Needs for large mass prototypic corium experiments the PLINIUS-2 platform

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NEEDS FOR LARGE MASS PROTOTYPIC CORIUM EXPERIMENTS: THE PLINIUS-2 PLATFORM

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ABSTRACT

Corium is the molten material formed after meltdown of a nuclear reactor core during a severe accident. In order to improve the understanding and modelling of corium behavior, experiments are needed both for LWRs and GenIV fast reactors. Experiments using low temperature simulant materials, thanks to lower costs and constraints, allow the testing of a larger number of configurations and the determination of correlations. But some crucial corium phenomena cannot be reproduced at low temperatures such as the importance of radiation heat transfer or the presence of a large (up to 1000 K) liquidus-solidus interval. Consequently, some experiments are performed with high temperature simulant materials: alumina thermite as well as refractory oxides. However, it is not feasible to simulate all the aspects of corium phenomenology, especially its high temperature physico-chemistry. Therefore, even though the use of depleted uranium implies a series of protective and regulatory measures, the need for prototypic corium experimented is supported through several examples
Another important aspect of experiment design deals with scaling. Small or medium scale corium experiments are easier to operate and only a few large scale (>100 kg) facilities have been built. Several effects are only visible with significant masses, as for instance, the formation of a corium cake during FCI or all the phenomena controlled by crust strength, such as underwater spreading or corium jet ablation. CEA is currently designing a new large prototypic corium platform: PLINIUS-2 for both LWR and SFR corium experimental research. Its main characteristics will be presented

KEYWORDS
Corium – Experimental Facility – Prototypic Material – Scaling – Severe Accidents

1. INTRODUCTION

Corium is the molten material formed after meltdown of a nuclear reactor core during a severe accident. In order to improve the understanding and modelling of corium behavior, experiments are needed both for LWRs and Gen IV fast reactors. This has been stressed for instance in Europe by SNETP in its Identification of Research Areas in Response to the Fukushima Accident [1].

Experiments using low temperature simulant materials (such as CORINE [2] for spreading or CLARA [3] for in-vessel pools) are widely used to test a larger number of configurations and the determination of correlations, thanks to lower costs and constraints. But some crucial corium phenomena cannot be reproduced at low temperatures such as the importance of radiation heat transfer or the presence of a large (up to 1000 K) liquidus-solidus interval. Consequently, some experiments are performed with high temperature simulant materials: alumina thermite is widely used (e.g. KATS [4] or EAGLE [5] facilities) but other high temperature oxides (as in DEFOR [6]) are also considered.
However, it is not feasible to simulate all the aspects of corium phenomenology, especially its high temperature physicochemistry. Therefore, even though the use of depleted uranium implies a series of protective and regulatory measures, we are convinced that experiments with prototypic material are absolutely needed.

Another important aspect of experiment design deals with scaling. Small or medium scale corium experiments are easier to operate and only a few large-scale (>100 kg) facilities have been built: FARO [7] in Europe, RASPLAV AW-200 [8] in Russia, VESTA [9] in Korea and ACE/MACE/CCI [10] in the USA. Several effects are only visible with significant masses, as for instance, the formation of a corium cake during FCI or all the phenomena controlled by crust strength, such as underwater spreading or corium jet ablation.

In a first section, the necessity to perform high temperature experiments with prototypic corium material will be discussed. Then, the issue of experimental scale will be considered. In a final section, the main characteristics of the future PLINIUS-2 prototypic corium experimental platform will be presented.

2. NEED FOR PROTOTYPIC CORIUM EXPERIMENTAL RESEARCH

2.1. Singularities of Uranium Oxide

The major component for the molten fuel is uranium oxide. The use of depleted uranium oxide for severe accident research allows being as close as possible to the real corium characteristics [11]. In this subsection, we are going to underline some specific properties of uranium oxide.

2.1.1. Electronic configuration and oxidation states of uranium

Uranium belongs to the actinide series. It has a specific electronic configuration: 5f3 6d 7s2 (Table 1). The existence of electrons in the 5f electronic band makes it possible to have located electrons. In the solid state, these electrons will determine the possible chemical bonds, particularly with oxygen. These chemical bonds control the chemical and physical properties of the uranium oxide. Uranium thus has many oxidation states: +3,+4,+5,+6 (Table1).

Table I. Electronic structure, oxidation states and ionic radii of uranium and some other elements [12]

Element	U	Al	**Zr**	**Hf**	Ce
Electronic structure	$5f^3$-$6d^1$-$7s^2$	$7s^2$-$6d^1$	$4d^2$-$5s^2$	$4f^{14}$-$5d^2$-$6s^2$	$4f^1$-5d1-$6s^2$
Oxidation state	0,+3,+4, +5, + 6	0, +3	0,+1,+4	0,+4	0,+1,+3,+4
Effective Ionic radius r (pm)	3+, r= 102.5 4+, r = 89 5+, r= 76 6+, r= 73	3+, r=53.5	+4, r=72	+4, r=71	3+, r=101 4+, r= 87

The diversity of the uranium electronic structure, oxidation states and ionic radii shows that it is one of the more complex elements of the periodic table, with no equivalent, except for some actinides such as plutonium. These specificities imply that uranium can form a great variety of oxides with different physical properties which strongly influence all possible states (e.g., gaseous species observed for example in VULCANO spreading experiment such as VE-U5 [13]).

2.1.2. Uranium-oxygen system

The uranium-oxygen phase diagram is quite complex [14]. Because of the numerous oxidation states, there are many possibilities for the formation of compounds.

- For the solid state, many compounds can exist. The main compounds are: UO_2, U_4O_9, U_3O_7, U_3O_8. Uranium dioxide actually present a large domain of non-stoichiometry from $UO_{1.70}$ to $UO_{2.25}$ in which the oxygen content may continuously vary. This diversity is really specific to the U-O system. All these compounds can be obtained during the course of a severe accident.

- For the liquid state, the experimental data are well-known in the UO_2-rich part. But for the oxygen solubility in liquid uranium, there are only a few data [14].

- For the gaseous state, the situation is also quite complex. The gas species which can exist are: UO, UO_2, UO_3 and U [14]. According to the conditions of pressure and temperature, and gas phase composition in a severe accident, it is possible to vaporize a significant amount of one or more of these species, especially UO and UO_3. This ability to form highly volatile species is specific to the uranium oxides whereas other oxides with high melting point are not volatile in the temperatures experienced during severe accidents.

These examples show the complexity and the specificity of the U-O system. The consequences on the studies of severe accidents phenomena must be properly assessed through specific

experiments (either separate effect test or integral) as it is not possible to reproduce this complexity by using simulants

2.1.3. Thermophysical properties of uranium oxide

To simulate the uranium oxide, some properties are fundamental for studies of spreading, cooling or stratification of corium: melting temperature, thermal conductivity, heat capacity and density. The best known non-radioactive high melting-point oxides that are considered as potential simulants of uranium oxide are Al_2O_3, HfO_2, CeO_2, ZrO_2

As an example, table 2 presents the density and the melting temperature of these oxides compared to UO2. As shown, the melting temperature and the density of HfO2 are very close to UO2. In general, the thermodynamic properties of these oxides, except ZrO_2, are not part of the nuclear material properties and thermodynamic databases. The densities of the other oxides are quite far from that of uranium dioxide.

It is possible to simulate one or the other of the properties of UO_2, but not all properties together. For instance, HfO_2 will be a good simulant of UO_2 hydrodynamics in mixtures with metals from the hydrodynamic point of view because of its high density. ZrO_2 could be a simulant in oxidic mixtures for some studies of spreading because it simulates the refractory behavior of UO_2 in the freezing process, from the thermodynamic point of view. However, both HfO_2 and ZrO_2 are very stable oxides and react differently with other materials and with the gas phase.

Table II. Physical properties of oxides in comparison with UO2.

Oxides	Melting point (K)	Density (293K) ($g.cm^{-3}$)	Specific heat ($J.K^{-1}.kg^{-1}$)	Latent heat fusion ($kJ.kg^{-1}$)
UO_2	Tm = 3120 ± 30	10.956	~300	260
CeO_2	3073	7.10	~300	465
HfO_2	3031 ± 25	9.68 (monocl.)	~300	457
ZrO_2	2973	5.68 (monocl.)	~300	810
Al_2O_3	2072	3.95	~700	1140

2.2 Experience with simulant and prototypic corium

Alumina (Al_2O_3) has often been used as a high temperature simulant of corium, either for pure oxidic melts or for oxide and metal melts obtained by aluminothermite. In the KROTOS facility, a large series of experiments have been made with alumina and with (UO_2-ZrO_2) melt interacting with water. It has been observed that alumina jets exhibit about one order-of-magnitude larger explosion efficiencies than prototypic corium jets [15, 16]. Different explanations have been proposed:

- A coherent corium melt pour tends to penetrate deeper into the water pool than an alumina pour [17];

- Comparisons of debris morphology in tests where no explosions were observed, show that the debris resulting from the alumina pours is significantly different from the finely fragmented corium debris with a mass mean diameter of about 1.7 mm (e.g. KROTOS 45) compared to larger alumina debris (e.g. 17 mm mass mean diameter in KROTOS 41) [17];
- Different droplet solidification patterns are expected between opaque corium droplets and semi-transparent alumina particles [18];
- The oxidation of UO_2 by steam produces incondensable gases that may blanket the droplets and prevent explosions, but these exothermal oxidation reactions may also increase the explosion yield [19, 20];

Unfortunately, these differences are not limited to alumina, but other potential simulants such as zirconia (ZrO_2) have shown different behavior with respect to drop solidification and debris shape, leading to differences in "triggerability" and explosion yields according to Park et al. [21].

All these experiences confirm the necessity to perform validation experiments with prototypic materials for Fuel Coolant Interaction. In the absence of contrary experimental evidence, we must assume that similar limitations on the use of simulant materials for fuel-sodium interactions.

Similar issues clearly arises for experiments where mass transfers between oxide and metal phases is important: studies about crust formation and solidification [22] , about stratification both in-vessel [23] and ex-vessel [24], about the evolution of metallic layer during Molten Core Concrete Interaction [25],… can also not be realized with simulants, since we do not have direct measurement of the corium properties (evolving with compositions) on which to apply any scaling law. This is also the case for all the phenomena which are mainly controlled by corium crust strength, such as underwater spreading [26, 27], since only limited data exists on the mechanical properties of such corium crusts [28].

3. NEED FOR LARGE SCALE EXPERIMENTS

At reactor scale, more than 100 t of corium (occupying more than 10 m^3) can be molten during a severe accident. Typical lower head diameters of the order of 4-5 m and reactor pit diameters of the order of 6 m, lead to corium pool depths between several decimeters and a few meters (including metal layer and dissolved concrete). It is clearly impossible to conduct experiments at such a large scale, especially with prototypic materials. Scaling is thus an important issue.

Due to the strong coupling between phenomena present during severe accidents, it is not possible to find a simple scaling law between laboratory scale and reactor scale. The role of computer codes and models is therefore important in order to transpose experimental results to reactor safety applications. Nevertheless, care must be given to ensure that the relative values of important characteristic times (or dimensions) are maintained so that similar regimes are considered during experiments and accidents.

3.1 Fuel Coolant Interaction

Large masses Fuel Coolant Interaction experiments are quite important to ensure that steady state jets are reached, while small scale experiments, although useful, are limited to the initial transients. For instance, whereas all the corium has been fragmented in small scale KROTOS experiments, a solid cake has been found in most of the 100-150 kg FARO tests [7].
Steady-state is achieved after the jet reaches the jet breakup length or the bottom of the test section. It is therefore recommended that the melt mass corresponds to a notional cylinder of the jet diameter section and of a few (typically 3) jet breakup length to analyze steady-state interaction.

Several correlations have been proposed to estimate the jet breakup length L_{brk}:

Saito et al. [29] $$\frac{L_{brk}}{D} = 2.1\left(\frac{\rho_j}{\rho_c}\right)^{0.5} Fr^{0.5} \quad (1)$$

Epstein & Fauske [30] $$\frac{L_{brk}}{D} = \frac{1}{2E_0}\left(\frac{\rho_j}{\rho_c}\right)^{0.5}; E_0 = 0.06 - 0.12 \quad (2)$$

Seiler & Payot [31] $$\frac{L_{brk}}{D} = \left(\frac{0.7^2}{3\pi}\right) V_j \frac{{\rho_j}^{0.5}}{(\rho_c.g.\sigma_j)^{1/4}} \quad (3)$$

where D is the jet diameter, ρ the density, V_j the jet velocity, Fr the Froude number $\frac{V_j}{\sqrt{gD}}$, σ the surface tension and the subscript j and c respectively refer to jet and coolant.

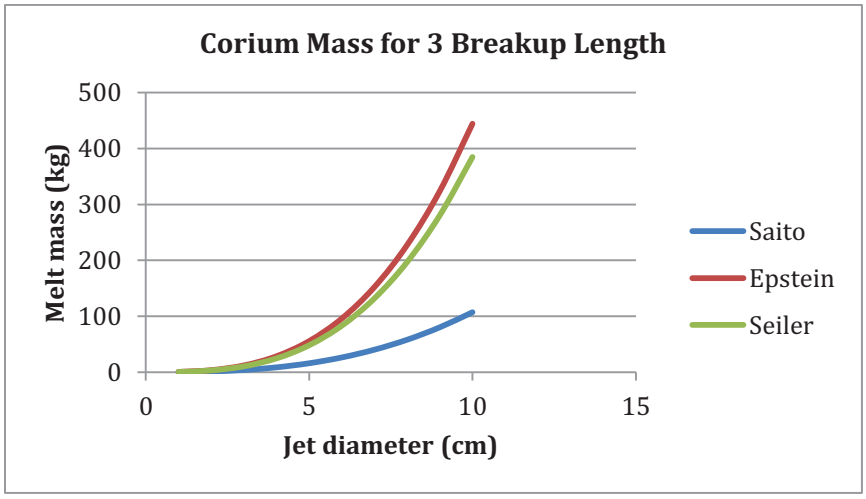

Figure 1. Estimation of masses (in kg) corresponding to 3 times break-up length of jets in water

Figure 1 presents the masses estimated from these 3 correlations which correspond to three times the breakup length in water. The calculation has been presented here for a typical jet velocity of 4 m/s, from FARO tests [32] and using a coefficient E_0=0.06 for Epstein & Fauske correlation (2). In order to study prototypic jet diameters around 10 cm, it appears that several hundred kilograms are necessary.

For corium-sodium interaction, calculations give slightly larger values, up to 500 kg for a 10-cm diameter jet with Epstein & Fauske correlation, due to the slightly heavier corium (fuel without zirconia) interacting with a slightly lighter coolant, sodium. It must be noted that 500 kg is also the order of magnitude of the mass of one subassembly (fuel + fertile + absorbers + structures) considered in current ASTRID core design [33].

To reach steady-state melt-coolant interaction, it is necessary that the corium jet is longer than the fragmentation length. As we plan to study jets of the order of 10 cm, melt masses of several hundreds of kilograms are necessary. Moreover, FARO and KROTOS experiments have shown than, whereas all the jet were fragmented in small mass corium-water experiments like KROTOS, a significant part of the jet formed a cake for jets above 100 kg [7]. This confirms of target of several hundred kilograms for PLINIUS-2.

3.2 Corium spreading experiments

For the dry spreading of corium-concrete mixtures, which is controlled by the increase of viscosity in solidifying melt, it has been shown that medium scale experiments (in the 40 kg range) provide sufficient information for transposition to reactor scale [36]. This will not be the case for underwater spreading which is controlled by the mechanical strength of surface crusts.
In this case, it is necessary that the spreading channel width is larger than the maximum corium height in order to prevent any significant influence of the horizontal dimension in crust resistance.
It is expected that underwater spreads will be thicker than spreads in air, due to thicker crusts. In FARO spreading tests L26S and L-32S, the average spread height was below 10 cm for more than 100 kg of $(U,Zr)O_2$ [34]. If larger thicknesses, around 20 cm are to be expected, a test section width of about 40 cm will be necessary. For a spreading length longer than the width, set to 75 cm, this leads to a volume of 60 L or 480 kg of corium having a density of 8000 kg/m^3.

3.3 Corium pool experiments

It has not been possible to draw scaling laws to be applied to corium pool experiments, due to the large number of phenomena which are coupled during these tests: natural or forced convection flows, heat transfer, crust formation/dissolution, thermochemistry, crust stability… One of the major concern lies with the risk of unprototypic anchoring of crusts. Unfortunately, it appeared that surface crust anchoring has been observed even in large scale test sections, such as in MACE test M3b [35] with a corium mass of 1950 kg in a 120 x 120 cm test section. Increasing the corium mass in order to guarantee prevention of crust anchoring would thus lead to very large masses (of several tons) requiring large decay heat simulation power and potentially generating large amounts of flammable gases. A preferred approached lies with the design of engineered features dedicated to the prevention of stable crust anchoring. Molten pool sizes of 30 cm, such as those of SSWICS tests [28] are considered necessary if mechanical crust breaking is considered. This corresponds to corium masses of at least 80-100 kg. Larger masses may be necessary for melts containing oxides and steel.

Nevertheless, corium pool size cannot be lowered to very small sizes. The presence of crusts at the interfaces (of the order of a few centimeters for typical heat fluxes around100 kW/m²) implies a minimum size of the test section so that the liquid phase dimensions are not smaller than those of the solid crusts. Moreover, it is advisable that the liquid pool contains several convection cells even along its surface smallest dimension. As convection cells have been observed on corium pool to measure a few centimeters, decametric liquid pools are needed. This leads to test sections of some decimeters in diameter, greater or equal to the size of the VULCANO MCCI test sections (~50 kg corium) [24]. Anyhow, the largest achievable mass would be advised. A 500 kg mass (about 60 L of melt) can correspond to a 42-cm diameter, 42-cm high cylindrical cavity.

4. THE PLINIUS-2 PROTOTYPIC CORIUM PLATFORM

CEA Direction of Nuclear Energy, considering the need for large-scale experiments with prototypic corium, both for the development of the ASTRID Sodium Fast Reactor demonstrator and for the safety of Gen II and Gen III Light Water Reactors has decided to launch the design of a new experimental platform, PLINIUS-2 [37]. This new platform will replace the current PLINIUS prototypic corium platform at CEA Cadarache.

A cold-crucible furnace in the 500-kg range will be a central facility in PLINIUS-2 platform as several hundred kilograms are necessary for the experimental program. It is currently under design by our partner ECM. This furnace can move to be installed above three experimental halls respectively dedicated to corium-sodium interaction, long term corium pool experiments (mainly for ablation or mitigation issues) and corium-water interaction.

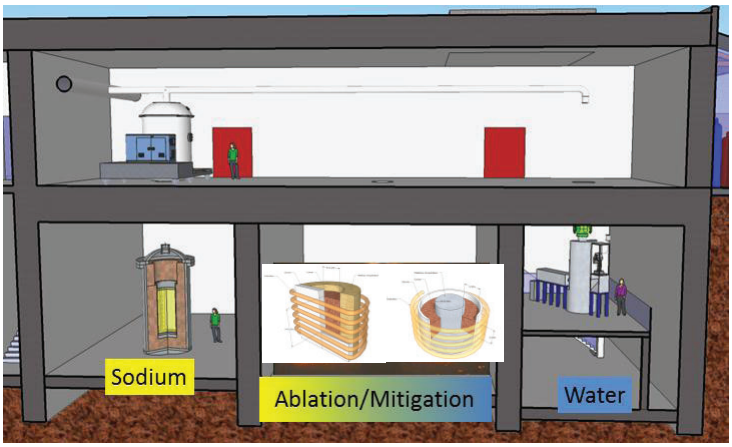

Figure 2. Schematic view of PLINIUS-2 Large-scale test facilities

Cold crucible induction heating is the most widely technology used for prototypic corium melting (RASPLAV [38], SICOPS [39], COMETA [40] SOFI [41], TROI [42], and VESTA[9]). Very high temperatures can be reached with this technology which also prevents any significant melt pollution thanks to the formation of a self-crucible. A complementary melting or pre-heating process has been selected: Self-propagating High-temperature Synthesis (SHS) usually named thermite reaction. This approach is routinely used to produce prototypic corium at ANL [10] and has already been tested in the current PLINIUS/VULCANO facility [43].

One of the advantages of the chosen technologies is the possibility to conduct experiments with molten oxides and molten steel.

4.1 Corium-Sodium Facility

The first objective of the corium-sodium facility will be to complete the FARO-TERMOS experimental program for Fuel-Sodium Interaction that had been terminated at JRC Ispra before conducting experiments with low-subcooling sodium [44]. Other points of interest lies with introduction of corium melt below the sodium free surface, simulation of ASTRID corium dispersion Channels and study of material transient ablation by jets.

It is planned to build two test sections:

- A large test section in which several hundred kilograms of corium will interact with about 1 m^3 of low-undercooling sodium (see section 3.1 for dimensioning rationale). The test section is made of three barriers (Figure 3):
  - A metallic liner to ensure sodium leak tightness;
  - A ceramic thermal barrier;
  - A pressure vessel.

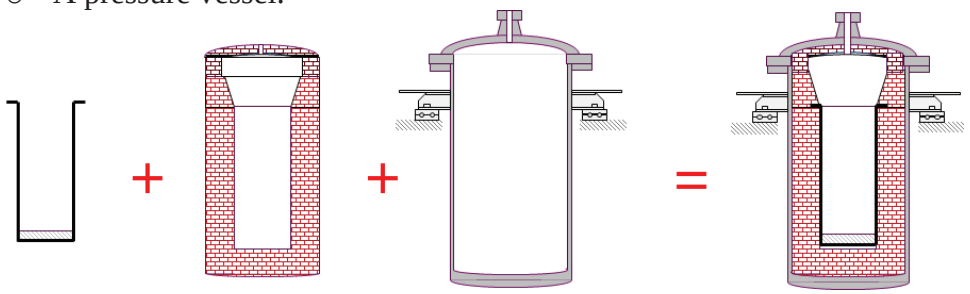

Figure 3. The three barriers of PLINIUS-2 Large-scale corium-sodium test section

- A smaller test section which will require a thinner vessel compatible with X-ray imaging (an adaptation of current PLINIUS/KROTOS imaging process [45] to the smaller corium-sodium debris) but will be limited to lower corium masses and/or larger subcooling.

Service devices to prepare deal with sodium and to eliminate sodium from corium debris will also be installed.

4.2 Corium-Water Facility

The corium-water interaction test facility (Figure 4 left) will roughly be a modern replica at a larger scale of the FARO facility at JRC-Ispra [7, 32]. A water filled vessel will be used for FCI experiments with steam and hydrogen treatment tanks (shared with the Material Interaction Hall). Two W-ray sources are planned for X-ray imaging.

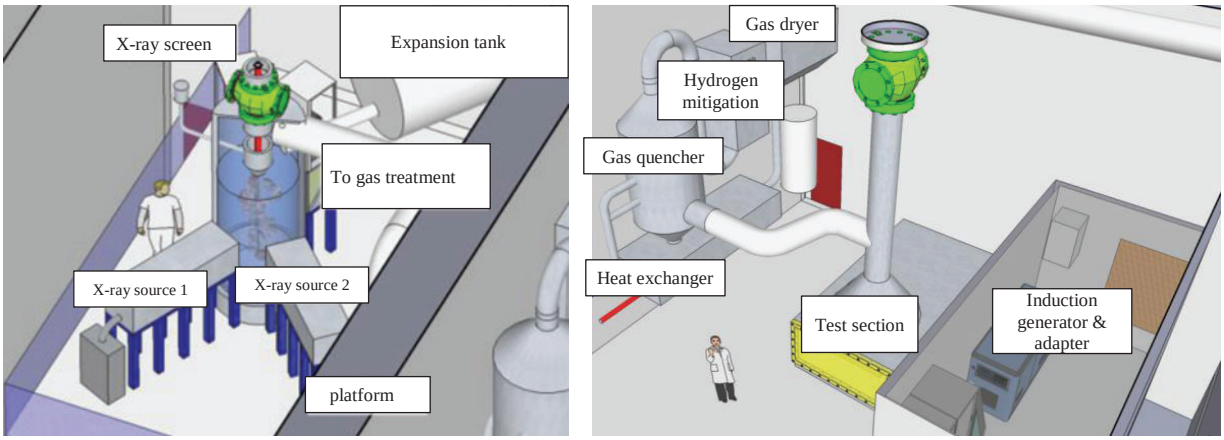

Figure 4. Left: Corium-Water Interaction hall – Right: Material Interaction Hall

Underwater spreading can also be performed in this hall, dedicated to all types of transient experiments with water.

4.3 Material Interaction Facility

This hall is dedicated to long term experiments in which, contrary to the transient experiments described above, the decay heat is larger than the thermal inertia and must be simulated. Induction heating has been selected to simulate decay heat in the oxidic phase, as in PLINIUS/VULCANO MCCI experiments [24]. Large masses, around 500 kg are the target of this facility to minimize border effects.

Experiments can also be conducted in this hall with water cooling. As steam and other non-condensable gases (hydrogen, carbon oxides from concrete,…) can be produced, a gas treatment system is being designed (Figure 4 left).

5. CONCLUSIONS

The experience from corium experimental research has confirmed that it is practically impossible to find a simulant of corium for integral experiments dealing with any phase of sever accident phenomenology. This has been supported by the peculiar chemical and physical properties of uranium oxides as well as from evidences from experiments with prototypic corium and with simulant materials, as for instance in the field of Fuel coolant Interaction.

Concerning scaling from laboratory to reactor, it has not been possible to derive scaling laws as for thermalhydraulic experiments. Nevertheless, it appears that large experiments with several hundred of kilograms are needed, in particular for Fuel Coolant Interaction and Underwater Spreading. Corium pool experiments also require large masses as a means to prevent border effects and to reduce the strength of surface crusts.

From this analysis, and an internal roadmapping of severe accident needs both for ASTRID and Light Water reactors, CEA has decided to launch a new large-mass prototypic-corium platform PLINIUS 2. It should be available around 2020 for experimental research programs on Fuel-

Coolant Interaction, corium coolability, Corium-Material Interaction (material being concrete, sacrificial ceramic,…) for both Sodium Fast Reactors and Light Water Reactors.

In order to achieve this, three test section rooms are necessary, one of them dealing with sodium experiments; they will be located below a furnace room where corium will be molten and transferred to dedicated test sections. Its modular conception will provide the necessary versatility to cope with new experimental programs.

It is proposed to operate PLINIUS-2 as an International User Facility in which national and international partners can contribute to dedicated experimental programs under the supervision of a steering committee representing PLINIUS2 stakeholders. When available, this new prototypical corium facility will be an asset for the European Research Area on Corium which is being organized within the SAFEST [46] consortium.

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