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Electroplating nickel onto uranium as fission-recoil barrier

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Abstract. Plating experiments have been performed in order to obtain fission recoil barriers on low-enriched uranium (LEU) metal foils used as targets suitable for the production of ^{99}Mo. The results of Ni deposition obtained in electroplating and electroless plating baths are presented. The best method of pretreatment uranium prior to plating is suggested. It was found that etching in solutions containing $NiCl_2$ gave better results on uranium rods, whereas electroplating produces coatings with the desired level of adherence and coverage on uranium foils. The thickness of the nickel plates, evaluated both microscopically and gravimetrically, is sufficient to act as a fission recoil barrier.

Key words: uranium • nickel • plating • fission-recoil barriers

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Introduction

Molybdenum-99 is produced mainly for medical application by irradiating high-enriched uranium targets (HEU). The replacement of such targets with low-enriched uranium (LEU) will require 5÷6 times more uranium for equivalent yield of ^{99}Mo.

The proposed target design [4] consists of a thin LEU metal foil sandwiched between the walls of Al tubes. The observed difficulties in removing the uranium foil from the target assembly has been attributed to bonding of the uranium to the Al tubes due to the ion mixing caused by fission fragments escaping from the uranium foil during neutron irradiation. The proposed solution [7] is to add fission-fragment absorbing barriers between the uranium foil and the Al tubes. The barrier layer thickness depends on the recoil range of fission fragments in a particular material. For nickel, the recoil distance amounts to ~ 7 μm [3]. For the sake of safety, the barrier layer thickness should be approximately twice the recoil distance. The barrier layer must also assure good coverage of the foil to preclude the possibility of localized bonding to the target and heat dissipation during irradiation.

The procedure of target assembling comprises wrapping of a uranium foil 140 μm thick between two sheets of a nickel foil 15 μm thick. Such a sandwich is collared around the inner Al tube and loaded into the Al outer tube. Finally, the target is expanded in a draw die by pulling the appropriately sized draw plug through the target.

If the Ni plating quality will assure good tightness, thickness and adherence to the uranium foil, this may considerably simplify the procedure of target assembling thus eliminating the troublesome step of sandwiching the foils.

Experimental

Methods

Pretreatment of uranium prior to plating

The principal difficulty in plating uranium is that it oxidizes readily in air and water, and the resulting oxide layer makes it impossible to obtain uniform and adherent deposits. Therefore, preparation of the uranium metal surface prior to plating was found to play a key role in the quality of the resultant coating [1, 2, 5].

For obtaining a good mechanical bond, the procedures applied comprised degreasing, cleaning the surface from most of the uranium oxide, and etching the uranium to obtain a rough surface. The most frequently used etchants have been nickel- and ferric-chloride solutions. Prior to etching, immersion in 8 M HNO_3 was applied.

Four methods of pretreatment were used.

Method I – with the use of $FeCl_3$ applied for uranium rod.

a) degreasing in xylene and ethanol – 1 min,
b) rinsing in distilled water,
c) pickling in 8 M HNO_3 – 7 min,
d) rinsing in distilled water,
e) etching in 5 M $FeCl_3$ – 2 min,
f) rinsing in distilled water,
g) pickling in 8 M HNO_3 – 6 min,
h) rinsing in distilled water,
i) pickling in 8 M HNO_3 – 5 min,
j) rinsing in distilled water.

Method II – with the use of $NiCl_2$ + HNO_3 applied for uranium rod.

a) degreasing in xylene and ethanol – 1 min,
b) pickling in 8 M HNO_3 – 10 min,
c) rinsing in distilled water,
d) etching in $NiCl_2$ + HNO_3 solution – 1.5 min,
   $NiCl_2 \cdot 6H_2O$ – 510 g/l ,
   HNO_3 – 340 g/l,
   temperature – 40°C,
   time – 50–120 s,
e) rinsing in distilled water,
f) pickling in 8 M HNO_3 – 15 min,
g) rinsing in distilled water,
h) treble repetition of the steps d) – g).

Method III – similar to method II except the repeated post-treatment h). This method was applied for a uranium foil whose surface was less oxidized than that of the rod.

Method IV – without etching in chlorides, applied for uranium foil.

a) degreasing in xylene and ethanol – 1 min,
b) pickling in 8 M HNO_3 – 15 min,
c) rinsing in distilled water,
d) pickling in 8 M HNO_3 – 15 min,
e) rinsing in distilled water.

Electroless plating

In this method electroless nickel deposits are obtained from a solution containing, except nickel, also some amount of phosphorus. The phosphorus is a result of the hypophosphite reducing agent used in the electroless nickel plating solutions.

The nickelizing conditions were: temperature – 93–96°C; pH – 4.3–4.5; bath ratio control – 1 dm^2/dm^3; plating rate – 15–25 μm/h; volume – 40 ml.

The deposition process was initiated by contacting the uranium rod with a steel plate immersed in the solution.

Galvanic plating

The Watts bath was used in these experiments [6].

$NiSO_4 \cdot H_2O$ – 34 g/100 ml; $NiCl_2 \cdot 6H_2O$ – 4.7 g/100 ml; H_3BO_3 – 3.8 g/100 ml.

Current density – 30 mA/cm^2; temperature – 50°C; time – 60 min; uranium cathode – 3.6 cm^2; nickel anode – 8 cm^2.

The nickel layer thickness was evaluated by weighing and microscopically. The cross section of nickel plated uranium was visualized by the use of a metallographic microscope (Material Research Laboratory of the Institute of Atomic Energy in Świerk).

Materials

For the plating experiments, natural uranium (NU) was used in the form of a rod 60 mm long and 5 mm in diameter as also NU foil strips 10 × 50 mm and 150 μm thick.

For the preparation of plating baths and etching and pickling solutions, analytical grade reagents were used. Commercially available bath (Chemonickel-95, GalwImp Ltd. PL) was used in electroless plating experiments.

Glass vials served as plating cells. The volume of the electrolyte was 40 ml. A pure nickel foil was applied as anode in the electroplating experiments.

Results and discussion

Uranium is one of the most difficult metals to plate because its surface has the tendency to oxidize and to become passive. It should be noted that since aqueous environments in which the uranium is pretreated react with uranium, it is not possible to prepare a completely oxide-free surface. The remaining oxide film on the uranium after pretreatment should be uniform to allow good plating conditions. It should also provide some protection from the plating solution but should not be bulky nor passivate the uranium surface. Deep valleys or high peaks on uranium surface can cause bad adherence of the nickel plates.

For the initial tests on electroless plating, ready available uranium rods were used. Long storage before use caused that their surface was highly oxidized and corroded. Pretreatment in accordance with method I resulted in a coarse, unevenly etched surface. This treatment had a detrimental effect upon subsequent nickel deposition.

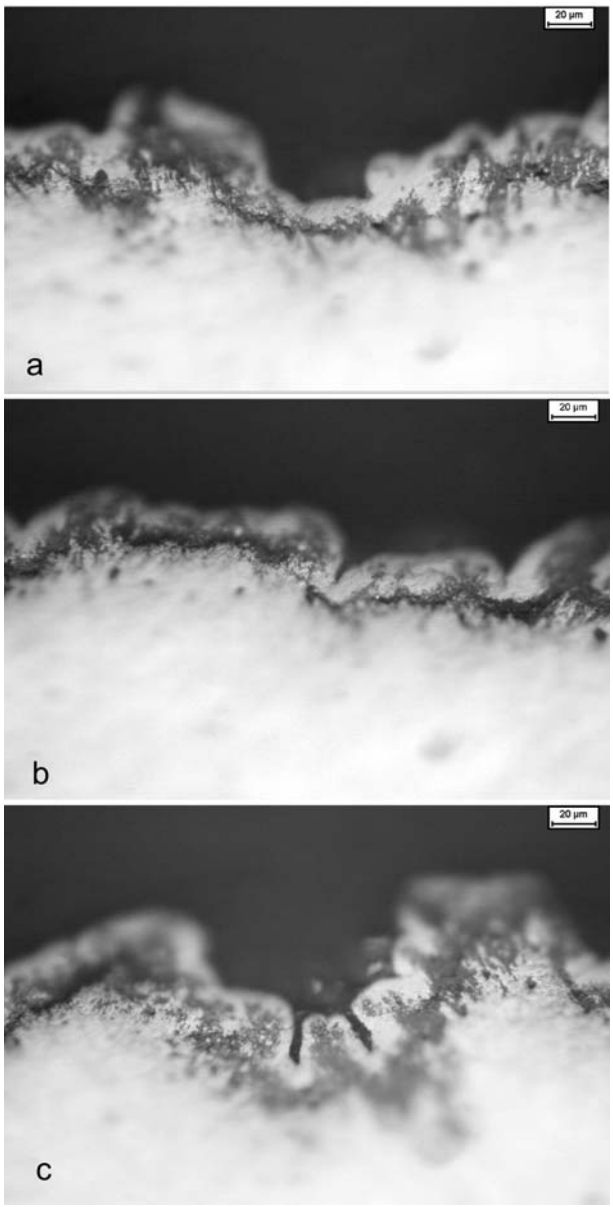

Fig. 1. Electroless nickel deposit on NU rods pretreated according to method II. a – relatively good coverage and adherence; b – localized lack of adherence; c – cracks and pores in the nickel plate (500X).

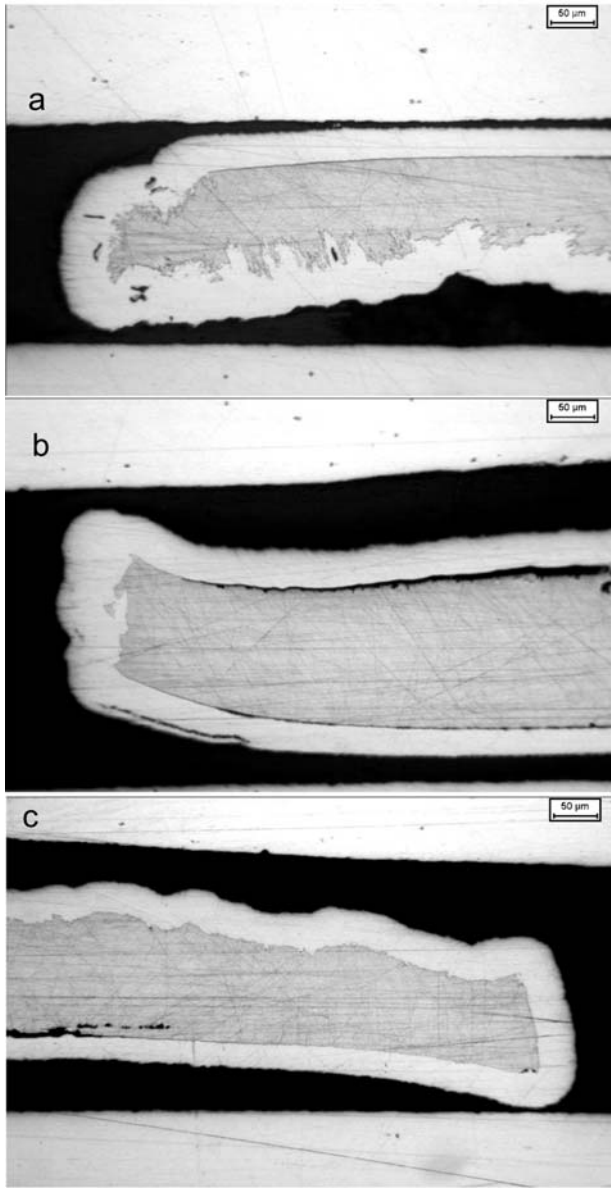

Fig. 2. Nickel plating on NU foils. a – foil etched prior to plating (method III); b and c – foil non-etched (method IV).

The results of electroless nickel deposition onto uranium rod pretreated according to method II are presented in Fig. 1 as cross section along the perpendicular axis of the rod. Figure 1a shows the relatively good coverage and adherence for nickel plating on NU substrate. In Fig. 1b, the localized loss of adherence can be observed. Figure 1c shows the pores in the nickel plate, apparently originating from areas on the uranium surface on which the nickel did not nucleate.

The results of galvanic plating of uranium foils are illustrated in Fig. 2. The nickel plating on NU foils etched according to method III is shown in Fig. 2a. Complete coverage and adherence can be observed. The nickel deposition occurred also inside the pores of the substrate, resulting apparently due to overetching. Figures 2b and 2c illustrate the deposited Ni layers on uranium substrates pickled according to method IV, without etching in chlorides. Some lack of adherence can be observed on one side of the foil (Fig. 2b), also a slight difference in thickness on opposite sides of the NU foil. Improvement of the adherence on both sides of the foil can probably be achieved by applying anode surrounding the cathode. Almost non overplating at the edges is observed (Fig. 2c).

Some additional results are illustrated in Figs. 3 and 4 concerning foils etched and non-etched (methods III and IV), respectively. Different thickness on the opposite sides, as also poor adherence can be observed (Fig. 3). Etching caused also some deterioration of the uranium substrate. It may cause that some uranium crystals are oriented in such a way that a large overvoltage occurs and nickel bridges over the pores. This does not happen in the case of the foil pickled in HNO_3 only (method IV). This is illustrated in Fig. 4. The plating shows better adherence and uniformity. The uranium surface has no pores, peaks or cavities.

The plating thickness was evaluated gravimetrically and microscopically. Both etched and unetched samples

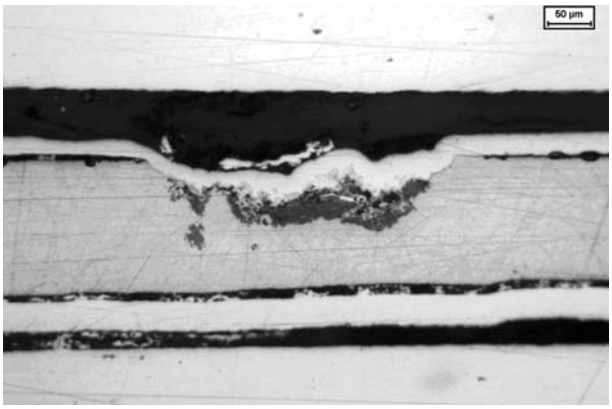

Fig. 3. Nickel plating on etched NU foil. Different thickness of plate and poor adherence. Localized deterioration of uranium substrate.

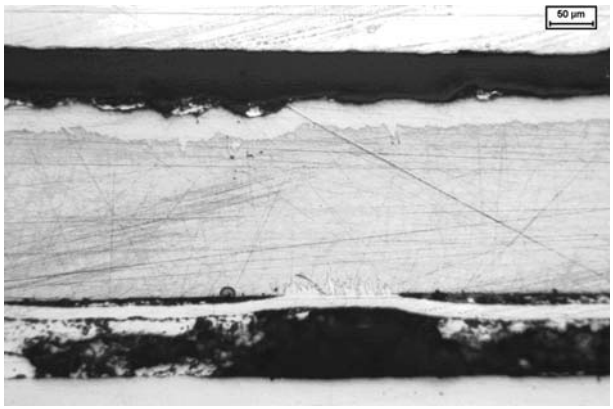

Fig. 4. Nickel plating on non-etched NU foil. Different thickness of Ni plate on opposite sides of the substrate and poor adherence on one side.

Table 1. Gravimetric and microscopic evaluations of Ni plating thickness

	Average gravimetric thickness	Average metallographic thickness
Etched foil	~ 20 µm	~ 35 µm
Non-etched foil	~ 30 µm	~ 45 µm

were used for this purpose. The results obtained are presented in Table 1. The average gravimetric thickness is considerably lesser than that resulting from the metallographic specimens. Nevertheless, the plates are sufficiently thick to act as fission recoil barriers.

Conclusions

The results of this study confirmed that pretreatment of uranium substrate prior to plating play a key role in the quality of the obtained plates. The success of chemical pretreatment was dependent to some extent on the purity of the uranium and on its metallurgical state (cast, rolled or wrought). Worse results were obtained for uranium rods than for the foil. Etching in $FeCl_3$ (method I) failed. Dark spots on the uranium surface remained unremovable even after repeated pickling in 8 M HNO_3 solutions.

Although the appearance of the plates obtained in electroless plating process seem to be fairly good, the same method applied to foil gave unsatisfactory results.

Fairly good results were obtained on etched in $NiCl_2$ solution (method III) and non-etched (method IV) uranium foils plated electrochemically. The thickness of the nickel plates obtained in this study is sufficient to act as a fission-recoil barrier. Important observation is that no considerable overplating at the edges of the foil occur, although it can change when a full-sized foil will be plated. Such an effect may excite the tolerance needed to maintain the desired fit within the target. In such a case, the option may include rolling of the plated foil to a uniform thickness prior to assembling the target.

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