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Nickel electrodepositing for uranium sealing

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Abstract

To its utilization as target for radioisotope production, uranium must be properly sealed before its irradiation in the reactor. For this purpose, aluminum has been a common choice, due to its chemical compatibility with uranium, reduced neutron cross section and mechanical stability in corrosive reactor pool environment. However, the use of aluminum in ⁹⁹Mo production comprises an alkaline dissolution route, and it not only generates considerable losses of ⁹⁹Mo, but produces large quantities of radioactive liquid as waste. In this sense, this work proposes an alternative route of producing uranium targets, based on replacing the aluminum with nickel. Uranium was electrodeposited over a nickel substrate and covered by successive electrodeposited nickel. Watts's bath and isopropanol were used as electrolytes, through pulsed electrodepositing methods. The samples were characterized by beta and gamma counter, XRD, SEM and OM. Although the electrodeposition of nickel over uranium still needs some progress, the results suggest a promising route of uranium target production.

Keywords: uranium target, nickel, uranium, electrodeposition.

Introduction

Tecnecium-99 metastable (^{99m}Tc) is formed by decay of Molibdenum-99 (⁹⁹Mo), a subproduct of ²³⁵U fission. It is the most common radiopharmaceutical, and it is used in many diagnostics, including bones, thyroid and breast cancer. As many isotopes, the ⁹⁹Mo is usually produced by neutron irradiation of ²³⁵U in a nuclear reactor. For this purpose, enriched uranium must be previously shielded and conformed (1).

Recently, IPEN has started developing a irradiation target, based on UAl_x dispersed in an aluminum alloy. This type of target is fabricated according to picture-frame technique outlined in Figure 1.

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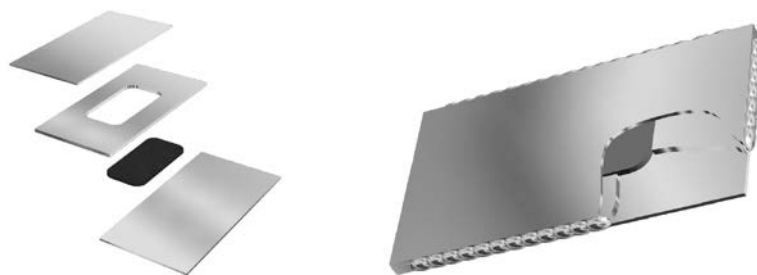


Figure 1 – Illustrating diagram of picture-frame technique to produce ^{99}Mo targets

Uranium targets from aluminum matrix, when submitted to post-radiation ^{99}Mo extraction, comprise an alkaline dissolution route. This route has disadvantages, since the aluminum and its alloys use an alkaline dissolution route, generating potentially losses of ^{99}Mo incorporated with solid waste, as well large quantities of high alkaline liquid radioactive produced as residues.

On the other hand, nickel-based targets use an acid route for ^{99}Mo extraction, generate lower volumes of waste and permit better yields. As the amount of nickel to be separated from uranium is relatively small, the post-processing of this target can be accomplished with relative ease and even can be separated during the dissolution, since nickel dissolves 8 times faster than uranium, as indicated Leonard et al. (2).

Concerning these data, the present study aims to develop a uranium target using electrodeposited uranium over nickel. After the layers of uranium were deposited, subsequent layers of nickel were deposited over it in order to seal the uranium for post irradiation (3,4,5,6).

Methodology

Aluminum substrates were covered with electrodeposited nickel, in order to provide pure nickel substrates for uranium deposition. The nickel electrodepositions were made using the Watts Bath (1 mol/L NiSO_4 + 0.2 mol/L NiCl_2 + 0.6 mol/L H_3BO_4) as electrolyte (7) and sodium dodecyl sulfate (sodium lauryl sulfate, 8.8×10^{-4} mol/L) as a wetting agent, nickel anode and pulsed galvanostatic mode (duty cycle of 50%).

After the nickel substrates were prepared, the cell electrolyte was changed to isopropanol containing 0.05 mol/L of uranyl nitrate, and pulsed current was applied again. As the uranium layers were deposited, the Watts bath was returned once again to the electrochemical cell and nickel was deposited as a sealant of uranium.

The reagents were all analytical grade. The electrodeposition steps were performed using a Metrohm Autolab 302N Potenciostat/Galvanostat with a $\text{Ag}/\text{AgCl}/\text{KCl}$ (sat.) standard electrode (+0.205 mV SHE). As counter electrode, platinum (for uranium deposition) and electrolytic nickel (for nickel deposition) were used.

The scanning electron microscope (SEM) images and energy dispersive X-ray spectroscopy data were obtained at a Hitachi TM3000 scanning electron microscope with a Bruker Quantax 70 EDS system module. All the images were obtained using backscattered electrons.

X-ray diffraction data (XRD) were acquired in a Rigaku Multiflex using Cu- α radiation. Reference diffraction peaks were extracted from ICDD (International Crystallography Diffraction Data) files.

For quantification of the uranium present in the sample, a reference graph of beta/gamma counts was plotted using known quantities of uranyl nitrate with a Ludlum 2929 Dual Scaler Counter.

Results and discussion

Electrodeposited nickel SEM images of the prepared substrates can be seen at figures 1 and 2. Due to the pulsed technique, during the electrodeposition the nucleation of new crystals is privileged in contrast with grain growth. The result is a fine grained, smooth and plain deposit, even though commercial organic brighteners were not used. Figure 2 shows the topography of the sample, confirming the homogeneity of the surface.

At figure 3, X-ray diffraction data of the electrodeposited nickel is presented. It shows the crystallinity of the deposited layer, and no other crystalline phases present at the sample.

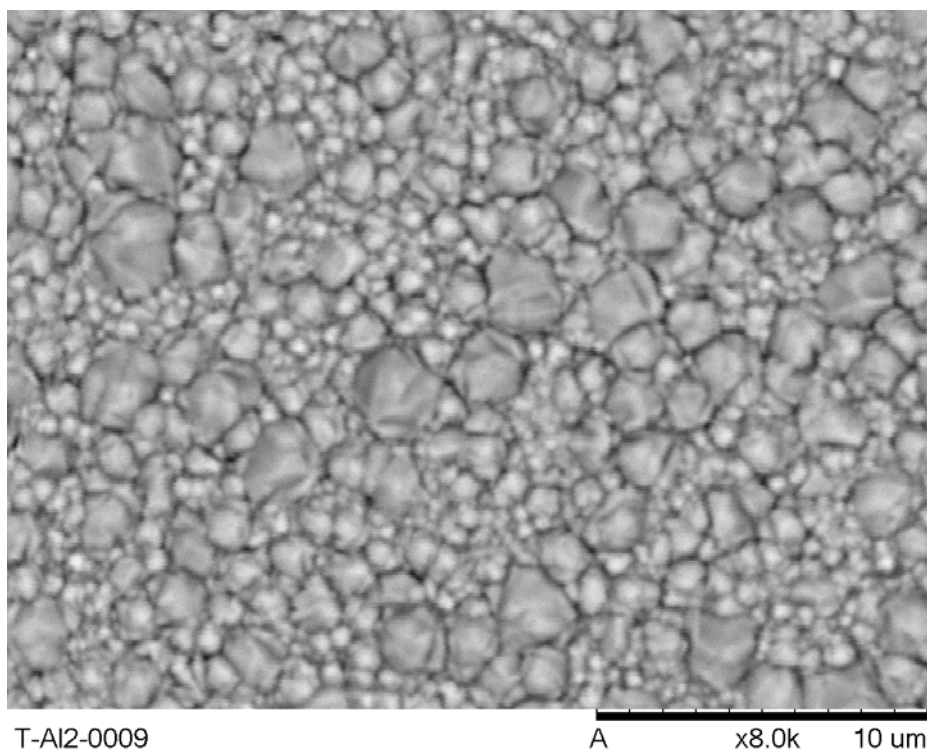


Figure 1 - SEM image of the electrodeposited nickel over aluminum.

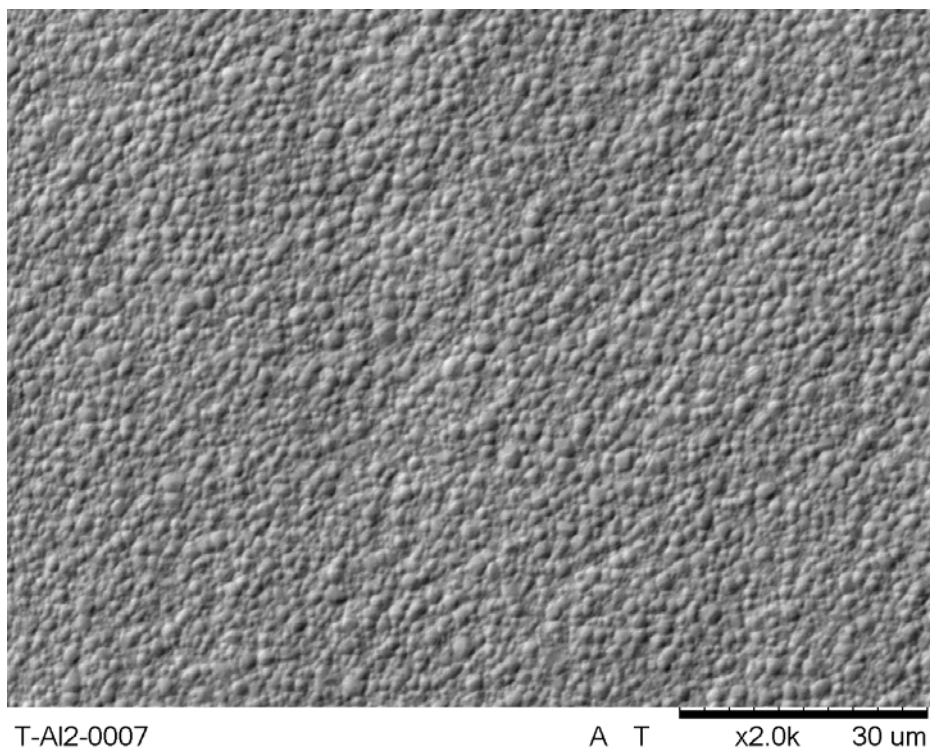


Figure 2 - SEM image of the electrodeposited nickel over aluminum (topographic mode).

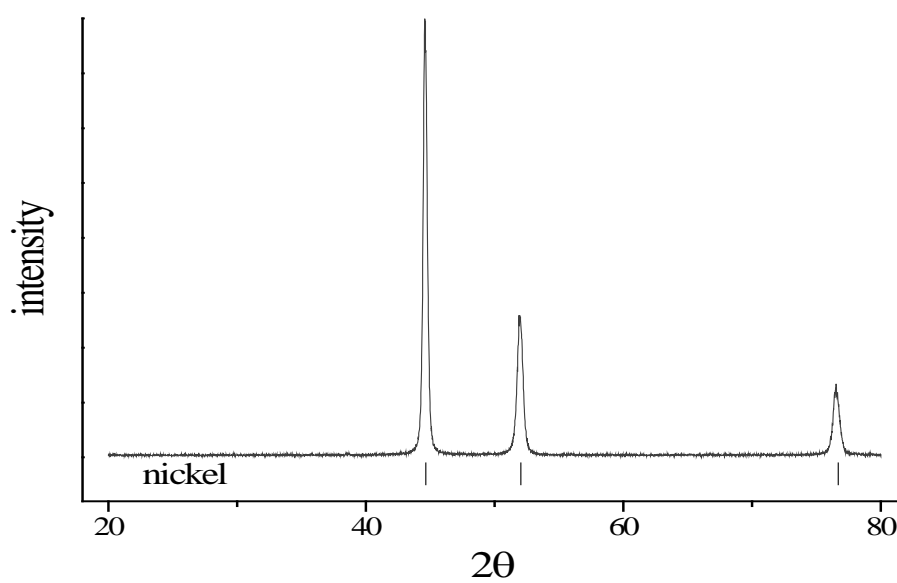


Figure 3 - XRD of the electrodeposited nickel over aluminum, with nickel reference diffraction peaks from ICDD files.

Figures 4 and 5 show the SEM images of subsequent electrodeposited of uranium over nickel. Its deposition starts preferentially at grain boundaries, probably in a form of mixed hydroxides of uranium without cristallinity. Due to its enormous difference of atomic weight compared to nickel, high contrast images could be obtained. These results are confirmed with the energy dispersive X-ray analysis (EDS). At figure 6, EDS was performed over the exactly area of figure 5, and its spectra is shown at figure 7.

During the electrodeposition steps of uranium, we noticed that after the cathodic current is stopped, the deposited uranium tends to redissolve, returning to the solution. It occurs because of the basic nature of the deposit and low pH of the solution. To avoid that, as soon as the electrodeposition reaction stops, the electrolyte must be readily removed from the cell.

The sample illustrated at figures 4 and 5 was submitted to a beta/gamma counter analysis and the result were 163175 counts for 20 minutes of analysis. Correlating this data to the calibration curve (figure 8), it is possible to estimate the quantity of deposited uranium as 0,067g.

In figure 9, is exhibited an optical microscopy image of a sample submitted to two uranium and subsequent nickel electrodepositing steps. It can be seen the different layers in microscopy. Even though the nickel coverage of uranium layers was satisfactory, these samples are still in development to reach better mechanical properties, in order to perform proper sealing of uranium to be irradiated immersed in the reactor pool. This sample had 65137 beta/gamma counts during 20 minutes, which is equivalent to an amount of 0.026g of uranium.

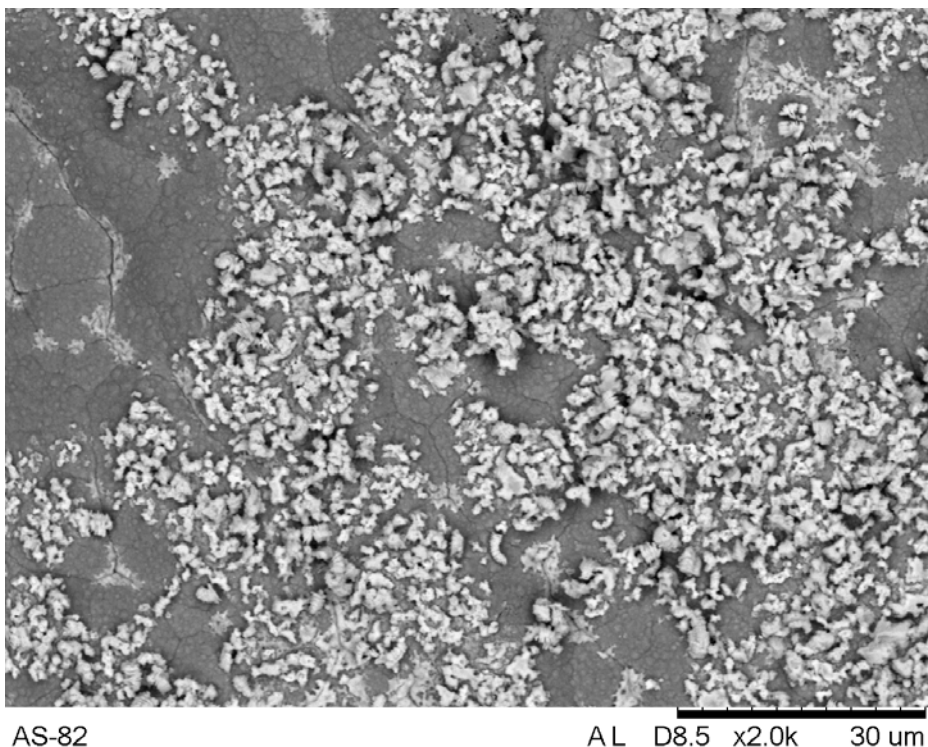


Figure 4 - SEM image of uranium deposited over nickel.

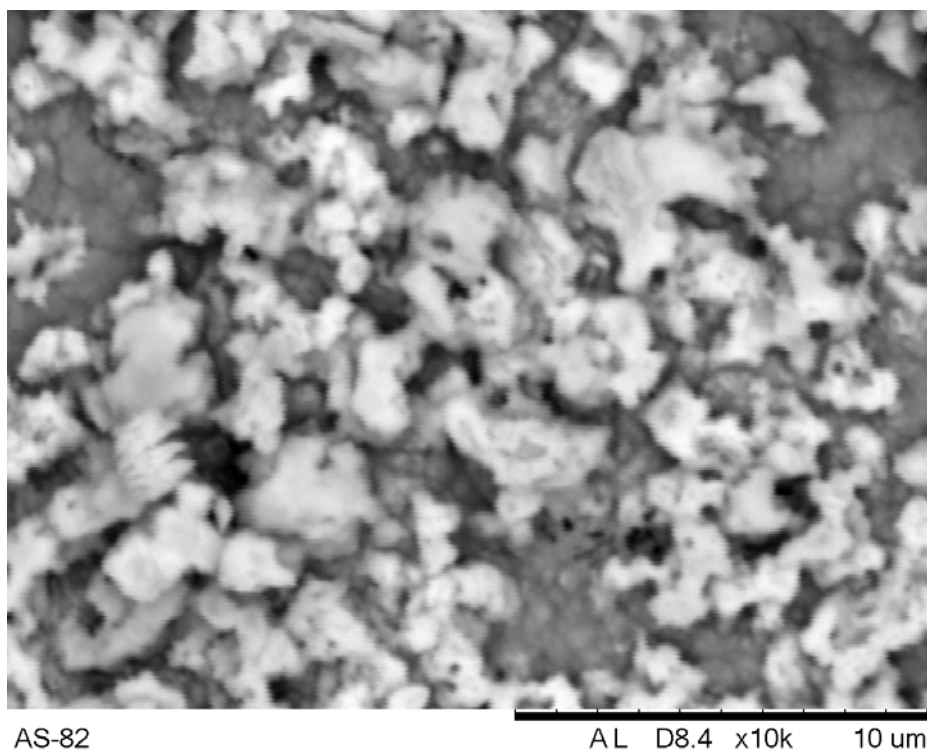


Figure 5 - SEM image of uranium deposited over nickel.

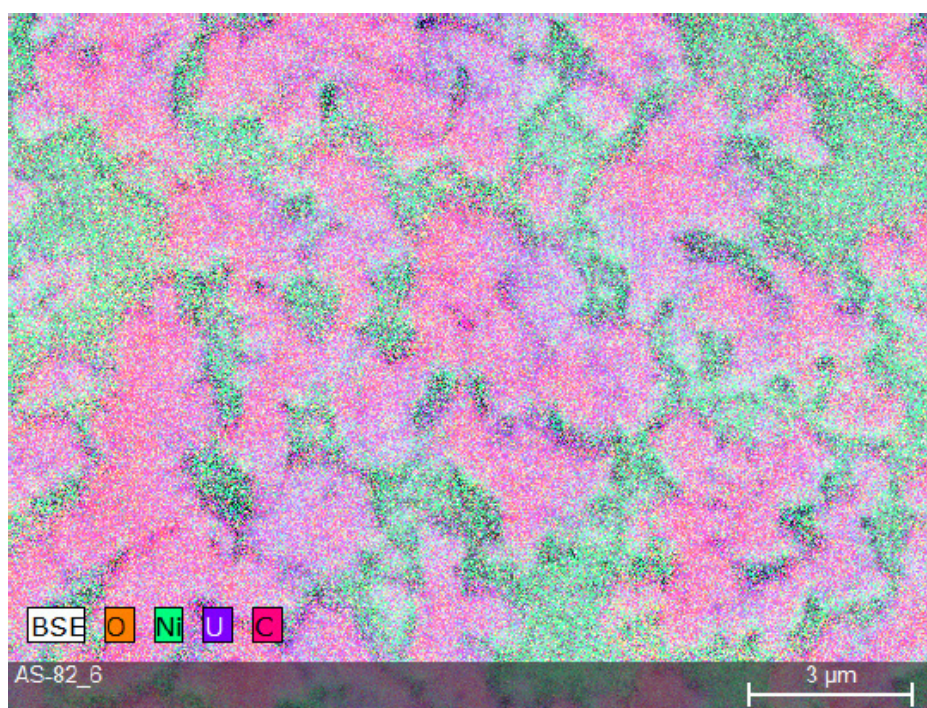


Figure 6 - EDS map plotted over the figure 5 micrography of uranium deposited over nickel.

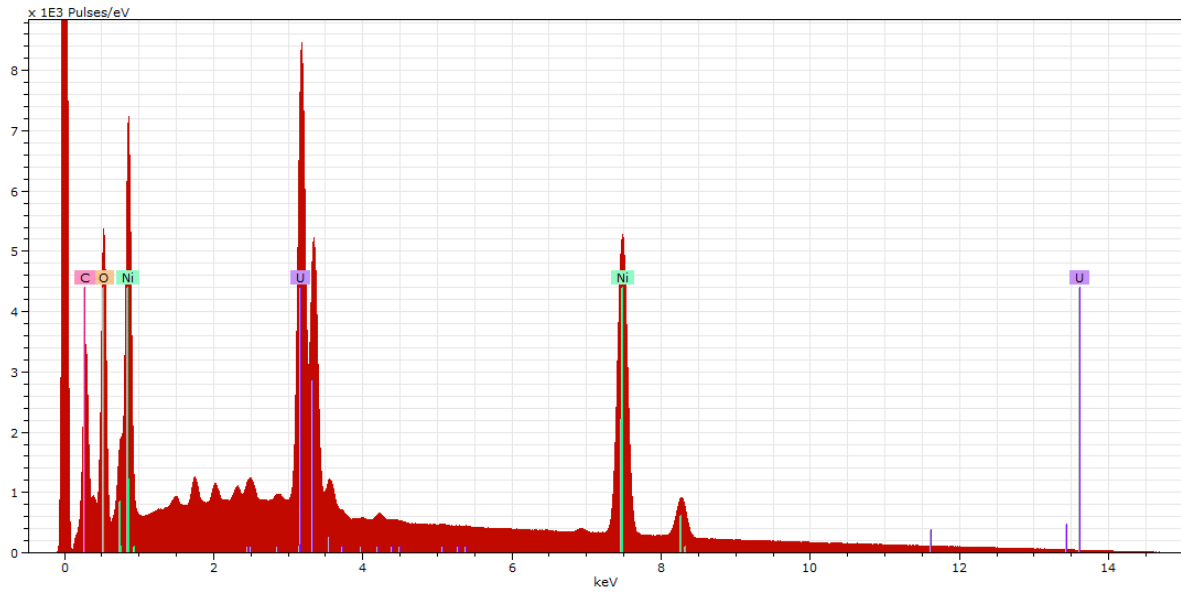


Figure 7 - EDS results from figure 6 analysis.

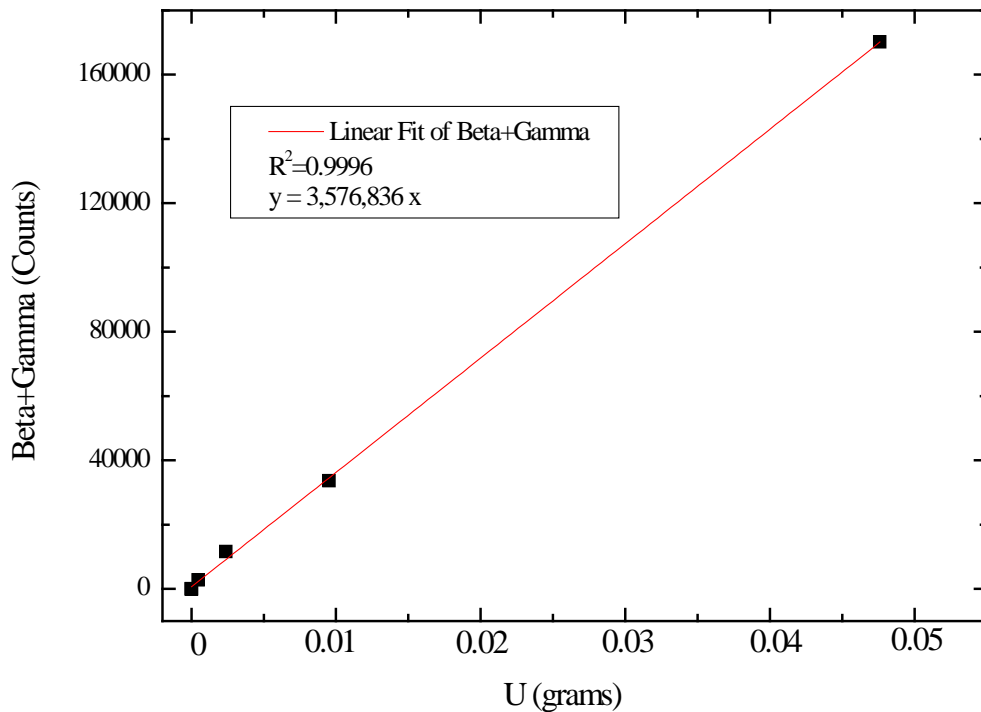


Figure 8 - Calibration curve with known quantities of uranyl nitrate and beta/gamma counts.

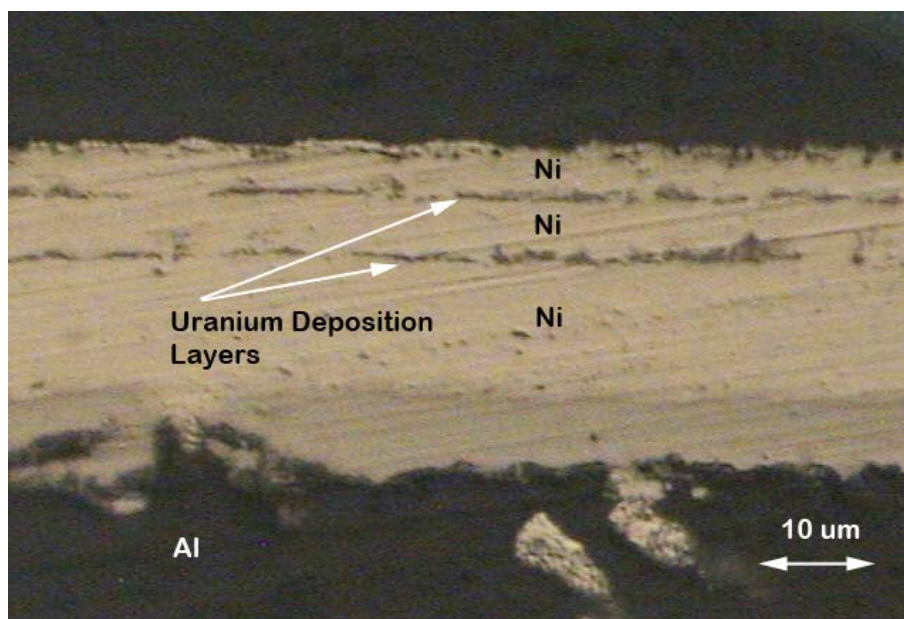


Figure 9 - Optical microscopy of a sample submitted to various Ni/U electrodeposition steps.

Conclusions

Nickel-covered aluminum substrates were used effectively to electrodeposit uranium, using isopropanol as electrolyte. The deposits were not homogeneous and appeared to exist in a mixed hydroxide form, therefore, not conductive. However, nickel could be electrodeposited successfully over uranium layers, and it represents a promising route for uranium targets manufacture, aiming radioisotopes production.

Bibliography

- (1) **National Research Council, Committee.** *Medical isotope production without highly enriched uranium.* [ed.] National Academies Press. 2009. <http://www.nap.edu/catalog/12569.html>. ISBN: 0-309-13040-9. Accessed on oct/2011.
- (2) LEONARD, R. A; CHEN, L.; MERTZ, C.,J.; VANDEGRIFT, J. F.. Progress in dissolving modified LEU Cintichem targets. [ed.] Argonne National Laboratory. **RERTR**. 1996. <http://www.rertr.anl.gov/99MO96/LEONAR96.PDF>. Accessed on oct/2011.
- (3) SULTAN, S.. Application of pulse plating for nickel electrodeposition from a formamide/water bath. **Technically Speaking**. 2008, 12, p.12-14.
- (4) WONBAEK, K.; WEIL, R.. Pulse plating effects in nickel electrodeposition. **Surface and Coatings Technology**, 1989, v.38, p.289-298.

- (5) SALIBA-SILVA, A.M.; GARCIA, R.H.L.; MARTINS, I.C.; URANO DE CARVALHO, E.F.; DURAZZO, M. Nickel electrodeposition over powder compact for irradiation target. In: Eight International Latin-American Conference on Powder Technology, 06-09 November, 2011, Florianópolis, SC – Brazil. **Proceedings of the Eight International Latin-American Conference on Powder Technology**, 2011.
- (6) SALIBA-SILVA, A.M.; GARCIA, R.H.L.; MARTINS, I.C.; URANO DE CARVALHO, E.F.; DURAZZO, M. . URANIUM BRIQUETTES FOR IRRADIATION TARGET. In: 2011 International Nuclear Atlantic Conference, 24-28 October, 2011, Belo Horizonte, MG - Brazil. **Proceedings Proceedings of the INAC 2011**, 2011.
- (7) WATTS, O. P. Rapid Nickel Plating. **Trans. Am. Electrochemical Society**, v.29, 1916, p. 395–403.

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