Experimental modelling of boric acid crystallization process during emergency core cooling of NPP with WWER

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Abstract. The results of experimental study of boric acid crystallization in WWER reactor in case of loss of coolant accident (LOCA) and operation of passive safety systems are presented. Description of the test facility and the experimental technique are given. The results of the visual observation of process of boric acid crystallization from the solutions with initial concentrations 371 and 466 g kg\(^{-1}\) H\(_2\)O are presented.

1. Introduction

Boric acid is used in nuclear power for several purposes. The main of them are compensation of changes in the neutron field in operational mode and ensuring the subcriticality of the core during refueling, outage and in the event of an emergency. During normal operation of the reactor, the boron concentration is maintained by the water treatment system operation. However, in case of a LOCA accident with a rupture of the main circulation pipeline (MCP), boric acid is carried away from the primary circuit with coolant through the break [1].

To compensate coolant losses in case of emergency, the design of the WWER-TOI reactor provides for the operation of the core flooding system from the hydro accumulators of the first, second and third stages [2]. In case of an accident with rupture of MCP, boric acid solution with a concentration of 16 g kg\(^{-1}\) H\(_2\)O from these systems provides the core cooling [3]. In addition, during the accident, the core is cooled by condensate from steam generators operating in condensation mode [4]. Due to the duration of the process (up to 72 hours), boiling of the coolant in the core, and low acid concentration in the vapor phase, an increase in the amount of boric acid in the solution in primary circuit is possible. This can lead to changes in its physical properties, including density, viscosity and surface tension [5]. This, in turn, can deteriorate heat transfer from the fuel rods. Moreover, if a certain concentration of acid in core is reached, its crystallization in the reactor is possible. This can lead to deterioration of heat transfer and, ultimately, to the melting of fuel.

According to a calculation carried out at IPPE, after 24 hours of the emergency process, the concentration of boric acid in the core is 311 g kg\(^{-1}\) H\(_2\)O and does not reach the solubility limit [6]. However, as it was mentioned above, in the projects of nuclear power plants of the new generation with WWER reactors, the core cooling by passive systems is provided for up to 72 hours. This implies the need for research of the process of boric acid precipitation in case of LOCA accident.

Some previous results of the study of boric acid crystallization processes during cooling of the reactor are presented in [7–9]. In the experiments presented in a series of papers, the state of the core in 30 minutes after the start of the accident was simulated [7, 8]. The absence of solution stratification
according to the height of the research facility was found. It was also found that the precipitation of boric acid on the walls of rods - imitators of fuel elements was not observed even with a significant excess of its maximum concentration in the solution. However, the accumulation of sediment in the cold areas of the facility was discovered.

In [9], the results of experimental studies conducted in Germany on the PKL facility are presented. In the tests, the possibility of crystallization and precipitation of boric acid during the first hours of the accident with parameters characteristic of PWR reactors with a capacity of 1300 MW was investigated. It was found that the deposition of boron is unlikely to lead to conditions that worsen the cooling of the core during the first six hours after the reactor shutdown.

The analysis of data showed that no experiments have been carried out to study the mass transfer of dissolved boric acid at concentrations characteristic of the primary circuit of a WWER reactor by the end of the 24 hours after the start of the accident in case of operation of passive core flooding systems.

2. Test facility

2.1. Description of the test facility

In IPPE, the test facility to study the process of boric acid crystallization in the WWER core in case of an accident was constructed. The test section of the facility is an electrically heated seven-rod simulator of a fuel assembly with a spacing grid. In the experiments performed, the linear density of energy on the surface of the fuel rod simulators was 70 W m⁻¹, which corresponded to the parameters of the core in 72 hours after the start of a LOCA accident.

Figure 1 shows the flowchart of the test facility. The main element of the facility is the solution preparation tank of boric acid (2) with a volume of 10.5 l. A filling line for H₃BO₃ solution is provided in the top cover of the tank. The shaft of the mixing device (1) also passes through the top cover. Figure 2 shows the main equipment of the facility: the solution preparation tank and the test section.

The fuel assembly simulator, equipped with thermocouples, is located in the center of the tank bottom. The rods assembly is equipped with a model of the spacer grid. The operation of fuel rods simulators is ensured by an electric heating element with an adjustable power. Observation of the boric acid crystallization processes inside the tank was performed through the viewing window.

The METRAN-150-DI gauge is used to measure the pressure (measurement error: up to ± 0.1% of the range). Cable thermocouples with a diameter of 1 mm are used to measure the temperature (measurement error of 1°C). The sampling frequency of the data acquisition system is 1 Hz.

![Figure 1](image1.png)

**Figure 1.** Schematic diagram of the test facility: 1 – mixing device; 2 – solution preparation tank; 3 – viewing windows; 4 – gearbox; 5 – gas cylinder containing nitrogen; 6 – fuel assembly simulator; 7 – heating element; T₁–T₇ – thermocouples, P₁ – pressure gauge (METRAN-150-DI).
2.2. Experimental technique
The task of the tests was to study the processes of crystallization and precipitation of boric acid on the surface of the test section and in the tank. The start of the boric acid precipitation process was achieved by gradually reducing the temperature of the solution at a constant H$_3$BO$_3$ concentration. This concentration was 5–10 g kg$^{-1}$ H$_2$O below the solubility limit at the initial temperature in the tank.

The experiments were carried out according to the following technique. First, by opening the valve, the tank is filled with distilled water up to 60 percent of the required volume. After that, the tank is heated to a temperature of 55°C. After reaching the required temperature, the necessary mass of boric acid powder is poured into the tank volume and the remaining portion of distilled water is added. The mass of boric acid added is measured using the electronic scale with an accuracy of 0.1 g.

Next, the upper volume of the tank is filled with nitrogen to establish the necessary pressure. Then, using electric heaters, the solution is further heated to the required temperature. After reaching it, the mixing device is turned on, and this ensures the complete dissolution of boric acid. Control of the state of the solution is carried out visually, through the windows on the tank. Boric acid temperature setpoints are maintained with an accuracy of 1 °C using a temperature relay. At the same time, the fuel assembly simulator is warmed up by the internal electric heater. When the boric acid solution reaches the specified temperature, heating of the tank is stopped, but the fuel assembly simulator is heated up throughout the whole test. Then the process of cooling the tank with a solution using a fan starts.

Through the illuminator, a visual observation of the solution in the tank volume and on the fuel assembly imitator is carried out. When the temperature in the volume of tank decreases, precipitation starts. At the same time photographing is performed. The experiment continues until the temperature of the solution decreases by 30–40 °C from the initial value. After completing the test, the solution is drained, then the tank is cleaned of boric acid precipitations by several washes with distilled water.

3. The experimental results
Two series of experiments with the power of the test section equal to 70 W with different pressures were performed at the test facility. The heat flux density on the surface of fuel rods simulators corresponded to this value in the WWER-TOI reactor in 72 hours after the start of a LOCA accident. The tests were carried out with initial temperatures of boric acid $T_0$ equal to 120 and 133°C, corresponding to a saturation pressure of 0.2 and 0.3 MPa, respectively. The main parameters of the tests are presented in Table 1.
Table 1. Parameters of the tests.

| Test number | $P$, MPa | $T_0$, °C | $C_{H2O}$, g kg$^{-1}$ | Heat loss $N_{HL}$, W |
|-------------|---------|-----------|------------------------|----------------------|
| 1           | 0.2     | 120       | 371                    | 79.2                 |
| 2           | 0.2     | 120       | 371                    | 133.7                |
| 3           | 0.3     | 133       | 466                    | 119.1                |
| 4           | 0.3     | 133       | 466                    | 179.4                |

In the tests, the process of the formation of boric acid crystals was investigated depending on the pressure, initial temperature, acid concentration and tank cooling rate. Figure 3 shows changes of the temperature of a solution of boric acid in the tank in all tests.

Figure 3. The change of temperature of the boric acid solution in the tests at different pressures: a) – 0.2 MPa; b) – 0.3 MPa; 1–4 – the number of the test.

Figures 4 and 5 show the view of inner surface of the solution preparation tank and fuel rod imitator at various stages of tests 1 and 2, respectively. As it can be seen from figure 4, in test 1, precipitation of boric acid crystals in the form of a needle-like structure on vertical and horizontal surfaces was observed. At the same time, the sediment was almost absent in the hot area of tank bottom (directly under the test section) and was completely absent on the surface of fuel rods simulators and the spacer grid. As the solution is cooled and the excess of boric acid concentration of solubility limit reaches 126 g kg$^{-1}$ H$_2$O, an increase in the amount of precipitated sediment is observed.

Figure 4. The view of the tank inner surface and fuel assembly simulator in test 1 at the moment when boric acid solubility limit is exceeded by 91 g kg$^{-1}$ H$_2$O (a) and 126 g kg$^{-1}$ H$_2$O (b).
In case of an increase in the rate of cooling of the solution in the tank (test 2), other processes of acid precipitation take place. As it can be seen from figure 5, the crystallization of boric acid is almost absent on the vertical surface, and the sediment on the horizontal surface has a loose, uniform structure. At the same time, in test 2, in contrast to the previous experiment, accumulation of acid in the hot lower area of tank bottom is observed. The precipitate formed looks like "snowflakes" of about 3-5 mm in size.

In addition, it should be noted that in test 2, there is a greater amount of precipitated acid, which can be seen from a comparison of figures 4a and 5a. This can be explained by the fact that the crystallization process also occurs at the interface between liquid and gas mediums. The formed film of boric acid, in spite of the greater density than that of the solution, is held on the liquid surface for a long time due to the forces of surface tension. However, as the temperature of solution decreases and the concentration of boric acid precipitated increases, the weight of the film increases too. At a certain point, the surface tension forces are not enough to hold the formed flakes of acid on the surface and they sink to the bottom of the tank. After that, a film of sediment starts to form again on the released surface and the process repeats.

Figures 6 and 7 show the view of inner surface of the solution preparation tank and fuel rod imitator at various stages of tests 3 and 4, respectively. In contrast to the previous tests, in experiments at pressure of 0.3 MPa, during the first 30 minutes of test, abundant precipitation is observed.
Figure 7. The view of the tank inner surface and fuel assembly simulator in test 4 at the moment when boric acid solubility limit is exceeded by 140 g kg^{-1} H_2O (a) and 177 g kg^{-1} H_2O (b).

As it can be seen from figure 6, during test 3, a significant increase in the height of the boric acid sediment layer at the bottom of the tank is observed. In test 4 (figure 7), in contrast to the previous experiment, an accumulation of acid on vertical surfaces inside the tank is observed.

Conclusions
The test facility with a seven-rod electrically heated fuel assembly simulator was constructed at IPPE. It has been used for the investigation of the processes of boric acid crystallization. The experimental results allow us to conclude that in case of an LOCA accident and operation of passive safety systems, H_3BO_3 crystals can form and accumulate on horizontal unheated surfaces in the volume of the core, for example on assembly bottom fittings. Also the formation of a boric acid film at the interface between the liquid and gas phases is possible. At the same time, the formation of boric acid crystals on the surface of fuel rod simulators and their accumulation in the gaps between the fuel rods have not been observed in the tests. The results obtained can be used to substantiate the operation of passive core flooding systems of new projects of NPPs with a WWER reactor.

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