



Estimation influence of boric acid drop entrainment to its accumulation in the VVER reactor in the case of accident*

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Abstract

Process of boric acid mass transfer during accidents accompanied with rupture of circulation pipelines in VVER reactors of new generation equipped with passive safety systems are examined. Results of calculation of variation of boric acid concentration in VVER-TOI reactor in case of accident development process are presented. Positive effects of boric acid droplet entrainment on the processes of acid accumulation and crystallization in the reactor core are demonstrated. The obtained results allow formulating the conclusion on the possibility of these processes in the reactor core which may lead to the disruption of heat removal from fuel pins. Review of available published reference data on physical properties of boric acid solutions (density, viscosity, thermal conductivity) is given. It is established that available information is of too general nature and fails to cover the whole range of parameters (acid temperature, pressure and concentration) typical for potential emergency situation on NPP equipped with VVER reactor. Necessity of experimental study of processes of droplet entrainment under parameters typical for VVER emergency operation conditions, as well as investigation of thermal physics properties of boric acid within wide range of acid concentration values is required.

Keywords

VVER, emergency operation mode, boric acid, accumulation, droplet entrainment, thermal physics properties of boric acid solutions, density, viscosity, thermal conductivity.

Introduction

Investigation of processes of boric acid accumulation and crystallization in reactor cores of new generation of VVER reactors conducted at present acquires special importance and expediency. This is first of all associated with safety requirements imposed on modern NPPs. In

pursuance with EUR requirements on the standard and expanded design of new generation of nuclear power plants equipped with LWR-type reactors cooling system for heat removal from reactor core (RC) must be designed to be functional during 72 hours of standalone operation in case of beyond-design basis accidents. As it is known, in case of development of emergency situation accompanied

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with rupturing of the main coolant pipeline incorporated in AES-2006 design project operation of passive safety systems (PSS) ensures long-term (not less than 24 hours) cooling of reactor core due to the supply in the core of boric acid solution with concentration equal to 16 g/kg from accumulator tanks of the first (GE-1) and second (GE-2) stages (Kalyakin et al. 2003, Remizov et al. 2009, Morozov and Remizov 2012, Kalyakin et al. 2014). In accordance with concept of safety assurance implemented in the VVER-TOI project cooling down the reactor core due to the supply of boric acid solution in the core must be continued during 72 hours. In order to accomplish this task, it is planned to use the third stage of accumulator tanks (GE-3). Taking into account the duration of the process, coolant boiling and low concentration of boric acid in the steam phase probability of boric acid crystallization on elements of reactor internal devices is not excluded.

Carrying of boric acid out of reactor core with steam or due to droplet entrainment may significantly reduce the risk of its crystallization. Therefore, investigation of processes of boric acid carry-over from reactor core is of significant supplementary importance for calculation of accident evolution processes for NPPs with new generation of VVER reactors equipped with passive safety systems. Besides that, significance of thermal physics properties of boric acid, such as density, viscosity, thermal conductivity for conducting the above calculations has to be noted.

Thermal physics properties of boric acid solutions

Knowledge of thermal physics properties of boric acid is of considerable importance for calculations of processes of accumulation and crystallization of boric acid in VVER reactor cores. At present the data on the density, viscosity and thermal conductivity of boric acid solutions are of fairly general nature and fail to cover the whole range of parameters (temperature, pressure, acid concentration) typical for emergency situation on NPP equipped with VVER reactors. However, information can be found in reference sources about experimental studies of thermal physics properties of aqueous solutions of boric acid.

Experimental data on density of aqueous solutions of boric acid covering wide range of temperatures (25–300°C) and pressure (10–50 MPa) are presented in (Azizov and Ahundov 1996). At the same time solutions with concentrations equal to 3.1–43.4 g/kg were examined in the study which is significantly lower than possible boric acid concentration in the reactor core in case of accident.

Dependence of density of boric acid solution with concentrations equal to 3.1; 18.6 and 43.4 g/kg and pressure equal to 10 MPa versus temperature is shown in Fig. 1 (Azizov and Ahundov 1996).

Values of density and viscosity of boric acid aqueous solutions with concentrations equal to 2.52; 25 and 45 g/kg at atmospheric pressure and temperatures equal to 65.6°C and 100°C are presented in (WCAP-17021-NP 2009). The results are represented in Fig. 2.

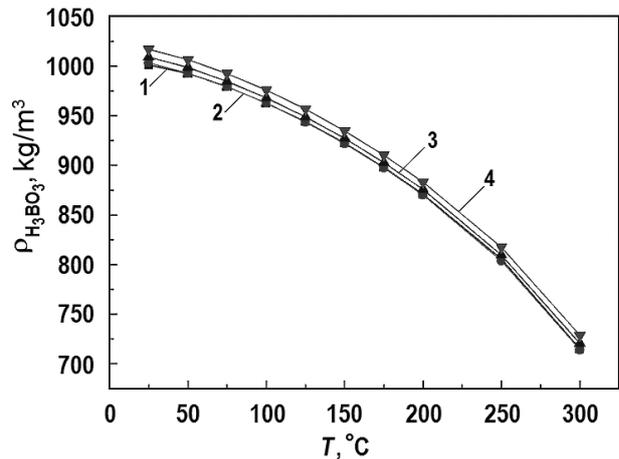


Figure 1. Dependence of density of boric acid solution on temperature at $P = 10$ MPa. Boric acid concentrations: 1–0 g/kg (H_2O); 2–3.1 g/kg; 3–18.6 g/kg; 4–43.4 g/kg

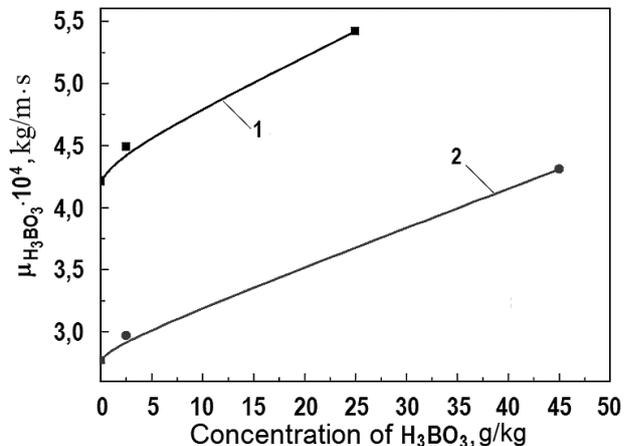


Figure 2. Dependence of density of boric acid aqueous solutions on concentration at pressure equal to 0.1 MPa (Azizov and Ahundov 1996). Temperature: 1–65.6°C; 2–100°C

Data on viscosity of boric acid aqueous solutions within the same range of concentrations are represented in Fig. 3.

Data on dynamic viscosity of boric acid aqueous solutions within the range of temperatures equal to 25–325°C and pressure range equal to 1–30 MPa are given in (Avanesyan and Ahundov 1980). Dependence of viscosity of boric acid aqueous solutions on temperature at pressure equal to 1 MPa is presented in Fig. 4 for concentrations equal to 2 and 20 g/kg.

Let us notice that studies (WCAP-17021-NP 2009, Avanesyan and Ahundov 1980) were conducted at low concentrations of boric acid solutions and within the pressure interval exceeding pressure characteristic in case of emergency process.

Results of studies of thermal conductivity of boric acid solutions are presented in (Gusejnov 2007). Measurements of heat conductivity were performed by the method of flat horizontal layer within the temperature interval of 290–430K, at temperatures equal to 0.1 and 10 MPa for solutions with concentration of boric acid equal to 1, 3 and 5 % (Fig. 5).

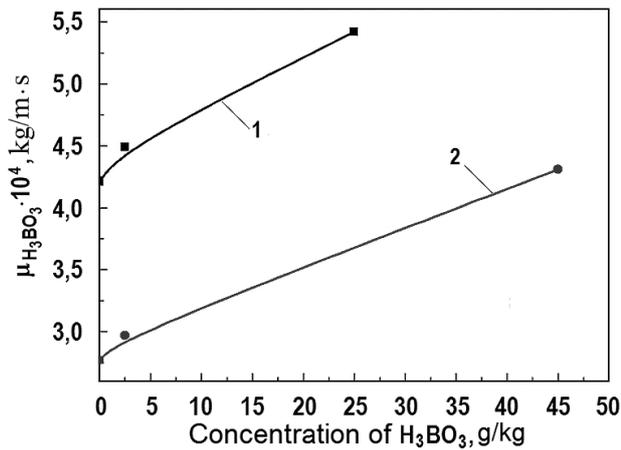


Figure 3. Dependence of viscosity of boric acid aqueous solutions on concentration at pressure equal to 0.1 MPa (Azizov and Ahundov 1996). Temperature: 1–65.6°C; 2–100°C

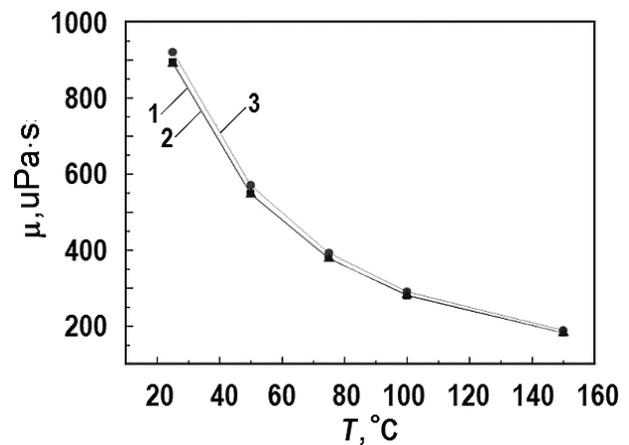


Figure 4. Dependence of viscosity of boric acid aqueous solutions on temperature for pressure equal to 1 MPa. Boric acid concentrations: 1–0 g/kg (H₂O); 2–2 g/kg; 3–20 g/kg

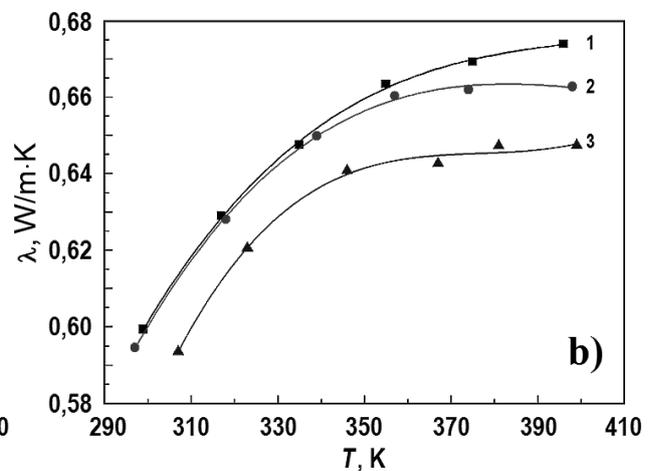
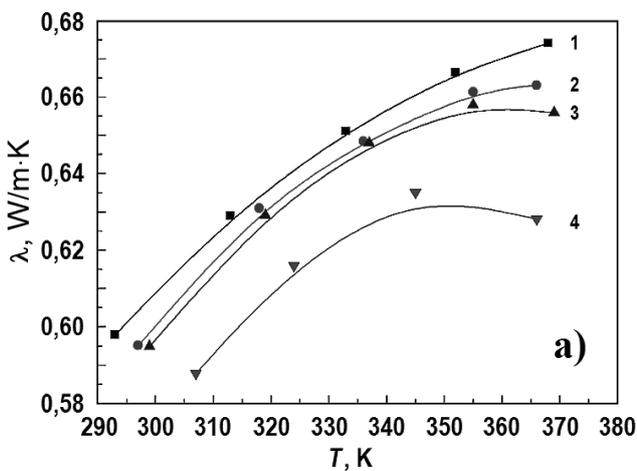


Figure 5. Dependence of thermal conductivity of aqueous solutions of boric acid on temperature and pressure: a)–0.1 MPa; b)–10 MPa. Concentration of boric acid: 1–0% (H₂O); 2–1%; 3–3%; 4–5%

A number of studies are dedicated to investigation of thermal physics properties of buffer solutions of boric acid where sodium phosphate and hydroxide were used as the alkalinizing agents (Yassin Hassan et al. 2015, Tuunanen et al. 1992). However, parameters of the investigated coolant do not correspond to water-chemistry operational conditions of NPPs equipped with VVER reactors and, besides that, similarly to the preceding papers investigation was conducted at fairly low concentrations of boric acid.

Despite the fact that in all the above reviewed papers results were obtained in experiments with low concentration of boric acid and in the conditions of performed experiments not corresponding to realistic parameters typical for emergency operation modes of NPPs with VVER reactors, the presented data demonstrate that thermal physics properties of H₃BO₃ solutions differ from the properties of water and this difference further increases with increased concentration of boric acid.

On the processes of mass transfer of boric acid in emergency operation modes of NPPs with VVER reactors

In order to evaluate the probability of crystallization of boric acid in reactor core it is necessary to address the processes of mass transfer of acid in case of development of emergency situation (Fig. 6).

Sequential activation of GE-1, GE-2 and GE-3 systems ensuring supply of boric acid in the reactor core takes place in case of rupture of the main coolant circulation pipeline. Significant amount of boric acid penetrating reactor core from accumulating tanks during the first moments after initiation of the accident will be carried away due to active steam generation, because in case of accident with rupture of the main circulation pipeline reactor is converted to the emergency evaporation operation mode.

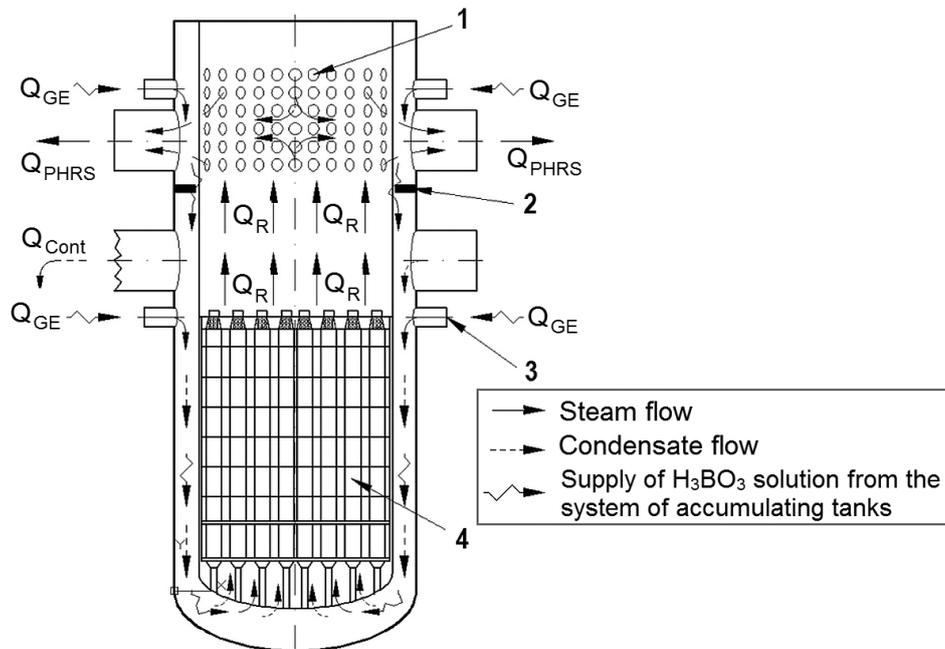


Figure 6. Mass transfer of boric acid in VVER reactor facility during accidents with rupture of the main coolant pipeline: 1 – reactor pit perforation; 2 – partitioning collar; 3 – flanged socket of the reactor core emergency cooling system; 4 – reactor core

Beside boric acid solution supplied from accumulating tanks of the PSS in the reactor penetration in the reactor core of condensate from steam generators of the three non-emergency cooling loops at saturation temperature in the primary cooling circuit begins after approximately 1.5 hours after initiation of the accident as the result of operation of passive heat removal system (Morozov and Remizov 2012a, Berkovich et al. 2010, Luk'yanov et al. 2010, Morozov and Shlyopkin 2016). Blending of flows of condensate with flows of boric acid solution supplied from the system of accumulating tanks takes place in the region of reactor bottom and, consequently, probability of crystallization of boric acid in the area appears to be low. Crystallization is possible in the reactor core due to the interaction of blended flows with cooler elements of reactor internal components. Certain fraction of boric acid carried away from reactor core with steam due to the droplet entrainment will penetrate steam generators where its precipitation will take place in contact with structural elements. Subsequently this process may lead to the poisoning of steam generator. Besides the above, certain part of boric acid will be carried away through the rupture of pipeline of the first cooling loop.

Reduction of boric acid concentration in the reactor core is possible as well as the result of its droplet entrainment. Estimation calculation of evolution of boric acid concentration in the reactor in emergency operation mode was performed for evaluating accumulation and crystallization of boric acid inside the VVER core and effects of droplet entrainment on the above processes. The main goal while performing the calculations was determination of the value of droplet entrainment at which concentration of boric acid in the reactor core will not exceed the limiting value.

Calculation of evolution of concentration of boric acid in the reactor core

A number of assumptions were made in the course of calculations the necessity of which is determined either by the complexity of processes taking place inside the cooling loop, or by the inadequacy of data on the properties of aqueous solutions of boric acid:

- Volumes of reactor pressure chamber (RPC), i.e. the space between the reactor core baffle and reactor vessel, were singled out in the system for performing calculations; at temperature values typical for accident accompanied with rupture of the main circulation circuit (120–140°C) gap is developed between the reactor vessel spacer ring and reactor pit allowing blending of boric acid solution supplied in the upper and lower reactor chambers and, therefore, a single unified volume, denominated as the RPC, from which boric acid is supplied in the reactor core is considered in the calculations;
- Properties of boric acid solutions in the reactor core and in the RPC are uniformly similar over the whole volume;
- All water supplied to the reactor core and in the RPC is at saturation temperature;
- Pressure in the system was accepted to be constant and equal to 0.3 MPa during the whole period of development of the emergency process;
- All boric acid originally located in the primary cooling loop, as well as acid supplied to the reactor from hydraulic accumulator tanks of the GE-1 system during the first several minutes after initiation of the accident is carried away in the reactor containment dome.

In connection with insufficiency of reference data thermal physics properties, such as density, viscosity and thermal conductivity were accepted to be equal to the values of parameters for water at corresponding conditions.

It was accepted in the calculations that nominal reactor power is equal to $3.2 \cdot 10^9$ W. Character of evolution of residual heat (N_{RC}) in the reactor core after its shutdown is represented in Table 1 (Kopytov et al. 2009).

Parameters of systems of GE-2 and GE-3 passive impoundment hydraulic accumulator tanks (Table 2) served as input data for the calculations (Schmal and Ivanov 2015).

Condensation capacity of steam generators was obtained from the results of experimental studies conducted on large-scale testing facility at the SSC RF-IPPE (Tuunanen et al. 1992).

$$N_{PHRS} = \begin{cases} 144.8 - 5.885 \cdot 10^{-4} \tau + 1.499 \cdot 10^{-9} \tau^2 & \text{at } \tau < 86400; \\ -131.25 + 7.619 \cdot 10^4 / \sqrt{\tau} & \text{at } \tau \geq 86400. \end{cases}$$

where N_{PHRS} is the condensation capacity of steam generator, kW; τ is the time, s.

Residual heat energy in the reactor core will be spent on evaporation of solution supplied in the core from RPC:

$$N_{RC} = G_N(h^2 - h\phi) + G_{12}(h\phi - h_{RPC}),$$

where G_N is the vented steam flow rate, kg/s; G_{12} is the cross-flow rate of boric acid from RPC in the reactor core, kg/s; r is the specific heat of evaporation, kJ/kg; $h\phi$ is the enthalpy of water at saturation temperature, kJ/kg; h_{RPC} is the enthalpy of boric acid solution in the RPC, kJ/kg.

Mass flow rate of boric acid exiting from the reactor core due to the processes of droplet entrainment is directly proportional to the steam mass flow rate (Sterman et al. 1982):

$$G_{Re} = k_{Re} G_N, \quad (1)$$

where G_{Re} is the mass flow rate of droplet entrainment of boric acid from reactor core in the RPC, kg/s; k_{Re} is the fraction of moisture carried away with steam (ratio of mass of droplets to the mass of dry steam).

Water-steam mixture exiting from reactor core volume and containing steam and droplets of boric acid solution is replaced by boric acid solution supplied from RPC and condensate supplied from steam generators:

$$G_{12} = G_N + G_{Re}. \quad (2)$$

Variation of boric acid mass (Dm_{RC}^B) in the reactor core volume during time interval Dt can be determined based on the above considerations as follows:

$$Dm_{RC}^B = (G_{12} C_{RPC} - G_{Re} C_{RC})Dt,$$

where C_{RPC} is the concentration of boric acid solution in the RPC, g/kg; C_{RC} is the concentration of boric acid solution in the reactor core, g/kg.

As it has been noted above, boric acid solution from RPC will crossflow in the reactor core volume as in the interconnected vessel with flow rate equal to G_{12} , while excess solution contained in the volume of reactor core will overflow in the volume of reactor vessel through the ruptured main circulation pipeline. Mass of solution overflowing in the volume of reactor containment during time interval Dt and variation of mass of boric acid in the RPC volume (Dm_{RPC}^B) during the same time interval can be calculated based on the above:

$$Dm_{RPC}^B = (G_{GE} C_{GE} + G_{PHRS} C_{RC} - G_{12} C_{RPC} - G_C C_{RPC})Dt.$$

In order to determine flow rate of vented steam we obtain expression for calculation of residual heat release taking into account (1) and (2) as follows:

$$N_{RC} = G_N [(h^2 - h_{RPC}) + k_{Re}(h\phi - h_{RPC})],$$

where h^2 is the enthalpy of dry steam at saturation temperature, kJ/kg.

Based on the above flow rate of vented steam is equal to

$$G_N = N_{RC} / [(h^2 - h_{RPC}) + k_{Re}(h\phi - h_{RPC})].$$

Variation of concentration of boric acid in the reactor core (DC_{RC}) and in the reactor pressure chamber (ΔC_{RPC}) will amount during the time interval Dt , respectively, to

$$DC_{RC} = Dm_{RC}^B / m_{RC}, \quad \Delta C_{RPC} = Dm_{RPC}^B / m_{RPC}.$$

Variation of enthalpy of the solution in the RPC (Dh_{RPC}) will be equal to the ratio of total enthalpy of flows entering the RPC to the sum of mass of boric acid solution

Table 1. Residual heat in the VVER core

Time, s	100	1000	$10 \cdot 10^3$	$28.8 \cdot 10^3$	$37.8 \cdot 10^3$	$50 \cdot 10^3$	$100 \cdot 10^3$	$130 \cdot 10^3$	$500 \cdot 10^3$	$147 \cdot 10^3$	$216 \cdot 10^4$
Residual heat, rel. units	0.0331	0.0206	0.0105	0.0077	0.0072	0.0067	0.0055	0.0053	0.0033	0.0022	0.0018

Table 2. Parameters of systems of passive VVER core impoundment hydraulic accumulation tanks

Parameter	Value					
	GE-2		GE-3			
Duration of the stage, s	100–5430		5431–10860	10861–29000	29001–86400	86401–259200
Boric acid solution flow rate per single channel, kg/s	10.0		5.0	3.3	1.6	1.6

already present in the RPC and mass of flows entering the volume in question per unit time. Boric acid solution is supplied to the RPC volume from the system of hydraulic accumulator tanks and from the reactor core via droplet entrainment. Besides the above, penetration of flows of condensate at saturation temperature from steam generators of the three non-emergency cooling loops must also be taken into consideration.

$$\Delta h_{RPC} = \frac{(G_{GE} h_{GE} + G_{PHRC} h' + G_{Re} h_{RC}) \Delta \tau}{m_{RPC} + (G_{GE} + G_{PHRC} + G_{Re}) \Delta \tau},$$

where G_{GE} is the flow rate of boric acid solution from GE, kg/s; h_{GE} is the enthalpy of boric acid solution in the GE, kJ/kg; G_{PHRC} is the condensate flow rate from steam generator, kg/s; h_{RC} is the enthalpy of boric acid solution in the reactor core, kJ/kg; m_{RPC} is the mass of water in the RPC, kg.

Calculation results

Results of calculation of variation of concentration of boric acid solution in the reactor core are presented in Fig. 7 depending on the value of droplet entrainment.

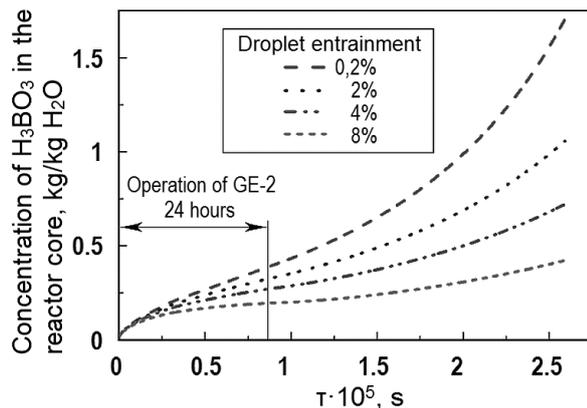


Figure 7. Variation of boric acid concentration in the reactor core at different values of droplet entrainment

The value of droplet entrainment was determined from calculations performed according to formula (1) taking into account different factors which can both increase content of moisture in steam and decrease it when steam-water mixture passes through the protective tube unit.

As it is clear from the above dependence the value of droplet entrainment may play significant role in the process of accumulation of boric acid. In the case when droplet entrainment will amount to 0.2% concentration of boric

acid in the reactor core will be equal to ~ 1700 g/kg, and at 2% to ~ 1060 g/kg; if, however, the value of droplet entrainment is equal to 4% then by the end of 72-hour period concentration of H_3BO_3 will be equal to ~ 722 g/kg. All these values significantly exceed the limiting concentration of boric acid which amounts to 415 g/kg for parameters of the accident.

In case when the value of droplet entrainment will amount to 8% concentration of boric acid in the reactor core will amount to ~ 423 g/kg which is insignificantly higher than the limiting value.

Thus, it can be stated that risk of crystallization of boric acid on different parts of internal core elements during the development of accident on VVER reactor facility is significantly reduced if the value of droplet entrainment of H_3BO_3 from the reactor core exceeds 8%. Let us note again that contribution of boric acid carrying away due to the solvent capacity of steam was not taken into account. The process in question can also positively affect the boric acid accumulation and crystallization.

Conclusion

Results were obtained indicating the possibility of exceedance of limiting concentration of boric acid during long-lasting process of accident development on NPP with VVER reactor (after 24-th hour of accident development). Risk of crystallization can be reduced due to the presence of processes of droplet entrainment of boric acid in the reactor core. Proof of that are the values of maximum H_3BO_3 concentration dependent on the value of moisture content of steam obtained in the calculations. Thus, positive effect of droplet entrainment on the processes of boric acid accumulation and crystallization was demonstrated. The implemented analysis of reference data demonstrated that the available information on the properties of boric acid are insufficient for performing comprehensive calculations of processes in VVER core in case of accident. This is associated, on the one hand, with low values of the investigated concentrations of H_3BO_3 and, on the other hand, with the mismatch between the experimental conditions and the parameters of emergency operation modes of VVER reactors. Consequently, knowledge of thermal physics properties of boric acid within wider range of concentrations is required for implementing more detailed calculation analysis of processes of accumulation and crystallization of boric acid. Expansion of this range is possible by conducting experimental studies.

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