Deliverable D6.2 (45 months)  
- RECONS WP6 -

Contract No. FP7-212287  
ReCosy


Objectives

➢ Sound understanding of redox phenomena controlling the long-term release/retention of radionuclides in nuclear waste disposal.

➢ Providing tools to apply the results to Performance Assessment/Safety Case.
The mission of the JRC-ITU is to provide the scientific foundation for the protection of the European citizen against risks associated with the handling and storage of highly radioactive elements.

Classification: No restriction
Unit: E05
Action No:51102

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Joint Research Centre
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Deliverable No. D6.2

NAME OF DELIVERABLE:
REPORT ON REDOX DRIVEN SPENT FUEL DISSOLUTION THROUGH GALVANIC COUPLING
REDOX PHENOMENA CONTROLLING SYSTEMS
ReCosy

COLLABORATIVE PROJECT (CP)

Submitting organizations: ITU, KTH
Due date of deliverable: 45 Project Months
Actual submission: 47 Project Months

Grant agreement N°.: FP7-212287

Start date of the project: 01 April 2008
Duration: 48 months

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Project co-funded by the European Commission under the Seventh Framework Programme of the European Atomic Energy Community (Euratom) for nuclear research and training activities (2007 to 2011)
Objectives

The objectives for project months 37 to 45 of the ReCosy project on redox driven spent fuel dissolution through galvanic coupling were the following. Galvanic coupling experiments on UO$_2$ and carbon steel under influence of hydrogen to clarify the uranium oxide reduction found in the experiments. It was also planned to finalise the studies on the surface properties of UO$_2$-Pd and UO$_2$-Mo thin film systems and on the effect of rare earth metal oxide doping on the redox reactivity of UO$_2$.

Status and results

Studies of the galvanic coupling of UO$_{2+x}$ with iron under influence of hydrogen have been carried out. Very low open circuit potentials have been measured under these conditions and consequently nearly no onset of corrosion was detected. Experiments with pre-corroded steel and uranium oxide layer show a heavy localised attack and a substantial release of uranium into the solution, but after 20 hours the remaining uranium oxide layer was identified as UO$_2$. Under all chosen experimental conditions a stabilization of the uranium oxide was found [1-4,12].

Conditions for formation of Mo particles in a UO$_2$ matrix were investigated. The refractory nature of Mo inhibits diffusion and agglomeration into particles at room temperature. High temperatures were selected, to enable Mo diffusion and particle formation, while still avoiding overlayer-substrate interdiffusion. The choice of appropriate substrate was important. The formation of U oxide in presence of reduced Mo particles was critical, because of the high oxygen affinity of Mo (compared e.g. to Pd) [1-6].

The studies on the effect of rare earth metal oxide doping on the redox reactivity of UO$_2$ lead to the following main findings [1-4,7-11]
- The dissolution yield in the reaction between H$_2$O$_2$ and UO$_2$ decreases with rare earth oxide doping.
- The catalytic decomposition of H$_2$O$_2$ on UO$_2$ initially produces hydroxyl radicals (adsorbed).
- The overall rate constant for the reaction between H$_2$O$_2$ and UO$_2$ is fairly insensitive to rare earth oxide doping.
- The rate constant for the catalytic decomposition of H$_2$O$_2$ on UO$_2$ is fairly insensitive to rare earth oxide doping.
- The redox reactivity of UO$_2$ pellets decrease significantly with upon rare earth oxide doping. This effect is more pronounced for weaker oxidants than for strong oxidants.

Dissemination

Publications, reports, or contributions in reports, proceedings:


Presentations:

Abstract
This report summarises the activities planned and performed in project months 37 - 45 in work package (WP) 6 of the ReCosy project. The main achievements in the fourth project year are given.
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