

Injectable Sacrificial Material Safety System to Mitigate Molten Corium in Containment

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INTRODUCTION

The catastrophic nuclear reactor accident at Fukushima created a loss of confidence in nuclear energy and a demand for new engineered safety features that could mitigate or prevent radionuclide releases to the environment. In a Fukushima's accident scenario, the core meltdown and subsequent reactor pressure vessel (RPV) failure could lead to molten corium being released to the drywell in a boiling water reactor (BWR) or reactor cavity in a pressurized water reactor (PWR) (Figure 1). For example, the molten corium released could further spread out to the BWR wet well and breach the containment. Hydrogen production from corium reacting with water or concrete could also occur, causing an explosion and contamination of the environment. Thus, future designs of nuclear reactors must include safety systems to ensure effective cooling, immobilization of the core melt, elimination or minimization of hydrogen gas production, and maximization of radionuclide retention. Some new reactor designs employ a core catcher and slab sacrificial material (SM), such as ceramic and concrete slab, to slow the corium flow [1]. Because existing reactors cannot easily be modified to include these SMs, it is therefore highly desirable that an injectable safety system (ISS) that uses a granular SM could be added without major retrofits.

As a part of a three-year laboratory directed research development (LDRD) project, a research study is being conducted at Sandia National Laboratories (SNL) to develop an ISS to utilize the highly endothermic decomposition of granular carbonate materials (e.g., CaCO_3 has a high decomposition energy of ~ 170 MJ/mole [2]) to absorb decay heat from the molten corium and to quickly solidify the molten material. This study includes two parts: Part 1 demonstrates this concept in series of surrogate experiments. Part 2 utilizes the SIERRA engineering modeling tool developed at SNL to model the surrogate experiments as well as molten corium spreading experiments reported in the literature. In Part 2, the proposed concept feature is also evaluated using MELCOR, a U.S. Nuclear Regulatory Commission severe accident code developed at SNL.

In this paper, a brief description of the ISS concept is provided, followed by the description of the surrogate experimental approach and the experimental progress to date. Finally, the modeling effort and its progress are provided.

INJECTABLE SAFETY SYSTEM

The motivation for developing an ISS using granular SM is based on how the molten corium flow could be stopped in the cavity (or drywell) region of the reactor containment. At

the same time, ISS should prevent the molten corium-concrete reaction (MCR).

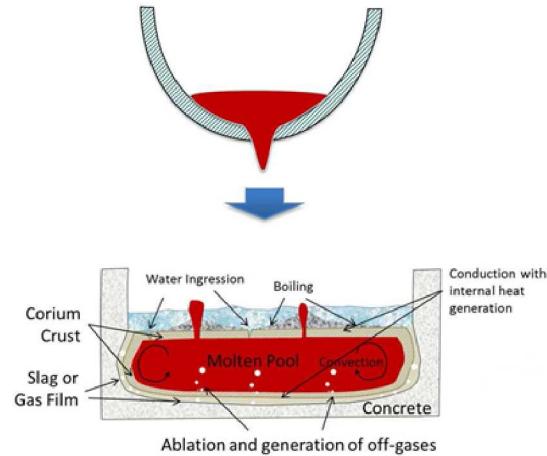


Figure 1. Possible reactions of molten corium with concrete in cavity.

To prevent MCR and corium spreading, an ISS must provide a barrier between the molten corium and the concrete floor without any significant retrofitting to the existing containment. Figure 2 shows the concept placement of the ISS in an existing BWR Mark I design. As shown in this figure, the ISS must deliver the granular SM onto the pedestal region before the molten corium breaches out the RPV. The interaction between the carbonate minerals (such as calcite) can prevent the molten corium from contacting the concrete floor. The decay heat from the corium could be absorbed by the carbonate decomposition and the CO_2 released from the endothermic decomposition reaction could create an open porous structure during corium solidification, thus facilitating further heat removal by subsequent injection of cooling water to this structure.

A preliminary study was conducted to estimate the carbonate needs for each of the existing reactor designs of six BWRs and PWRs in the U.S. In this study, we assumed that the material property values were constant. The evaluation was performed for a temperature of 1000 K. The carbonate material was assumed to have a grain size of 5 mm forming an SM bed with a porosity of 0.37. The granular material was assumed to be injected into the cavity (or drywell pedestal) at 298 K. Nominal leakage rate during operation in the plant is ignored. Table 1 shows the quantity of carbonate material required and the amount of CO_2 generated from the interaction of molten corium and carbonate for corium cooling.

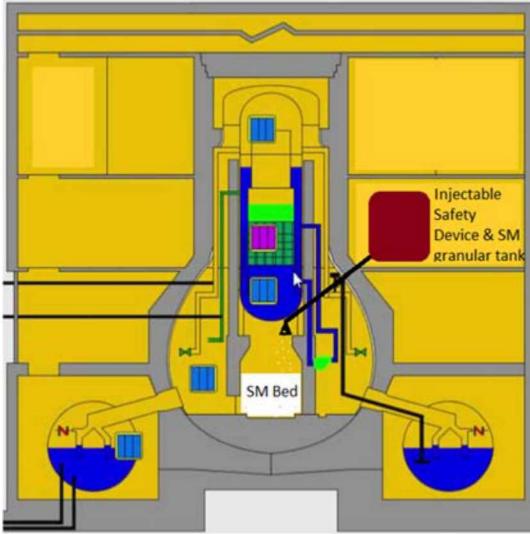


Figure 2. Conceptual design of the injectable SM safety system in a BWR Mark I design.

Table 1. Quantity of carbonate material to be injected and amount of CO₂ Generated [3].

Containment Type (Reactor Name)	CaCO ₃ Injected to Cavity (kg)	CO ₂ Generated (kg)
Large Dry PWR(Zion)	40,000	3,023
Sub-atmospheric PWR (Surry)	60,000	1,951
Ice Condenser PWR (Sequoyah)	75,000	3,567
Mark I BWR (Peach Bottom)	280,000	13,400
Mark II BWR (LaSalle)	250,000	4,310
Mark III BWR (Grand Gulf)	350,000	5,430

As shown in Table 1, a significant amount of carbonate material is required for the BWR Mark I design. To provide a more accurate estimate of the carbonate needs and the CO₂ release, code simulations are needed, as discussed in the modeling section.

EXPERIMENT

The temperature of molten corium could be more than 2500 K. The experiments at such high temperatures are difficult to perform. For a proof-of-principle, an appropriate surrogate material system with a moderate melting temperature was chosen. The surrogate for molten corium (herein as referred as “melt”) was chosen to have a melting temperature higher than the decomposition temperature of carbonate mineral (MCO₃), which ranges from 350 to 750°C [4,5], and have a relatively high density to mimic an actual fuel density (~11 g/cm³ for UO₂). Given its melting temperature of 880°C, and high density, 9.53 g/cm³, PbO was chosen as the surrogate material. The density of PbO is high enough to allow for density inversion during melt solidification.

Based on the MCO₃-PbO system identified, a series of benchtop experiments were conducted. In each test, a small quantity in the of tens of grams of PbO was first melted in a

furnace at > 1000 °C. The melt was then poured into a quartz dish containing a known grain size and amount of MCO₃. Real-time monitoring using a remote temperature gun and video camera were conducted for the tests. Post-testing characterization was performed to determine microstructures, chemical composition variations, and CO₂ bubble distributions in the solidified materials.

Over 20 sets of SM-melt interaction benchtop experiments have been conducted with different SMs (such as calcite and dolomite) and melt masses. In these experiments, a SM was either preheated to 550 °C or not preheated. In either case, a small quantity (~10 to 20 grams) of PbO was melted at 950 to 975 °C and then poured onto a MCO₃ granular bed. The reaction between MCO₃ and the melt was evident: the melt quickly solidified, within tens of seconds (not a violent reaction though), resulting in an open porous structure. It is postulated that the formation of porous PbO/MO/MCO₃ cake was caused by CO₂ generation from carbonate decomposition, which was confirmed by a measurable mass loss. Figure 3 shows the results of two samples reacted with preheated (550 °C) CaCO₃ powder and with room temperature (22 °C) dolomite with the particle size ranging between 0.3 and 1 mm. As shown in Figure 3(a), the end-product contains large connected pores. Careful visual examination of the mixture shows structures similar to those observed in the COMET experiment [6]. Also shown in Figure 3(b), the larger particle size and the experiments with no pre-heating result in denser solid structures. Migration of the particles on the surface of the mixture was evident, which indicates that the density-induced convection of the melt during cooling.

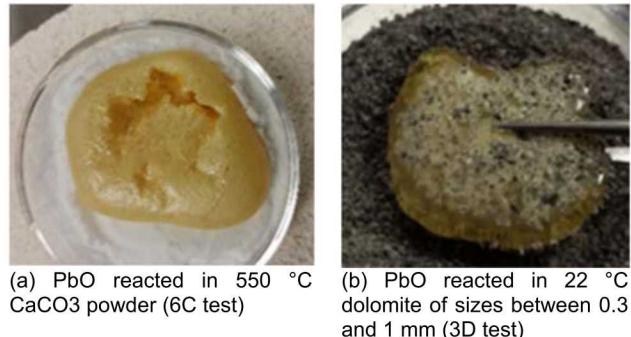


Figure 3. MCO₃-PbO melt interaction with CaCO₃/Dolomite.

In these tests, we found that the particle size of a material could significantly affect the extent of mixing between the carbonate particles and melt and therefore the extent of SM decomposition. The best performance for the melt solidification was obtained for the calcite with particle sizes ranging from 45 to 75 µm. Too fine particles may not be conducive to the mixing of SM and melt; the optimal particle size is also expected to be scale-dependent.

To better understand the pore structure formation, micro-X-ray CT was performed. The analysis further confirms the generation of CO₂ bubbles from the melt-SM interface during

the cooling process, as indicated by the extensive presence of distinct pores. It is evident that gas bubbles were nucleated at the melt-carbonate mixing front and tended to be collected at the top surface of the melt by convection. At the bottom of the melt, SM particles tended to intrude into the melt through density inversion [3].

To confirm this concept in a larger-scale, similar tests have been performed at SNL's SURTSEY facility. Figure 4 shows a ~1kg molten melt (at 950 °C) being poured onto a granular calcite (CaCO_3) material at room temperature. As shown in this figure, the red molten melt flows onto the 1mm sized calcite granular bed generating white smoke and a large number of bubbles in the mixture. Thus, the mixture swells upward to the surface while the reactions continue. Once the temperature falls below the decomposition of the calcite, the bubbling stops and the color of the mixture changes from red to a yellowish color.



Figure 4. 1 kg PbO at 950 °C pouring onto < 1mm calcite bed on a pan (090418 test).



Figure 5. By-Product samples from 090418 Test (see Figure 4).

Post-examination of the mixture shows evidence of large pore formation (see Figure 5). The reactions were mainly at the surface of the carbonate bed. Additional tests are being performed and the composition of the white smoke will be identified. Furthermore, to support the modeling effort as described in the next section, the experiments with better controlled physical configurations will be performed to

provide better constraints on modeling parameters and to provide validation data.

MODELING

We are developing computational models of corium flow, reaction, and solidification after a nuclear meltdown accident. To accomplish this, we use the thermal/fluids code Aria which is part of the SIERRA code base at SNL. Due to the complexity of the thermal environment for these problems, elevated temperature (2500 K), internal heat generation by radionuclide decay, and the heat transfer and multicomponent phase-change of the composite material, the accurate prediction of the melt solidification front is difficult. Similar to a flow of lava, molten corium released from the vessel breach onto the containment floor starts to spread out and solidify as heat losses to the environment occur (radiation, convection to air, and conduction with the floor). As the corium spreads and cools, it begins to vary in thickness leaving a complex, uneven surface, which makes the location of the corium front difficult to predict. A corium spreading model, ignoring any chemical reaction was developed in Aria using the conformal decomposition finite-element method and control volume finite-element method, which solves the full Navier-Stokes and heat transfer equations for multiphase problems. The details of the computational methods and model selection used here can be found in [7].

Validation of our computations was accomplished by comparing our modeled results with the experimental measurements conducted in the FARO L26 experiments [8, 9]. In these experiments, 160 kg of corium composed of 80% UO_2 and 20% ZrO_2 heated to approximately 2950 K was gravity-fed and allowed to spread on a steel plate. To model solidification, a pseudo-solidification approach was used where the corium viscosity is modified using the Ramacciotti viscosity model $\mu_c = \mu_L \exp(2.5 \cdot C \cdot f)$, where μ_L is the liquidus viscosity of the corium, $C = 6$, and f is the solid volume fraction based on the liquidus and solidus temperature of the corium [10].

As the hot corium begins to spread on the plate, it has a relatively low viscosity and spreads quickly. As the corium spreads along the plate and cools, the material touching the steel plate begins to solidify and decelerate at the corium front. However, because hot, fast moving corium is still being introduced at the inlet, mass begins to accumulate behind the front until it gives way and begins to flow again. Figure 6 shows a snapshot of a three-dimensional simulation colored by temperature, which shows that the corium near the walls has cooled considerably, but the interior of the corium remains hot. This balance of heating, cooling, and mass accumulation causes the undulating behavior of the corium front observed in the FARO-L26 experiments.

The undulation of the corium front observed in the experiments is captured well in the current simulations shown in Figure 7, which plots the corium front location (in meters) as a function of time. For the 2D case, the predicted front location is in excellent agreement with the experiment after t

≈ 5 s, although the speed of the corium front was over-predicted at earlier times. The 3D case shows only qualitative agreement as the undulating behavior of the front is correctly captured, however the spreading front location is somewhat under-predicted at later times. This can be explained through various choices of model constants selected for the 2D case which may not be applicable to the 3D case, where the span wise spreading of the corium and introduction of side-walls changes the heat transfer characteristics quite substantially. For a more thorough explanation of the discrepancies between the 2D and 3D cases, as well as more in-depth insight into the selection of model constants and results we refer the reader to [7].

Time = 7.920

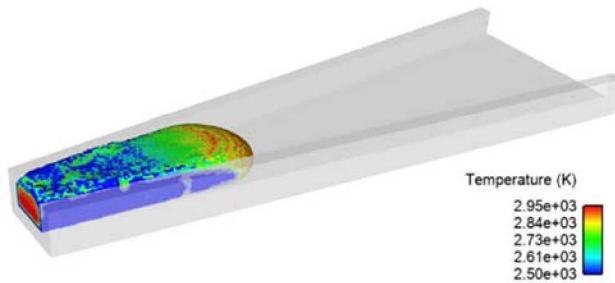


Figure 6. 3D simulation of FARO L26 experiment at $t = 7.92$ seconds.

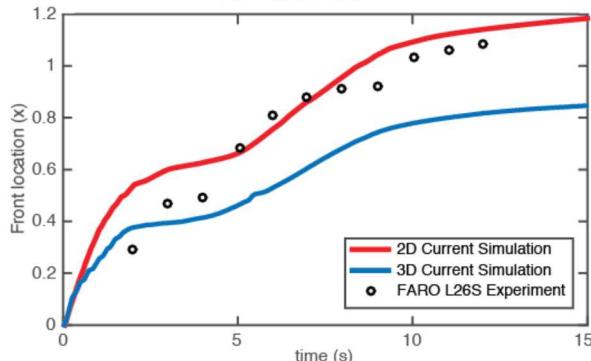


Figure 7. Simulation results compared to experimental data.

CONCLUSIONS

This paper has demonstrated an ISS concept to contain and cool the ex-vessel molten corium from a severe nuclear accident using granular carbonate minerals as SMs. We believe the use of granular SM's could be added to existing operating reactor plants without significant cost. These dry SMs would not affect any safety-related electrical equipment that are necessary to mitigate and monitor the progression of the accident.

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REFERENCES

BWR	Boiling water reactor
ISS	Injectable safety system
LDRD	Laboratory direct research and development
MCO3	Carbonate mineral
MCR	Molten corium-concrete reaction
MO	Mineral oxide
PWR	Pressurized water reactor
RPV	Reactor pressure vessel
SM	Sacrificial material
SNL	Sandia National Laboratories

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