JRC Scientific and Technical Reports

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

Contract No. FP7-212287
ReCosy

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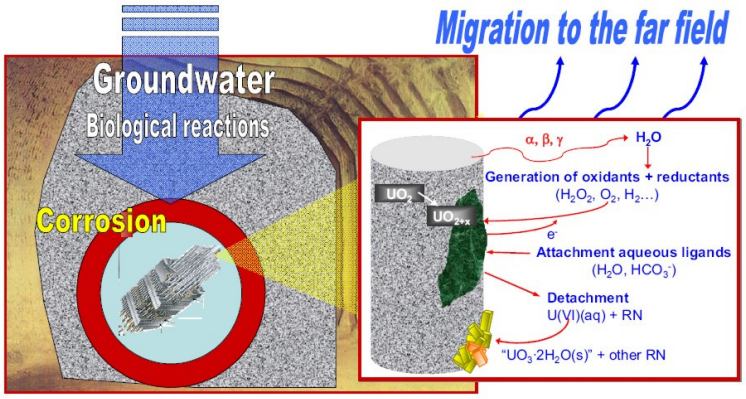
Objectives

- 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.



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Classification: No restriction
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Joint Research Centre
Institute for Transuranium Elements

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EURATOM


Reosy

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

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)		
Dissemination Level		
PU	Public	PU
RE	Restricted to a group specified by the partners of the project	
CO	Confidential, only for partners of the project	

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_2O_2 and UO_2 decreases with rare earth oxide doping.
-The catalytic decomposition of H_2O_2 on UO_2 initially produces hydroxyl radicals (adsorbed).
-The overall rate constant for the reaction between H_2O_2 and UO_2 is fairly insensitive to rare earth oxide doping.
-The rate constant for the catalytic decomposition of H_2O_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:
[1] G. Buckau, B. Kienzler, L. Duro, M. Grivé, V. Montoya (2009): Collaborative Project “Redox Phenomena Controlling Systems” - 1st Annual Workshop Proceedings, Forschungszentrum Karlsruhe, Wissenschaftliche Berichte, FZKA 7466, Karlsruhe 2009, Germany.

[2] G. Buckau, B. Kienzler, L. Duro, M. Grivé, V. Montoya (eds.) (2010): 2nd Annual Workshop Proceedings of the Collaborative Project “Redox Phenomena Controlling Systems” (EC 7th FP CP RECOSY), Karlsruhe Institute of Technology, KIT Scientific Reports 7557, Germany.

[3] M. Altmaier, B. Kienzler, L. Duro, M. Grivé, V. Montoya (eds.) (2011): 3rd Annual Workshop Proceedings of the Collaborative Project "Redox Phenomena Controlling Systems" (EC 7th FP CP RECOSY), Karlsruhe Institute of Technology, KIT Scientific Reports 7603, Germany.

[4] D.H. Wegen, P. Carbol, A. Seibert, T. Gouder, M. Jonsson, M. Trummer, A. Loida, B. Kienzler, D. Cui, K. Spahiu, D. Dobrev, A. Vokál, P. Brůha, R. Červinka (2011): REDOX PROCESSES AFFECTING THE SPENT FUEL SOURCE-TERM. Summary report WP6 in: Proc. RECOSY 4th ANNUAL MEETING, 23rd – 26th January 2012, Karlsruhe, Germany.

[5] T. Petersmann, A. Seibert, D.H. Wegen, T. Gouder, S. Stumpf, Th. Fanghänel: (2011): ELECTROCHEMICAL INVESTIGATIONS ON DOPED AND UNDOPED UO_2 SPENT FUEL MODEL SURFACES, RECOSY 3rd Annual Workshop Proceedings of the Collaborative Project "Redox Phenomena Controlling Systems" (EC 7th FP CP RECOSY), Karlsruhe Institute of Technology, KIT Scientific Reports 7603, Germany, pp. 231 – 240.

[6] T. Petersmann, T. Gouder, A. Seibert, Th. Fanghänel (2011): ELECTROCHEMICAL INVESTIGATIONS ON DOPED AND UNDOPED UO_2 SPENT FUEL MODEL SURFACES, RECOSY 3rd Annual Workshop Proceedings of the Collaborative Project "Redox Phenomena Controlling Systems" (EC 7th FP CP RECOSY), Karlsruhe Institute of Technology, KIT Scientific Reports 7603, Germany, p. 291.

[7] R. Pehrman, M. Trummer, C. Lousada, M. Jonsson (2011): REDOX REACTIVITY OF DOPED UO_2 - EFFECTS ON THE REACTIVITY TOWARDS H_2O_2, RECOSY 3rd Annual Workshop Proceedings of the Collaborative Project "Redox Phenomena Controlling Systems" (EC 7th FP CP RECOSY), Karlsruhe Institute of Technology, KIT Scientific Reports 7603, Germany, pp. 241 – 249.

[8] M. Trummer, B. Dahlgren and M. Jonsson (2010): ON THE DYNAMICS OF OXIDATIVE DISSOLUTION OF Y_2O_3 DOPED UO_2. J. Nucl. Mater. 407 (2010) 195-199

[9] C.M. Lousada, M. Trummer, M. Jonsson (2011): REACTIVITY OF H_2O_2 TOWARDS DIFFERENT UO_2-BASED MATERIALS. THE RELATIVE IMPACT OF RADIOLYSIS PRODUCTS REVISITED. J. Nucl. Mater. In press doi: 10.1016/j.jnucmat.2011.06.003

[10] R. Pehrman, M. Trummer, C.M. Lousada, M. Jonsson (2012): ON THE REDOX REACTIVITY OF DOPED UO_2 PELLETS – INFLUENCE OF DOPANTS ON THE H_2O_2 DECOMPOSITION MECHANISM. Submitted to J. Nucl. Mater.

[11] M. Trummer (2012): THE EFFECT OF SOLID STATE INCLUSIONS ON THE REACTIVITY OF UO_2. A KINETIC AND MECHANISTIC STUDY. Doctoral Thesis in Chemistry, KTH, Stockholm, Sweden

Presentations:

[12] D.H. Wegen, T. Gouder, A. Seibert (2012): STUDIES ON THIN FILM MODEL SYSTEMS AND ON UO_2 IN PRESENCE OF CORRODING IRON, RECOSY 4th Annual Workshop, 23-26.01.2012, Karlsruhe, Germany.


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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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