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Experimental Measurements of Thermophysical Properties of Several Corium Compositions and Influence on Fuel-Coolant Interaction

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ABSTRACT

During a nuclear severe accident, the melting of the fuel leads to the formation of a complex mixture, the so-called corium. Corium thermophysical properties are of major importance for all severe accident phenomena and their knowledge is still partial today. To reduce uncertainties in severe accident codes for simulation, a more reliable knowledge of corium properties is necessary; the impact of corium thermophysical properties on severe accident phenomena is illustrated through the influence of surface tension on Fuel-Coolant Interaction (FCI) physical mechanisms.

Concerning corium thermophysical properties, reliable existing data are scarce. For example, for surface tension, some data are available for pure zirconium and very few for the main corium mixture $U_{1-x}Zr_xO_{2-y}$. The PLINIUS severe accident platform (CEA-IRESNE), located at CEA Cadarache, is devoted to experimental studies concerning corium phenomenology and its modelling. It involves several specifically designed facilities, able to reach high temperature. In particular, the experimental device -so-called VITI- is dedicated to analytical studies of thermophysical properties at liquid state. In order to improve knowledge on corium thermophysical properties, a specific experimental device -VITI-Maximum Bubble Pressure- has been developed.

In this work, the experimental approach, based on the Maximum Bubble Pressure (MBP) method, to measure the surface tension and the density at very high temperature (T > 2000 °C) is presented. In-vessel corium compositions have been considered for this study with different rate of oxidation representative of reactor cases: from corium C0 -no oxidation of zirconium- till corium C100 -all zirconium has been oxidized. Results on density and surface tension of four in-vessel corium compositions and their assessment are presented.

1 INTRODUCTION

In the case of nuclear severe accident, the involved high temperature (T > 2000 °C) leads to the melting of the core and the formation of so-called “corium”. The corium composition evolves throughout accident progression and contains a mixture of nuclear fuel, melted metals and oxides and fission products. Numerous coupled multi-physical phenomena are involved during the severe accident progression. Some remain partially understood, even if the overall phenomenology is rather well known.

However, data on liquid corium thermophysical properties are scarce as measuring them at high temperature is an experimental challenge. Common measurement methods need to be adapted and raise the issue of chemical compatibility between materials, whereas instability and vaporisation of samples may occur with levitation methods. As high temperatures are involved, the composition of the sample may change during the test due to partial evaporation or contamination from the environment. As corium composition can be volatile according to experimental conditions, levitation methods are difficult to apply.

In this work, the “Maximum Bubble Pressure” (MBP) method is used at high temperature to measure the density and the surface tension of in-vessel corium compositions ($U_{1-x}Zr_xO_{2-y}$) with varying oxidation degree of zirconium, corresponding to several accident scenarios. After each test at high temperature, reliable measurements with SEM (Scanning Electronic Microscope) / EDS (Energy Dispersion Spectrometer) allow to know the real composition.

2 SEVERE ACCIDENT PHENOMENOLOGY AND CORIUM THERMOPHYSICAL PROPERTIES

2.1 Impact of thermophysical properties on severe accident phenomenology

In the frame of international projects, several nuclear safety assessment analyses have been performed after Fukushima Daiichi accidents in 2011, including response to severe accident conditions, mitigation tools, coping time. Results of these studies have shown that residual uncertainties still exist in the understanding and reliable modelling of severe accident. For example, the OECD/NEA/BSAF [1] and BSAF-Phase 2 [2] programs have shown still some discrepancies concerning the results of severe accident progression assessment for each reactor (Fukushima Daiichi 1-F1, 1-F2, and 1-F3).

Even if the overall severe accident phenomenology is relatively well known for both in and ex-vessel configurations, modelling of some physical phenomena needs to be enhanced to reduce uncertainties on severe accident calculations for reactor applications.

Several international sensitive analyses studies have been devoted to identify the main sources of uncertainties in severe accident code calculation and nuclear safety assessment. Among the identified sources, uncertainties on corium thermophysical properties have been assessed as being one important source of uncertainty on simulation as it has been shown in the OECD/NEA TCOFF-1 project [3]. Corium thermophysical properties uncertainties have impact on severe accident code simulation.

The uncertainties of corium thermophysical properties are coming from the lack of reliable knowledge due to the few experimental data and modelling that currently exist. Corium thermophysical properties measurement is highly challenging due to the temperature domain (T > 2000 °C) and the strongly corrosive corium behavior.

2.2 Application to Fuel-Coolant Interaction

Let us illustrate with an example the importance of thermophysical properties for a nuclear severe accident phenomenon. Fuel-Coolant Interaction (FCI) with its possible consequence steam explosion can occur during the progression of the severe accident. In this case, there is a first step of coarse fragmentation jet (3-phases) that can be followed by a fine fragmentation before a possible triggering and steam explosion [4]. Size distribution and Sauter diameter of the droplet will have huge impact on steam explosion energetics, and so to possible reactor damages. Among thermophysical properties, the surface tension has a key role in this phenomenon as it affects the corium jet fragmentation and more particularly the size of the droplets. The jet fragmentation directly depends on the critical Weber's number:

$$We = \frac{\rho V^2 D}{\sigma} \tag{1}$$

Where ρ, V, D and σ represents the density [kg.m^{-3}], the speed [m.s^{-1}], the droplet diameter [m], and the surface tension [N.m^{-1}], respectively.

In particular, for a low jet injection speed, corresponding to the nuclear reactor case, calculations have been performed with several values of surface tension. It has been recently demonstrated that the size of the droplets is divided by 3 when the surface tension is decreasing from 1 to 0.4 N.m^{-1} [5]. These results show the importance of surface tension for the jet fragmentation process.

2.3 Corium thermophysical properties measurement techniques

Reference data about corium density were measured during RASPLAV project [6]. Depending on the corium composition, two experimental methods were used: MBP for the less oxidized composition, and pycnometer method for totally oxidised corium (C100). In this last method, the volume of the crucible is precisely known, and the mass of the melt inside is determined after crystallisation. This method is rather simple, but there is an uncertainty on the volume due to the unknown shape and volume of the meniscus when the sample is liquid.

For less oxidised corium compositions (C22 and C32), the density obtained with MBP method ranges between 7630 and 8200 kg.m^{-3} depending on corium composition and temperature (Table 1). The surface tension of these compositions was also measured. MBP method gives precise results for surface tension but is less precise for density, thus the uncertainties on this property are higher than that obtained with the pycnometer method. Despite the discrepancy of the values and their high uncertainties (up to 25 %), this property seems to decrease when the zirconium oxidation degree $(n_{ZrO_2}/(n_{Zr}+n_{ZrO_2}))$ increases (Table 2). More recently, thermophysical properties of corium C32 (32 mol.% of oxidised zirconium among total zirconium content, U/Zr ratio = 1.2) have been measured [7] with MBP method. Surface tension value (Table 2), measured at liquidus temperature (2470 °C) is higher than the results from the RASPLAV project (+0.100 N.m^{-1}, which represents around 11 %) [6]. The sample composition is a source of uncertainty as a small change may have a significant effect on surface tension: for example, even a very slight oxygen pollution has been shown to be enough to drastically decrease the surface tension of metals [8,9]. Due to the high temperature involved, volatilisation and crucible-sample interaction are likely to occur during the test. A modification of the composition during the test could explain the discrepancies between values measured with the same method, especially as the sample composition was not assessed after test in these references.

Table 1: Experimental density data on corium in the literature.

Corium	Zr oxidation degree	U/Zr ratio	Temperature [°C]	Density [kg.m^{-3}]	Experimental method	References
C22	22 %	1.63	2600	8200 ± 1000	MBP	[6]
C32	32 %	1.2	2500-2600	7630 ± 900		
			2470	7850 ± 240		[7]
C100	100 %	1.52	2700	7410 ± 20	Pycnometer	[6]
			2900	7310 ± 20		
			3100	7230 ± 20		

Table 2: Experimental surface tension data on corium in the literature.

Corium	Zr oxidation degree	U/Zr ratio	Temperature (°C)	Surface tension (N.m^{-1})	References
C22	22 %	1.63	2600-2630	0.668 ± 0.100	[6]
			2585-2630	0.750 ± 0.117	
C32	32 %	1.2	2450-2600	0.510 ± 0.135	
				0.593 ± 0.127	
			2470-2850	0.663 ± 0.063	
			2470	0.749 ± 0.015	[7]
C100	100 %	1.63	2685-2700	0.573 ± 0.150	[6]
		1.8	2790	0.569 ± 0.046	[10]

3 MATERIALS AND METHODS

3.1 Experimental layout of VITI facility

At CEA Cadarache, PLINIUS platform is dedicated to nuclear severe accident studies with prototypic corium -i.e. with depleted uranium dioxide- and contains several experimental devices. One of them, VITI facility is very versatile thanks to its various configurations [7, 10-12]. This device, shown in Figure 1, is composed of a water-cooled sealed steel enclosure (8). The heating is ensured by an inductor (6) connected to a power generator. Indirect induction is used for measurements: an electromagnetic field coupling happens between the inductor and the graphite susceptor (3). The crucible and the sample are then heated by thermal radiation from the susceptor. Thermal shield (5) limits radiative heat loss. Two bi-chromatic pyrometers (11) allow to measure the temperature of the crucible through dedicated windows (7) and holes in the susceptor and in the thermal shield.

The configuration used in this work is VITI-MBP, based on maximum bubble pressure method to measure density and surface tension of materials at high temperature (Figure 1).

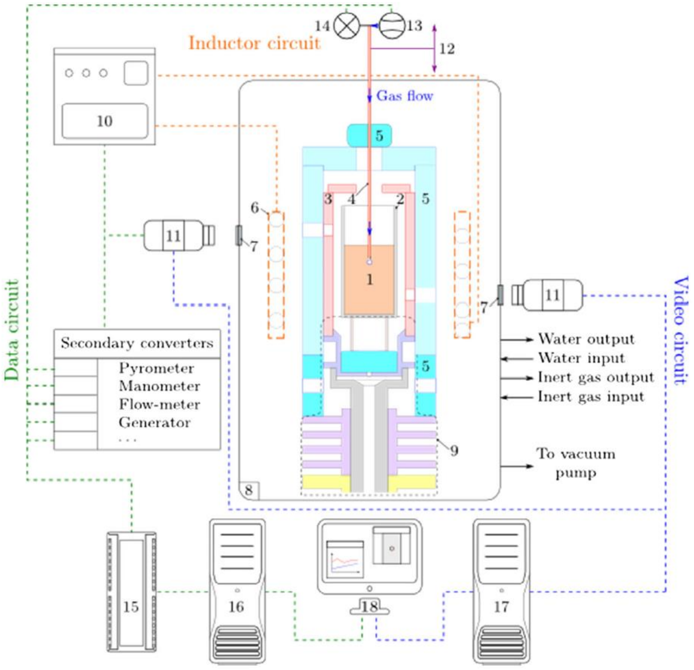


Figure 1: VITI-MBP experimental device: (1) liquid sample, (2) crucible, (3) susceptor, (4) capillary tube, (5) thermal shield, (6) inductor, (7) windows, (8) confinement vessel, (9) crucible support system, (10) generator, (11) bichromatic video-pyrometers, (12) micrometer translation stage, (13) flowmeter, (14) pressure sensor, (15-18) data acquisition [6]

Argon gas, controlled by a flowmeter (13), flows through a capillary tube (4), immersed in the liquid material. The position of this capillary tube is precisely controlled using a micrometric translation table (12). A pressure sensor (14) is measuring the pressure of the capillary tube. Collected data are transmitted to a dedicated computer (16) through a data bus (15). This computer also controls experimental devices. Another computer (17) is dedicated to video data, obtained by the video-pyrometers.

3.2 Data post-treatment

According to Young-Laplace equation, the immersion depth of the capillary inside the liquid sample, h [mm], is related to the pressure Δp [mbar] in the following manner:

$$\Delta p = \frac{2\sigma}{r_b} + \rho g h \tag{2}$$

where r_b represents the bubble radius [mm], g the standard acceleration [m.s^{-2}] ρ the density [kg.m^{-3}], and σ the surface tension [N.m^{-1}].

3.3 Sample preparation and test conduct

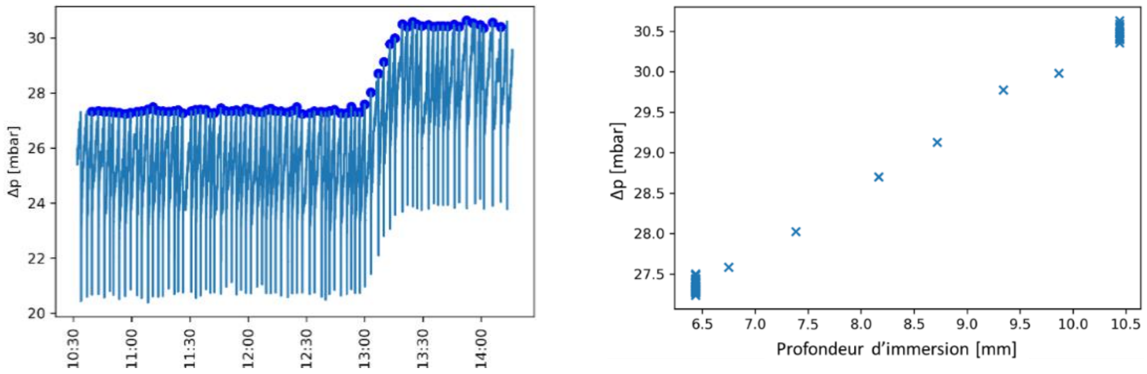
For each corium composition considered, one sample was tested. It was obtained by mixing zirconia powder, zirconium pellets and depleted-uranium dioxide powder. Thermodynamic calculations, performed with ThermoCalc software and TAF-ID and NUCLEA databases, allowed to estimate liquidus temperatures of our corium compositions and thus to deduce testing temperatures, just above the liquidus.

Typical test conduct consists in heating the sample above the estimated liquidus temperature and to insert the capillary tube inside the liquid sample. During the bubble formation and growth, the pressure increases and reaches its maximum when the bubble radius, r_b, is equal to the capillary radius, r_{cap}. By measuring this pressure increase at several immersion depths of the capillary tube inside the liquid sample, it is then possible to derive its density and surface tension by using the Young-Laplace equation. As two properties are measured, at least two immersion depths of the capillary tube are required; the pressure increase due to bubble formation during the path in between is also recorded to confirm the slope of the curve obtained.

4 CORIUM THERMOPHYSICAL PROPERTIES RESULTS

For each temperature, the overpressure peaks measured for a given immersion depth of the capillary were quite reproducible, as shown Figure 2a. This can also be deduced from the limited spread of the pressure points for an immersion depth (Figure 2b at both ends of the curve). During the descent of the capillary between two fixed depths, the measured points were well aligned in a straight line. This ensures the quality of the test performed and of the results.

After each test, SEM/EDS analysis were performed in order to determine the exact corium composition: at these high temperatures (T > 2000 °C), volatilisation may occur. Initial and final compositions of each corium are given in Figure 3. For less oxidized compositions (C0 and C30), the U/Zr ratio decreased, indicating uranium loss, and the zirconium oxidation degree increased during the test. The composition of the C50 corium was the only one that did not change significantly. For the C70 corium, the U/Zr ratio increased, indicating a loss of zirconium, and the zirconium oxidation degree increased. To compare the density evolution with the temperature between corium compositions, the test temperature is divided by the liquidus temperature of each corium, estimated with ThermoCalc and TAF-ID database.



a) b)

Figure 2: Capillary tube pressure a) depending on time, with peaks detection, and b) according to its immersion depth, during a test on C0 corium at 2592 °C

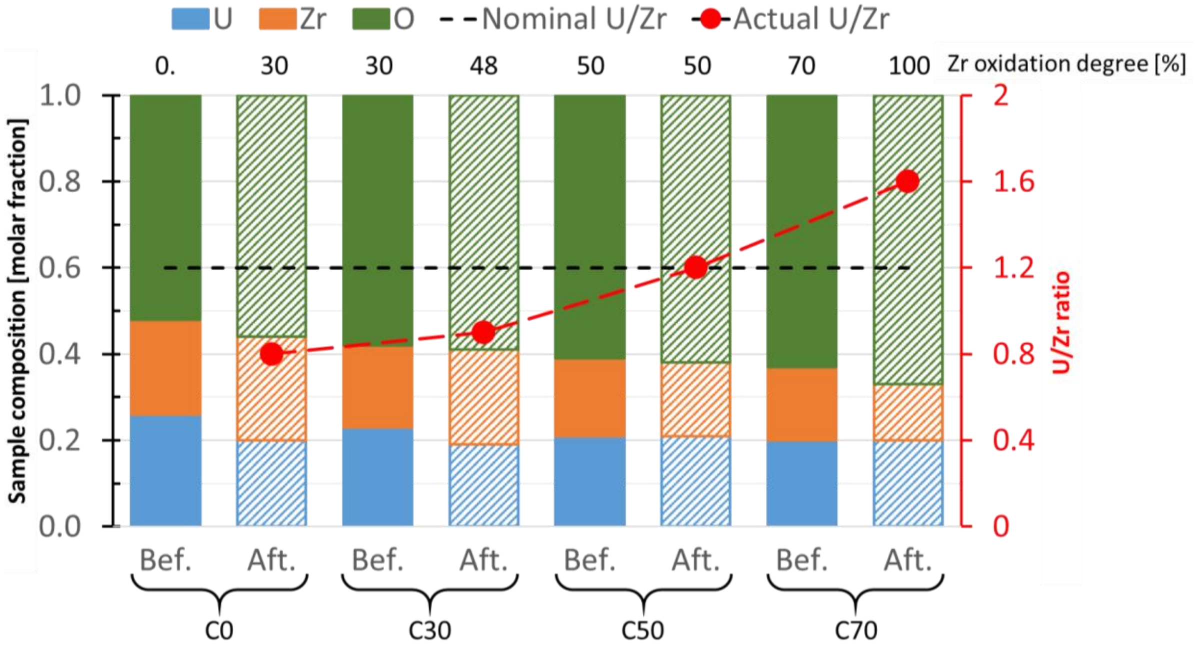
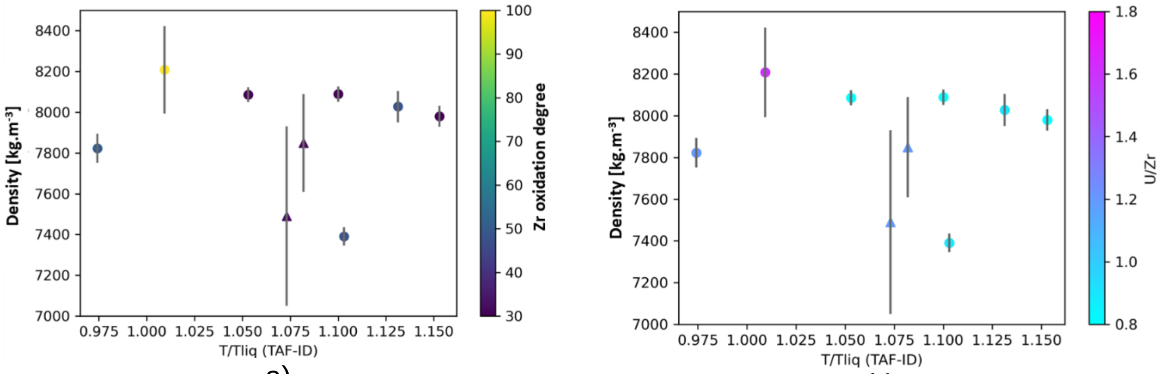


Figure 3: Corium sample composition, before and after test

For the C0 corium composition, data are acquired at several temperatures and a density decrease can be observed with temperature increase (Figure 4). For the other compositions, as data were acquired for only one or two temperatures, the density dependency on temperature was not clearly observed. Values measured for the density of corium range between 7400 and 8400 kg.m^{-3}. Moreover, the increase in oxidised zirconium content seems to make the corium density decrease (Figure 4a), which would be expected as zirconium oxide has a lower density than metallic zirconium. However, as the U/Zr ratio also changes, it may increase the density when the uranium content increases as well (Figure 4b).

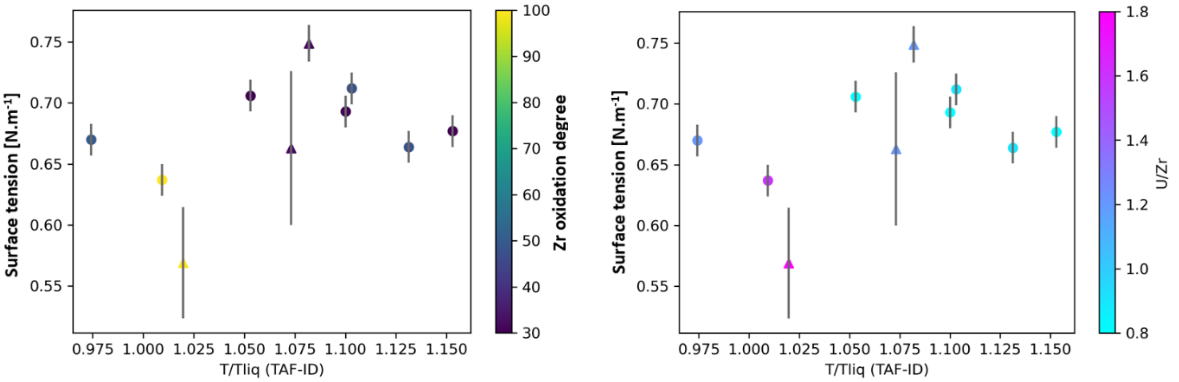


a)

b)

Figure 4: Measured (●) and literature (▲) density of corium depending on temperature and a) zirconium oxidation degree, b) U/Zr ratio; liquidus temperature calculated with TAF-ID database

Regarding the surface tension, measured values range between 0.72 and 0.63 N.m^{-1} and a decrease is observed with temperature increase. The increase of zirconium oxidation degree also seems to make the surface tension decrease (Figure 5). This behaviour would be expected as oxygen is known for its tension-active role [8-10]. In addition, it can be observed the higher U/Zr ratio the lower surface tension and the U/Zr ratio seems to have a stronger impact on the surface tension than that of the oxygen content. Nevertheless, U/Zr ratio and oxygen content may have linked effects as corium with higher oxide content also have higher U/Zr ratio in our measurements.



a)

b)

Figure 5: Measured (●) and literature (▲) surface tension of corium depending on temperature and a) zirconium oxidation degree, b) U/Zr ratio; liquidus temperature calculated with TAF-ID database

5 CONCLUSION

In this work, density and surface tension of corium at liquid state were measured at high temperature with the MBP method. Some corium compositions were considered, with fraction of oxidised zirconium ranging from 0 to 70 % of total zirconium content. Post-test analysis revealed a modification of the composition during the test, with an increasing U/Zr ratio along with the zirconium oxide content. This source of uncertainty has been evaluated for thermophysical properties of corium for the first time.

The density of liquid corium seems to decrease with the temperature increase. The U/Zr ratio and the zirconium oxidation degree also have an effect on this property, even if their effect could interfere: an increase in zirconium oxidation degree tends to lower the density while an increasing U/Zr ratio increases the density. Concerning the surface tension, it lowers with the temperature, the zirconium oxidation degree and the U/Zr ratio; this later seems to have the strongest impact. As for the density, the effects of these parameters may interfere.

However, these results expand significantly the existing data on corium thermophysical properties. More experimental data would be required to confirm these trends, to dissociate the effect of U/Zr ratio and zirconium oxidation degree on thermophysical properties and to obtain reliable temperature variation laws for these properties. Implementation of reliable thermophysical properties laws would be necessary for severe accident codes to decrease the level of uncertainty.

ACKNOWLEDGMENTS

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