Experimental Observations of Boric Acid Precipitation Scenarios

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Abstract

During a Loss of Coolant Accident (LOCA) in Light Water Reactors (LWR), borated water is injected into the core through the safety injection system. The continuous vaporization of the water from the core may increase the concentration of boric acid in the core that, under certain conditions may reach the solubility limit and precipitate. This includes scenarios where the liquid water supply to the core if affected blockages due for example to debris accumulation. Questions have been raised on the effects of the precipitate in the core on the flow behavior, including the possibility of additional blockages produced by precipitate accumulation. A simple experimental facility was constructed to perform simple experimental observations of the behavior of borated water under the combined effects of the boiling and the boric acid precipitation (BAP). The facility consists of a transparent polycarbonate vertical pipe where forty-five heated rods have been installed to supply the power to the water to reach the saturation temperature and maintain a given boil-off rate. The layout and geometry of the experimental apparatus were design to emulate a simplified core of a Pressurized Water Reactor (PWR). Experimental observations have been conducted under two different conditions. The first test was executed to observe the behavior of the water and the boric acid precipitate during a boil-off scenario with without external borate water addition (decreasing water level). During the second test, borated water was constantly injected from the top of the test section to maintain a constant mixture level in the test section. Both tests assumed no flow from the bottom of the test section which may be the case of PWR LOCA scenarios in presence of debris-generated core blockage. The observations performed with a set of three cameras installed around the test section, showed interesting effects of the vapor bubbles on the boric acid precipitate migration and accumulation in the test section. The results may help addressing some basic questions regarding the effect of the precipitation of boric acid during core blockage LOCA scenarios and, provide the basis for further experimental or computational analysis required to resolve the BAP issue as part of the Generic Safety Issue 191.

Keywords: Flow observations, boric acid precipitation, Hot Leg Injection

1. INTRODUCTION

The Emergency Core Cooling System (ECCS) in a Pressurized Water Reactor (PWR) is design to provide the required coolant flow to removes the decay heat from the reactor core during a postulated Loss of Coolant Accident (LOCA) scenario, bringing the system to the cold shutdown condition. During the first phase of the accident, often identified as safety injection phase, the ECCS draws cold borated water from the Refueling Water Storage Tank (RWST) injecting into the cold legs. During this phase, depending on the pressure of the primary system, cold borated water from the accumulators is also injected into the primary system. Boron (in the form of boric acid) is used as a soluble neutron absorber in the primary coolant. The main functions of boron are to compensate for fuel burnup and xenon poisoning with the required reactivity margin during normal operation, and to provide the necessary subcriticality of the core during refuelling and maintenance [1]. During normal reactor operation, the specified boron concentration is maintained by the volume control system. In case of a LOCA, borated water is injected by the ECCS as previously described. Injection continues into the same locations (cold legs) after the sump switchover (SSO) during the cold leg recirculation phase. This phase differs in that the coolant is now drawn from the containment sump compartment and recirculated back into the primary system. This conditions of water recirculation from the containment sump requires to be maintained for several days in order to remove the decay heat from the core and achieve a cold shutdown condition. Hot leg switchover (HLSO) is a procedure initiated as a manual action by the operators at a time determined by plant design to insure adequate core flushing and prevent or mitigate precipitation of boron in the core. Generally this involves simultaneous injection through cold and hot legs. Understanding the local behavior of the borated water under boiling conditions and the possible effects of the presence of boric acid in the solution is of paramount importance for the correct actuation or revision of the manual operation procedures currently in place for LOCA scenarios of different break sizes and locations. Of particular interest would be scenarios where precipitation of boric acid occurs, to study the effects of the precipitate on the water cooling capabilities and the overall core coolability during the long-term cooling phase.

2. MOTIVATION

The industry and the research community has posed questions on the behavior of the coolant in the core in the presence of boric acid and the effect of the boric acid precipitation on the core coolability. Properties of the water (in particular viscosity and density) are expected to change as the concentration of boric acid increases [2]. Concentration of boric acid may also increase and reach the solubility limit, causing precipitation of boric acid in the core. The major concern during this phase of the accident is that solid precipitate remaining in the core may deposit or accumulate in different regions and impact the coolant flow and the core coolability. This condition combined with possible presence of debris penetrated through the sump strainers may seriously impact the coolant flow in the core, leading to conditions for core damage. Under hypothetical debrisgenerated full core blockage at the bottom of the core, the coolant may only reach the core from alternative flow paths (usually the top of the core [3] or lower elevations through the pressure relief holes [4]. Change (increase) in density may also change the natural circulation patterns in the core and possibly create a stratification where denser borated water may tend to collect at the bottom of the core, thus preventing colder water coming from the alternative flow path to reach the bottom of the core.

A very simple boil-off experiment using DI-water with boric acid was conducted to qualitatively observe the behavior of boiling water in presence of high concentration of boric acid and the possible effects of precipitation on the water behavior. The experiment was conceived in such way that solution of water and boric acid was left boiling in the beaker. No liquid water was added during the test to compensate the water boil-off producing a continuous increase of the boric acid concentration from the initial value, until boric acid saturation was reached. The test was conducted with the aid of (Figure 1):

- A 2-liter glass beaker
- A small water heater (max power 1 kW)

Temperature was continuously monitored during the experiment using a class 1 k-type thermocouple connected to a Fluke® 52II thermocouple reader (0.3 °C accuracy) and periodically checked with an immerged thermometer.



Figure 1. Experimental Setup - Beaker

2.1 Beaker test Preparation

The test solution was prepared by mixing in the beaker 275 g of boric acid (Optibor B Orthoboric Acid, H₃BO₃), in 2 l of DI-water at room temperature. This initial concentration (C₀ = 137.5 g/l)

was imposed in order to reach the saturation limit $C_s = 275$ g/l when approximately 1 l of water had evaporated¹. C_s is the solubility limit of boric acid in water at 100 °C found in [5].

The heater was turned on and the solution was continuously steered using a stainless steel steering rod, allowing the dissolution of the boric acid as the temperature increased toward the water saturation temperature. This phase of the experiment was recoded with a camera placed in front of the experimental setup. Video and pictures were taken throughout the entire experiment.

The mixture was observed boiling as the liquid level decreased down to the point where boric acid solubility limit was expected to be reached. Once of the first visible effect that was observed at this point was the formation of a very thin layer of precipitate at the free surface off the liquid as result of the precipitation of the boric acid near the vaporization surface. As the quantity of liquid evaporated increased, small white particles of precipitate were observed fluctuating in the beaker but entrained by the rising bubbles toward the top section of the boiling liquid (Figure 2). At this time, water still appeared clear through the glass beaker.



Figure 2. Early Stage - Formation of Precipitate

As the evaporation continued, the formation of precipitate at the surface and the entrainment of other precipitate in the water toward the top of the liquid contributed to the formation of a growing layer of boric acid on the liquid surface (Figure 3a). The presence of larger amount of precipitate in the liquid water increased the visual opacity of the water (Figure 3b). As the process continued, larger pieces of boric acid were observed to be released from the growing layer (not reaching a distinguishable thickness) affecting even more the water opacity (Figure 3c). In a later phase, as the liquid level drastically decreased, the effect of precipitate entrainment toward the top layer and the subsequent release of large pieces from the top layer was observe to cycle, becoming easily distinguishable to the observation:

- Water became clearer as the entrainment of the precipitate in the remaining liquid by the rising bubbles took place, and the top layer grew (Figure 4a)
- Water reached its maximum clearness as big pieces of boric acid appeared clearly visible. The top layer appeared detached from the liquid level perhaps due to the increased pressure under the layer (Figure 4b)

¹ Due to the boric acid carryover in the vapor, the estimation of the volume of water to be evaporated to reach the desired concentration C_s is approximate.

- Top layer reached a maximum thickness and collapsed in a visible explosive manner releasing a large amount of boric acid into the liquid water which is now visible opaque, as the cycle started again.

The described phenomenon was observed even at very low liquid (approx. 300 ml). The experiment was terminated after a few cycles were recorded.



Figure 3. Middle Stage - Growing Layer and Reduced Transparency



Figure 4. Late Stage – Cycling Pattern

Three main features of the observed behavior of the water under the described experimental conditions were of particular interest:

- When reaching the solubility limit, boric acid starts precipitating at the liquid free surface
- Small size precipitate is released and/or formed in the liquid water. The amount of the boric acid carried by the liquid phase was visibly tracked by the turbidity of the water (turbidity not physically measured). The increasing amount of precipitate as the liquid level decreased seemed not affect the liquid behavior in the beaked, even during the later phase of the experiment.
- At very high concentrations, a thick layer of precipitate formed at the top of the liquid. Based on development of the vapor overpressure at the bottom of the layer, this was observed to collapse releasing large amount of precipitate into the liquid. This effect cycled until the end of the experiment.
- Large pieces of solid precipitate were observed to float during the later phase of the experiment but no appreciable deposition of solid precipitate was observed at the bottom of the beaker.

The behavior of the solution observed during this simple experiment raised other basic questions:

- 1) What would be the behavior of the solution when a different geometry and/or heat configuration is used? (heat from the bottom horizontal plate may induce a different boiling/recirculation patterns and transport the precipitate in other ways compared to heated vertical rods.
- 2) The large open space in the beaker may not allow the deposition of precipitate at the bottom since bubbles formed at the base of the beaker may produce a much larger buoyancy forces

and turbulent flow at the bottom plate compared to the regions between vertical adjacent rods.

These basic questions motivated the design and construction of a small and simple experimental facility with a more realistic geometry (closer to a reactor fuel configuration) to conduct similar qualitative visualization of boric acid solutions.

3. THE PRECIPITATION AND STRATIFICATION TEST APPARATUS

An experimental facility was design to reproduce a geometry "as close as possible" to a typical PWR core. Even though rigorous scaling laws were not followed, the design was conceived to provide the facility with a specific set of geometrical features to conduct qualitative visualization of boric acid solutions and effect of precipitation in the flow of boiling water in such geometries. Description of these features is provided in the section.

A detailed view of the facility, the test section, and the other components is shown in Figure 5a. The test facility consist of a polycarbonate cylindrical enclosure (which may be assimilated to the core barrel of a typical PWR design). Two polycarbonate flanges were welded to the cylindrical section to create the facility test section (Figure 5a, b, and c). Two stainless steel plates were fabricated with the scope of mounting the heated rods with a give pattern (bottom plate), and to hold the rods in a straight parallel position avoiding vibrations during the experiments (top plate). The top plate was also equipped with venting holes to allow flow through the top region (Figure 5c). The current design of these plates using for this first phase of the experimental activity do not reflect the realistic design of upper and lower plates of the PWR core. Forty-five threaded holes were machined on the plates to allow the placement of an equal number of heater rods (Figure 5e). In the experiment reported in this paper, four rods at the corner of the rods array were removed to allow the insertion from the bottom of two k-type thermocouples (to monitor the temperature inside the test section at two different locations) and two sampling ports (Figure 5d) to collect samples of boric acid solutions for future concentration measurements². The thermocouples were connected to a Fluke® 52II thermocouple reader (Figure 5a).

The test section is connected at the top end with a polycarbonate cylindrical extension where four ports equipped with valves were installed (Figure 5a). These ports were mainly used for injection of borated solution during the experiment described in this paper, simulating the injection from the top of the core. The bottom section of the test facility is the section where heated rods are connected. No flow is possible through the bottom plate in the configuration presented. The flow added to the test section through the injection ports is controlled by a metering pump (MityFlex 913) located below the test section (Figure 5a). The suction of the pump is immerged into a metered glass beaker (reservoir) containing the solution to be added during the experiment. The use of a glass beaker allowed the estimation of the injection rate to be maintained during the experiment to supply the amount of water to compensate the evaporation rate, maintaining the liquid inventory into the test section approximately constant. The solution in the beaker was maintained at an approximate constant temperature with the use the heater (mentioned in section 2) and an immerged thermocouple.

The top of the extension section was closed with a polycarbonate lid connected to the top flange to prevent uncontrolled discharge of vapour (entraining boric acid). The vapour was instead

² The present paper does not show measurements results of the boric acid concentration versus time during the experiments. Sampling and measurement techniques are currently being investigated and will be the topic of future publications.

discharged through a 1" hose exhaust connected to the top lid and diverted upward toward a filtration system for safety purposes.

Heated rods, placed in a square lattice configuration, are connected to dedicated power supplies and can be powered independently for future use. In the experiment presented, 41 heated rods were grouped and connected to ten independent power supplies with power readers (each power supply alimenting 4 rods except one where 5 rods were connected). The setting of the power supplies were chosen in order to give an approximate total power of 2.5 kW uniformly distributed to the installed rods. The value of the power used was chosen arbitrarily below the maximum power installed to test the thermal-mechanicals capabilities of the facility. Higher power will be used in future experimental activity.



Figure 5. Experimental Facility Overview

Table 1 summarizes the main dimensions and other features of interest of the test section and its relevant components. The values reported for a typical PWR core configuration are displayed only for reference.

Feature	Reference PWR	Test Facility
Diameter	3.35 m	0.1524 m
Core Diameter/Height Ratio	0.785	0.785
Rod Diameter	0.91 cm	0.95 cm
Bundle Pitch/Diameter Ratio	1.33	1.75
Max Power	75 MW ³	20.5 kW

Table 1. Main Features of the Experimental Faci	lity
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The diameter of the test section (0.1524 m, corresponding to a 6 inch inner diameter pipe) was mainly dictated by the availability of the piping and related flanges in the laboratory, to optimize the construction time. The height of the test facility was imposed in order to preserve the core diameter/height ratio. The closest heated rods diameter that was found available at the time of design was considered to be acceptably close to the reference diameter for the purpose of the experimental analysis to be conducted. The center-to-center pin distance (and subsequently the bundle Pitch/Diameter ratio) could not be preserved from the reference configuration due to the size of the rods' head (see Figure 5e) and the mounting configuration at the bottom plate. Modifications of the facility with the selection of different heated rods may be implemented in future developments. Each rod is rated to a maximum power of 500 W, providing a maximum power installed of approximately 20 kW. This power is larger than the power that would be required if the reference power density or heat flux would be preserved.

3.1 Test Conditions and Preparation

The facility was initially loaded with a solution prepared with 660 g of boric acid (Optibor ® Orthoboric Acid, H₃BO₃), in 21 of warm DI-water (approximately 50 °C). This initial solution was poured into the test section from the top. An additional quantity (0.5 1) of DI-water at the same temperature was used to clean the boric acid accumulated at the top flange. The final solution in the test section when the test was initiated contained 660 g of boric acid in 2.51 of DI-water ($C_0 = 264 \text{ g/l}$). This concentration was chosen to be slightly below the solubility limit of boric acid in water at 100 °C ($C_s = 275 \text{ g/l}$) so that the saturation could be reached sometime after the water started boiling and evaporating from the test section. The total initial volume corresponded to a liquid level right above the top plate. The temperature of 50 °C for the initial solution allowed the dissolution of most part of boric acid, limiting thermal stresses to the test facility (which was usually slowly pre-heated). The heated rods were turned on when the solution in the test section was quiescent (Figure 6) with the excess of boric acid deposited at the bottom of the test section.

³ The reference power was assumed to be the decay power at approximately 30 minutes after the reactor shutdown, corresponding to a typical sump switchover time for a Large Break LOCA [6].



Figure 6. Test Preparation – Injection of the Initial Solution (660 g of H₃BO₃ + 2.5 l of DI-water)

The injection solution was prepared with similar techniques described above, with a concentration of $C_a = 60$ g/l. This concentration is not prototypical for any plant conditions and was selected arbitrarily based on solubility of boric acid at the preparation temperature. The solution addition was prepared in batches added to the reservoir beaker when the remaining solution reached the 300 ml mark. The addition of each batch was recorded during the experiment.

4. **RESULTS**

As the power (2.5 kW) was turned on, the temperature was observed to increase until reached the boiling point. The temperature achieved and the high turbulence in the test section due to the bubble formation and migration to the top produce a very rapid dissolution of the entire quantity of boric acid previously deposited at the bottom of the test section. When complete dissolution was achieved, no visible traces of boric acid between rods or in the region between the outer rods and the test enclosure were observed. At this point the metering pump was turned and setup to the rate to compensate the boil-off from the test section. The setting of the metering pumps was defined during previous shakedown of the facility and manually fine-tuned during the experiment. The observations conducted showed similarity with the beaker test described in section 2.

- Water remained clear during the early stage of the experiment, even after the solubility limit was expected to be achieved (Figure 7a).
- Small visible particles started appearing in the solution but mainly entrained or maintained in suspension by the violent bubbles departed from the surface of the rods, preventing any deposition at this time.
- As the mass of boric acid in the liquid increased, the turbidity of the water visibly increased while a layer of boric acid on top of the mixture level was observed to form and grow (Figure 7b).



Figure 7 – Experimental results (Early Stage – Left; Middle Stage – Right)

At very high concentrations, bigger particles were observed and deposition of solid precipitate started accumulating in specific locations at the bottom of the test facility (Figure 8):

- The outer region of the rod bundle, between the outer rods and the enclosure
- The corners of the bundle near thermocouples and sampling ports
- The walls of the enclosure



Figure 8 – Experimental results (End of the Test)

The test was continued for a few minutes after turning off the injection pump, letting the liquid water to evaporate to acceptable limits. As the water level decreased, the top layer of boric acid grew rapidly similar to what observed during the beaker experiment. When deposition of boric acid impeded any neat visualization through the walls of the test section, the test was terminated.

5. DISCUSSION

An experimental apparatus was contracted to conduct experiments with water and boric acid and observe the behavior of the mixture under boiling conditions with precipitation in a geometry characterized by similarities with a typical PWR core. The observations conducted put in evidence the interesting phenomena that can be summarized as follows:

- Even at high concentrations of boric acid, perturbation of the water flow induced by the precipitation were not appreciably observed.
- Violent bubble generation, departure, and migration to the top of the test section prevented deposition of precipitation between the rods and along the heated test section.
- No stratification was observed due to change in water density cause by increased concentration. Instead well mixed conditions were observed, allowed by the boiling in the test section.
- Deposition of the precipitate was observed in regions of lower power density, such as the outer region of the test section, or in "colder" surfaces (the test enclosure)

Even if a rigorous scaling approach was not followed to determine the power applied and other specific geometrical features (in particular the center-to-center pin distance), the results summarized in this paper may be a more realistic basis to make hypotheses on the behavior of the water in the reactor core during the long term cooling phase of a LOCA scenario, or be used as reference for future calculations and experiments required to developed or revise methodologies to estimate the time to HL switchover time. With the use of this simple and small experimental facility, the authors also intended to develop techniques for visualization and measurements of boric acid, to be applied in future experimental activities using the described facility or new scaled experimental apparatus.

6. NOMENCLATURE

- BAP Boric Acid Precipitation
- C₀ Initial Concentration
- Ca Concentration of Added Solution
- C_s Concentration at Saturation (Solubility Limit)
- DI De-Ionized
- ECCS Emergency Core Cooling System
- HLSO Hot Leg Switvhover
- LOCA Loss Of Coolant Accident
- LWR Light Water Reactor
- PWR Pressurized Water Reactor
- RWST Refueling Water Storage Tank
- SSO Sump Switchover

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