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# Carbon-14 Source Term

## CAST



## Description of Zircaloy-4 dissolution experiment in a shielded box (D3.8)

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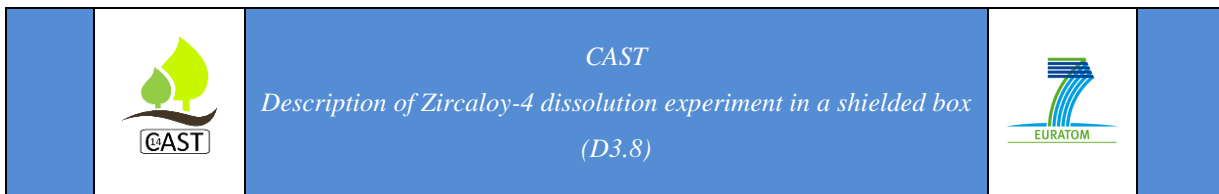
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### Dissemination Level

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## **CAST – Project Overview**

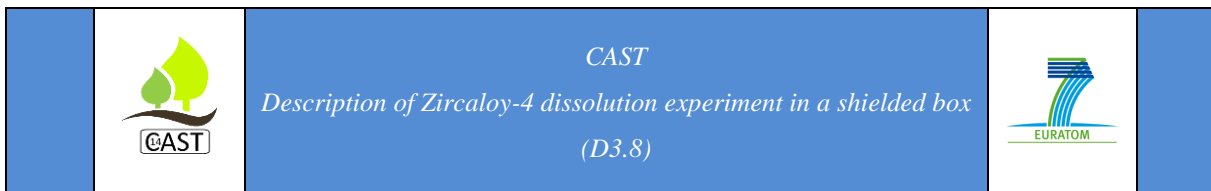
The CAST project (CARbon-14 Source Term) aims to develop understanding of the potential release mechanisms of carbon-14 from radioactive waste materials under conditions relevant to waste packaging and disposal to underground geological disposal facilities. The project focuses on the release of carbon-14 as dissolved and gaseous species from irradiated metals (steels, Zircaloys), irradiated graphite and from ion-exchange materials.

The CAST consortium brings together 33 partners with a range of skills and competencies in the management of radioactive wastes containing carbon-14, geological disposal research, safety case development and experimental work on gas generation. The consortium consists of national waste management organisations, research institutes, universities and commercial organisations.

The objectives of the CAST project are to gain new scientific understanding of the rate of release of carbon-14 from the corrosion of irradiated steels and Zircaloys and from the leaching of ion-exchange resins and irradiated graphites under geological disposal conditions, its speciation and how these relate to carbon-14 inventory and aqueous conditions. These results will be evaluated in the context of national safety assessments and disseminated to interested stakeholders. The new understanding should be of relevance to national safety assessment stakeholders and will also provide an opportunity for training for early career researchers.

For more information, please visit the CAST website at:

<http://www.projectcast.eu>



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## Executive Summary

The amount and chemical form of C-14 as well as the inventories of Sb-125 and Cs-137 are determined in Zircaloy cladding from an irradiated UO<sub>2</sub> fuel rod segment. Experimentally measured radionuclide contents are compared to the theoretically predicted inventory of the Zircaloy-4 obtained by means of MCNP-X calculations. The cladding was sampled from the plenum of a fuel rod segment which experienced an average burn-up of 50.4 GWd/t<sub>HM</sub> in the Gösgen pressurised water reactor (PWR). These cladding samples were dry cut and digested in dilute acid. <sup>14</sup>C was separated from other radionuclides in gaseous and aqueous aliquots and analysed by liquid scintillation counting (LSC). The amount of Sb-125 and Cs-137 was determined by  $\gamma$ -spectrometry.

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## 1 Introduction

Due to its half-life of about 5730 years and assumed mobility, C-14 is one of the crucial radionuclides with respect to estimated doses arising from release to the environment in a canister failure scenario for spent nuclear fuel (SNF). In long-term safety assessments of SNF disposal facilities the following scenario is taken into consideration: upon contact with water, C-14 bearing species are released from the fuel rods into aqueous solution and gas phase as inorganic or organic compounds. C-14 species will also be released from Zircaloy in packaged intermediate-level wastes from the reprocessing of SNF. Solubility, sorption behaviour and distribution between solution and gas of C-14 depend strongly on its chemical form.

Only very few previous studies dealing with C-14 quantification and/or speciation from irradiated materials from nuclear reactors are available. [NEEB *et al.*, 1980] and [BLEIER *et al.*, 1984] determined the inventory and distribution of C-14 and H-3 in fuel rod specimen irradiated in boiling water reactor (BWR) and pressurised water reactor (PWR). [BLEIER *et al.*, 1988] investigated the chemical state of C-14 after leaching of cladding material from spent PWR and BWR fuel rods in a salt solution. [YAMAGUCHI *et al.*, 1999] determined the inventory and chemical form of C-14 in irradiated Zircaloy hull waste from spent nuclear fuel reprocessing. [ZHANG, 1993] performed leaching experiments with irradiated pebbles of a pebble-bed reactor and [STROES-GASCOYNE *et al.*, 1994] determined the inventory of C-14 in irradiated CANDU fuel released from gap and grain boundaries to aqueous solutions. Very recently a study published by [SCHUMANN *et al.*, 2014] deals with the determination of the C-14 content in activated steel components from a neutron spallation source and a nuclear power plant (NPP). There are several studies on C-14 quantification and/or speciation in spent ion exchange resins or NPP process water, an over-view is given in [MAGNUSSON, 2007].

KIT-INE have developed methods that allow the separation and quantification of inorganic and organic C-14 species in gaseous and aqueous samples taken from dissolution experiments with irradiated claddings of a PWR fuel rod segment. The sample

characteristics, sample preparation in a shielded box-line and dissolution experiments performed with these irradiated Zircaloy-4 cladding samples, as well as the preliminary results obtained from these dissolution experiments, are described in the following chapters.

## 2 Materials and Methods

### 2.1 Materials

The studied Zircaloy-4 plenum cladding was sampled from fuel rod segment N0204 of fuel rod SBS1108. The fuel rod was irradiated during four cycles in the PWR Gösgen (KKG, Switzerland) and discharged in May 1989. During reactor operation an average burn-up of  $50.4 \text{ GWd/t}_{\text{HM}}$  was achieved. The fuel of segment N0204 and similar segments were fabricated by Kraftwerk Union AG (KWU) using the NIKUSI short-term fast sintering process [STRATTON *et al.*, 1991]. Except two  $\text{UO}_2(\text{nat.})$  pellets, all fuel pellets of segment N0204 were initially enriched with 3.8 wt.% of U-235. In the plenum of the fuel rod segment a stainless steel spring was inserted. Relevant characteristic parameters of the irradiated material are given in Table 1 as well as photos in Fig. 1.

In total 177 fuel assemblies, each with a  $15 \times 15$  lattice, form the reactor core of PWR Gösgen. 205 of the 225 positions of the lattice geometry are occupied with fuel rods, the 20 remaining guide tubes are available for absorber rods (AgInCd) [KERNKRAFTWERK-GÖSGEN, 2010]. Fuel rod SBS1108 was inserted in the central region of the  $15 \times 15$  fuel assembly. For irradiation data see Table 1. The N0204 segment was stored gas tight until 2012. Then the segment was transported to JRC-ITU for characterisation, gas sampling (puncturing), cutting and sampling of fuel pellets, plenum cladding and the stainless steel spring. Results of the non-destructive characterisation and gas sampling are given in reports of JRC-ITU and KIT-INE [GONZALEZ-ROBLES *et al.*, 2012; WEGEN *et al.*, 2012a; WEGEN *et al.*, 2012b, c], respectively. After puncturing and successive cutting of the fuel rod segment, gas samples, fuel, plenum Zircaloy cladding, as well as the stainless steel spring, were returned to KIT-INE for further investigations.

**Table 1: Characteristics and irradiation data of the studied material [METZ *et al.*, 2014].**

Reactor	type: PWR fuel type: UO <sub>2</sub> thermal power: 3002 MW
Zircaloy specimen data	material: Zircaloy-4 (Zry-4) rod diameter: 10.75 mm wall thickness: 0.725 mm length: 75.3 mm
Plenum spring data	material: X7CrNiAl17-7 (1.4568) weight: 10.4 g
UO <sub>2</sub> fuel pellet	initial enrichment: 3.8% of <sup>235</sup> U two UO <sub>2</sub> (nat.) pellets at top and bottom of the pellet stack pellet diameter: 9.3 mm pellet height: 11 mm pellet density: 10.41 g·cm <sup>-3</sup>
Irradiation data	average burn-up: 50.4 GWd/t <sub>HM</sub> number of cycles: 4 average linear power rate: 260 W/cm maximal linear power rate: 340 W/cm date of discharge: 27 May 1989 irradiation duration: 1226 days

The radionuclide inventory of the Zircaloy-4 plenum samples was calculated by means of the Monte Carlo N-Particle code version MCNP-X [MCNP, 2011] and the JEFF-3.1 database [JEFF, 2006]. Calculations were performed based on the known burn-up history of fuel rod SBS1108 and the dimensions, weight, density and nominal chemical composition of the Zircaloy-4 (see Table 2). Also important is the direct surrounding of the material and the position in the fuel assembly and nuclear reactor. Limited information on the Zircaloy-4 cladding irradiated in the commercial PWR provided by the utility and manufacturer which is in agreement with data of a publication related to the irradiation of adjacent test fuel rods [STRATTON *et al.*, 1991]. Unfortunately the real chemical composition of the irradiated Zircaloy-4 material used in this study is not available therefore nominal chemical compositions are used in the calculations (0.005% N-14, 0.016% O-17 and 0.012% C-13). Normative specifications usually indicate a range of composition (for alloying elements) or

maximum values (for impurities). Especially the content of the C-14 precursor elements N-14, O-17 and C-13 are important for the calculations and the inventory of C-14. The use of those maximum levels in activation calculations gives excessive values for C-14 and other activation/fission products in the material. [SAKURAGI *et al.*, 2013; GRAS, 2014] recommend 40 ppm N-14 content for Zircaloy-4 for C-14 activation calculations. This value is 25% below the initial nitrogen content in the irradiated Zircaloy-4 which was derived from information of the manufacturer of the fuel rods.

**Table 2: Nominal composition of Zircaloy-4 and values used for calculation.**

	Zircaloy-4 [wt.%] [RUDLING <i>et al.</i> , 2007]	Zircaloy-4 [wt.%] [BAILLY <i>et al.</i> , 1999] [NEEB, 1997]	Zircaloy-4 used for calculation [wt.%]
<b>Alloying elements</b>			
Zr	balanced	balanced	balanced
Sn	1.20–1.70	1.20–1.70	0.8 (considered as Zyr-4 DX ELS 0.8*)
Fe	0.18–0.24	0.18–0.24	0.24
Cr	0.07–0.13	0.07–0.13	0.13
O	0.09–0.16	0.11–0.14	0.16
Si	0.005–0.012	< 0.012	
<b>Impurities:</b>			
Al	< 0.0075	< 0.0075	
B	< 0.00005	< 0.00005	



Cd	< 0.00005	<0.00005	
C	< 0.0270	< 0.0270	0.0120
Co	< 0.0020	< 0.0020	0.0020
Cu	< 0.0050	< 0.0050	
Hf	< 0.0100	< 0.0100	
H	< 0.0025	< 0.0025	
Mg	< 0.0020	< 0.0020	
Mn	< 0.0050	< 0.0050	0.0050
Mo	< 0.0050	< 0.0050	
Ni	< 0.0070	< 0.0070	0.0070
N	< 0.0080	< 0.0065	0.0050
Ti	< 0.0050	< 0.0050	
U	< 0.00035	< 0.00035	0.00035
W	< 0.01	< 0.01	

\* Zyr-4 DX ELS 0.8: A duplex (DX) cladding which consists of an Extra-Low Sn 0.8 wt.% (ELS 0.8) outer corrosion resistant layer with a thickness < 100 μm and the rest of the thickness is Zircaloy-4 to provide the mechanical strength [RUDLING *et al.*, 2007].



**Figure 1: The upper two pictures show the whole irradiated Zircaloy-4 plenum specimen available at KIT-INE. The lower left picture shows the bottom half of the specimen, where the plenum was cut from the fuel rod segment whereas the lower right picture shows the end cap of the plenum specimen.**

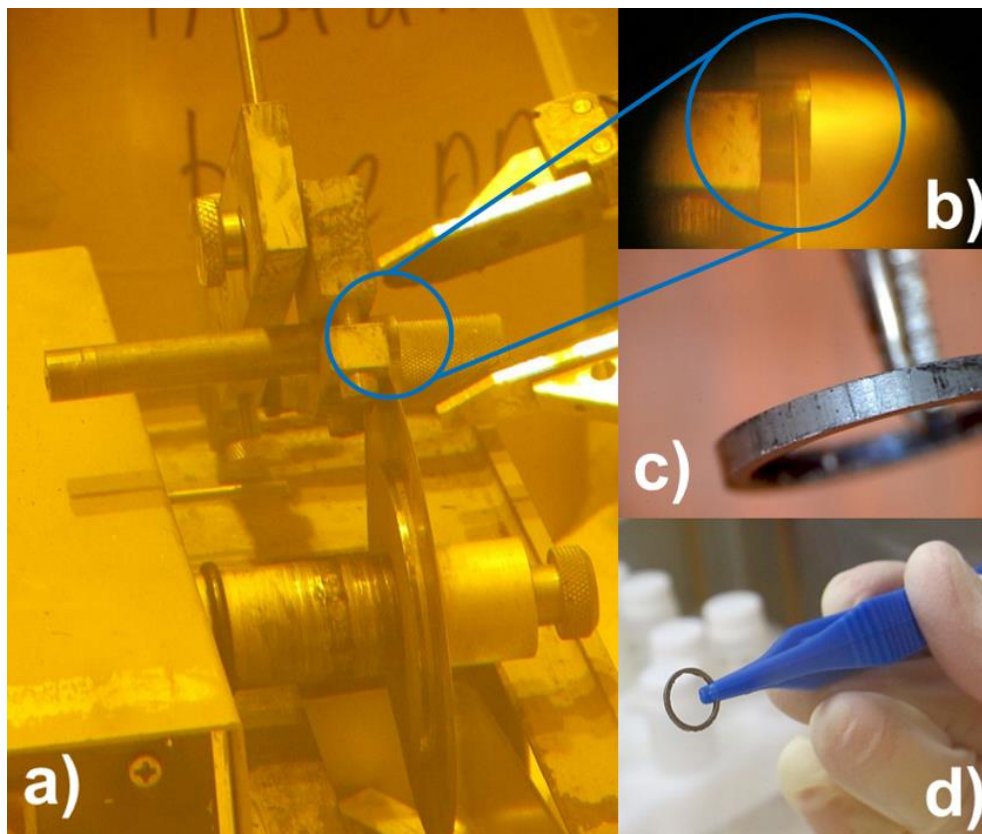
## ***2.2 Sampling and sample preparation***

Preparation of Zircaloy-4 samples from the irradiated plenum tube was conducted in the KIT-INE shielded box line. The samples (in total five) were dry cut using a low speed saw (Isomet 11-1180, Buehler Ltd.) equipped with a diamond saw blade. The cutting was very slow (30–40 minutes per sample) to prevent overheating of the samples. Characteristics of the samples are given in Table 3 and photos in Fig. 2. Finally a surface analysis was carried out using an optical microscope (DigiMicro 2.0, Drahtlose Nachrichtentechnik, dnt) available inside the shielded box to determine the geometric surface area of the samples. Due to the relative low dose rate ( $\leq 280 \mu\text{Sv/h}$ ) of each sample and in accordance with German Radiation Protection Ordinance [BGBL, 2005] and the technical advisory board of

KIT-INE, the samples were removed from the shielded box and dissolution experiments including the samples were conducted in a specifically manufactured glove box.

**Table 3: Characteristic data of the irradiated Zircaloy-4 samples.**

Sample	#1	#2	#3	#4	#5
mass [mg]	184.4	130.1	119.7	177.5	189.0
dose rate [ $\mu\text{Sv/h}$ ]	$\leq 270$	$\leq 150$	$\leq 170$	$\leq 260$	$\leq 280$



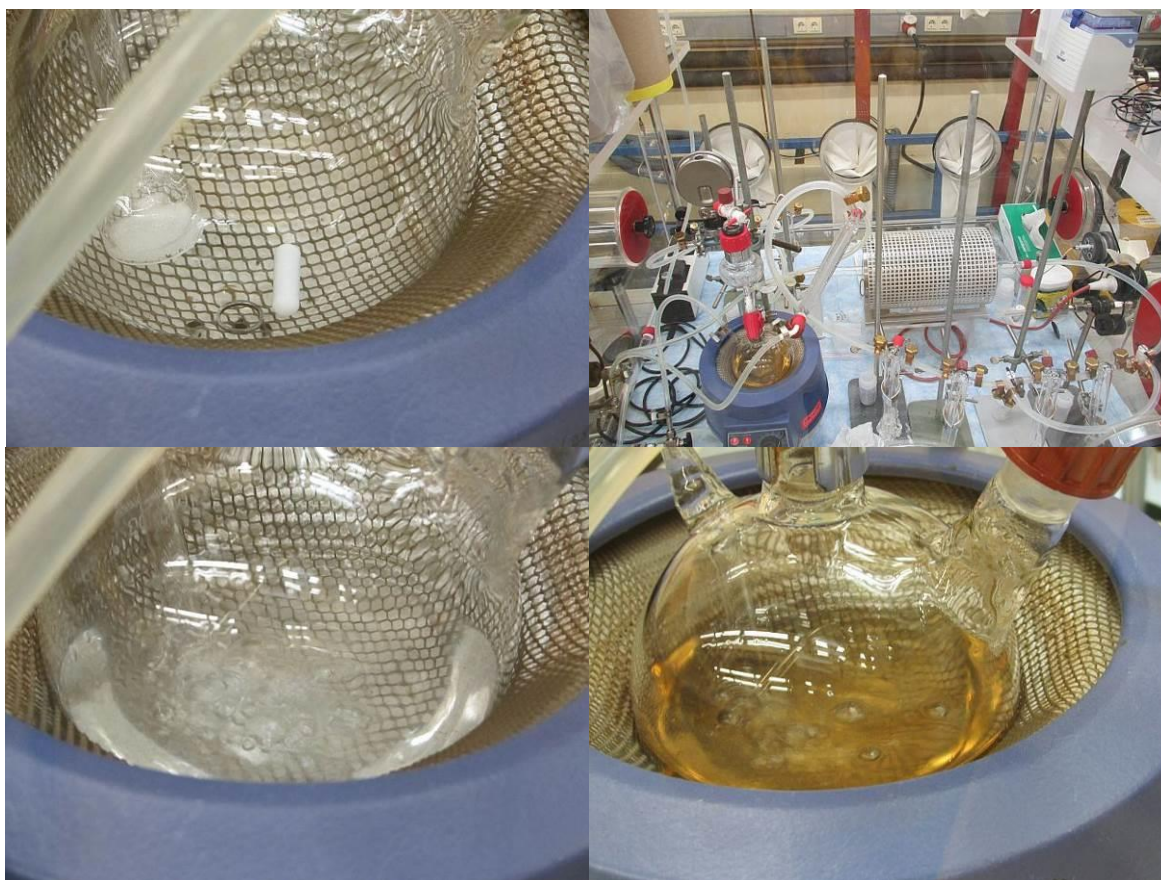
**Figure 2: Figure 2a) shows the low speed saw and the Zircaloy-4 plenum cutting half through a sample (figure 2b). The photo in figure 2c) was made using the optical microscope available inside the shielded box and figure 2d) shows the preparation of the sample for the dissolution experiments inside a specifically manufactured glove box.**

## 2.3 Sample dissolution

Two types of digestion experiments were conducted with the Zircaloy-4 samples. Three cladding samples (#1 to #3) were digested in 16% H<sub>2</sub>SO<sub>4</sub> + 3% HF in a flask of the C-14 extraction apparatus in a specifically manufactured glove box and one cladding sample (#4) was digested using an autoclave equipped with a glass liner and a gas collecting cylinder with two valves. Sample #5 was digested and analysed by  $\alpha$ -,  $\gamma$ -, ICP-MS-spectrometry and LSC to demonstrate that the radioactive inventory of the samples is within the regulatory limits for working in a glove box. An overview on the experimental program including extraction and analytical techniques is given in [HERM *et al.*, 2014].

In the experiments with samples #1, #2 and #3, the cladding specimen was placed in the flask (Fig. 3) connected to the CO<sub>2</sub> gas absorption system after evacuating the system to 0.2 bar below atmosphere and setting the nitrogen gas flow rate to 300 mL/min, 100 mL 24% H<sub>2</sub>SO<sub>4</sub> was added immediately to the sample container using the dropping funnel. Then slowly 50 mL 10% HF was added through the dropping funnel to the flask and the Zircaloy-4 sample was digested within 30 minutes. Gases evolving during the digestion process (H<sub>2</sub>, HT, CO, CO<sub>2</sub>, gaseous hydrocarbons) were absorbed in 2 M NaOH directly or after oxidation to CO<sub>2</sub> in the catalytic furnace. The remaining carbon fractions (liquid hydrocarbons e.g. alcohols, aldehydes, ketones, carboic acids...) in solution were then released as CO<sub>2</sub> by wet oxidation and absorbed in 2 M NaOH. Then samples were collected from all washing bottles and flasks and analysed by  $\alpha$ -,  $\gamma$ -, ICP-MS-spectrometry and LSC.

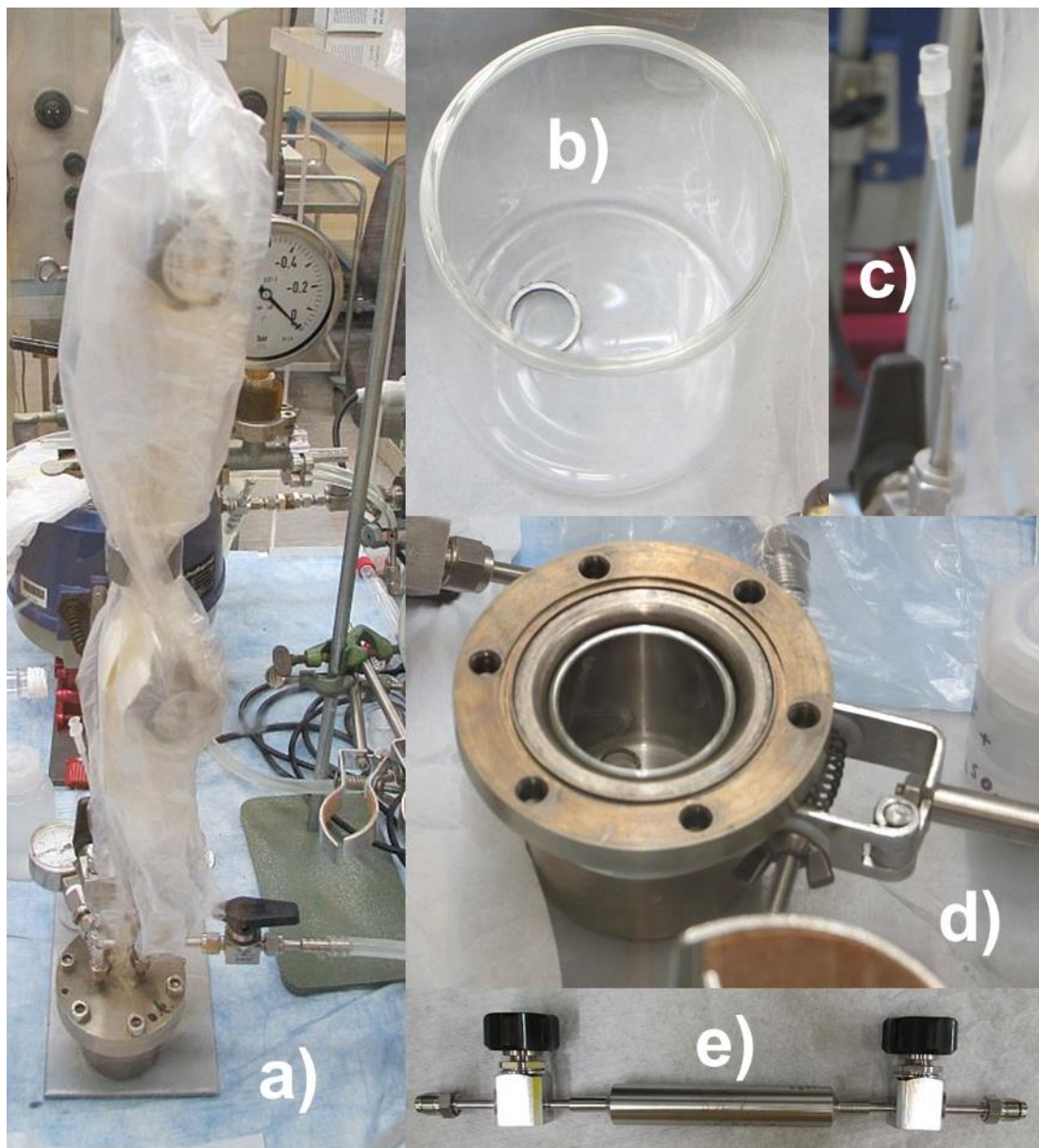




**Figure 3: The upper pictures show the Zircaloy-4 sample inside the flask (left) and the whole CO<sub>2</sub> absorption apparatus including the catalytic furnace (right). The lower left pictures shows the digestion of the sample at room temperature and the lower right picture shows the wet oxidation process during which the solution turns gold/brownish before decoloring again.**

For the autoclave digestion (see Fig. 4) , sample #4 was placed in a glass liner inside the autoclave and the autoclave was sealed air tight. A gas collecting cylinder with two valves was mounted on top of the autoclave. The autoclave was flushed with nitrogen and 20 mL of a 16% H<sub>2</sub>SO<sub>4</sub> + 3% HF mixture were added through a long tube. After the addition all valves in the lid of the autoclave were closed. During the digestion process the autoclave developed a pressure of about 1.4 bar. After in total five hours and ensuring constant pressure, the gas collecting cylinder was opened and the gas phase collected. The final pressure (after opening the 50 mL gas collecting cylinder) in the whole system was about

0.5 bar. The gas phase was analysed by gas-MS. The aqueous solution as well as the remaining gas phase in the gas collecting cylinder were also analysed for C-14 in the CO<sub>2</sub> gas absorption system as described in [HERM *et al.*, 2014].

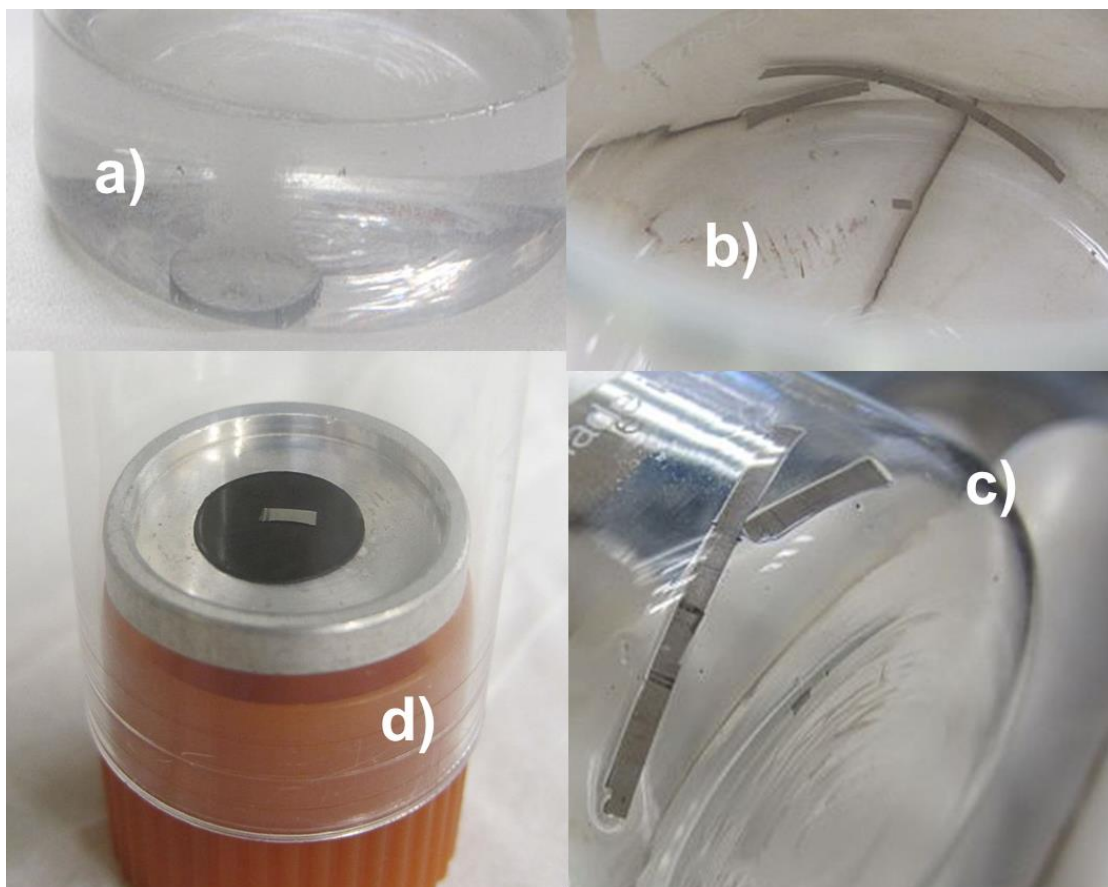


**Figure 4:** Figure 4a) shows the sealed autoclave with gas collecting cylinder mounted on top. The glass liner with the Zircaloy-4 sample is seen in figure 4b) and the glass liner with sample in the open autoclave in 4d). Figure 4c) shows the tube used for the addition of the acid. The gas collecting cylinder with two valves is seen in figure 4e).

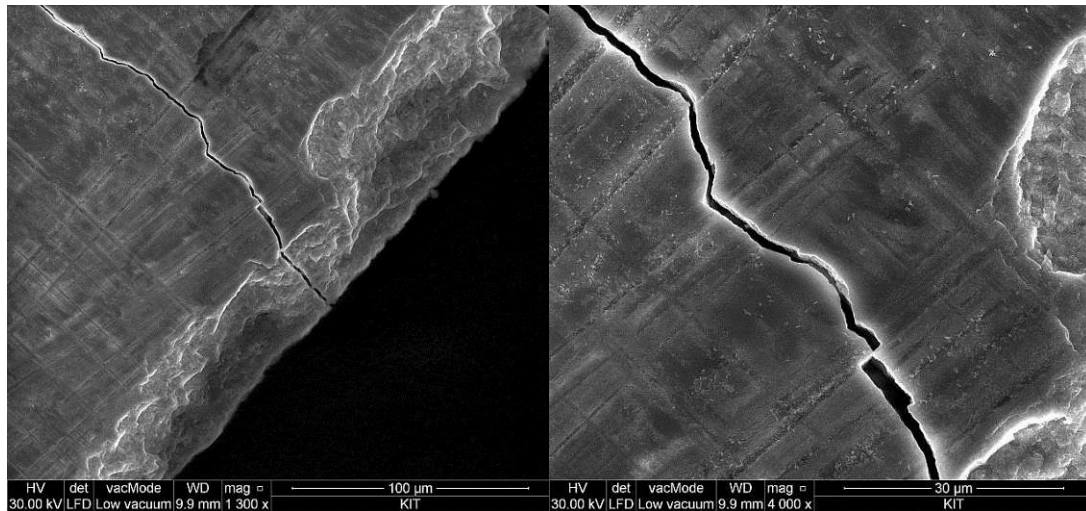


### 3 Preliminary results of the dissolution experiments

In the digestion experiment with sample #5 it was observed, that the oxide layer of the Zircaloy-4 remains physically intact after the complete digestion of the alloy at room temperature without stirring, purging or heating the solution (see Fig. 5). However it is assumed that radionuclides present in the oxide layer are released during the digestion of the Zircaloy. Therefore a sample of the oxide remnant was taken and analysed by scanning electron microscopy – energy dispersive X-ray spectroscopy (SEM-EDS) (see Fig. 6). The oxide layer gradually dissolves during the C-14 extraction process as the solution in the flask is stirred, purged with nitrogen and heated during the wet oxidation step.



**Figure 5: Figure 5a) shows the Zircaloy-4 sample 5 during digestion whereas figure 5b) and c) show the remaining oxide layer after the digestion and in figure 5d) the oxide layer prepared for the SEM-EDS measurements is shown.**



**Figure 6: SEM images of the irradiated Zircaloy-4 oxide layer.**

During the optical examination of the Zircaloy-4 samples, using the microscope available in the shielded box-line, black/blueish precipitates were found on the inner surface of the cladding (see Fig. 7). According to previous studies on similar precipitates on the inner surface of plenum claddings these precipitates consist of cesium. To determine the characteristics of these precipitates XAFS investigations at the INE Beamline for Actinide and Radionuclide Science at ANKA are foreseen with the irradiated and a non-irradiated Zircaloy-4 cladding.



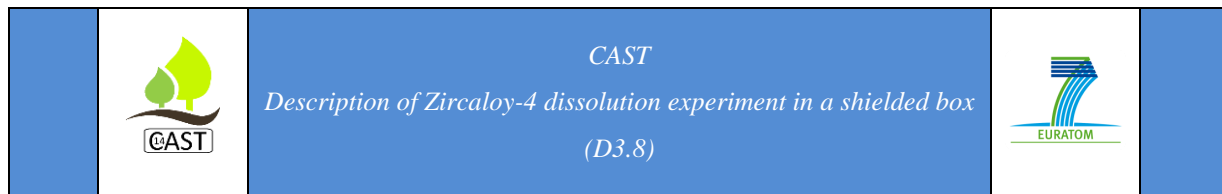
**Figure 7: Black/blueish precipitates seen on the inner surface of the irradiated Zircaloy-4 cladding.**



Table 4 summarizes the experimentally determined inventories of C-14, Cs-137 and Sb-125. These were further compared to our MCNP-X calculations. [YAMAGUCHI *et al.*, 1999] studied a hull specimen of a spent PWR fuel rod with a burn-up of 47.9 GWd/t<sub>HM</sub> which is similar to the burn-up of the PWR fuel rod segment used in this study (50.4 GWd/t<sub>HM</sub>). The experimental results obtained in this study for C-14 and Sb-125 are in good agreement with our calculations and in the case of C-14 also with literature data of [YAMAGUCHI *et al.*, 1999]. The experimental Cs-137 inventory exceeds by a factor of 117 the calculated value. The excess of Cs-137 is related to the precipitation on the inner Zircaloy-4 surface. The volatile cesium is released during reactor operation from subjacent UO<sub>2</sub> pellets and precipitates in the cooler plenum part of the fuel rod segment. The additional Cs-137 inventory is not taken into account in the MCNP-X calculations.

**Table 4: Results of the inventory analyses.**

radionuclide	C-14 [Bq/g]	Cs-137 [Bq/g]	Sb-125 [Bq/g]
measured contents in Zyr-4 of segment SBS1108–N0204 (50.4 GWd/t <sub>HM</sub> ) [p.w.]	$3.7(\pm 0.4) \times 10^4$	$3.4(\pm 0.3) \times 10^6$	$2.4(\pm 0.2) \times 10^5$
calculated contents in Zyr-4 of segment SBS1108–N0204 (50.4 GWd/t <sub>HM</sub> ) [p.w.]	$3.2 \times 10^4$	$2.9 \times 10^4$	$2.6 \times 10^5$
measured contents in Zyr-4 of spent PWR fuel rod hull specimen (47.9 GWd/t <sub>HM</sub> ) [YAMAGUCHI <i>et al.</i> , 1999]	$3.0 \times 10^4$		



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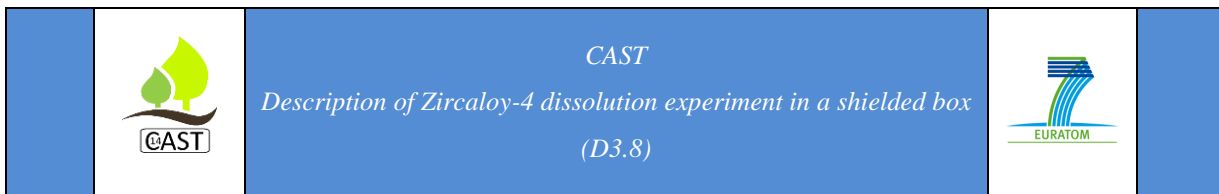
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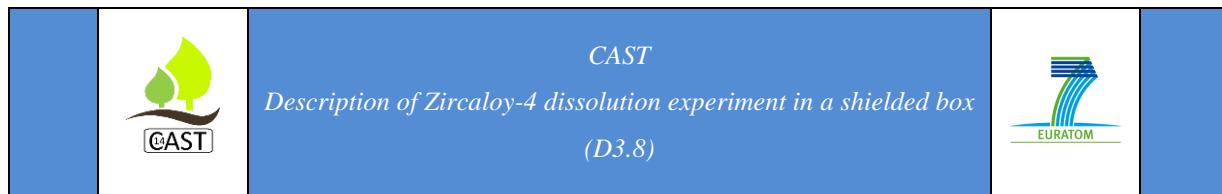
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