

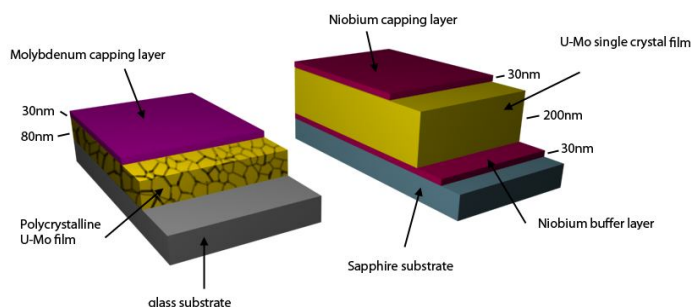
# URALLOY

## Next generation nuclear fuel materials

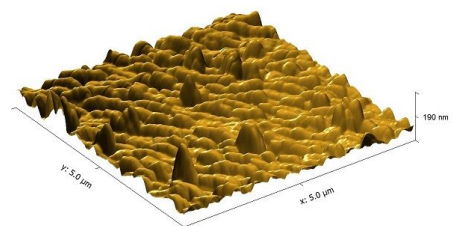
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Energy demand is continuing to rise and, with worldwide initiatives to reduce carbon dioxide emissions and find reliable energy sources that can replace fossil fuels while producing sufficient energy, nuclear energy is coming more and more to the forefront. Nuclear power is becoming more reliable and cost effective as the technology further matures, yielding improvements in its safety and efficiency. In addition, nuclear power plants are CO<sub>2</sub> emission-free sources of electricity. The 151 nuclear reactors operating within the EU provide over 30% of our electricity needs and avoid the emission of 700 million tonnes of CO<sub>2</sub> per year. However, in the wake of the Fukushima Daiichi nuclear power plant incident, there is an increased emphasis on developing improved and inherently safe materials for fuels, cladding and structural materials. The preferred choice of nuclear fuel materials is limited not only by the characteristics of the reactor cores in which they are deployed but also by their subsequent behaviour as a waste-form. Whilst early fission reactors used natural uranium metal as the fuel material, uranium dioxide (UO<sub>2</sub>) quickly became the predominant nuclear fuel in later reactors and remains so today. UO<sub>2</sub> is corrosion resistant but it will slowly dissolve in water when stored for long period of time. In addition, its thermal performance in a reactor is relatively poor as a result of its very low thermal conductivity. Metal fuels offer a significant improvement in thermal performance, but historical data tells us that when pure uranium metal also has its performance limitations. When heated up in a reactor core U metal will expand. Unfortunately this expansion is deleterious because its thermal expansion coefficient is different in different crystal directions, leading to substantial swelling and degradation. However, we can avoid this phenomenon whilst retaining the high thermal conductivity by stabilizing a different phase-structure of uranium, which has far more homogenous thermal expansion properties. This beneficial alteration can be achieved via alloying with a small amount of another metal and has the added benefit of imbuing stainless properties on the alloy. Certain alloys have already been deployed in test reactors e.g. KURRI, Japan, and are viable candidates for Gen(IV) reactor fuels.

In the present project we have tested uranium alloying with a range of transition metals, such as molybdenum, zirconium and tantalum. This has been achieved using an entirely novel and inherently safe approach - **thin film synthesis** via DC magnetron sputtering. Using this process thin film samples of many different binary alloy combinations can be prepared as polycrystals (materials consisting of many grains of varying size and orientation) or single crystals (monocrystalline solid grown in one, particular, direction with no grain boundaries or defects), depending on the type of substrate used; see Fig. 1. In order to avoid oxidation/contamination of the film a capping layer can be used for protection. The purity and pristine flatness of the thin film samples provide a simplified model surface (Fig. 2), which can subsequently be exposed to different environmental conditions during characterisation testing. The main objective of the current project was to investigate the oxidation and corrosion properties of possible next generation uranium-alloy fuel materials. In particular, we were interested in the performance of such materials in interim storage and under expected disposal conditions.



**Fig. 1.** Comparison of polycrystalline (left) and single-crystal (right) U-Mo alloy thin films [1].



**Fig. 2.** Surface topography of U-Zr alloy thin film [2].

The project successfully synthesised both, polycrystalline and single-crystal U-Mo, U-Zr and U-Zr thin film samples. In the case of U-Mo system, relatively low molybdenum content allowed to stabilize the single high-temperature uranium phase. The U-Zr system exhibited mixture of low- and high-temperature uranium phases. The reason for this may be related to the insufficiently high temperature during the fabrication process or the Zr alloying concentrations for this system differ from those identified for bulk alloy samples. The U-Ta system does not tend to form an alloy at high temperatures. Different analytical techniques, employing photons and electrons, were used to probe the crystal structure (a unique arrangement of atoms in a crystalline solid), the microstructure (including grain orientations) and surface composition and properties.

This work is of major interest to the nuclear industry, in light of the current plans to build new high-temperature nuclear reactors based on next generation fuel materials. The current work has developed a significant capability for the synthesis and subsequent experimentation on uranium alloy materials. The novel thin-film approach has been demonstrated as a viable route for synthesising samples which have sufficient pedigree and quality to be ideal surfaces for fundamental corrosion studies. The fact that the samples have such low mass and associated radioactivity, means that the thin-film approach to actinide materials science makes it relatively easy for academics at almost any other EU institution to access, transport and experiment upon these nuclear materials without the need for designated infrastructure. This is an incredibly valuable outcome for the project and already we have used this capability to grow and disseminate thin-film samples to the Universities of Cambridge, Manchester and Liverpool.

We must also recognise that this study is a pilot for further study of the synthesis and corrosion behaviour of uranium alloy fuel materials. This subject remains highly important for the next generation of nuclear reactors and hence for the population of the EU. As part of an evidence-based cost-benefit analysis for selecting next-generation nuclear fuel materials there needs to be an associated waste storage strategy for each fuel type, which takes into account the physiochemical stability of the arising waste in numerous storage and disposal settings. Further work will continue to be led by Bristol (T B Scott and A M Adamska) as part of (i) the Diamond Light Source 'Long-Duration Experiment' programme starting in October 2014 and running for an initial 2 year period funded by the STFC and (ii) The EPSRC-funded PACIFIC programme devoted to research on synthesis and performance of nuclear fuel materials. It is expected that numerous further publications will arise from the work completed in the present fellowship and from the upcoming work on these two aforementioned projects.

## References

- [1] A M Adamska, R Springell, T B Scott, Characterization of poly- and single-crystal uranium-molybdenum thin films, *Thin Solid Films* 550 (2014) 319-325.
- [2] A M Adamska, R Springell, A Warren, L Pico, O Payton, T B Scott, Growth and characterization of uranium-zirconium alloy thin films, accepted for publication in *J. Phys. D: Appl. Phys.* on 12<sup>th</sup> of June 2014, Ref: JPhysD-101676.R1.