium targets are prepared by DC constant current electrolysis of the metal from alkaline plating solutions. Copper plates coated with nickel are used as target carriers.

TeO_2 (7.3 g l^{-1}) was added to a 1000 ml beaker, followed by the addition of KOH (15.5 g l^{-1}) and deionized water (250 ml). The homogenized solution was filtered through fine glass filter (0.45 μm) to remove any residual particles, if necessary, to be ready for electroplating.

The non-reactive plating vessels used were hollow Perspex cylinders (diameter 6 cm, height 20 cm) fitted with an axial Pt anode wire mounted at the bottom by means of a tube-end fitting though a perforated septum. Four symmetrical windows (22.36 or 11.69 cm^2) on the vertical sidewalls allow positioning of up to four copper target backings. Each slot is sealed by an O-ring fitted-window. The slot geometrical shape determines the actual target electrodeposition area. Windows liquid-tight sealing is realized by stainless steel mechanical pestles mounted on a PVC ring surrounding the plating vessel which press the copper backing against O-ring seal. An external PVC ring is fitted with four supporting pins, which hold a motor–stirrer combination in position. The stirrer is a hollow perforated POM cylinder mounted on the axis of a DC motor and surrounding the platinum anode. The stirrer rotation speed is set at 1000 rpm during the process and its rotating direction is reversed after 8 s, improving deposit homogeneity. To keep the desired temperature at a preset level, a heater (a series of six isolated 1 Ω/1 W resistors, through which an appropriate DC-current is forced (1.1 A—40 °C up to 1.8 A—60 °C), is mounted circularly at the bottom of the vessel. An insulated sensor, introduced through the stirrer support-plate, monitors the plating bath bulk temperature. As electrolysis to depletion requires long-time plating (up to 24 h), evaporation of the plating solution occurs. To maintain a constant liquid volume of 450 ml, a glass/graphite conductivity sensor monitors the solution level and actuates a peristaltic pump at the required rate, supplying distilled water to compensate evaporation losses. The rack-mounted home-made electronics includes a motor/stirrer control, an adjustable DC voltage generator card and four V/I converters coupled to current boosters.

The freshly prepared solution of tellurium was poured into the plating vessel. This refined procedure is a result of several repeated experiments with different concentrations of KOH and electroplating currents. All electrodeposited Te target layers were examined in morphology by a scanning electron microscopy (SEM) technique (using a Joel model JSM 6400 at an accelerating voltage of 20 kV). The thermal shock tests were also conducted which involve the heating of the target up to 250 °C (the temperature that the Te layer can experience during a high current irradiation) for 1 h followed by submersion of the hot target in cold water (15 °C).

3. Results and discussion

In order to optimize the Te electrodeposition, the experimental conditions were investigated as follows.

3.1. Pretreatment of the Cu-backing prior to the tellurium plating

To ensure a good adhesion, the copper plate was cleaned using 1000 grade sandpaper followed by rinsing the copper plate with deionized water.

The copper plate was placed in the plating vessel with 450 ml of nickel sulfate plating solution. The copper plate was connected to the power supply as the cathode whiles the platinum electrode as the anode. The power supply was turned on and the current adjusted to 50 mA cm^{-2} and allowed to plate for 3 min under vigorous stirring (1000 rpm, 8/8 cycle), giving a nickel-electroplated target. The nickel-electroplated copper plates were thoroughly rinsed with water followed by acetone and then paper-dried and weighed. The obtained layers had a thickness in excessive of the copper of approximately 10 μm.

3.2. Influence of the applied voltage

The electrodeposition experiments were carried out with different applied voltages (DC, chopped sine (CS) and asymmetric square wave (SW) using a constant current at room temperature, 25 °C. All the electrodepositions were performed under vigorous stirring with a 11.29 mA cm^{-2} current density. The voltages alternatives to DC were believed to reduce or eliminate dendrite formation in the deposits. The results are summarized in Table 1.

Since the current efficiency (η) obtained using DC current was the highest, it was therefore used for the remainder of the electrodeposition experiments.

3.3. Influence of current density

Application of a very low current density results in a rather poor-quality deposit. Alternatively, a very high