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Experimental contribution to the corium thermodynamic modelling – The U–Zr–Al–Ca–Si–O system

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ABSTRACT

During a severe accident in a nuclear reactor, extreme temperatures may be reached ($T > 2500$ K). In these conditions, the nuclear fuel may react with the Zircaloy cladding and then with the steel vessel, forming a mixture of solid–liquid phases called in-vessel corium. In the worst scenario, this mixture may penetrate the vessel and reach the concrete underneath the reactor. In order to develop the TAF-ID thermodynamic database (www.oecd-nea.org/science/taf-id) on nuclear fuels and to predict the high temperature behaviour of the corium + concrete system, new high temperature thermodynamic data are needed. The LM2T at CEA Saclay centre started an experimental campaign of phase equilibria measurements at high temperature (up to 2600 K) on interesting corium sub-systems. In particular, a heat treatment at 2500 K has been performed on two prototypic ex-vessel corium samples (within the U–Zr–Al–Ca–Si–O system) with different amounts of CaO and SiO_2. The results show that depending on the SiO_2-content, the final configuration of the samples can be significantly different. The sample with the higher CaO-content showed a dendritic structure representative of a single quenched liquid phase, whilst the sample richer in SiO_2 exhibited a microstructure which suggests the presence of a liquid miscibility gap. Furthermore a new laser heating setup has been conceived. This technique allows very high temperature measures ($T > 3000$ K) limiting the interactions between the sample and the surroundings.

1. Introduction

During a severe accident in a nuclear power reactor, a mixture of solid and liquid phase called corium may form inside the reactor vessel due to the chemical interaction between the fuel UO_2, the Zircaloy cladding and the steel vessel. The high temperature behaviour of the chemical system U–Zr–Fe–O strongly affects the accidental scenario proceedings. In the case of loss of the mechanical strength of the reactor steel vessel, the molten corium may reach the concrete (Al_2O_3–CaO–SiO_2) in the cavity underneath the vessel: in this scenario the more complex U–Zr–Fe–Ca–Si–Al–O system is concerned. Many phenomena take place during Molten Corium Concrete Interaction MCCI (Journeau and Piluso, 2012): high temperature concrete decomposition, heat transfer due to gas bubbles agitation, formation of several phases, oxidation of metals, etc. Several studies on the interaction between corium and concrete were published starting from the 1980s; in particular, numerous large-scale experiments were performed on this subject. Journeau and Piluso (2012) reported an exhaustive list of the early MCCI experiments as well as more recent MCCI projects. All these studies gave valuable data for a better comprehension of the phenomena occurring after the interaction between molten corium and concrete. However extensive uncertainties still exist on various aspects of MCCI scenarios. The complexity of the concerned chemical system, the extreme temperature and pressure conditions and the elevated number of possible accidental scenarios make the MCCI studies rather intricate.

Although large-scale experiments give information on macroscale phenomena, such as ablation profile, corium flooding behaviour, coolability of the molten core, density and viscosity evolution, the interpretation of the sample microstructure is challenging. In fact, the large dimensions of the samples cause temperature and composition gradients between the centre and the periphery of the sample as shown for example by Journeau et al. (2001).

In this framework a campaign of small-scale experiments is ongoing at the Laboratoire de Modélisation, de Thermodynamique

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et de Thermochimie (LM2T), CEA Saclay. At the moment, two series of experiments were performed:

- The first series was dedicated to the study of the liquid immiscibility in the U–Zr–O ternary system. The liquid miscibility gap has been studied in the past (Juenke and White, 1970; Guéneau et al., 1998). However, some experimental and modelling discrepancies still exist, in particular on the extension of the immiscibility region (Guéneau et al., 1998; Chevalier et al., 2004). Novel experimental results allowed to obtain two tie-lines in the ternary miscibility gap. Details on this series of experiments will be soon published in a separate paper (Quaini et al., submitted for publication).
- The second series of small-scale experiments was devoted to the investigation of the corium/concrete interaction. In particular, two samples have been investigated for a better comprehension of the interaction between a simplified corium and concrete. The experimental details and the results on this study are reported in the present paper.

In parallel the development of TAF-ID thermodynamic database is ongoing. This database is developed in the framework of an OECD–NEA project (Organisation for Economic Co-operation and Development–Nuclear Energy Agency). Several groups of research gather their calculation tools and thermodynamic database into the TAF-ID project. At the present stage Canada, France, Japan, The Netherlands, Republic of Korea and USA actively contribute to the development of the TAF-ID database. The main objective of the TAF-ID project is to assure a comprehensive and reliable international thermodynamic database for the calculation of phase diagram and thermodynamic properties of advanced nuclear fuel, fission products and structural materials (steel, Zircaloy, concrete). The TAF-ID database can be coupled with severe accident codes to predict the high temperature behaviour of the corium + concrete system during an accidental scenario.

Finally, the innovative high temperature setup called ATTILHA, conceived and developed at the LM2T will be presented. This experimental facility will allow precise measurement in a wide temperature range (1500–3000 K). The coupling of a laser heating technique with an aerodynamic levitation setup will allow investigating systems whereof contactless conditions are paramount.

2. Materials and methods

During the present work, heat treatments were performed in a Joule effect furnace (W-resistor) represented schematically in Fig. 1. This furnace is usually employed for high temperature mass spectrometer measurement (Baichi et al., 2001). The annealing tests are performed under very high vacuum ($p \sim 10^{-7}$ Pa). The temperature is monitored by using a calibrated IMPAC ISR-900 two-channel optical pyrometer.

Two samples within the U–Zr–Al–Ca–Si–O system were investigated. These samples are constituted by UO_2–ZrO_2–Al_2O_3–CaO–SiO_2 to simulate the interaction between a simplified corium (UO_2–ZrO_2) and concrete (Al_2O_3–CaO–SiO_2). During the present work, Fe was not considered.

The initial compositions of the samples were calculated by A. Boulin (LPMA, CEA Cadarache) with TOLBIAC-ICB software (Spindler et al., 2006). The calculation was performed assuming an oxidised corium only composed by UO_2 (80 tons) and ZrO_2 (20 tons). The whole mass of molten corium at 2950 K pours onto the concrete slab. Two types of concrete were considered: a limestone concrete (rich in CaO) and a siliceous concrete (rich in SiO_2). The composition of the samples corresponds to that of the liquid pools (CORIUM_1 for the interaction with limestone concrete,

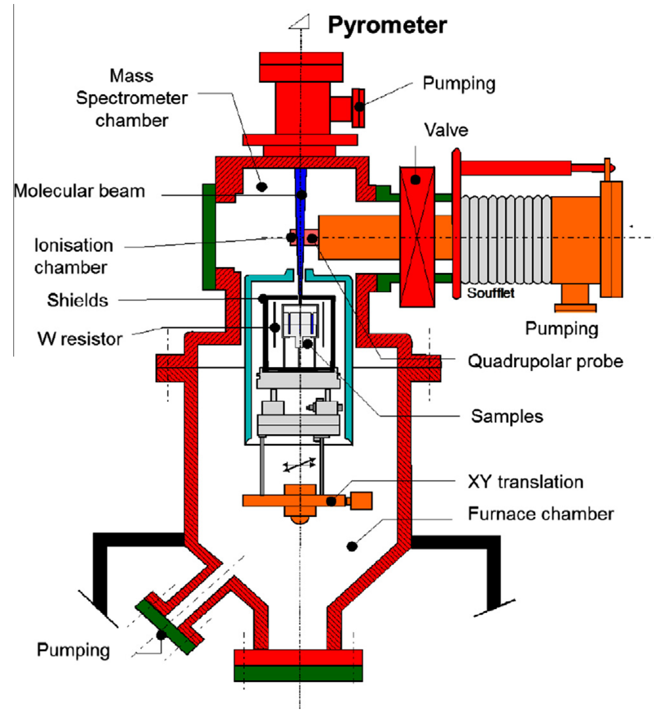

Fig. 1. Schematic view of the furnace employed for the annealing tests.

CORIUM_2 for the interaction with siliceous concrete) 24 h after the corium/concrete interaction (CCI). The aim is to study the effect of the concrete composition (i.e., the SiO_2 content) on the final configuration of the samples.

The samples were directly fabricated by mixing the starting materials in W-crucible (with screwed lid). The starting materials were Al_2O_3 powder (Alfa Aesar, 99.997%), CaO powder (SCM, 99.9%), SiO_2 spheres (SCM, 99.995%), UO_2 powder (CEA supply, 99.996%) and ZrO_2 powder (Goodfellow, purity 99.9%). The compositions of the samples are reported in Table 1.

The samples underwent a heat treatment for 30 min at 2500 ± 25 K and then they were cooled (3 K/s). The samples were then analysed at the LMAC, CEA Marcoule. For the characterisation a Carl Zeiss Merlin scanning electron microscope equipped with a Field Emission Gun (SEM-FEG) was used. It is coupled with energy dispersion spectroscopy (EDS, Oxford Instruments X-MAX 80 mm^2) and a wavelength-dispersive X-ray spectroscope (WDS, SX 100 CAMECA). The standards used for the analyses were UO_2 for uranium, $CaSiO_3$ for calcium and oxygen, SiO_2 for silicon, Zr for zirconium, W for tungsten and Al for aluminium.

3. Results and discussion

In the following, the experimental results obtained on the two investigated samples are reported. The differences observed in the samples will be discussed.

Table 1
Compositions of the in vessel corium + concrete samples (in mass percentage).

	CORIUM_1	CORIUM_2
Al_2O_3	1.8	2.1
CaO	32.4	11.0
SiO_2	24.6	64.0
UO_2	30.8	15.4
ZrO_2	10.4	7.8