Surface science lab station for in-situ corrosion studies of actinide model systems

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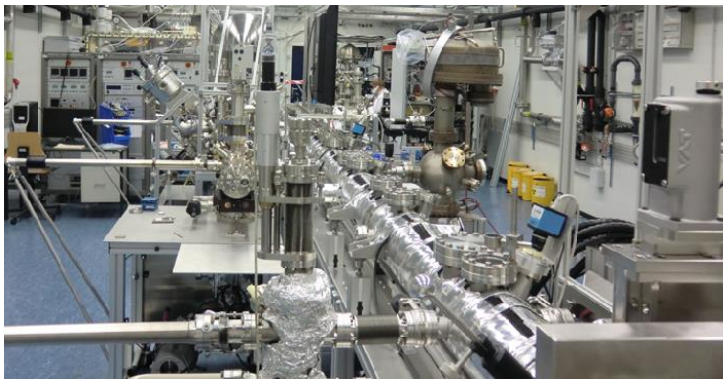
Abstract

Surface reactions play an important role in the handling of nuclear fuel. In particular, knowledge of the reaction mechanisms is important for prediction of the fuel behaviour under special conditions and under long term storage. One promising route for establishing the reaction mechanisms on the complex fuel is to replace it by simplified model systems. These allow addressing part-aspects of the overall process in single effect studies. In our approach, the fuel matrix is represented by sputter deposited thin films of actinide oxides or simulants for these (UO_2, ThO_2, PuO_2, CeO_2, mixed oxides, etc.) into which fission products, minor actinides, cladding or waste container materials are incorporated by co-deposition. The surface reactivity of these systems is studied in a two-step process.

- Single surface-gas interactions are investigated by studying the gas adsorption under UHV. While being different from real world corrosion conditions, the reaction parameters can be closely controlled (gas dosage, mixing, temperature…).
- Performing plasma reaction allows reaching high reaction stages (e.g. high oxides) and simulating the effect or radiolysis products on spent nuclear fuel.

The contribution presents the Surface Science Lab Station (Fig. 1) installed at JRC- Karlsruhe which is dedicated to such studies. The set-up integrates sample preparation, analysis and surface reactions in one single, well controlled environment to ensure unaltered, representative surfaces (avoiding e.g. contact with lab atmosphere). The laboratory is available to external users and its flexible design enables it fulfilling a great number of different tasks.



It allows carrying out sample preparation, and reactivity studies under UHV and ambient pressure. Special emphasis is given to the combination of spectroscopy techniques (XPS, UPS, BIS, TPD, HREELS, Kelvin probe) probing the average surface. The system has a modular setup, where the various techniques are mounted in individual chambers, which can be connected to the station on demand. Separation into independent devices ensures robust operation of such complex facility.

As example, we will report on the study of surface oxidation and reduction of uranium by oxygen, hydrogen and water, present either as molecular gases or as atomic gases (generated by ECR plasma). Films are analysed in-situ by X-ray and Ultra-Violet Photoelectron Spectroscopy. Pure U oxidation states of +4,+5 and +6 are observed and they correspond to $U5f^2$, $U5f^1$ and $U5f^0$ configuration respectively [1]. The gradual oxidation of U under corrosive conditions can thus be followed in detail.

References

1 G. El Jamal, T. Gouder, R. Eloirdi, and M. Jonsson, J. Nucl. Mater. 2022 (560), 153504.