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DELIVERABLE (D-N°: 2.2) Status of Rim and Grain Boundary Diffusion Experiments (12 months)

FIRST-Nuclides

(Contract Number: FP7-295722)

D.H. Wegen, P. Carbol, H. Curtius,
J. Vandenborre

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Status of Rim and Grain Boundary Diffusion Experiments (12 months)

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Objectives

The second component "Rim and grain boundary diffusion" of work package 2 (WP2) deals with investigations on oxygen diffusion in spent UO_2 fuel. The examination of diffusion mechanisms will result in the quantification of water penetration into the fuel (grain and grain boundaries) structures and subsequently couple the diffusion/corrosion phenomena. Furthermore, investigations on irradiated and unirradiated fuel kernels separated from high temperature gas cooled reactor (HTR) fuel are planned which are complementary to those on light water reactor (LWR) fuel. The experimental part of WP2 started in project month 4 and will end in project month 33 [1], [4].

The JOINT RESEARCH CENTRE – INSTITUTE FOR TRANSURANIUM ELEMENTS (JRC-ITU) is the leading organization of WP2. The investigation of diffusion effects will start in the first project year with the characterisation and preparation of spent fuel samples, which will be used for corrosion experiments in $H_2{}^{18}O$ water at room temperature during the second project year. In the last year the $^{18}O/^{16}O$ depth profiles will be determined using a shielded SIMS (secondary ion mass spectrometry) to quantify the oxygen diffusion into SNF.

FORSCHUNGSZENTRUM JÜLICH GMBH (JÜLICH) is working on spent high temperature reactor fuel. Within the first half of the project the radionuclide inventory in the fuel kernel and in the coatings will be determined and compared to calculated values as well. Further on investigations of the microstructure and of the elemental distribution of the fuel kernel and of the coatings will be performed before (first half of the project) and after leaching (second half of the project). After cracking of the tight coatings the fission gas release fraction will be measured in the first 18 months. Then static leaching experiments with the separated fuel kernels and coatings will start in year two in order to determine the fast instant radionuclide release fraction.

Unirradiated tristructural-isotropic (TRISO) fuel particles are investigated by the CENTRE NATIONAL DE LA RECHERCHE SCIENTIFIQUE (CNRS) at the ARRONAX cyclotron. The particles are irradiated using a He^{2+}-beam in the dose rate range of 0 - 100 Gy/min. The corrosion of UO_2 TRISO particles is investigated in view of grain boundary effects and secondary phase formation and the influence of hydrogen. The experiments will start in the beginning of the second project year with studies on the role of grain boundaries, followed by investigations of fuel particle corrosion under hydrogen and in varying dose rates.

Status and results

After six month experimental work the outcome is coined by: preparatory work, testing of new experimental set-ups and, characterisation of materials and samples.

JRC-ITU has started the preparation of the oxygen/water diffusion study. The spent fuel was selected and the transfer of ownership is on-going. ITU's shielded SIMS has been prepared for measurements on spent fuel fragments (holder, procedures etc.). An amount of 400 g of ^{18}O-labelled water (>98 at.-% $H_2{}^{18}O$) was purchased. This was difficult because the global supply is limited and most of the produced ^{18}O is reserved for the production of ^{18}F, which is widely used in PET (positron emission tomography) [6].

JÜLICH has started to analyse the microstructure and elemental distribution before leaching as well as the radionuclide inventory in the fuel kernel and in the coatings. The surface of the fuel kernel shows large grains and the metallic precipitates appear as hexagonal platelets. The elements Cs, O, U, Mo, Xe, Zr, and Tc were identified whereas higher amounts of the volatile elements Cs and Xe were detected in the surrounding porous carbon layer (the so-called buffer). Furthermore, the elements Am, Pu, Cm, U, Eu, Ce, Sr, Tc, and Pr were found quantitatively within the kernel. On the other hand, about 95 % of the Cs-activity was found in the coatings [2], [7].

CNRS has started to set-up their analytical tools. The first *in situ* tests were carried out. Solid UO_2 samples were immersed in ultra pure water and irradiated at the ARRONAX facility with a He^{2+} beam. During irradiation first Raman spectra were measured [3], [8].

Dissemination

Publications, reports, or contributions in reports, proceedings:

[1] D.H. Wegen (2012). *FIRST-Nuclides 1st Annual Workshop - WP2 Summary Report - Contribution to WP2 of the collaborative project FIRST Nuclides*. JRC Scientific and Policy Reports, JRC76116, European Atomic Energy Community, Germany, 2012.

[2] H. Curtius, E. Müller, H.W. Müskes, M. Klinkenberg, D. Bosbach (2012). *HTR Spent Fuel - Microstructure and Radionuclide Inventory-*. 1st Annual Workshop Proceedings, 7th EC FP – FIRST-Nuclides, 9th-11th October 2012, Budapest, Hungary.

[3] J. Vandenborre, A. Traboulsi, G. Blain, J. Barbet, M. Fattahi (2012). *Radiolytic Corrosion of Grain Boundaries onto the UO_2 TRISO Particle Surface*. 1st Annual Workshop Proceedings, 7th EC FP – FIRST-Nuclides, 9th-11th October 2012, Budapest, Hungary.

Presentations:

[4] D.H. Wegen (2012). *WP2: Fission Gas Release & Rim and Grain Boundary Diffusion.* 1st Annual Workshop, 7th EC FP – FIRST-Nuclides, 9th-11th October 2012, Budapest, Hungary.

[5] D.H. Wegen, D. Papaioannou, W. de Weerd (2012). *Sampling and Measurement of Fission Gas from Spent Nuclear Fuel.* 1st Annual Workshop, 7th EC FP – FIRST-Nuclides, 9th-11th October 2012, Budapest, Hungary.

[6] P. Carbol, I. Marchetti (2012). *Oxygen and Water Diffusion into 42 GWd/tHM UO_2 Fuel under Reducing Conditions*. 1st Annual Workshop, 7th EC FP – FIRST-Nuclides, 9th-11th October 2012, Budapest, Hungary.

[7] H. Curtius (2012). *HTR Spent Fuel - Microstructure and Radionuclide Inventory-*. 1st Annual Workshop, 7th EC FP – FIRST-Nuclides, 9th-11th October 2012, Budapest, Hungary.

[8] J. Vandenborre et al. (2012). *Radiolytic Corrosion of Grain Boundaries onto the UO_2 TRISO Particle Surface: WP2- First In Situ Raman Tests under He^{2+} Irradiation.* 1st Annual Workshop, 7th EC FP – FIRST-Nuclides, 9th-11th October 2012, Budapest, Hungary.

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Abstract: This report summarises the activities planned and performed in project months 1 - 12 by the beneficiaries collaborating in the component "*Rim and grain boundary diffusion*" of work package 2 (WP2) of the CP – FIRST-Nuclides project in 2012. The main achievements in the first project year are given.

The research leading to these results has received funding from the European Union's European Atomic Energy Community's (EURATOM) Seventh Framework Programme FP7/2007-2011 under grant agreement no. 295722 (FIRST-Nuclides project).