

**INFLUENCE OF FILM FORMATION ON  
THE ANODIC DISSOLUTION OF  
NUCLEAR FUEL (UO<sub>2</sub>)**

Bill Santos\*, Dave Shoemsmith, Jamie Noël.  
Department of Chemistry,  
The University of Western Ontario,  
London, Ontario, Canada N6A 5B7  
Email: [bsantos@uwo.ca](mailto:bsantos@uwo.ca)

**INTRODUCTION**

The Canadian Nuclear Fuel Waste Program has developed a geological disposal scenario for fuel wastes resulting from the operation of nuclear generating stations. The fuel waste consists of CANDU (CANada Deuterium Uranium) fuel bundles sealed in a corrosion resistant metallic container, and placed in a disposal vault in granite rock. Models to predict the behavior of used nuclear fuel under permanent waste disposal conditions must take into account the long term evolution in the condition of the fuel. Since corrosion will be occurring into neutral ground waters in which UO<sub>2</sub><sup>2+</sup> has a low solubility, corrosion product deposits will accumulate on the fuel surface. We are using a combination of electrochemical and surface analytical techniques to investigate film formation processes on UO<sub>2</sub> surfaces.

**EXPERIMENTAL**

The electrodes used in this study were 1.5at% SIMFUEL, natural UO<sub>2</sub> pellets doped with non-radioactive elements. Films were grown under constant potential conditions, for 36 hours in 0.1 mol/L NaCl (pH=9.5). Subsequently the electrode was either removed for surface analysis (XPS) or the potential was scanned back to -1.2V at 10 mV.s<sup>-1</sup> and the cathodic stripping voltammogram (CSV) recorded.

**RESULTS AND DISCUSSION**

Figure 1 shows the log I - log t plots recorded potentiostatically over the range 100 mV to 500 mV (vs SCE). Behaviour can be divided into three distinct regions:

(a) Region 1: the slope of log I - log t (~ -1) is consistent with an initial film growth process:



(b) Region 2: a shoulder in the plots occurs earlier at higher currents, suggesting solution saturation leading to deposition of a corrosion product:



(c) Region 3: rapid dissolution at localized surface sites within the corrosion product leads to local acidity due to hydrolysis:



At the lowered pH, the UO<sub>3</sub> · 2 H<sub>2</sub>O corrosion product deposit is partially dissolved, allowing a higher UO<sub>2</sub> dissolution current. These claims were subsequently investigated by CSV or SEM/XPS. Figure 2 shows a series of SEM photomicrographs, showing a transition from minor surface oxidation at 200 mV to extensive pitting at 500 mV.

**CONCLUSION**

In summary, the formation of oxide films (UO<sub>2.33</sub>, UO<sub>3</sub>·2H<sub>2</sub>O) suppresses oxidation and dissolution up to ~ 300 mV. Also oxidation at higher potentials leads to local under-deposit acidity.

There was no evidence of grain boundary attack consistent with the absence of non-stoichiometry in the SIMFUEL.

**ACKNOWLEDGEMENTS**

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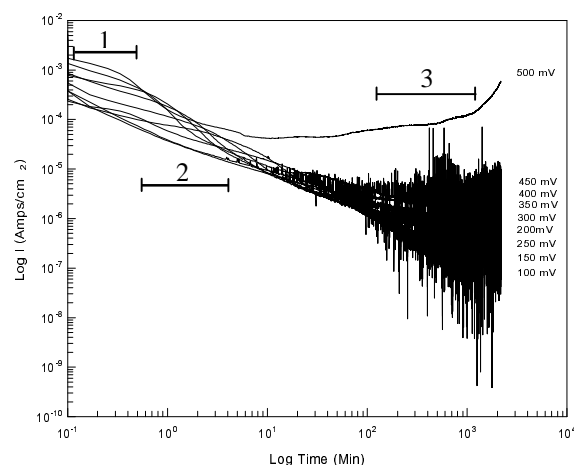


Fig.1: Plot of log current as a function of log time for anodic oxidation at various potentials from 100 to 500mV in 0.1 mol·L<sup>-1</sup> NaCl (pH = 9.5).

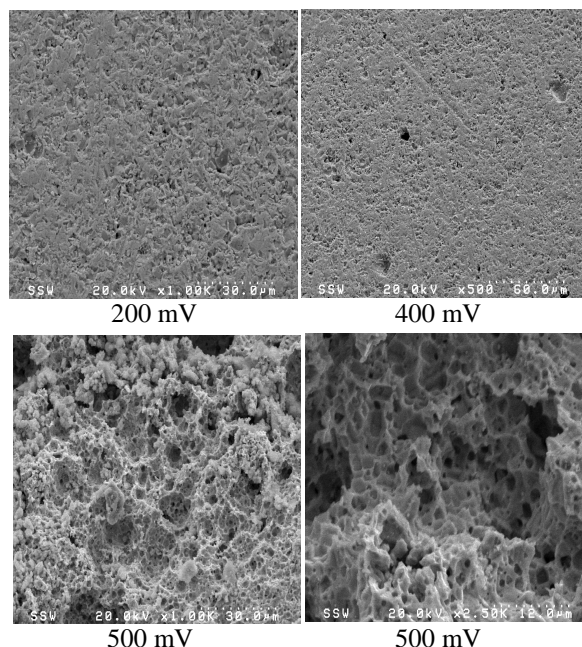


Fig. 2: SEM images of a 1.5at% SIMFUEL electrode after various anodic oxidations for 36 hours.