Effect of mass transfer processes on accumulation and crystallization of boric acid in WWER core in emergency cases

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Abstract. In this paper the processes of boric acid mass transfer in a WWER-TOI nuclear reactor in case of the accidents with main coolant circuit rupture and operation of passive safety systems (the hydro accumulators systems of the first, second and third stages, as well as the passive heat removal system) are considered. The results of the calculation of changes in the boric acid solution concentration in the core for the WWER emergency mode are presented. According to the results of the calculation a significant excess of the ultimate concentration of boric acid in accidents with main coolant circuit rupture after 43 hours of emergency mode is observed. The positive influence of the boric acid droplet entrainment on the processes of its crystallization and accumulation in the core is shown. The mass of boric acid deposits on the internals is determined. The received results allow concluding that the accumulation and crystallization of boric acid in the core may lead to blocking the flow cross section and to deterioration of heat removal from fuel rods. The necessity of an experimental studies of the processes of boric acid drop entrainment under conditions specific to the WWER emergency modes is shown.

1. Introduction

Currently, because of the modern international requirements to ensure safety of a new generation nuclear power plants, issues related to the possibility of a long-term (up to 72 hours) autonomous cooling of the nuclear reactor core in case of a beyond design basis accidents are of high priority. In the Russian Federation design of the WWER-TOI reactor plant installation, developed on the basis of the NPP-2006 project. In this project, nuclear reactor core cooling in case of a beyond design basis accidents with main circulation pipeline (MCP) rupture and loss of all AC sources within 72 hours is provided by the functioning of passive safety systems (PSS). The hydro accumulator systems of the first, second and third stage (HA-1, HA-2, HA-3) included in the PSS, due to the successive supply of boric acid solution with concentration of 16 g/kg to the reactor, jointly with passive heat removal system (PHRS), prevent draining of the nuclear reactor core and enable removal of residual heat from the core in case of loss of coolant accident (LOCA) with total loss of AC power [1-4].

Considering duration of the emergency process, boiling of boron-containing coolant and low content of boric acid in the steam, leaving the reactor, conditions of possible accumulation and subsequent crystallization of boric acid can occur in nuclear reactor core. These processes can lead to
the appearance of flakes of boric acid in the coolant or deposit formation on the surfaces of fuel rods, which can lead to abnormality of core heat removal process. Consequently, studies related to the processes of mass transfer of boric acid in the WWER core in case of emergency take on important practical significance [5]. To estimate possibility of accumulation and crystallization of boric acid in the WWER reactor core, a calculation analysis of the change in boric acid concentration in the reactor in emergency mode has been done.

2. Determination of droplet entrainment value

Removal of boric acid from the reactor core in case of emergency can be carried out in two ways: by droplet entrainment in the steam-water mixture and due to steam solvent ability. There are a number of dependences that make it possible to determine the value of moisture of steam, leaving the reactor [6]. However, these dependences cover a limited range of parameters and are not always applicable for calculating droplet entrainment of moisture from a WWER core in case of emergency mode. For example, in [6] there is the following formula for calculating water content of steam:

\[ \omega = 2.75 \cdot 10^8 \frac{N^{2.3}}{Ga^{1.1} \left[ \rho' \left( \rho - \rho'' \right) \right]^{0.25}}, \]  

where \( N = \frac{w_0^2}{(\varphi g H)} \), \( w_0 \) – superficial steam velocity, \( \varphi \) – actual volumetric steam content, \( \rho' \) – fluid density, \( \rho'' \) – steam density, \( \omega \) – water content of steam, \( Ga \) – Galilean number, \( H \) – height of steam rise, \( g \) – gravity factor.

However, this dependence is applicable for calculating water content of steam only when the steam rise is less than or equal to 1 m, which is significantly lower than in the WWER reactor. In addition, there are dependencies that allow us to calculate the steam content in steam-water mixture flow [6]:

\[ x = 0.26(w_0/\sqrt{\varphi g/\sigma})^{0.36} \left( \frac{\rho''}{\rho - \rho''} \right)^{0.12}, \]

where \( \sigma \) – liquid tension

The comparison shows that when calculating water content of steam and steam content at the WWER core outlet, operating in emergency mode, according to the above dependences, divergence of results can reach 30%. Moreover, you must consider that calculation of the parameters of the steam-water mixture according to these formulas is possible only at a certain evaporation rate. In [6] boundary conditions are defined for which the dependences are valid, however, when beyond the boundary parameters, the calculating mechanisms are not proposed. The analysis showed that in the case of LOCA, evaporation rate is beyond the satisfiability limits of dependencies (1) and (2).

3. Calculation of the processes of mass transfer of boric acid in the nuclear reactor core

With regard to all stated above, use of dependences (1) and (2) to calculate the droplet entrainment processes at the early stage of the accident is not possible. Consequently, during the calculation analysis, the entire emergency process was divided into two stages. During the early stage of an accident (Figure 1a), a very high steam content is observed in core, which causes the foamed liquid level to rise above the upper perforation of the reactor barrel, and the steam-water mixture splashes out from the nuclear reactor core into the reactor volume and then into the containment through the pipeline rupture. Duration of this stage is ~ 7 hours. Further, the intensity of the residual energy release decreases, the steam content in the liquid volume decreases and the level of the boric acid solution is set at the height of the broken pipe of MCP (the second stage of the accident - Figure 1b).

During the calculation, a number of assumptions were made, needed due either to the complexity of the processes occurring in the circuit or to inadequacy of data on the properties of boric acid solutions:
Figure 1. Mass transfer of boric acid in WWER reactor vessel in case of LOCA. a) – the initial stage of an accident, b) - the second stage of the accident: 1 – reactor core; 2 – emergency core cooling system nozzle; 3 – separation collar; 4 – reactor barrel perforation; 5 – liquid level; \( \rightarrow \) – steam flow, \( \leftarrow \) – condensate and coolant flow; \( \downarrow \) – inflow of boric acid solution from the hydro accumulator systems; \( Q_R \) – heat energy released in the reactor; \( Q_{SC} \) – energy of steam going through a leak into the containment; \( Q_{HA} \) – energy removed from the core by the liquid from hydro accumulators; \( Q_{PHRS} \) – heat energy removed from the core due to operation of the PHRS.

1. During the calculations, two volumes were allocated in the system: the volume of the reactor core and the volume of the reactor pressure chamber (RPC);
2. The thermal physical properties of boric acid solution, such as density and viscosity, were assumed to be equal to water parameters under appropriate conditions;
3. All water entering the core and RPC is at saturation temperature;
4. Pressure in the system was assumed to be constant and equal to 0.3 MPa;
5. All boric acid, initially located in the primary circuit, as well as entered into the reactor from the accumulators of the HA-1 system is carried out into a containment.

When calculating, it was assumed that the rated power of the reactor is \( 3.2 \cdot 10^9 \) W. The change in residual heat removal \( (N_{RC}) \) in the reactor core is presented in [7]. The initial data for the calculations were the parameters of the accumulators of the passive flood of HA-2 and HA-3 [8].

At the initial stage of the emergency, when foamed liquid level is above the upper perforation of reactor barrel, intensity of the flow from the core into the RPC will be high, and mass of evaporated water during a period of time \( \Delta \tau \) will be as follows:

\[
\Delta m_{H_2O} = \frac{N_{RC} \cdot \Delta \tau}{h_{p} - h_{RPC}},
\]

where \( h^s \) – enthalpy of steam at saturation temperature (kJ/kg), \( h_{RPC} \) – enthalpy of boric acid solution in RPC (kJ/kg).

Boric acid solution mass entered into the reactor from the hydro accumulators and mass of the condensate formed in the steam generator (SG) due to the operation of PHRS will be as follows:

\[
\Delta m_{s}(\text{solution}) = (G_{HA} + G_{PHRS}) \Delta \tau,
\]

where \( G_{HA} \) – flow rate of boric acid solution from HA (kg/s), \( G_{PHRS} \) – condensate flow rate from SG (kg/s).

Mass of boric acid entered the reactor from hydro accumulators will be as follows:
\[ \Delta m_{HA}(H_2BO_3) = G_{HA} \cdot C_{HA}(H_2BO_3) \cdot \Delta \tau, \]

where \( C_{HA}(H_2BO_3) \) - concentration of boric acid in the hydro accumulators (kg/kg).

Since flow rate of the coolant entering the reactor exceeds the evaporation rate in the core, part of the solution will come into the containment. Mass of boric acid solution entering the containment is following:

\[ \Delta m_{n}(\text{solution}) = \Delta m_{n}(\text{solution}) - \Delta m_{n}(H_2O). \]

Accordingly, concentration of boric acid in the reactor core in the first stage of the emergency process is determined by the following formula:

\[ C_{i}(H_2BO_3) = \frac{(m_{RC} + m_{RPC})C_{i-1} + \Delta m_{n}(H_2BO_3)}{m_{RC} + m_{RPC} + \Delta m_{n}(\text{solution}) - \Delta m_{n}(H_2O)}, \]

where \( m_{RC} \) and \( m_{RPC} \) - mass of boric acid solution in reactor core and RPC respectively (kg).

Over time, the residual heat removal will decrease, and at the same time foamed liquid level in the reactor will also fall. The distance from the top of the core to the upper perforation at core barrel is 2.77 m, and elevation difference between the lower generatrix of the "cold" branch of primary circuit and the upper row of perforation holes in the reactor barrel is 1.845 m. Then the critical steam content when foamed liquid level will stand above the upper row of holes in the barrel in case of emergency with a "cold" leg break is:

\[ \phi_{cr} = 1.845/2.77 = 0.666. \]

Steam content is determined by the formula from [6]:

\[ \phi = 0.26 \left( \frac{w''^2}{g \sqrt{\frac{\sigma}{g}} (\rho'' - \rho''')} \right)^{1/3} \left( \frac{\rho''}{\rho'''} \right)^{0.12}, \]

where steam velocity \( w'' = N_{RC}/F_{CS} \).

In this model, the second stage of the emergency process begins after steam content drops below the critical one. At the second stage of the calculation, volumes of the reactor core and RPC are considered separately.

On the second stage of an accident energy of the residual heat will be spent on heating boric acid solution, entering the core from the RPC and evaporating it:

\[ N_{RC} = G_{N}r + G_{12} \left( r + h' - h_{RPC} \right), \]

where \( G_{N} \) - steam flow rate (kg/s); \( G_{12} \) - flow rate of a solution of boric acid from RPC to the core (kg/s), \( r \) - specific heat of evaporation (kJ/kg), \( h' \) - enthalpy of water at saturation temperature (kJ/kg), \( h_{RPC} \) - enthalpy of boric acid solution in RPC (kJ/kg).

Mass flow rate of boric acid leaving the core and RPC due to droplet entrainment processes \( G_{Re} \) is:

\[ G_{Re} = \omega G_{m}, \quad (3) \]

where \( G_{m} \) - steam-water mixture flow rate (kg/s).

Steam-water mixture leaving the reactor containing steam and drops of boric acid is replaced by a solution of boric acid coming from the RPC:

\[ G_{12} = G_{N} + G_{Re}. \quad (4) \]

A change in the mass of boric acid in the volume of the core per time interval \( \Delta \tau \) will be:

\[ \Delta m_{RC}(H_2BO_3) = (G_{12}C_{RPC} - G_{Re}C_{RC}) \Delta \tau. \]
where $C_{\text{RPC}}$ – concentration of boric acid solution in RPC (kg/kg), $C_{\text{RC}}$ – concentration of boric acid solution in the reactor core (kg/kg).

As noted above, solution of boric acid from RPC will flow into the core with a flow rate $G_{12}$, and the excess of solution will be poured through the ruptured MCP into the containment. On the basis of this, it is possible to determine mass of the solution that has poured out into the the containment in time $\Delta \tau$ and a change in mass of boric acid in the volume of RPC for the same time interval:

$$\Delta m_{\text{RPC}} (H_3BO_3) = (G_{\text{HA}} C_{\text{HA}} + G_{\text{RC}} C_{\text{RC}} - G_{12} C_{\text{RPC}} - G_{\text{sc}} C_{\text{RPC}}) \Delta \tau$$

To determine evaporation rate, taking into account (3) and (4), we obtain formula for calculating the residual heat:

$$N_{\text{RC}} = G_N \left[ (h^* - h_{\text{RPC}}) + \omega (h^* - h_{\text{RPC}}) \right].$$

Hence the steam flow rate is the following:

$$G_N = \frac{N_{\text{RC}}}{(h^* - h_{\text{RPC}}) + \omega (h^* - h_{\text{RPC}})}.$$ 

Change in boric acid concentration in the nuclear reactor core over the $\Delta \tau$ will be the following:

$$\Delta C_{\text{RC}} (H_3BO_3) = \frac{\Delta m_{\text{RC}} (H_3BO_3)}{m_{\text{RC}}},$$

4. Calculation results

The above stated dependencies are used to determine changes in boric acid concentration in RPC and the core. Variations in the parameters at the $i$-th step of the calculations over the time interval $\Delta \tau$ are calculated sequentially. The results are used for calculations at the next time interval. The results of calculation of change in boric acid solution concentration in the reactor core are shown in Figure 2.

**Figure 2.** Change of boric acid solution concentration in the core (I – duration of the first stage of the emergency process).

As the graph shows, concentration of $H_3BO_3$ at the end of 72 hours will be ~ 1,13 kg/kg $H_2O$, which significantly exceeds the limiting concentration of boric acid ($C_{\text{lim}}$), which at water saturation temperature under consideration is about 0,415 kg/kg.

After reaching the solubility limit of boric acid in the reactor core, the process of $H_3BO_3$ crystallization starts (after about 43 hours of the initial of emergency process). As noted above, deposits of boric acid in the core has significant impact on heat removal during the emergency process, therefore, the calculation of the boric acid deposit mass was also performed during the calculation. Deposit mass ($m_{\text{dep}} (H_3BO_3)$) was calculated using the following formula:
\[ m_{\text{dep}}(\text{H}_3\text{BO}_3) = m_{\text{RC}} \cdot \left( C_{\text{RC}}(\text{H}_3\text{BO}_3) - C_{\text{lim}} \right). \]

The calculation results are shown in Figure 3. As the graph shows, mass of boric acid deposits in the core to the end of 72 hours of emergency process is about 7150 kg.

5. Conclusion
Calculation of the processes of mass transfer of boric acid is of great practical importance in the analysis of emergency processes in WWER reactors of a new generation. The calculated estimation showed a significant excess of the limiting concentration of boric acid in the WWER-TOI reactor core in ~ 43 hours after start of emergency. This process can cause its crystallization on the fuel rods surface and deterioration of heat removal from the core. It should be noted that this calculation has been carried out using a number of conservative assumptions and did not take into account removal of boric acid from the nuclear reactor core due to dissolving ability of steam.

During the calculation analysis it was found that at the moment there is an uncertainty in the available dependencies for calculating moisture of steam and steam content of the medium at the outlet of the WWER core in case of emergency. These parameters are crucial for the analysis of the processes of mass transfer of boric acid. To eliminate existing uncertainties, experimental studies of the mass transfer processes of \( \text{H}_3\text{BO}_3 \) at the parameters specific to for emergency modes of NPPs with WWER are needed.

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