RERTR 2010 — 32nd International Meeting on
Reduced Enrichment for Research and Test Reactors

October 10-14, 2010
SANA Lisboa Hotel
Lisbon, Portugal

SPUTTERING AS A COATING TECHNIQUE FOR MONOLITHIC U-MO FUEL FOILS

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ABSTRACT

The application of diffusion preventive coatings onto fuel and the proper bonding of fuel to cladding are important issues for the fabrication of monolithic U-Mo fuel plates. Sputtering, a common coating technique in microelectronics, is introduced in this context as a reliable and simple method, both to coat monolithic U-Mo fuel foils with different diffusion preventive materials and to possibly overcome bonding problems via coating the fuel foils with cladding material prior to the actual cladding application.

As an example, the cleaning and coating of monolithic U-10Mo foils in small size (60 mm x 30 mm) and full size (600 mm x 60 mm) will be presented. In a first step, Zry-4 and Ti were deposited on these foils as diffusion preventive materials, in a second step, the cladding materials Al and AlFeNi were applied onto the coated foils. Cross sections reveal the homogeneity and apparently good adhesion of the coated material.

1. Introduction

The formation of an interaction diffusion layer (IDL) between U-Mo and Al was identified to be the major obstacle in the utilization of U-Mo alloys as nuclear fuels. Several measures against IDL formation were therefore suggested and tested in the last years. The application of a monolithic fuel design turned out to be a very effective measure, as it results in a strong reduction of IDL formation due to the reduced U-Mo / Al interface. It is believed, that a barrier layer between the U-Mo fuel core and the surrounding Al cladding could further reduce or even completely avoid IDL formation in the monolithic fuel design. Therefore this option is currently under investigation.

In 2009, TUM and AREVA-CERCA launched a common R&D program on monolithic U-Mo fuel [1]. Amongst other objectives, the program has the aim to identify an appropriate barrier material and to provide a feasible process for barrier layer application. The sputter coating technique turned out to be a convenient and promising barrier application method. As sputter coating is being studied and used at TUM since several years in the field of nuclear materials processing, it was chosen for further investigation in the context of the program.

2. Setup and materials

Two different experimental setups were used for sputter coating. Basic feasibility tests were all conducted in a small and simple tabletop sputtering device (see figure 1 (b)) with substrates of 10 mm x 10 mm or 25 mm x 25 mm dimension. For the sputter coating of full sized foils, a specially designed sputtering plant has been used (see figure 1 (a)), which is mounted inside an argon filled glove box that allows to perform all steps of sample handling without oxidation ([1], [2]).

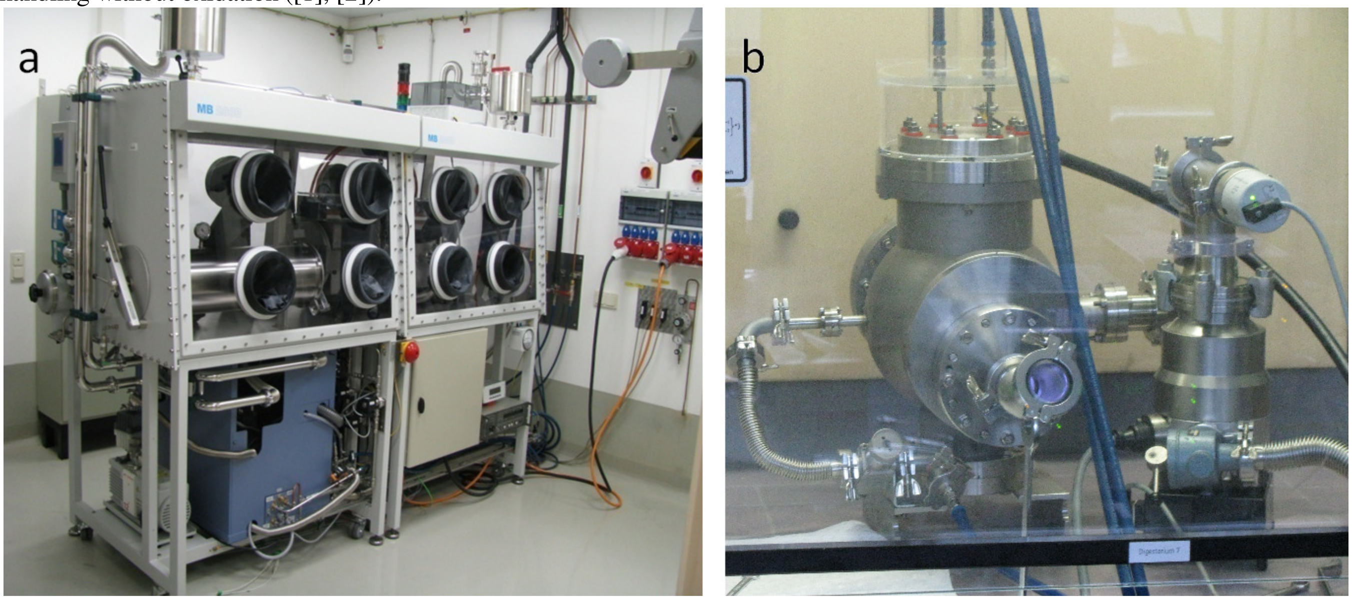

Figure 1: (a) Sputtering plant for coating full size foils. (b) Tabletop sputtering setup for coating smaller samples.

According to the material pre-selection made for the AREVA-CERCA R&D program, UO_2 as well as Zr, Ti, Nb and Ta were regarded as suitable materials for a diffusion preventive barrier layer [1]. However, oxide layers can comparatively easy be applied to monolithic fuel using heating and subsequent oxidation of the fuel surface under air. Therefore, the focus of the investigation was on sputter coating of fuel with Zr, Ti, Nb and Ta. As a measure for the minimal coating thickness required for each of these materials, the mean ranges of the typical fission fragments ^{90}Sr and ^{140}Xe in the different materials were used (see table 1).

material	**mean range of ^{90}Sr [µm]**	**mean range of ^{140}Xe [µm]**
Zr	8.2	7.4
Ti	8.6	7.8
Nb	6.2	5.6
Ta	5.7	5.2

Table 1: Ion range of the typical fission products ^{90}Sr and ^{140}Xe at 80 MeV in the different materials (calculated by SRIM).

Feasibility studies on the processing of these materials were conducted in the context of sample production for heavy ion irradiation tests [3] and tensile tests [4]. It could be shown, that the application of barrier layers of all mentioned materials in the necessary range of thickness and quality by sputter deposition is technically easy to realize for samples with size 10 mm x 10 mm and 25 mm x 25 mm. Scaling up the process to full-sized fuel foils thus was regarded as feasible.

3. Coating of surrogate foils

Several DU-10Mo full size foils were provided to TUM by courtesy of Y-12 National Security Complex. Each foil is approximately 600 mm x 60 mm in size and ~ 350 µm in thickness (see figure 2 (a)). The surfaces of all foils were completely oxidized when delivered, so each foil had to be roughly cleaned by grinding before it could be coated. The grinding is done in air and without chemical aids. After grinding, the foil is inserted and stored in the high purity argon atmosphere of the sputtering plants´ glove box.

Each coating process requires the mounting of a sputtering target of required material, positioning of the foil to be coated, evacuation of the sputtering chamber to a base vacuum of ~ 10^{-4} Pa and injection of a steady stream of argon to

reach a working pressure of ~ 10^{-1} Pa. To start the sputtering process, a voltage of several hundred volts has to be applied to the sputtering target to ignite the glow discharge plasma.
In the first processing step, the target is used as an anode. If the plasma is ignited in this configuration, the surrogate foil is continuously bombarded by ions, which results in a sputter cleaning of the foils surface on atomic scale (see figure 2 (b)). This cleaning process is maintained for approximately 10-15 minutes at a sputtering power of ~ 100 W. After this cleaning step, the polarity of the target is switched to cathode and the plasma is ignited again. Now the target is bombarded by ions and the coating of the foil starts (see figure 2 (c)). The coating process is maintained for several hours at a plasma power of ~ 1000 W until the desired layer thickness is reached. When finished, the sputtering chamber is ventilated and the foil can be removed.

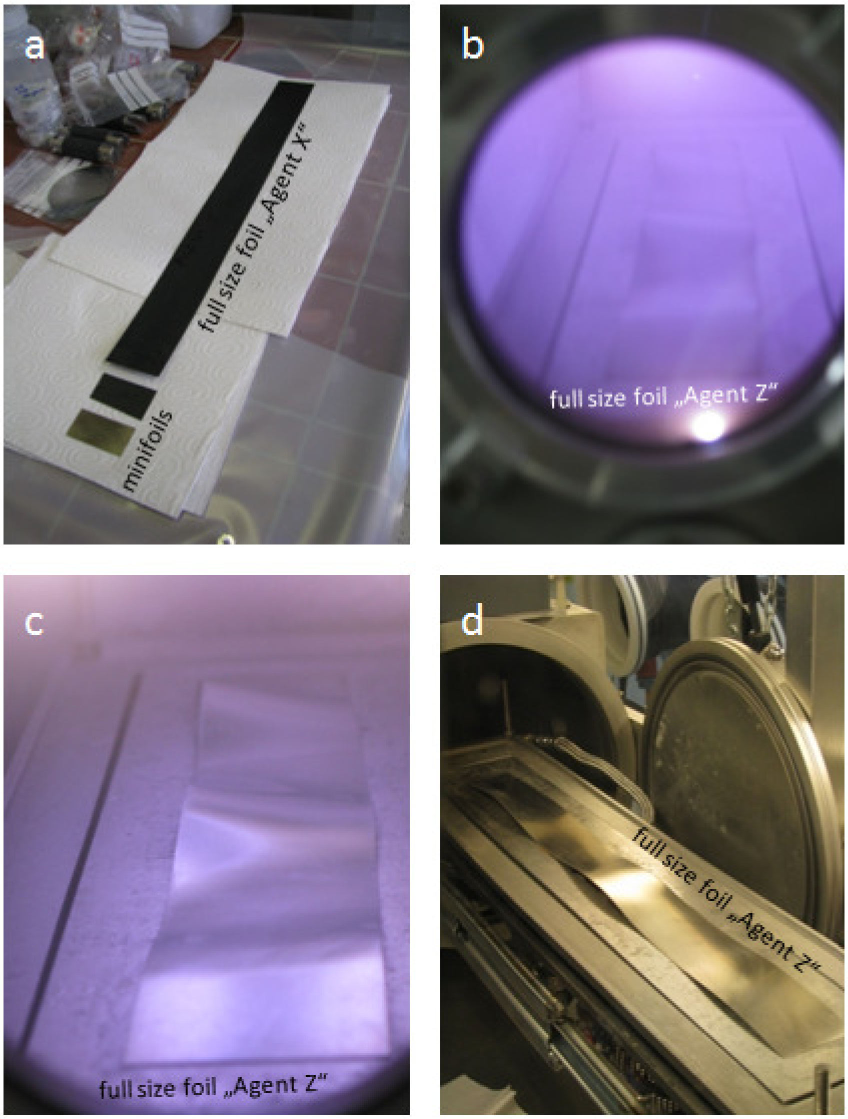

Figure 2: (a) The full size foil “Agent X” was cut into several minifoils, which were used for the first coating tests. (b) The full size foil “Agent Z” during the sputter cleaning and (c) during the sputter coating process. The ripples in the foil do not result from cleaning or sputtering, but were already present in the foil when delivered. (d) “Agent Z” after the coating with Zry-4 and AlFeNi.

By the described process, the configurations listed in table 2 have been realized.

number & description	size [mm x mm]	coated with
2 minifoils (from Agent X)	30 x 30	Zr, Zry-4 + AlFeNi
6 minifoils (from Agent X)	60 x 30	Zry-4 + AlFeNi
1 full size foil (Agent Z)	600 x 60	Zry-4 + AlFeNi
1 full size foil (INL 4 - 2A)	600 x 60	Ti + Al

Table 2: DU-10Mo minifoils and full size foils that have been coated so far by sputtering.

The term “minifoils” denotes foil pieces with a size of 30 mm x 60 mm respectively 30 mm x 30 mm, that were cut out of a full size foil. Minifoil coatings with Zr and Zircalloy 4 (Zry-4) were performed prior to the full size coatings, to clarify whether a stable coating upon a grinded surface can successfully be achieved. This was not self-evident, as all coating tests before had used polished substrates, but the resulting coatings turned out to be stable indeed.
Six further minifoils with size of 30 mm x 60 mm and one full size foil were coated with a first layer of Zry-4 and on top of that a second layer of the aluminum alloy AlFeNi (see figure 3). The first layer is intended to serve as a diffusion preventive barrier as described before. The purpose of the second layer is to avoid bonding difficulties between barrier layer and cladding, which eventually may appear during clad bonding. By sputtering a thin layer of cladding material, a “pre-cladding”, onto the barrier layer, the clad bonding does not have to be achieved between two different materials (barrier and cladding) but between two layers of identical material (pre-cladding and cladding), which should not impose a problem.
Furthermore, the pre-cladding serves as an oxidation preventive barrier, as far as barrier coated foils with an oxygen sensitive barrier material as Zr have to be handled in air.
The last coating so far has been another full size foil, which was coated with Ti as diffusion preventive barrier and Al as pre-cladding.

4. Examination

All sputter coated foils and minifoils listed in table 2 look excellent and don´t show any signs of delaminating. One of the minifoils sized 30 mm x 30 mm, coated with Zry-4 and AlFeNi, was cut and the cross section was examined by SEM (see figure 3 (a),(b)). The applicated layers are very plane and homogenous, no voids, cracks or delaminating are visible. In EDX measurements, no diffusion layers between fuel surrogate, barrier and pre-cladding could be observed. XRD measurements of the sample are currently prepared.
A series of tensile tests with coated minifoils of 30 mm x 30 mm size is also in preparation at TUM. Its´ aim is to examine the bond strength of the sputter deposited layers to the grinded DU-10Mo foils. These tests will complement the tensile tests conducted by Dirndorfer [4], who measured an excellent adhesion of our sputter deposited layers on polished substrates.
In parallel, AREVA-CERCA has received 6 minifoils coated with Zry-4 and AlFeNi to perform roll bonding tests. Results of these tests are expected in the next few weeks.

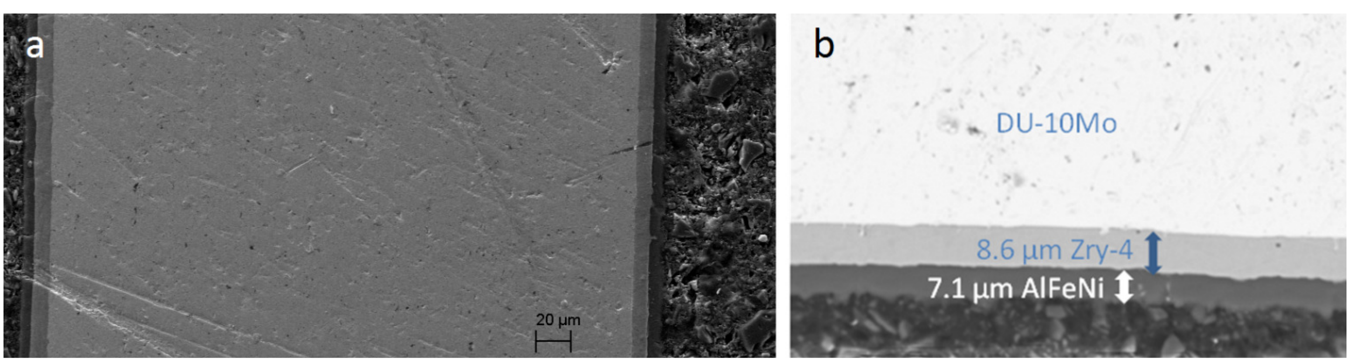

Figure 3: (a) Cross section of an Agent X minifoil. (b) Edge of fuel zone with barrier layer.

5. Conclusion

It has been shown, that small and full size DU-10Mo foils can easily be coated with the barrier materials Zr respectively Zry-4 and Ti as well as the cladding materials Al and AlFeNi in a thickness of 8-10 microns. The quality of the produced coatings and their adhesion to the substrate is excellent. Further examinations are currently performed to verify this. Much thicker deposits with identical quality were fabricated in the same sputter coating setup on other substrates before [2][3][4], therefore we expect thicker coatings could easily be realized on full size foils if required. Similar results are expected for coatings of Nb and Ta, which were successfully tested on different substrates as well [4], and will be the next to test on full size DU-10Mo foils. Coatings of other relevant materials, as UO_2, Si, ZrN and Mo, as well as layers or coatings of neutron poisons like Cd or B (respectively chemical compounds of these materials) and even coatings of U-Mo alloys to reach gradients in the Mo content on the surface of the U-Mo fuel core seem feasible as well, but haven´t been tested yet.
It has been shown, that coatings of two layers on top of each other can successfully be realized on full size foils by sputter coating. The examples that have been fabricated were layers of 8-10 microns of AlFeNi on a DU-10Mo foil coated with Zry-4 barrier as well as 8-10 microns of Al on top of a DU-10Mo foil coated with Ti barrier. The additional coating, the so-called pre-cladding, is intended to simplify bonding of barrier coated fuel foils and cladding. Furthermore, we expect the pre-cladding to work as an oxidation preventive barrier that could simplify the handling of barrier coated foils in air. Roll bonding tests by AREVA-CERCA will give information, whether both effects can actually be verified, and by that, if a pre-cladding layer is useful or not.
We are convinced, that sputter coating is a simple method to apply diffusion preventive barriers and other functional coatings onto fuel foils. An up-scaling of the process from laboratory scale to industrial level seems feasible and promising.

6. Acknowledgements

This work was supported by a combined grant (FRM0911) from the Bundesministerium für Bildung und Forschung (BMBF) and the Bayerisches Staatsministerium für Wissenschaft, Forschung und Kunst (StMWFK). The DU-10Mo full size foils and minifoils were provided by courtesy of Y-12 National Security Complex.

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