The calculational modeling of impurity mass transfer in NPP circuits with liquid metal coolant

V Alexeev, F Kozlov, V Kumaev, E Orlova, Yu Klimanova, I Torbenkova
SSC RF-IPPE, Russian Federation

E-mail: alexeev@ippe.ru

Abstract: The authors create three levels of models (one-dimensional, two-dimensional and three-dimensional) for estimation of impurity mass transfer in sodium circuits units as well as applicable computational programs. In the one-dimensional model the flow path elements are simulated by annular channels. The Lagrange coordinate system is used in the mathematical description of processes in channels.

The two-dimensional model is based on the porous body approximation and enables one to simulate global spatial distributions of coolant flow velocity fields, coolant and fuel rods temperatures, and concentration distribution of transferred substances. The mathematical description of passive multicomponent impurity transfer is carried out using the system of the differential equations with sources and impurity diffusion, written for each component. The equations are solved by the finite-difference method.

The developed version of the three-dimensional code is based on a general approach of the spatial three-dimensional description of thermal-hydraulic and mass-transfer processes in fuel rod bundles. The determining system of finite-difference equations of hydrodynamics and heat exchange is obtained using the control volume approach.

As a result of the performed calculations, valuable data on corrosion products transfer in the primary circuit of the BN-600 reactor are obtained.

1. Introduction
The mass transfer of impurities in circuits with liquid metal coolant is determined by a complex of simultaneous hydrodynamic and physical-chemical processes including convective and diffusive transfer of dissolved impurity, reactions at the interphase, phase transitions in coolant volume as well as transport of suspended particles by coolant flows and their deposition on the circuit surface.

In non-isothermal sodium circulating circuits in the regions with increased temperature the ingress of corrosion products of structural materials in the form of a solution or particles (under certain conditions) in coolant occurs, which are then transferred through the circuit by hydrodynamic forces. At coolant cooling in the regions with lower temperature, the concentration of dissolved impurities can become supersaturated, which results in spontaneous formation of solid-phase nucleus in coolant volume. The presence of fine-dispersed suspended particles in coolant at high volumetric density of particles causes their coagulation, as a result of which the particle sizes increase. In some cases this can be the determining factor for the whole mass transfer process.

To estimate the effect of particle deposition on the circulating path surfaces on thermohydraulics of the installation, it is necessary to know both the amount (thickness) of depositions and their influence on the change of channel surface roughness.
The mass transfer of corrosion products in FR sodium circuits is simulated for the purpose of determining the conditions that provide effective performance of coolant and structural materials, including evaluating allowable increase of coolant temperature level and impurity concentration in coolant; obtaining quantitative data on ingress and accumulation of solid-phase impurities in the elements of the hydraulic circuit and in gas cavities of NPP sodium circuits.

The questions of modeling of impurity mass transfer in circuits with liquid metals are considered in [1-3].

2. One-dimensional model

The one-dimensional mathematical model of impurity mass transfer in sodium circuits gives a comprehensive description of the process, including the following stages.

- ingress of components of structural materials (Fe, Cr, Ni) in coolant;
- transfer of structural material components by coolant flow;
- formation of solid-phase nucleus at coolant supersaturation by metal impurities (in the cooled regions);
- particle coagulation in the formed fine disperse system;
- particle deposition on the flow path surface;
- particle sedimentation in stagnant zones;
- crystallization of dissolved impurity on surface of particles or dissolution of particles;
- crystallization of dissolved impurity on surface of flow path channels.

Density of particle size distribution (Bennett law)

\[ \varphi(l) = \frac{a}{l_0} \left( \frac{l}{l_0} \right)^{a-1} \exp \left( -\left( \frac{l}{l_0} \right)^a \right), \]  

where \( l \) – the particle size;
\( l_0 \) and \( a \) – constants for the given disperse system.

Concentration of particles from \( l \) up to \( l + dl \) in size

\[ d n(l) = \frac{c \varphi(l) dl}{m(l)}, \]  

where \( c \) – concentration of all particles;
\( m(l) \) – weight of one particle.

Density of particle mass distribution (mass spectrum)

\[ f(m) = \frac{d n(m)}{dm} = \frac{c \varphi(l) l}{3 m^2}. \]  

Figure 1 presents the comparison of the curves for various density of particle size distribution. As it is seen from Figure 1, using Eq. (1) and varying the factors \( a \) and \( l_0 \) the distribution curves can be obtained with various scatter and position of maximum point.
Figure 1. Density of solid phase nucleus distribution by Eq. (1) for various determining parameters:
1 – $a = 9$, $l_e = 1.77 \times 10^{-9}$ m;
2 – $a = 6$, $l_e = 2.5 \times 10^{-9}$ m;
3 – $a = 4$, $l_e = 3 \times 10^{-9}$ m.

The evolution of disperse system (the basic kinetic equation of coagulation) is described by the equation

$$
\frac{d f(m)}{d t} = \frac{1}{2} \int_{0}^{\infty} \beta(m, m') f(m) f(m') dm' - \int_{0}^{\infty} \beta(m, m') f(m) f(m') dm' + \frac{I_1(m) + I_2(m)}{m \cdot d m}
$$

$$
- ko(m) f(m) \frac{S}{V} - kz(m) f(m),
$$

where $m, m'$ – weights of colliding particles;
$S$ and $V$ – the area of the surface washed by coolant and volume of the section of the circuit flow path being considered;
$ko(m)$ – factor of particles deposition on channel surface;
$kz(m)$ – the factor describing sedimentation of particles in stagnant zones;
$\beta$ – kernel of the kinetic equation;
$I_1(m), I_2(m)$ – intensity of volumetric and erosive sources of suspension for particles of mass $m$, kg/(m$^3$ c).

The time variation of concentration of dissolved corrosion products equation is obtained (for cylindrical channel)

$$
\frac{d c^f_{D}}{d t} = \frac{4 \beta_{l} (c^w_{D} - c^f_{D})}{D} - \sum_{m} \frac{6 \beta_{p} f(m) \cdot m}{\rho_{p} l} \frac{c^f_{D} - c^{w}_{D}}{\rho_{m} l} \cdot d m,
$$

where $c^w_{D}$ and $c^f_{D}$ – concentration at wall of the channel and saturation in flow;
$\beta_{l}$ and $\beta_{p}$ – mass transfer coefficients between channel wall and particle surface;
$\rho_{p}$ and $l$ – particle density and size;
$\rho_{m}$ – mass of the largest particle.

The closure equations include the equations describing mass transfer between a particle and turbulent liquid flow, between coolant flow and channels walls, deposition of particles on channel surface, heat exchange between coolant flow and channel walls.

In the one-dimensional mathematical model the following approach to the description of mass transfer of corrosion products in the BN-600 primary circuit was applied.
• The hydraulic circuit with circulation of main coolant flow (no less than 94% of maximal
   flow rate in the primary circuit) is selected, figure 2.
• The primary circuit is conventionally divided into a number of the sections with constant
geometrical and hydrodynamical parameters.
• As a first approximation the sections are modeled by straight annular channels.
• The Lagrange axis is used in the mathematical description of processes (the coordinate axes
are connected to coolant flow).
• Time of coolant transfer, geometrical parameters, hydrodynamical and temperature regimes
in each section of the circuit are determined. The duration of sodium transfer through the
whole circuit is 48.37 s.

Figure 2. The schematic of BN-600 primary hydraulic circuit: 1 – core; 2 – breeding zone; 3 –
assembly storage; 4 – top chamber + mixing chamber + collector of intermediate heat
exchanger (IHE); 5 – IHE; 6 – collector of IHE + overflow chamber + pump suction; 7 –
pressure pipeline; 8 – head chamber.

3. Two-dimensional model
The two-dimensional model is based on porous body approximation and allows the simulation of
global spatial distributions of velocity fields of coolant flow, coolant and fuel rods temperatures,
and concentration distribution of transported substances.

The mathematical description of transfer of passive multicomponent impurity can be carried out
by means of a system of differential equations of transfer with sources and diffusion, which are
written for each impurity component [4, 5]. The system of the equations of impurity component
transfer involves immediately the convective terms dependent on the distribution of velocity vector
in coolant flow as well as the sources dependent on medium temperature. It is assumed that there is
no direct inverse effect of mass transfer processes on coolant flow dynamics and heat exchange,
which allows the system of transfer equations to be solved independently, but only after performing
the thermal-hydraulic calculation that defines the distribution of coolant flow velocity and
temperatures in the circuit.

The system of the equations describing the specified processes takes the following form in the
Cartesian coordinate system.

Equation of transfer:

$$\frac{dC^n}{d\tau} + \sum_{p=1}^{N} J^{np} e = \frac{d}{dx_k} \left( D^n \frac{dC^n}{dx_k} \right), \quad (6)$$

where \( u_k \) – components of velocity vector, \( k=1,2; \)
\( C^n \) – concentration of \( n \) component of impurity;
\( D^n \) – diffusion factor of \( n \) component;
\( \varepsilon \) – volumetric porosity of coolant environment;
\( J^{np} \) – mass source of \( n \) component at its formation from \( p \) component.

The equations of coolant flow and continuity in porous medium simulating the structure and geometry of the circuit element being considered are of the form:

\[
\frac{du_i}{dt} + u_k \frac{du_i}{dx_k} = -\frac{1}{\rho_0} \frac{dp}{dx_i} + \frac{1}{\varepsilon} \frac{d}{dx_k} \varepsilon \frac{du_i}{dx_k} - \Lambda \frac{1}{2d_h} \rho \mu_i + g_i \frac{\rho(t)}{\rho_0},
\]

\[
\frac{d}{dx_k} u_i \varepsilon = 0,
\]

where \( \Lambda \) – medium resistance coefficient estimated per unit volume of the medium;
\( \rho_0 \) – density of medium estimated for average temperature;
\( \nu \) – coefficient of kinematic viscosity;
\( d_h \) – hydraulic diameter of porous body;
\( g_i \) – projections of vector of free fall.

Energy equations for coolant and porous body:

\[
\frac{d}{dt} \frac{d}{dx_k} \varepsilon p_c \rho \frac{d}{dx_k} + \frac{d}{dx_k} \varepsilon p_c \rho u_k \frac{d}{dx_k} = \frac{d}{dx_k} \varepsilon \lambda_c \frac{dt_s}{dx_k} + \varepsilon q_v + K(t_s - t)S,
\]

\[
\frac{d}{dt} \frac{d}{dx_k} \varepsilon s \rho_s \frac{d}{dx_k} + \frac{d}{dx_k} \varepsilon s \rho_s u_k \frac{d}{dx_k} = \frac{d}{dx_k} \varepsilon_s \lambda_s \frac{dt_s}{dx_k} + \varepsilon_s q_v + K(t_s - t)S,
\]

where \( t \) – coolant temperature;
\( t_s \) – temperature of a porous body;
\( \varepsilon_s = 1 - \varepsilon \) – volume fraction of porous body (solids) in the medium;
\( \rho_c, \rho_s \) – density of coolant and porous body;
\( c_p, c_{ps} \) – specific heat capacity of coolant and porous body;
\( \lambda_c, \lambda_s \) – thermal conductivity of coolant and porous body;
\( q_v, q_{vs} \) – volumetric density of heat generation in the coolant and porous body;
\( S \) – heat-transfer surface between coolant and porous body in unit volume of the medium;
\( K \) – heat transfer coefficient between coolant and porous body.

The equations are solved by means of finite-difference methods:

- the equation of transfer and energy – by the method of alternating directions;
- the equation of flow dynamics - by the semi-implicit sweep local one-dimensional method.

4. Three-dimensional model

The three-dimensional model is also based on porous body approximation [6].

The developed version of the three-dimensional code is based on a general approach to the spatial description of three-dimensional thermal-hydraulic and mass-transfer processes in rod bundles. The determining system of finite-difference equations of hydrodynamics and heat exchange is obtained using the control volume method, which consists in approximating the integral laws of conservation of mass, momentum and energy for the elements of control volume system covering the
computational domain. As such system the system formed by a triangular-prismatic mesh is used, with the mesh points coinciding with the centers of fuel rods (see Figure 3). A triangular prism is a system element; it can simulate both stationary (solid) and flowing (coolant in interspace of fuel rods) medium. The mesh is employed that divides the fuel element in its cross-section into 6 triangular elements and the interspace of fuel elements into 6 triangular elements as well.

Figure 3. Distribution of control volumes in the cross section of the computational domain with the highlighted sub-regions simulating fuel rods and hexagon shell (the diagram presents the central part of the computational domain in the x-y plane for a 127-rod assembly).

The equations of hydrodynamics for “solid” and “liquid” elements are uniformly approximated similar to that in the initial version of the code. This allows the procedure of end-to-end calculation to be used. The zero velocity of solid movement is obtained automatically from solving the equations of motion with the given volume resistance, which is considered to be a large value for solids. For the mesh cells describing the inter-space of fuel rods with coolant, the volume resistance is set according to the data on hydraulic resistance in rod bundles at longitudinal flow. For the mesh cells situated outside the region of the rod bundle, the volumetric resistance is assumed to be equal to zero.

5. The results of calculations

5.1 One-dimensional model
The calculations have shown that in approximately 400 circulation cycles in the BN-600 primary circuit the equilibrium disperse system is formed in sodium, with its parameters (density of distribution and concentration of particles) not changing at further increase of circulation cycles. The
parameters of the formed disperse system do not depend on the shape of nucleus distribution (initial curves in Figure 1).

The calculation allowed the quantitative characteristics to be obtained of the formed self-preserved disperse system of corrosion products in the BN-600 primary circuit. This curve is approximated by analytical relation (11) and the Bennett equation with the parameters \( l_e = 1.95 \times 10^{-8} \) m and \( a = 2.7 \), which are presented in Figure 4.

\[
g(m) = 10^{13.924 - 11.3 \times 10^{-15} m^{0.384} - 1100 m^{0.16}}
\]

The average size of corrosion product particles in the BN-600 primary circuit is about 0.02 \( \mu \)m; the sizes of a great bulk of dispersed-phase particles do not exceed 0.04 \( \mu \)m. The concentration of the whole spectrum of suspended particles in sodium is about 0.0004 ppm. That is what together with dissolved components of structural material basically determines mass transfer of corrosion products in the circuit.

As it follows from the calculation, in the regions with maximum flux density, the deposition rate is about 0.38 microns/year in the core (with the dissolution of depositions not considered) and 0.08 microns/year in the IHE at a 50 % porosity of depositions.

On the average for fuel assemblies, the amount of soluble material is 3 times greater than the amount of deposited particles. In the core the depositions are formed on surfaces at the inlet region. At the core outlet there takes place the dissolution of structural material in sodium.

The density distributions of corrosion products mass flux along the BN-600 intermediate heat exchanger and core are shown in Figure 5 and Figure 6.

The obtained data testify that under the steady-state operation conditions (at nominal parameters) the mass transfer of structural material corrosion products in the BN-600 primary circuit will not result in the change of operational characteristics of the primary circuit during the whole lifetime of the reactor.
5.2 Two-dimensional model

For the numerical simulation of mass transfer in the BN-600 primary circuit a two-dimensional mesh consisting of ~18000 cells was used. The computational domain representing a cylinder of ~3 m in radius and ~13 m in height was simulated by a porous medium with parameters of a real reactor medium in terms of porosity, heat generation and hydraulic resistance. The computational domain presents the core with reflector, pressure head chamber, mixing chamber, downcomer channel and heat exchanger zone. The temperature of all components and total flow through the reactor were specified as initial conditions to calculate the steady-state mode.

The central part of the reactor involving the core with reflectors, top and bottom chambers, central column were simulated by cylindrical elements with porosity and heat generation corresponding to the real values for this region. In this case the geometrical structure of the region and spatial pattern of thermal-hydraulic processes were not disarranged in the main.

The peripheral area of the hydraulic path involving heat exchangers, downcomer, inlet and outlet nozzles is of three-dimensional structure that cannot be described by a two-dimensional model. Therefore, an approximation method was used and this zone was modeled by parallel annular channels with the heat-and-mass-transfer surfaces and coolant velocity similar to those of a real structure.
The operability of the code for performing calculations in two-dimensional approximation is confirmed by the data obtained: the ingress of corrosion products from structural material surface into coolant is observed in the circuit sections with power generation; the flow of dissolved impurities is observed in the circuit sections with relatively low temperatures (the zone of heat exchanger, bottom pressure chamber). The concentration distribution of particles and dissolved corrosion products in the cross section of the primary hydraulic path was also obtained (see Figure 7 and Figure 8).

5.3 Three-dimensional model
The test calculations were performed of the concentration of dissolved corrosion products in sodium in a 127-rod assembly of the BN-600, which make it possible to estimate the concentration levels at fuel rod surface and in primary sodium flow.

Figure 9 presents the distribution of corrosion products concentration with height at the fuel rod wall: on the left - in the center of the fuel assembly, on the right – at the periphery. As it follows from the figure, the central rods show rather higher increase of corrosion products concentration in the near-wall region as compared with the peripheral ones. The values of concentrations can differ several times. A similar difference was obtained for corrosion products flux from the fuel rod surface to coolant, see Figure 10.

The calculations performed allowed the estimation of concentration distribution of corrosion products in sodium along the fuel rod perimeter, see Figure 11. The concentration of corrosion products in sodium along the perimeter of fuel rods are non-uniform for peripheral rods, and as judged by the data obtained the near-wall concentrations of impurity are considerably higher than its volume concentrations in sodium; a non-uniform nature of near-wall concentrations along the perimeter was not revealed for central fuel rods.

6. The conclusion
The results presented in the report on the development of mathematical models of impurity mass transfer in circuits with liquid metal coolant and applicable computer codes of different levels indicate that some progress in this area of research has been achieved. The adequate mathematical models have been developed; the operability of the developed software has been confirmed. As a result of the performed calculations, valuable data on corrosion products mass transfer in the primary circuit of the BN-600 fast reactor were obtained. The use of these data will improve the degree of the reactor safety.

At the same time, this line of investigations is at its initial stage and requires further innovative development. It is essential to substantiate a comprehensive simulation of physical-chemical, thermal-hydraulic and technological processes in sodium circuits of NPP, including the development of associated three-dimensional codes.
Figure 7. Distribution of dissolved corrosion products concentration

Figure 8. Distribution of particles concentration
Figure 9. The corrosion products concentration in the near-wall region of rods, distribution along the rod height: on the left – bundle center, on the right – peripheral rods

Figure 10. The corrosion products flux from wall: on the left – bundle center, on the right – peripheral rods
Figure 11. The corrosion products concentration in coolant flow at the core outlet \((10^{-8} \text{ kg/m}^3)\). Figure shows 1/4 parts of section.

References

[1] Alexeev V, Kozlov F, Zagorulko Yu, etc 1996 Research of dynamics of suspensions behaviour in sodium circuits of fast reactors Preprint of SSC RF IPPE/№2576

[2] Alexeev V, Kozlov F, Zagorulko Yu, Kraev N 1997 Research of mass transfer of nickel and chromium in not isothermal site of circulating sodium circuit Preprint of SSC RF IPPE/№2666

[3] Alexeev V 2002 The tritium and structural materials corrosion products mass transfer in circuits with sodium coolant The dissertation on a scientific degree of Dr.Sci.Tech. /SSC RF IPPE, Omsk

[4] Kumaev V, Lebezov A, Alexeev V 2005 development and application of MASKA-LM code for calculation of thermal hydraulics and mass transfer of lead cooled fast reactors The 11th International Topical Meeting on Nuclear Reactor Thermal-Hydraulics (NURETH-11), Paper: 191. (Avignon, France, October 2-6, 2005) pp 191/1-191/6

[5] Patankar S 1984 Numerical methods of the solution of problems of heat exchange and dynamics of liquid (Moscow: Energoatomizdat)

[6] Kumaev V, Leonchuk M, Dvortcova L 1985 Technique of numerical calculation of three-dimensional flows of coolant in rod bundles Preprint of SSC RF IPPE/№1733