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Spent nuclear fuel experiments: Final results of dissolution experiments

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1 Introduction

The EURATOM H2020-project DisCo (*Modern spent nuclear fuel Dissolution in failed container Conditions*) dealt with dissolution of modern-type spent nuclear fuel (SNF) which contain additives (such as Al and Cr) as well as the dissolution of mixed-oxide fuel containing a certain amount of Pu (MOX fuel). The main goal of Work Package 3 of DisCo was to study experimentally the matrix dissolution of irradiated MOX fuels and irradiated UO_x fuels, containing Al and Cr as dopants, under relevant disposal conditions, i.e. strongly reducing conditions imposed by hydrogen overpressure. The research groups that contributed with SNF dissolution experiments were Fundacio CTM Centre Tecnologic – now Fundacio Eurecat (EURECAT) in conjunction with Universitat Politècnica de Catalunya (UPC), Joint Research Centre (JRC-KA), Karlsuher Institut für Technologie (KIT-INE) and Studsvik Nuclear AB (STUDSVIK).

Results of the SNF dissolution experiments gained in Work Package 3 (WP3) were published in detail by EURECAT, JRC-KA, KIT-INE and STUDSVIK in the Proceedings of the DisCo annual meetings in May 2019 (onsite meeting in Köln), April 2020 (online meeting) and October 2021 (hybride meeting, online and onsite in Barcelona). Digital data sheets with results of the respective experiments were provided to partner institutions of Work Package 5 of DisCo for input of their modelling studies. Furthermore, each WP3 partner institution intends to publish their outcomes in international scientific journals with peer-review. Since all details of the SNF experiments and their data are available in other publications, however dispersed to different literature sources, this report summarizes the principal findings of the SNF dissolution experiments of WP3 and deals with overarching results.

2 Spent UOx / MOX Fuel Samples and Characteristics of Dissolution Experiments

All partners of WP3 dealt with spent nuclear fuel (SNF), which had been irradiated in commercial light water nuclear reactors. Dissolution experiments under oxic, anoxic andstrongly reducing conditions were performed with irradiated standard UO_x fuel, MOX fuels and UO_x fuels doped with Cr and with Cr+Al, i.e. one standard and one doped BWR UO_x fuel with average burnups of 57 and 59 MWd·(kg $_{HM}$)$^{-1}$, resp., two PWR MOX fuels with average burnups of 38 and 54 MWd·(kg $_{HM}$)$^{-1}$, resp., one standard PWR UO_x fuel with an average burn-up of 73 MWd/kg$_{HM}$ and one doped PWR UOx fuel with an average burn-up of 58 MWd/kg$_{HM}$. At the Hot Cells and Shielded Box-Line of the laboratories in Karlsruhe and Nyköping, the irradiated fuel rods had been cut into segments and two types of fuel samples had been prepared: (a) fuel segments with their Zircaloy claddings, denoted as "cladded segments"; (b) fragments of the fuel matrix without any cladding material, denoted as "fragments". For most experiments, preparation of cladded segments and fragments from the fuel rods as well as preparation of the dissolution experiments were performed in the framework of Work Package 2 of DisCo. Samples of the two BWR UO_x fuels (Metz et al., 2012; Roth et al., 2016) and the PWR UO_x fuel of 73 MWd·(kg $_{HM}$)$^{-1}$ local burnup had been prepared already before the onset of DisCo. Details on sample characterisation and experimental set-ups are presented in the D2.1 report of WP2 (Farnan et al., 2021).

Characteristic data of the studied spent UOx fuel and spent MOX fuel samples (average and local burn-ups, fission gas release measured in puncturing tests), solution types and gas atmospheres of dissolution experiments are reported in Table 1. Most dissolution experiments were conducted in presence of hydrogen or argon/hydrogen atmosphere under strongly reducing conditions. Moreover, four dissolution experiments with a standard PWR UO_x fuel and MOX fuel had been conducted in presence of argon (anoxic conditions) or air (oxic conditions). Except two experiments, in all experiments, fragments and cladded segments of these irradiated fuel samples were exposed to diluted NaCl solution with 1 to 10 mM $NaHCO_3$ (pH ~ 8, denoted as NaCl-BIC). In two other experiments, a standard PWR UO_x fuel was exposed to so-called "Young Cement Water with Calcium" (denoted as YCWCa), which is a hyper-alkaline diluted NaOH, $Ca(OH)_2$ solution, containing 77 mM Na_2CO_3 and other minor constituents.

sample name and authors of expt.	fuel type	reactor type	average burn-up [GWd/t]	linear power [W/cm]	fission gas release in puncturing	sample type	solution	gas atmo-sphere
MOX-H (Serrano Purroy et al.)	MOX	PWR	54 (56 local)	180	2.5%	cladded segment	NaCl-BIC	Ar
MOX-L (Serrano Purroy et al.)	MOX	PWR	54 (48 local)	180	2.5%	cladded segment	NaCl-BIC	Ar
KWU11.38-5810 (Herm et al.)	MOX	PWR	38	200	6.0%	fragments of pellet	NaCl-BIC	H2 / Ar
KWU11.38-5810 (Herm et al.)	MOX	PWR	38	200	6.0%	cladded segment	NaCl-BIC	H2 / Ar
AC-73BIC (Kokinda et al.)	standard UOx	PWR	60 (73 local)	255	13.6%	cladded segment	NaCl-BIC	H2 / Ar
AC-73YCWCa (Kokinda et al.)	standard UOx	PWR	60 (73 local)	255	13.6%	cladded segment	YCWCa	H2 / Ar
5A2 (Barreiro Fidalgo et al.)	standard UOx	BWR	57 (66 local)	n.g.	2.5%	fragments of pellet	NaCl-BIC	H2
C1 (Barreiro Fidalgo et al.)	Al-Cr-doped UOx	BWR	59 (63 local)	n.g.	1.4%	fragments of pellet	NaCl-BIC	H2
AC5-Cr (Carbol et al.)	Cr-doped UOx	PWR	58	n.g.	n.g.	fragments of pellet	NaCl-BIC	H2
73BIC (Kokinda et al.)	standard UOx	PWR	60 (73 local)	255	13.6%	cladded segment	NaCl-BIC	**air**
73YCWCa (Kokinda et al.)	standard UOx	PWR	60 (73 local)	255	13.6%	cladded segment	YCWCa	**air**

***Table 1**: Characteristic data of studied spent UOx fuel and spent MOX fuel samples, solution types and gas atmospheres of dissolution experiments. In addition, names of the lead authors of the experimental working teams are given. "n.g." denotes not given data.*

3 Main Results of Spent UOx / MOX Fuel Dissolution Experiments

In the dissolution experiments with irradiated UOx and irradiated MOX fuel samples, aqueous concentrations of actinides (americium, curium, neptunium, plutonium and uranium) and fission products (barium, cesium, gadolinium, iodine, molybdenum, rubidium, strontium, technetium and zirconium) were analysed. Concentrations of these elements are given in the appendix of this report (Table A-1 and Table A-2) and in the contributions of EURECAT, JRC-KA, KIT-INE and STUDSVIK to the Proceedings of the DisCo annual meetings. Since radionuclide concentrations in experiment AC5-Cr were measured only in the first 24 days, these data are not presented in this report. The amount of xenon and krypton released into the gas phase were measured in four dissolution experiments with irradiated PWR MOX fuel (38 MWd/kg) and irradiated BWR UOx fuels (57 and 59 MWd/kg). Data on the fission gas release during fuel dissolution are not presented in this report. Yet they are presented in the respective contributions to the Proceedings of the 3rd and 4th DisCo annual meetings (Barreiro-Fidalgo et al., 2020; 2021; Herm et al., 2020; 2021).

For comparing the overarching results of the SNF dissolution experiments, aqueous concentrations of five elements are chosen:

- Uranium as major element of the matrix of dissolving spent UOx fuels and spent MOX fuels (Figure 1);
- plutonium as redox-sensitive element of the matrix (Figure 2);
- americium as non-redox-sensitive element of the matrix (Figure 3);
- strontium as non-redox-sensitive fission product with high solubility under the studied geochemical conditions (Figure 4);
- cesium as another non-redox-sensitive fission product with high solubility under the studied geochemical conditions (Figure 5).

Based on release pattern of Am, Cm, Np, Pu and U (Tab. A-1; Fig. 1, Fig. 2., Fig. 3) it is obvious that all SNF dissolution experiments are characterized by a considerable scatter in measured concentrations of these radioelements within the first year. In contrast to the actinides' release pattern, measured concentrations of non-redox sensitive fission products show less pronounced fluctuations in the initial stage of the experiments.

In SNF dissolution experiments under anoxic (MOX-H seg., MOX-L seg.) and oxic conditions (UOx-73BIC seg., UOx-73YCWCa seg.) uranium concentrations continuously increase. At the end of these experiments after 210 to 623 days, still a steep gradient of concentration versus time ($\Delta C/\Delta t$) is observed, and U concentrations in the anoxic and oxic experiments are orders of magnitude above those measured in experiments in presence of hydrogen (Fig. 1). In contrast to the observations in the oxic and anoxic experiments, aqueous U concentrations tend to approach a steady state level after one year in the experiments with hydrogen overpressure. In these experiments under strongly reducing conditions, measured U

concentration are progressing to approach to a so-called “concentration plateau” between 1 to $5 \cdot 10^{-8}$ mol/L in the long run (Fig. 1).

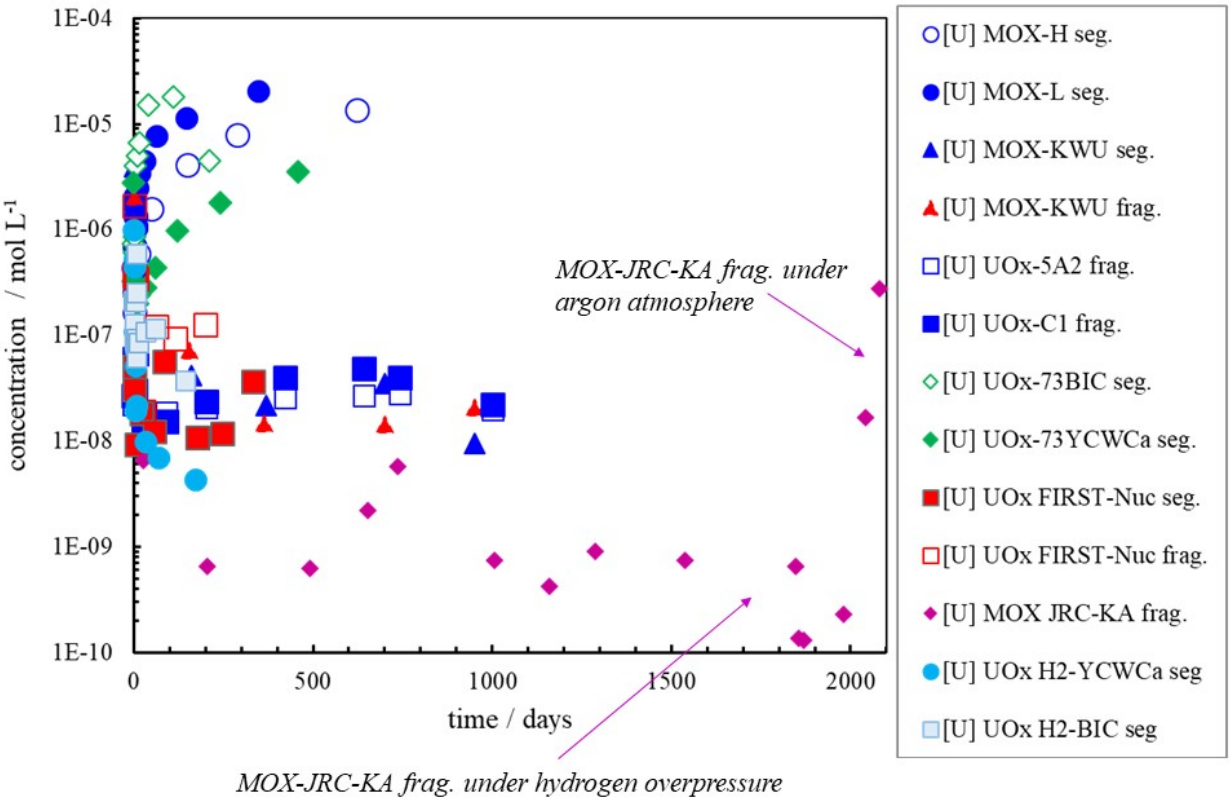

Figure 1: *Concentration of uranium released into solution versus time in dissolution experiments with irradiated UOx and MOX fuels.*

In most experiments under anoxic, oxic and reducing conditions, aqueous concentrations of Am, Cm, Np and Pu are relatively low in comparison to concentrations of U and concentrations of the major fission products Cs, I and Sr (Tab. A-1 and Tab. A-2; Fig. 2 and Fig. 3). The highest Am and Pu concentrations are observed in two experiments under oxic and anoxic conditions (MOX-L seg. and UOx-73BIC seg.). Surprisingly, relatively high Am and Pu concentrations are measured in the dissolution experiments with the two BWR UOx fuels (UOx-5A2 frag. and UOX-C1 frag.). Except experiments UOx-5A2 frag. and UOX-C1 frag., dissolution experiments under strongly reducing conditions display lower Am and Pu concentrations (i.e. below $4 \cdot 10^{-9}$ mol/L) than Am and Pu concentrations under oxic and anoxic conditions. In contrast to considerable difference in U concentrations between reducing and anoxic/oxic experiments, the differences in Am and Pu concentrations are less influenced by the redox conditions.

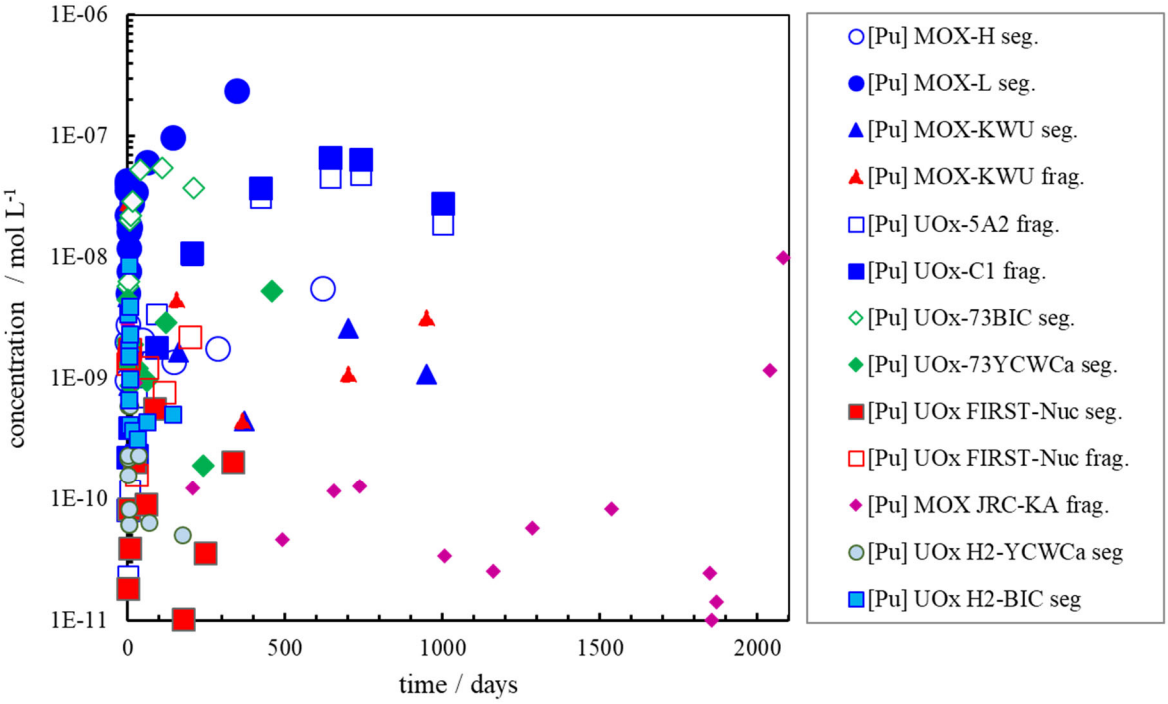

Figure 2: *Concentration of plutonium released into solution versus time in dissolution experiments with irradiated UOx and MOX fuels.*

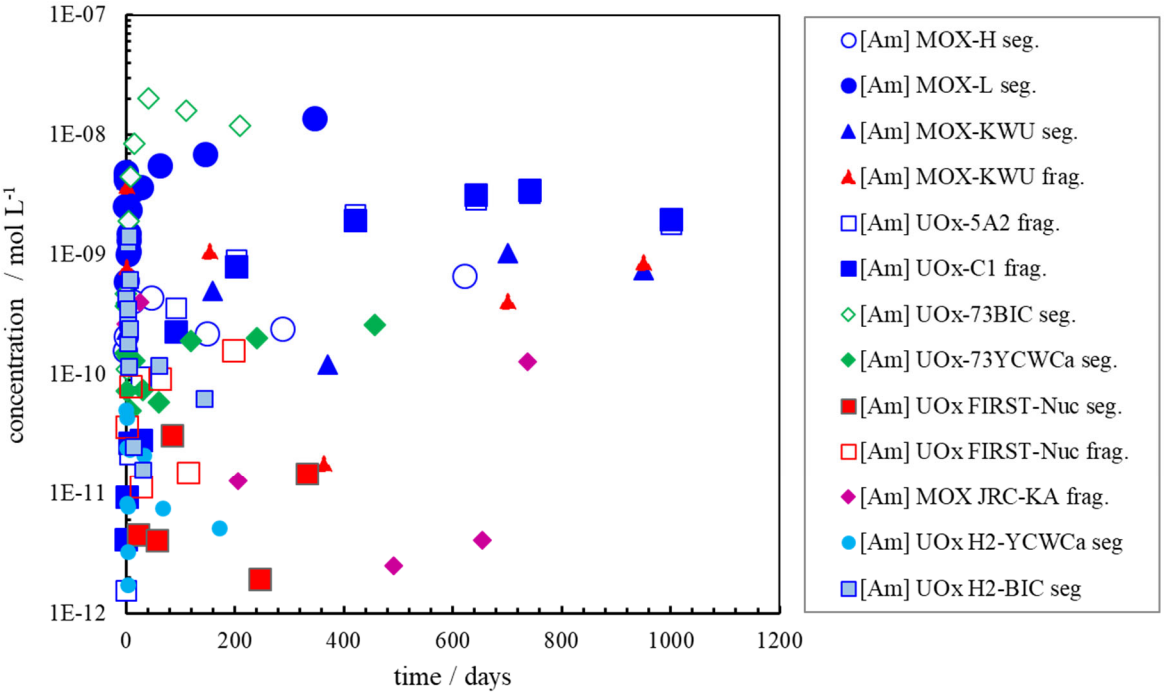

Figure 3: *Concentration of americium released into solution versus time in dissolution experiments with irradiated UOx and MOX fuels.*

In all experiments, release patterns of fission products differ significantly from the release patterns of the actinides. For this summary, measured concentrations of cesium and strontium are selected because they are prominent fission products and have been measured in most experiments of WP3. Aqueous concentrations of both elements increase throughout all dissolution experiments except the last samplings of experiments UOx-5A2 frag., UOx-C1 frag. and MOX-KWU seg. (Fig. 4 and Fig. 5). The last two samplings of UOx-C1 frag. have virtually the same Sr concentration, and Cs concentrations in the last two samplings of experiments UOx-5A2 frag., UOx-C1 frag. and MOX-KWU seg. are about the same, too. The general trend of the SNF dissolution experiments is characterized by fast Cs and Sr release rates (in terms of $\Delta C/\Delta t$) in the initial stages of the experiments. Thereafter, a drastic slow down of the release rates is observed in the long term of the experiments with hydrogen.

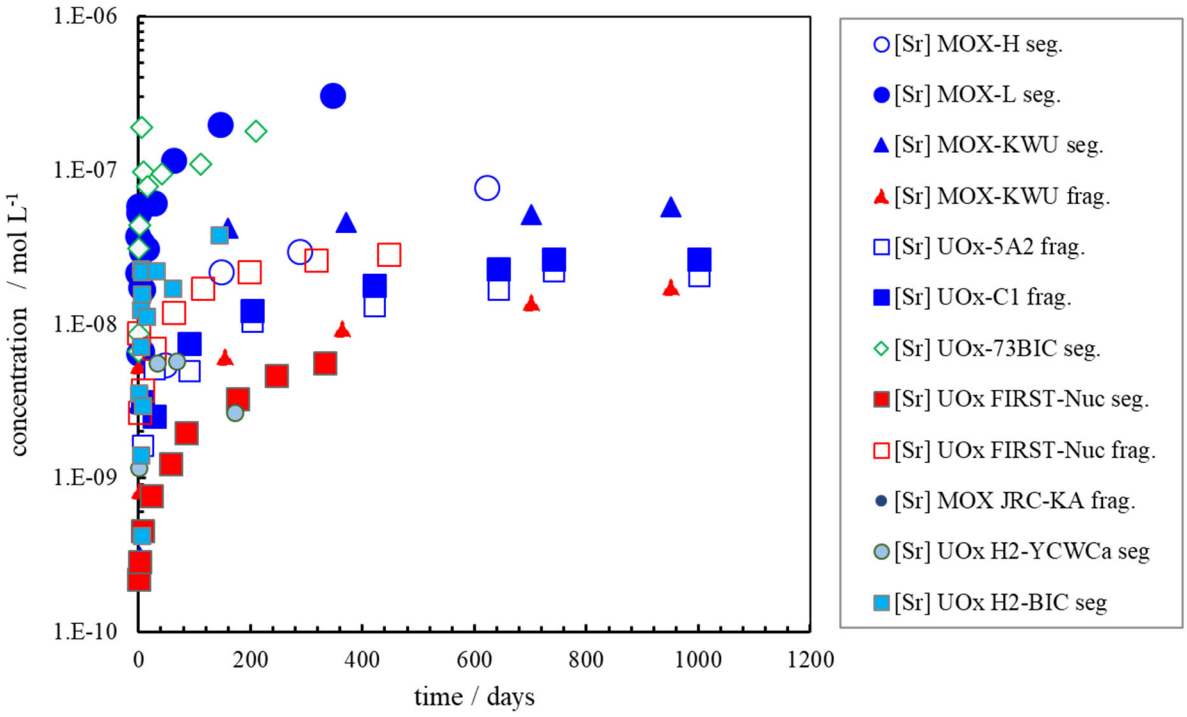

***Figure 4**: Concentration of strontium released into solution versus time in dissolution experiments with irradiated UOx and MOX fuels.*

Sr release patterns measured in experiments under oxic / anoxic conditions differ distinctly from those of experiments under strongly reducing conditions (Fig. 4). After an initial stage of some months, experiment MOX-L seg. (anoxic conditions) and experiment UOx-73BIC seg. (oxic conditions) show Sr concentrations continuously increasing above 10^{-7} mol/L with relatively steep concentration vs. time gradients ($\Delta C/\Delta t$). Sr concentration in the last sampling of experiment MOX-H seg. (anoxic condition) is slightly lower ($8 \cdot 10^{-8}$ mol/L) than concentrations in the two other experiments under oxic / anoxic conditions. Nevertheless, in MOX-H seg. the Sr release is characterized by a steep $\Delta C/\Delta t$ gradient, too. In contrast, all SNF dissolution experiments conducted under hydrogen overpressure display relatively low Sr concentrations, approaching towards "concentration plateau" between 1 to $6 \cdot 10^{-8}$ mol/L in the long run. Unlike the Sr release pattern in experiment MOX-H seg. and the other

anoxic/oxic experiments, Sr concentration vs. time gradients (ΔC/Δt) in the experiments under hydrogen are characterized by quite low values of about 10^{-11} mol/(L·d).

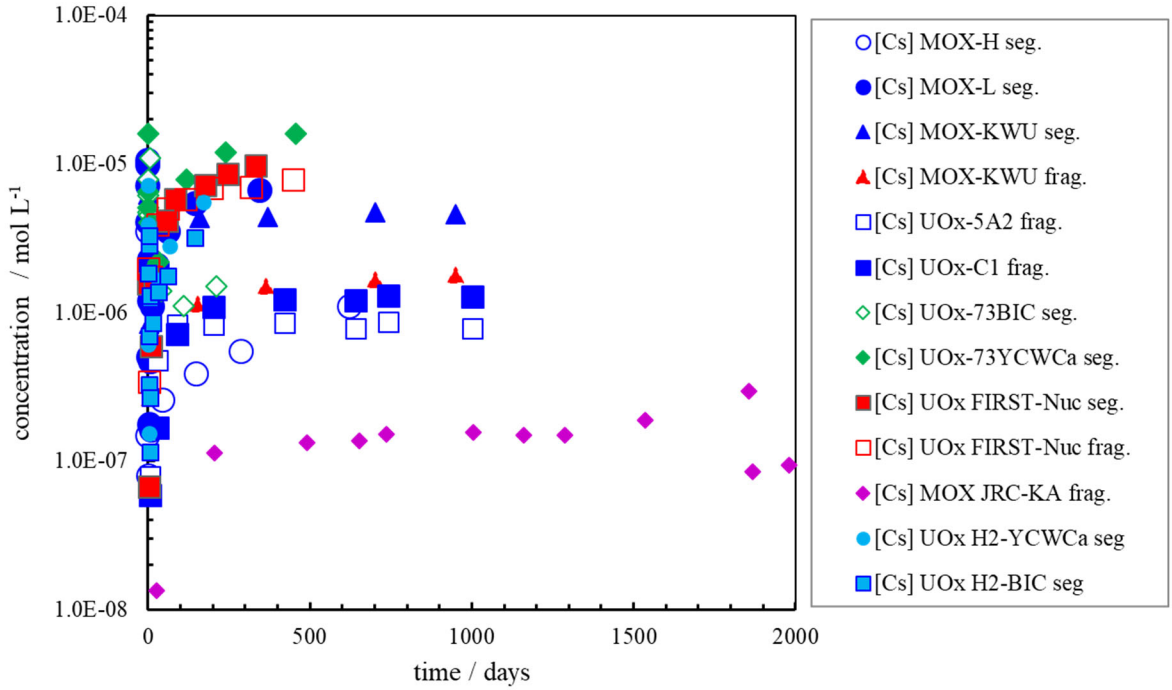

***Figure 5**: Concentration of cesium released into solution versus time in dissolution experiments with irradiated UOx and MOX fuels.*

Generally, aqueous concentrations and release rates of Cs are significantly higher than the respective values of Sr. In experiments with a duration longer than 500 days, Cs approaching towards various "concentration plateaus", where different concentration levels are observed (Fig. 5). Similar to Sr, release rates of Cs in terms of ΔC/Δt are characterized by quite low values in the range of 10^{-11} to 10^{-10} mol/(L·d) after about two years. However, the different levels of the Cs "concentration plateaus" are apparently not (mainly) controlled by the presence of hydrogen. The Cs "concentration plateau" of experiment MOX-KWU seg. (performed with a cladded segment) is considerably higher than that of MOX-KWU frag. (performed with fragments), although both experiments were conducted with the same solution and at the same hydrogen overperessure. Cs concentrations and release rates (ΔC/Δt) of experiments under oxic and anoxic conditions (MOX-H seg., MOX-L seg., UOx-73BIC seg.) are virtually the same as Cs concentrations and release rates measured in experiments UOX FIRST-Nuc seg. and UOX FIRST-Nuc frag. with hydrogen overpressure (Fig. 5); the latter were performed in the collaborative EURATOM project FIRST-Nuclides (Kienzler et al., 2016; Lemmens et al. 2017).

4 Discussion and Conclusions

The measured radionuclide concentrations (Tab. A-1 and Tab. A-2) together with the characteristic data of the irradiated UOx and MOX fuels used in the SNF dissolution experiments of the DisCo project provides a very informative data-set with respect to radionuclide release both from the matrices of the irradiated fuels and from the so-called "instant release fraction". However, interpretation of features of the "instant release fraction" is out of scope of this workpackage. Here, the focus is set on the understanding of SNF matrix dissolution in the strongly reducing conditions representative of repository environments. Results of the DisCo dissolution experiments are compared to radionuclide concentrations measured in three SNF dissolution experiments conducted in presence of hydrogen overpressure, i.e. an experiment with irradiated MOX fragments (Carbol et al., 2009; denoted as MOX JRC-KA frag.) and two experiments with a cladded segment and fragments of an irradiated high-burnup UOx fuel (Kienzler et al., 2016; Lemmens et al. 2017; denoted as UOx FIRST-Nuc seg. and UOx FIRST-Nuc frag., respectively). In the foreseen publications of the WP3 partners in international scientific journals, results of the WP3 experiments will be compared to a more comprehensive data-set of results from SNF dissolution experiments with hydrogen overpressure.

In the initial stage of all studied SNF dissolution experiments, a similar release behaviour of uranium is observed. In the first samplings relatively high U concentrations were measured, which scatter over a wide concentration range, followed by a decrease in uranium concentration approaching a concentration level of about $2 \cdot 10^{-8}$ mol L^{-1} in the experiments in presence of hydrogen overpressure (Fig. 1). The observed initial scatter of U concentrations in solution and the significant decrease of U in the following about twelve months is a common observation in SNF dissolution experiments under disposal conditions (reducing conditions, near neutral pH to (weakly) alkaline conditions). Often, the initial scatter of U concentration - and the initial scatter in concentrations of other matrix bound elements - is related to dissolution of pre-oxidized surfaces of the used spent nuclear fuel samples. As a consequence, the interpretation of the radionuclide release from dissolving SNF matrix is based mainly on the outcomes of long-term dissolution experiments, running for at least one year.

As mentioned in the previous chapter, measured U concentration approach towards a so-called "concentration plateau" between 1 to $5 \cdot 10^{-8}$ mol/L in the long run of the SNF dissolution experiments in presence of hydrogen overpressure. Between one and two years of dissolution, the U release rates slow down to quite low levels. When considering the last three samplings of experiments MOX-KWU frag., UOx-5A2 frag. and UOx-C1 frag., one may argue that the net uranium release rate is close to zero. Since the uranium "concentration plateau" is distinctly above the solubility of UO_2(am, hyd) (defined by the reaction UO_2(am, hyd) + $2H_2O$ (l) $\leftrightarrow$ $U(OH)_4$(aq)), and the ambiguity whether the U release rate has approached to zero, it is concluded that dissolution of the UO_2 matrix of the studied spent nuclear fuel samples continues even in presence of hydrogen overpressure, though the dissolution rate slows down drastically with time. With other words, an equilibrium state between spent nuclear fuel and the aqueous solution, where dissolution rate equals precipitation rate, has not been achieved within the duration of the experiments.

Nevertheless, the experiments of DisCo WP3 together with the data of experiments MOX JRC-KA frag. (Carbol et al., 2009), UOx FIRST-Nuc seg. and UOx FIRST-Nuc frag. (Kienzler et al., 2016; Lemmens et al. 2017) demonstrate clearly a strong hydrogen inhibition effect on the SNF matrix dissolution. Generally, long-term U concentrations in the SNF dissolution experiments under hydrogen overpressure are orders of magnitude below those in experiments under anoxic or oxic conditions (Fig. 1). Whereas the U release rates are quite slow in experiments with hydrogen overpressure ($\Delta C/\Delta t \approx 10^{-11}$ mol/(L·d)), experiments under oxic and anoxic conditions display relatively steep concentration vs. time gradient in the time interval of one to two years. Similarly, the direct comparison of experiments UOx H2-YCWCa seg. and UOx H2-BIC seg. (conducted in presence of hydrogen overpressure; U $< 4 \cdot 10^{-8}$ mol/L after five months) with experiments UOx-73YCWCa seg. and UOx-73BIC seg. (conducted under oxic conditions; U $> 1 \cdot 10^{-6}$ mol/L after five months) demonstrates the inhibition of uranium release by hydrogen.

Am is a non-redox sensitive actinide, whereas Pu is a redox-sensitive actinide. Notably, the predominance field of aqueous Pu(IV) species / $Pu(OH)_4$(am) is much wider than the predominance field of aqueous U(IV) species / UO_2(am, hyd). Therefore, it is not expected that an effect of hydrogen is considerably reflected (with a huge difference between oxic/anoxic versus strongly reducing experiments) by the Am and Pu release patterns. Still, relatively high Am and Pu concentrations are observed in experiments MOX-L seg. and UOx-73BIC seg., which were conducted under anoxic and oxic conditions, respectively (Fig. 2 and Fig. 3). The general trend of the Pu and Am release data in experiments under hydrogen overpressure indicate that aqueous concentrations of these actinides tend towards concentration plateaus at levels of PuO_2(am, hyd and $Am(OH)_3$(s) solubility levels (log [Pu]tot = –(9 ± 1) and log [Am]tot = –(9 ± 1), respectively).

Aqueous concentrations and release rates of Cs are significantly higher than the respective values of Sr (Fig. 4 and Fig. 5). The observed differences in Cs and Sr release rates decrease in the long run of the experiments. The relatively fast Cs release, in particular in the first two years of the experiments, is related to the so-called “instant release fraction” of cesium. The relatively slow Sr release in the long-term of experiments under hydrogen overpressure (with $\Delta C/\Delta t$ values around 10^{-11} mol/(L·d)) is related to release of strontium from the UO_2 matrix of the spent nuclear fuel samples. With exception of the last Sr measurements of experiment UOx-C1 frag. and the last measurements of experiments UOx-5A2 frag., UOx-C1 frag. and MOX-KWU seg., a continuously slow release of the two fission products is observed even after two years of exposure of the spent nuclear fuel samples to the aqueous solutions. Together with the level of the uranium “concentration plateau” above the UO_2(am, hyd) solubility limit, these observations indicate that dissolution of the UO_2 matrix of SNF continues even in the presence of hydrogen overpressure. Similar to release of uranium, the strontium release is strongly inhibited by hydrogen. As shown above, in the SNF dissolution experiments with hydrogen lower Sr concentrations and slower Sr release rates are observed compared to the experiments under oxic and anoxic conditions. This similarity in the dependence of the U and Sr release on hydrogen is interpreted as a indication of a strong

inhibition effect on the SNF matrix dissolution. In contrast, the Cs release appears to be more governed by fuel inherent properties rather than the presence of hydrogen.

Based on the release patterns of uranium, cesium and strontium in experiment UOx-C1 frag. with other experiments under hydrogen overpressure it is concluded that Al+Cr as dopants in the UOx fuel do not significantly change the release of U, Cs and Sr under these strongly reducing conditions. In particular the Cs release in experiment UOx-C1 frag. (with fragments of a Al+Cr doped UOx fuel) and in experiments UOx-5A2 frag. (with fragments of a "standard" UOx fuel) and MOX-KWU frag. (with fragments of a MOX fuel) are quite similar. A closer look to the uranium and strontium release data reveals that the release of these elements are to some extent higher in the experiment with Al+Cr doped UOx fuel compared to the release of this elements in experiments with non-doped UOx fuel and MOX fuel under hydrogen overpressure. There are no indications of a significant difference in the dissolution behaviour between irradiated MOX fuel and irradiated non-doped UOx fuel under strongly reducing conditions in NaCl-BIC solution.

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Appendix

Experiment / *sample*	time / days	[U]	[Pu]	[Np]	[Am]	[Cm]
MOX-H seg.	0.2	4.3E-07	2.0E-09	1.0E-10	1.6E-10	6.9E-12
MOX-H	1.0	1.6E-07	9.6E-10	5.1E-11	2.1E-10	9.3E-12
	3.0	2.6E-07	2.8E-09	7.4E-11	5.9E-10	2.5E-11
	13.0	5.9E-07	1.6E-09	1.6E-10	4.0E-10	2.3E-11
	48.1	1.6E-06	2.0E-09	3.1E-10	4.3E-10	2.8E-11
	150.0	4.1E-06	1.3E-09		2.2E-10	1.3E-11
	289.0	7.9E-06	1.7E-09		2.4E-10	1.6E-11
	623.0	1.3E-05	5.5E-09		6.5E-10	5.3E-11
MOX-L seg.	0.2	6.9E-07	2.2E-08		2.5E-09	3.6E-10
MOX-L	1.0	1.3E-06	3.5E-08		4.2E-09	6.1E-10
	1.95	1.7E-06	3.9E-08		4.5E-09	6.6E-10
	1.97	2.1E-06	4.2E-08		4.8E-09	7.0E-10
	2.1	3.5E-07	4.9E-09		5.9E-10	1.0E-10
	5.2	7.1E-07	1.2E-08		9.9E-10	1.7E-10
	6.9	1.1E-06	1.6E-08		1.3E-09	2.2E-10
	7.0	1.4E-06	1.8E-08		1.5E-09	2.5E-10
	7.0	1.0E-06	7.6E-09		1.0E-09	1.7E-10
	8.9	2.5E-06	1.7E-08		2.3E-09	4.0E-10
	16.0	3.4E-06	2.7E-08		3.4E-09	5.9E-10
	29.0	4.5E-06	3.4E-08		3.6E-09	6.2E-10
	64.2	7.6E-06	6.0E-08		5.5E-09	1.1E-09
	147.0	1.1E-05	9.6E-08		6.8E-09	1.3E-09
	348.0	2.1E-05	2.3E-07		1.3E-08	2.8E-09
MOX-KWU seg.	1.0	4.0E-06	4.7E-09	1.1E-09	4.2E-09	1.3E-10
KWU11.38-5810	2.0	4.0E-07	8.7E-10	1.5E-10	2.3E-10	2.5E-11
	160.0	4.2E-08	1.6E-09	3.4E-11	5.0E-10	2.8E-11
	370.0	2.2E-08	4.5E-10	2.2E-11	1.2E-10	2.6E-11
	721	3.5E-08	2.6E-09	1.4E-11	1.0E-09	3.7E-11
	988	9.5E-09	1.1E-09	1.0E-11	7.4E-10	2.9E-11
MOX-KWU frag.	1.0	2.0E-06	2.8E-08	1.7E-09	3.7E-09	1.0E-10
KWU11.38-5810	2.0	1.3E-07	1.6E-09	8.4E-11	7.8E-10	4.6E-11
	154.0	7.2E-08	4.5E-09	6.5E-11	1.1E-09	3.3E-11
	364.0	1.5E-08	4.4E-10	1.7E-11	1.8E-11	1.9E-11
	700.7	1.4E-08	1.1E-09	8.1E-12	4.0E-10	1.9E-11
	950.0	2.1E-08	3.2E-09	3.9E-11	8.6E-10	4.7E-11
UOx-5A2 frag.	0.1	2.2E-08	2.3E-11	2.1E-11	1.6E-12	2.1E-12
5A2	1.0	3.1E-08	8.4E-11	4.5E-11	4.2E-12	6.3E-12
	7.0	3.1E-08	1.2E-10	3.1E-11	2.2E-11	1.4E-11
	28.0	1.9E-08	7.1E-10	1.7E-11	9.5E-11	4.2E-11
	91.0	1.8E-08	3.4E-09	3.5E-11	3.6E-10	2.4E-10
	203.0	2.1E-08	1.0E-08	5.6E-11	9.1E-10	5.6E-10
	421.0	2.6E-08	3.1E-08	1.4E-10	2.2E-09	1.3E-09
	642.0	2.7E-08	4.5E-08	1.8E-10	2.9E-09	1.6E-09
	741.0	2.8E-08	4.8E-08	1.9E-10	3.3E-09	1.8E-09
	1001.0	2.0E-08	1.9E-08	9.8E-11	1.8E-09	9.1E-10
UOx-C1 frag.	0.1	2.7E-08	8.2E-11	2.3E-11	4.2E-12	3.7E-12
C1	1.0	6.0E-08	2.2E-10	2.2E-10	9.4E-12	5.3E-12
ADOPT	7.0	6.8E-08	3.9E-10	1.6E-10	2.7E-11	2.0E-11
	28.0	1.3E-08	2.3E-10	5.5E-11	2.8E-11	1.5E-11
	91.0	1.5E-08	1.8E-09	7.6E-11	2.3E-10	1.8E-10
	203.0	2.4E-08	1.1E-08	1.4E-10	8.0E-10	4.5E-10
	421.0	4.0E-08	3.7E-08	3.6E-10	1.9E-09	1.1E-09
	642.0	4.8E-08	6.6E-08	5.6E-10	3.1E-09	1.5E-09
	741.0	4.0E-08	6.5E-08	3.9E-10	3.5E-09	1.7E-09
	1001.0	2.2E-08	2.8E-08	1.3E-10	2.0E-09	9.5E-10

Table A-1: Aqueous concentrations of actinides (Am, Cm, Np, Pu and U) measured in dissolution experiments with irradiated UO_x fuels and irradiated MOX fuels.

Experiment / *sample*	time / days	[U]	[Pu]	[Np]	[Am]	[Cm]
UOx-73BIC seg.	0.1	7.4E-07	5.8E-09	6.6E-10	4.7E-10	1.4E-10
73BIC	0.2	3.3E-07	1.9E-09	3.7E-10	1.5E-10	3.0E-11
	1.1	6.0E-07	2.0E-09	1.3E-09	1.1E-10	2.7E-11
	2.1	8.5E-07	6.3E-09	7.1E-10	3.9E-10	8.8E-11
	5.2	4.0E-06	2.0E-08	1.6E-09	1.9E-09	5.7E-10
	9.0	5.0E-06	2.2E-08	9.9E-10	4.5E-09	1.4E-09
	16.0	6.6E-06	2.9E-08	2.1E-09	8.5E-09	2.7E-09
	41.1	1.5E-05	5.3E-08	3.2E-09	2.0E-08	6.5E-09
	111.0	1.8E-05	5.5E-08	4.1E-09	1.6E-08	5.9E-09
	210.1	4.5E-06	3.7E-08	4.4E-09	1.2E-08	4.8E-09
UOx-73YCWCa seg.	0.1	2.8E-06	4.5E-09	4.9E-10	3.7E-10	9.2E-11
73YCWCa	0.2	3.2E-07	2.1E-09	2.3E-10	1.5E-10	3.8E-11
	1.1	2.3E-07	1.3E-09	2.8E-10	7.3E-11	1.1E-11
	4.1	2.9E-07	2.1E-09	2.5E-10	1.4E-10	3.2E-11
	9.1	2.0E-07	8.7E-10	1.1E-10	5.0E-11	1.4E-11
	16.1	2.4E-07	1.9E-09	1.6E-10	1.3E-10	3.5E-11
	31.2	2.8E-07	1.2E-09	1.0E-10	7.4E-11	2.0E-11
	60.1	4.4E-07	9.5E-10	1.0E-10	5.9E-11	1.2E-11
	120.2	9.7E-07	2.9E-09	2.3E-10	1.9E-10	5.0E-11
	240.1	1.8E-06	1.9E-10	1.9E-10	2.0E-10	5.0E-11
	457.1	3.5E-06	5.3E-09	3.6E-10	2.6E-10	6.8E-11
AC(YCWCa)	0.1	1.0E-06	5.9E-10	5.0E-11	5.1E-11	1.7E-11
AC-73YCWCa	0.9	5.3E-07	2.2E-10	2.9E-11	2.4E-11	4.9E-12
	0.9	4.5E-07	2.3E-10	4.2E-11	4.4E-11	8.6E-12
	1.1	1.1E-07	1.6E-10	1.4E-11	8.3E-12	5.5E-12
	3.9	7.4E-08	6.2E-11	1.0E-11	1.7E-12	1.6E-12
	3.9	5.1E-08	6.2E-11	2.0E-11	7.8E-12	6.0E-13
	4.1	1.9E-08	8.3E-11	1.1E-11	3.3E-12	0.0E+00
	6.9	2.2E-08	6.0E-10	2.1E-11	2.4E-11	2.6E-12
	32.9	9.8E-09	2.3E-10	7.3E-12	2.1E-11	2.9E-12
	68.0	7.0E-09	6.5E-11	3.1E-12	7.5E-12	2.6E-12
	172.1	4.3E-09	5.1E-11	2.9E-12	5.1E-12	9.1E-13
AC(BIC)	0.1	2.1E-07	3.4E-09	3.8E-10	4.3E-10	7.5E-11
AC-73BIC	3.0	9.0E-08	1.8E-09	4.4E-10	2.7E-10	4.6E-11
	3.0	1.3E-07	1.5E-09	6.4E-10	3.5E-10	6.7E-11
	3.2	2.1E-07	8.6E-09	2.1E-10	1.3E-09	1.6E-10
	4.0	8.3E-08	6.6E-10	1.0E-10	1.8E-10	4.0E-11
	6.0	6.1E-08	4.1E-10	1.2E-10	1.2E-10	2.7E-11
	6.0	5.9E-07	3.9E-09	3.3E-10	1.4E-09	2.9E-10
	6.1	2.5E-07	2.3E-09	1.6E-10	6.2E-10	1.3E-10
	7.0	9.3E-08	9.8E-10	7.9E-11	2.4E-10	5.6E-11
	14.1	8.5E-08	3.7E-10	1.6E-10	2.5E-11	1.1E-11
	31.0	1.1E-07	3.1E-10	2.9E-10	1.6E-11	5.2E-12
	61.0	1.2E-07	4.4E-10	2.0E-10	1.2E-10	2.8E-11
	144.0	3.7E-08	5.0E-10	3.6E-11	6.3E-11	9.2E-12

Table A-1: Aqueous concentrations of actinides (Am, Cm, Np, Pu and U) measured in dissolution experiments with irradiated UO_x fuels and irradiated MOX fuels (continued).

Experiment / sample	time / days	[Tc]	[Sr]	[Mo]	[I]	[Cs]
MOX-H seg.	0.2	3.6E-09		7.8E-08		3.5E-06
MOX-H	1.0	2.0E-09		2.0E-08		1.5E-07
	3.0	2.3E-09		1.5E-08		7.9E-08
	13.0	6.7E-09	2.8E-11	5.7E-08		1.6E-07
	48.1	1.9E-08	5.3E-09	1.8E-07		2.5E-07
	150.0	5.7E-08	2.2E-08	3.9E-07		3.9E-07
	289.0	1.3E-07	2.9E-08	5.2E-07		5.4E-07
	623.0	3.3E-07	7.6E-08	8.1E-07		1.1E-06
MOX-L seg.	0.2	7.9E-09	2.1E-08	6.0E-08		4.1E-06
MOX-L	1.0	1.7E-08	3.7E-08	1.5E-07		7.1E-06
	1.95	3.0E-08	5.2E-08	2.5E-07		9.8E-06
	1.97	3.3E-08	5.8E-08	2.8E-07		1.0E-05
	2.1	2.9E-09	6.4E-09	3.2E-08		5.0E-07
	5.2	1.3E-08	1.7E-08	8.2E-08		1.2E-06
	6.9	3.0E-08	2.9E-08	1.4E-07		2.0E-06
	7.0	3.5E-08	3.3E-08	1.6E-07		2.2E-06
	7.0	3.4E-09	6.5E-09	4.5E-08		1.7E-07
	8.9	1.2E-08	1.7E-08	7.5E-08		4.6E-07
	16.0	4.7E-08	3.0E-08	1.3E-07		1.1E-06
	29.0	1.4E-07	6.0E-08	2.5E-07		2.1E-06
	64.2	4.1E-07	1.2E-07	5.2E-07		3.5E-06
	147.0	9.5E-07	2.0E-07	9.9E-07		5.4E-06
	348.0	1.4E-06	3.0E-07	1.5E-06		6.6E-06
MOX-KWU seg.	1.0		3.1E-09			6.2E-06
KWU11.38-5810	2.0		3.1E-10			8.4E-07
	160.0	9.8E-09	4.2E-08		7.6E-06	4.3E-06
	370.0	4.5E-09	4.6E-08		8.8E-06	4.4E-06
	721	6.0E-09	5.1E-08		9.5E-06	4.7E-06
	988	2.5E-08	5.8E-08		9.1E-06	4.6E-06
MOX-KWU frag.	1.0		5.3E-09			4.5E-06
KWU11.38-5810	2.0		8.2E-10			2.6E-07
	154.0	7.5E-09	6.1E-09		3.3E-06	1.1E-06
	364.0	4.0E-09	9.3E-09		5.5E-06	1.5E-06
	700.7	4.5E-09	1.4E-08		4.7E-06	1.7E-06
	950.0	3.3E-08	1.7E-08		4.3E-06	1.8E-06
UOx-5A2 frag.	0.1	4.6E-11			2.5E-09	4.4E-10
5A2	1.0	9.4E-11		1.1E-09	2.8E-08	2.3E-09
	7.0	4.4E-11	1.6E-09	5.1E-09	0.0E+00	7.9E-08
	28.0	1.5E-09	5.1E-09	5.5E-08	2.9E-07	4.8E-07
	91.0	7.2E-10	5.0E-09	7.3E-08	5.1E-07	8.3E-07
	203.0	1.4E-09	1.0E-08	1.1E-07	4.7E-07	8.3E-07
	421.0	3.6E-09	1.3E-08	1.5E-07	4.4E-07	8.6E-07
	642.0	2.7E-09	1.7E-08	1.4E-07	7.9E-07	7.8E-07
	741.0	2.5E-09	2.2E-08	1.6E-07	4.7E-07	8.6E-07
	1001.0	1.8E-09	2.1E-08	1.8E-07	4.8E-07	7.9E-07
UOx-C1 frag.	0.1	5.9E-11			2.5E-09	1.0E-09
C1	1.0	1.3E-10		6.5E-10	3.7E-09	2.6E-09
	7.0	1.0E-10	3.1E-09	8.4E-09		5.9E-08
	28.0	1.1E-09	2.5E-09	9.4E-09	6.7E-08	1.7E-07
	91.0	8.8E-10	7.5E-09	1.6E-08	1.5E-07	7.2E-07
	203.0	1.1E-09	1.2E-08	3.3E-08	2.5E-07	1.1E-06
	421.0	3.5E-09	1.8E-08	4.4E-08	2.9E-07	1.2E-06
	642.0	3.0E-09	2.3E-08	6.6E-08	3.1E-07	1.2E-06
	741.0	2.7E-09	2.7E-08	7.3E-08	3.0E-07	1.3E-06
	1001.0	2.2E-09	2.7E-08	8.1E-08	3.2E-07	1.3E-06

Table A-2: Aqueous concentrations of fission products (Ba, Cs, Gd, I, Mo, Rb, Sr, Tc and Zr) measured in dissolution experiments with irradiated UO_x fuels and irradiated MOX fuels.

Experiment / sample	time / days	[Ba]	[Gd]	[Zr]	[Rb]
MOX-H seg.	0.2			1.1E-09	
MOX-H	1.0			4.8E-10	
	3.0			2.0E-09	
	13.0			7.5E-10	
	48.1			3.9E-10	
	150.0			8.7E-10	4.6E-09
	289.0			2.7E-10	1.6E-08
	623.0			2.2E-10	4.0E-08
MOX-L seg.	0.2			8.4E-09	
MOX-L	1.0			1.4E-08	
	1.95			1.8E-08	
	1.97			1.8E-08	
	2.1			5.9E-09	
	5.2			1.1E-08	2.2E-09
	6.9			1.2E-08	6.8E-09
	7.0			1.3E-08	8.4E-09
	7.0			9.1E-09	1.1E-09
	8.9			1.7E-08	3.5E-09
	16.0			2.3E-08	8.4E-09
	29.0			2.7E-08	2.1E-08
	64.2			3.3E-08	5.8E-08
	147.0			3.7E-08	1.5E-07
	348.0			5.2E-08	2.4E-07
MOX-KWU seg.					
KWU11.38-5810					
MOX-KWU frag.					
KWU11.38-5810					
UOx-5A2 frag.	0.1	6.4E-10	1.2E-11	5.5E-10	
5A2	1.0	2.6E-10	1.6E-11	7.7E-10	
	7.0	3.1E-08	8.7E-11	8.0E-10	3.7E-09
	28.0	4.4E-08	3.0E-10	1.5E-09	2.0E-08
	91.0	4.8E-08	1.4E-09	8.5E-09	3.0E-08
	203.0	5.6E-08	3.5E-09	5.8E-08	1.6E-08
	421.0	7.0E-08	6.0E-09	1.4E-07	1.7E-08
	642.0	7.6E-08	7.0E-09	2.0E-07	1.6E-08
	741.0			1.3E-07	2.1E-08
	1001.0			1.0E-07	2.2E-08
UOx-C1 frag.	0.1	1.1E-09	1.5E-11	8.2E-10	
C1	1.0	6.9E-09	4.1E-11	9.7E-10	4.4E-10
	7.0	3.8E-08	8.5E-11	1.1E-09	7.1E-09
	28.0	2.9E-08	1.5E-10	7.8E-10	6.1E-09
	91.0	3.8E-08	1.3E-09	9.6E-09	1.1E-08
	203.0	5.1E-08	3.5E-09	7.3E-08	
	421.0	7.3E-08	6.4E-09	1.7E-07	
	642.0	9.5E-08	8.5E-09	2.9E-07	
	741.0			1.8E-07	
	1001.0			1.5E-07	

Table A-2: Aqueous concentrations of fission products (Ba, Cs, Gd, I, Mo, Rb, Sr, Tc and Zr) measured in dissolution experiments with irradiated UO_x fuels and irradiated MOX fuels (continued).

Experiment / sample	time / days	[Tc]	[Sr]	[Mo]	[I]	[Cs]
UOx-73BIC seg.	0.1	1.8E-08	6.7E-09	1.3E-07		6.1E-06
73BIC	0.2	8.4E-09	8.7E-09	5.7E-08		4.7E-06
	1.1	7.8E-09	3.1E-08	1.0E-07		7.8E-06
	2.1	5.3E-09	4.4E-08	5.5E-08		4.5E-06
	5.2	4.5E-09	1.9E-07	3.9E-08		1.1E-05
	9.0	3.4E-09	9.8E-08	2.3E-08		4.0E-06
	16.0	4.0E-09	7.8E-08	2.8E-08		1.8E-06
	41.1	1.1E-08	9.5E-08	8.7E-08		1.4E-06
	111.0	5.9E-08	1.1E-07	2.4E-07		1.1E-06
	210.1	1.4E-07	1.8E-07	9.7E-08		1.5E-06
UOx-73YCWCa seg.	0.1	4.2E-08		6.7E-07	1.0E-06	1.6E-05
73YCWCa	0.2	5.6E-08		8.5E-07	3.0E-07	6.5E-06
	1.1	1.9E-07		2.1E-06	1.3E-07	5.1E-06
	4.1	4.7E-07		5.9E-06	3.5E-07	4.0E-06
	9.1	6.3E-07		7.1E-06	8.1E-07	1.9E-06
	16.1	5.9E-07		6.7E-06	1.1E-06	1.4E-06
	31.2	9.2E-07		8.4E-06	1.5E-06	2.2E-06
	60.1	1.1E-06		8.2E-06	8.6E-07	4.3E-06
	120.2	1.6E-06		1.1E-05	1.1E-06	7.9E-06
	240.1	3.0E-06		1.8E-05	1.9E-06	1.2E-05
	457.1	2.9E-06		2.3E-05	4.2E-06	1.6E-05
AC(YCWCa)	0.1	2.3E-08	1.0E-27	7.8E-07		7.2E-06
AC-73YCWCa	0.9	1.3E-08	9.7E-28	4.2E-07		3.9E-06
	0.9	1.1E-08	1.2E-09	3.5E-07		3.1E-06
	1.1	1.6E-09	1.1E-27	1.1E-07		6.1E-07
	3.9	1.4E-09	1.1E-27	1.8E-07		7.0E-07
	3.9	1.3E-09	8.6E-28	2.1E-07		6.9E-07
	4.1	2.2E-10	1.1E-27	4.6E-08		1.6E-07
	6.9	3.3E-10	1.5E-08	9.0E-08		3.3E-07
	32.9	4.5E-10	5.6E-09	2.1E-07		1.7E-06
	68.0	5.6E-10	5.7E-09	2.5E-07		2.8E-06
	172.1	7.1E-10	2.7E-09	3.8E-07		5.5E-06
AC(BIC)	0.1	5.0E-09	3.6E-09	5.1E-08		1.8E-06
AC-73BIC	3.0	5.4E-09	1.3E-08	5.8E-08		2.9E-06
	3.0	6.0E-09	2.3E-08	7.0E-08		3.3E-06
	3.2	7.8E-10	1.4E-09	1.4E-08		3.3E-07
	4.0	1.0E-09	7.2E-09	1.9E-08		6.9E-07
	6.0	1.2E-09	1.6E-08	3.0E-08		1.2E-06
	6.0	1.7E-09	2.2E-08	2.9E-08		1.3E-06
	6.1	3.3E-10	4.2E-10	4.5E-09		1.1E-07
	7.0	3.5E-10	3.0E-09	6.9E-09		2.7E-07
	14.1	9.4E-10	1.1E-08	1.7E-08		8.6E-07
	31.0	1.7E-09	2.2E-08	3.0E-08		1.4E-06
	61.0	1.8E-09	1.7E-08	4.5E-08		1.8E-06
	144.0	3.0E-10	3.8E-08	4.2E-08		3.2E-06

Table A-2: Aqueous concentrations of fission products (Ba, Cs, Gd, I, Mo, Rb, Sr, Tc and Zr) measured in dissolution experiments with irradiated UO_x fuels and irradiated MOX fuels (continued).

Experiment / sample	time / days	[Ba]	[Gd]	[Zr]	[Rb]
UOx-73BIC seg.	0.1	4.9E-09		7.6E-09	1.7E-07
73BIC	0.2	4.0E-09		1.9E-09	1.5E-07
	1.1	2.8E-08		1.3E-09	2.5E-07
	2.1	7.6E-08		1.9E-09	3.9E-08
	5.2	3.4E-07		1.1E-09	7.6E-08
	9.0	1.7E-07		1.5E-09	8.4E-08
	16.0	1.4E-07		2.0E-09	6.7E-08
	41.1	1.6E-07		2.7E-09	7.7E-08
	111.0	1.5E-07		2.4E-09	8.3E-08
	210.1	2.0E-07		2.0E-09	1.5E-07
UOx-73YCWCa seg.	0.1			1.4E-07	1.7E-07
73YCWCa	0.2			1.7E-08	5.6E-08
	1.1			2.3E-09	3.2E-08
	4.1			1.6E-09	3.3E-08
	9.1			5.4E-10	4.9E-08
	16.1			5.8E-10	7.3E-08
	31.2			2.3E-10	1.6E-07
	60.1			1.0E-10	4.3E-07
	120.2			7.3E-11	8.4E-07
	240.1			7.7E-11	1.2E-06
	457.1			2.9E-11	1.6E-06
AC(YCWCa)	0.1			6.9E-09	2.4E-07
AC-73YCWCa	0.9			4.9E-09	1.2E-07
	0.9			2.9E-09	1.0E-07
	1.1			3.8E-09	1.7E-08
	3.9			2.9E-09	1.5E-08
	3.9			1.9E-09	1.2E-08
	4.1			4.5E-09	1.5E-09
	6.9			3.3E-09	1.3E-27
	32.9			1.8E-09	1.3E-27
	68.0			3.4E-09	8.5E-28
	172.1			2.1E-09	7.9E-28
AC(BIC)	0.1			9.5E-09	6.1E-08
AC-73BIC	3.0			4.3E-09	6.5E-08
	3.0			6.2E-10	6.7E-08
	3.2			1.1E-08	5.5E-09
	4.0			2.2E-09	3.9E-09
	6.0			1.3E-09	1.8E-09
	6.0			1.6E-08	8.3E-10
	6.1			9.5E-09	3.0E-10
	7.0			4.0E-09	3.0E-09
	14.1			4.2E-10	2.1E-08
	31.0			3.2E-10	3.9E-08
	61.0			1.5E-10	5.2E-08
	144.0			1.3E-09	4.6E-08

Table A-2: Aqueous concentrations of fission products (Ba, Cs, Gd, I, Mo, Rb, Sr, Tc and Zr) measured in dissolution experiments with irradiated UO_x fuels and irradiated MOX fuels (continued).