An investigation of tritium transfer in reactor loops

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Abstract. The work is devoted to the important task of the numerical simulation and analysis of the tritium behaviour in the reactor loops. The simulation was carried out by HYDRA-IBRAE/LM code, which is being developed in Nuclear safety institute of the Russian Academy of Sciences. The code is intended for modeling of the liquid metal flow (sodium, lead and lead-bismuth) on the base of non-homogeneous and non-equilibrium two-fluid model. In order to simulate tritium transfer in the code, the special module has been developed. Module includes the models describing the main phenomena of tritium behaviour in reactor loops: transfer, permeation, leakage, etc. Because of shortage of the experimental data, a lot of analytical tests and comparative calculations were considered. Some of them are presented in this work. The comparison of estimation results and experimental and analytical data demonstrate not only qualitative but also good quantitative agreement. It is possible to confirm that HYDRA-IBRAE/LM code allows modeling tritium transfer in reactor loops.

1. Introduction

Nowadays the fast reactor technology is one of the most promising ways for liquid-metal reactor development. In a view of great importance to provide radiation safety in these reactors, it is necessary to simulate the main physical processes of radionuclide behavior during normal operation. To carry out such simulations it should be indispensable to develop computer codes for modeling the main physical and chemical processes.

Tritium is a radioactive isotope of hydrogen and its half-life is 12,32 years. The nucleus of a tritium atom consists of a proton and two neutrons. Helium, beta-particle and antineutrino are generating during a radioactive decay of tritium:

\[
^3H \rightarrow \ ^3He + \beta + \nu^-
\]

Tritium mostly comes from the reactor core by ternary fission and its production rate depends on the reactor power. Due to its good permeability, tritium can leave the reactor core, transfer in channels and permeate through pipe walls even during normal operation. These properties of tritium (good permeability and radioactivity) make computational simulation of tritium transfer very important problem.

In this paper we consider the problem of modeling of the tritium transfer behaviors, such as tritium transport, permeation, leakage, etc. Simulation was carried out by HYDRA-IBRAE/LM code, which is being developed in Nuclear Safety Institute of the Russian Academy of Sciences (IBRAE RAN). The code is based on thermal-hydraulic two-fluid model, containing three one-dimensional conservation equations for two phases (gas and fluid). There are some modules in HYDRA-
IBRAE/LM code for special computational problems, for tritium transfer simulation a module named TRITIUM was developed. TRITIUM module includes following models:

- Tritium source in the primary loop;
- Tritium and hydrogen transfer in the primary loop;
- Tritium and hydrogen source in the secondary loop;
- Tritium and hydrogen source in the tertiary circuit;
- Tritium and hydrogen permeation to the outside through the walls of components and piping;
- Tritium and hydrogen leakage;
- Tritium and hydrogen removal by the purification system installed in the primary and secondary loops.

2. Mathematical model

In HYDRA-IBRAE/LM code, tritium mass transfer processes are considered in channel and basic mass-balance equation is

\[
\frac{dM_{ij}}{dt} = \sum_{k} \frac{M_{ik} \cdot \tau}{\tau_{k,j}} - \frac{M_{ij} \cdot \tau}{\tau_{j}} + S_{ij} - R_{ij}
\]  

(1)

Where \(M_{ij}\) - the mass of chemical \(i\) in node \(j\), \(t\) - time, terms \(\sum_{k} \frac{M_{ik} \cdot \tau}{\tau_{k,j}}\) and \(\frac{M_{ij} \cdot \tau}{\tau_{j}}\) are source and sink in the node, \(S_{ij}\) and \(R_{ij}\) - mass source of chemical and mass sink rate of component in the node. Source and sink terms are determined by the phenomena taking place in the node, e.g. tritium permeation, leakage or tritium removal by purification system. Terms \(S_{ij}\) and \(R_{ij}\) depend on coolant properties, pipe material and geometrical properties. For the same reactor items a type of the equations (1) is similar.

HYDRA-IBRAE/LM code computes equation coefficients dynamically; it means that the coefficients and values depend on system characteristics changing over time, e.g. temperature, pressure, etc.

3. Analytical tests

3.1. Test 1 (Initial tritium concentration in a channel)

Analytical test on tritium transport in a channel was computed by HYDRA-IBRAE/LM code. There was initial tritium concentration in a horizontal sodium-filled circle tube. The inlet sodium flow rate was constant. Initial and boundary conditions for simulation are listed in Table 1.

| Parameter               | Value  | Unit |
|-------------------------|--------|------|
| Channel length          | 0,1    | m    |
| Diameter                | 0,03   | m    |
| Inlet flow rate         | 2·10^{-4} | m³/s |
| Sodium temperature      | 835    | K    |
| Sodium pressure         | 1,52·10^{5} | Pa   |
| Initial tritium mass    | 2·10^{12} | kg   |

Table 1. HYDRA-IBRAE/LM initial and boundary conditions.

Assuming that there is one node in the channel, the equation (1) is simplified in following way:

\[
\begin{align*}
\frac{dM_{1}}{dt} &= -\frac{M_{1}}{\tau} \\
 t = 0 & : M_{1} = M_{0}
\end{align*}
\]
Where \( \tau \) is the characteristic time, which is computed by equation:

\[
\tau = \frac{V_{ch}}{G_V} = \frac{L}{U}
\]

(in the cylindrical channel),

Where

- \( V_{ch} \) – channel volume \((m^3)\);
- \( G_V \) – coolant flow rate \((m^3/s)\);
- \( L \) – channel length \((m)\);
- \( U \) – coolant velocity \((m/s)\).

Analytical solution of this equation is expressed by

\[
M_i(t) = M_o \exp\left(-\frac{t}{\tau}\right)
\]

This test was computed using HYDRA-IBRAE/LM code. The computational results are shown in the Fig. 1. The red line is related to HYDRA-IBRAE/LM results, the black one is related to analytical solution.

![HYDRA-IBRAE/LM results in comparison with analytical solution.](image)

**Figure 1.** HYDRA-IBRAE/LM results in comparison with analytical solution.

Figure 1 demonstrates very good agreement between analytical solution and HYDRA-IBRAE/LM computations. An average relative error for the analytical test was smaller than 0.01%.

### 3.2. Test 2 (Tritium source in a channel)

The second analytical test is devoted to the tritium transfer in a channel, which has an initial tritium source. The inlet sodium flow rate was constant. In the first part of the channel there is a tritium source. Initial and boundary conditions for simulation are listed in Table 2. During the estimation, tritium mass in the channel was calculated.

| Parameter          | Value | Unit |
|--------------------|-------|------|
| Channel length     | 0,2   | m    |
| Diameter           | 0,45  | m    |

**Table 2.** HYDRA-IBRAE/LM initial and boundary conditions.
Inlet flow rate | 2·10^{-2} | m³/s
---|---|---
Sodium temperature | 835 | K
Sodium pressure | 1.52·10⁵ | Pa
Initial tritium source | 4·10^{-12} | kg/s

Assume that there are two equal parts in the channel. The main equation is simplified:

\[
\begin{align*}
\frac{dM_1}{dt} &= Q - \frac{M_1}{\tau} \\
\frac{dM_2}{dt} &= \frac{M_1 - M_2}{\tau} \\
M_1(0) &= M_2(0) = 0,
\end{align*}
\]

where \( M_i \) – tritium mass in node \( i \) and \( \tau \) is the characteristic time, which is computed by equation:

\[
\tau = \frac{V_{ch}}{G_v} = \frac{L}{U} \quad \text{(in the cylindrical channel)},
\]

Where

\( V_{ch} \) – channel volume (m³);
\( G_v \) – coolant flow rate (m³/s);
\( L \) – channel length (m);
\( U \) – coolant velocity (m/s).

Analytical solution of this task is expressed by

\[
\begin{align*}
M_1(t) &= Q\tau \left( 1 - \exp \left( -t / \tau \right) \right) \\
M_2(t) &= Q\tau \left[ 1 - \frac{1}{1 + t / \tau} \exp \left( -t / \tau \right) \right].
\end{align*}
\]

Figures 2-3 show the comparison between analytical solution and HYDRA-IBRAE/LM results. The red line is related to HYDRA-IBRAE/LM results, the black line is related to the analytical solution.
Figure 2. HYDRA-IBRAE/LM results in comparison with analytical solution (node 1).

Figure 3. HYDRA-IBRAE/LM results in comparison with analytical solution (node 2).

Average relative error is no more than 0.01%. This result demonstrates good qualitative and quantitative agreement.

4. Comparative computations with THYTAN code

4.1. Modeling of purification system
Unfortunately, suitable experimental data for verification of purification system does not exist, so the purification model was verified by analytical tests and comparative computations with another computer codes. The numerical analysis code, THYTAN (Tritium and HYdrogen Transportation
ANalysis [1], was developed by the Japan Atomic Energy Research Institute (a former organization of the Japan Atomic Energy Agency). THYTAN was developed in the late 1990s as a tool for estimating tritium transport in the HTGR (high-temperature gas-cooled reactor) hydrogen production system.

We consider a helium-filled channel with a flow into a cold trap. In THYTAN code there is a similar mass balance equation [1]:

\[ V_j \frac{dC_{ij}}{dt} = \sum (F_{\text{total, }i-1} \cdot C_{i-1, j}) - F_{\text{total, }j} \cdot C_{i, j} - S_{i, j}, \]

where

\( V_j \) – the volume of node (m³);

\( F_{\text{total, }j} \) – the volumetric flow rate of all chemicals in node \( j \) (m³/s);

\( C_{i, j} \) – the volume fraction of chemical \( i \) in node \( j \) (m³/m³);

\( S_{i, j} \) – the total amount of volume change rate of chemical \( i \) in node \( j \) by considering generation, release, permeation, removal, leakage, and isotope exchange reactions (m³/s).

Tritium behavior in the purification system is expressed by the following equation:

\[ \frac{dC}{dt} = -k_{ct} C \cdot \frac{F_{PF, He}}{V_j}, \]

where

\( C \) – tritium concentration in node (m³/m³);

\( k_{ct} \) – fractional efficiency of purification system;

\( F_{PF, He} \) – helium flow rate at purification system (m³/s);

\( V_j \) – the volume of node (m³).

THYTAN boundary conditions for this test are listed in Table 3.

| Parameter                  | Symbol | Value  | Unit   |
|----------------------------|--------|--------|--------|
| The volume of node         | \( V_j \) | 1      | m³     |
| Helium flow rate at system | \( F_{PF, He} \) | 1,0 \times 10^{-3} | m³/s  |
| Fractional efficiency      | \( k_{ct} \) | 0,9    | –      |
| Initial tritium conc.      | \( C_{0, j} \) | 1,0 \times 10^{-9} | m³/m³ |

HYDRA-IBRAE/LM input parameters for modeling this test are listed in Table 4.

| Parameter       | Value | Unit |
|-----------------|-------|------|
| Channel length  | 10    | m    |
| Diameter        | 0,799 | m    |
| Number of cells | 5     | –    |
|                      |       |     |
|----------------------|-------|-----|
| Cold trap coordinate | 4,47  | m   |
| Helium flow rate at purification system | 1,336·10^{-4} | kg/s |
| Helium temperature in channel | 723 | K   |
| Helium temperature in cold trap | 303 | K   |
| Initial tritium mass in channel | 5,0·10^{-12} | kg   |
| Fractional efficiency of purification system | 0,9 | – |

Comparison between results of HYDRA-IBRAE/LM and THYTAN are shown in Figure 4. Red and black lines are related to THYTAN and HYDRA-IBRAE/LM, respectively.

Figure 4. Comparison between THYTAN and HYDRA-IBRAE/LM results.

The results are nearly identical. Both codes uses the similar model and this agreement is predictable. According to this comparison it is possible to say that purification model of HYDRA-IBRAE/LM is used correctly.

4.2. Modeling of tritium leakage
The second comparative test with THYTAN code is related to tritium leakage to the atmosphere. A problem formulates in the following way: in the primary loop with helium inside, there is a leak to the containment vessel. The containment vessel has a leak to the atmosphere. During the simulation the tritium mass in the primary loop and in the containment vessel were computed.

This task was considered by authors of [1]. Figure 5 shows a nodalization scheme of THYTAN for calculation of leakage.
Figure 5. Nodalization scheme (THYTAN) for verification of the leak model.

The same task was simulated by HYDRA-IBRAE/LM code. Parameters for simulation are listed in Table 5.

| Parameter                                      | Value       | Unit     |
|------------------------------------------------|-------------|----------|
| Initial tritium mass in primary loop           | $9.6 \times 10^{-10}$ | kg       |
| Tritium source in primary loop                 | $11.08 \times 10^{-17}$ | kg/s     |
| Primary loop coolant                           | Helium      |          |
| Primary loop volume                            | 232         | m$^3$    |
| Primary loop temperature                       | 809         | K        |
| Primary loop pressure                          | $2.33 \times 10^6$ | Pa       |
| Leak rate from primary to containment vessel   | $3.7 \times 10^{-5}$ | kg/s     |
| Containment vessel coolant                     | Nitrogen    |          |
| Containment vessel temperature                 | 809         | K        |
| Containment vessel pressure                    | $3.93 \times 10^7$ | Pa       |
| Containment vessel volume                      | $1.56 \times 10^4$ | m$^3$    |
| Leak rate from containment vessel to atmosphere| $2.3 \times 10^{-3}$ | kg/s     |

Figures 6–7 show a comparison between estimation results.
According to Figures 6-7, the computational results demonstrate not only qualitative but also good quantitative agreement. The standard deviation of computational results is no more than 1% and 6% for primary loop and containment vessel, respectively. Thus, HYDRA-IBRAE/LM code allows simulating tritium leakage to atmosphere.

5. Experiment simulation
Peach Bottom [2] was the first installation of a High-Temperature Gas-Cooled Reactor (HTGR) in the United States. Its power operation started in 1967 and finished in 1974, when it was shut down for decommissioning. During the operation period the some tritium properties in the steam generator was periodically monitored.
The Peach Bottom End-of-Life (PBEOL) program started in March 1975. The main object of this program was to give data for validation and verification of specific codes for predicting physics, thermal, fission product. After shutting down of the Peach Bottom reactor in October 1974, there was an interest in carrying out measurements of the tritium permeation rates in the steam generator tubes. Measurements were carried out in the superheater, the evaporator, and the economizer sections of the Peach Bottom steam generator. Measurements of the tritium permeation rates were made in the operating temperature ranges of these sections:

- 673 – 973 K in the superheater;
- 573 – 694 K in the evaporator;
- 494 – 623 K in the economizer.

The experimental section is shown in Figure 8 in the simplified form. There were two coaxial channels with wall between them, the initial tritium concentration was in the inner channel and there was no tritium in the outer channel.

![Figure 8. Experimental section.](image)

In the experiments the permeation rates were measured in the different sections. The superheater walls were made from Incoloy-800, evaporator and economizer tubes were made from low carbon steel.

Simulation of this experiment was carried out by HYDRA-IBRAE/LM code. In this paper we will present results for the superheater section. The permeation rate $J$ was computed by

$$J = \frac{M_2^{HT}}{S \cdot t},$$

Where

- $M_2^{HT}$ – tritium mass in the outer channel (kg);
- $S$ – permeation area (taken from experimental data [2], m$^2$);
- $t$ – time.
Initial parameters are shown in Table 6.

**Table 6. HYDRA-IBRAE/LM input parameters for simulation.**

| Parameter                        | Value      | Unit   |
|----------------------------------|------------|--------|
| Inner channel radius             | 6.35·10⁻³ | m      |
| Wall radius                       | 9.525·10⁻³ | m      |
| Outer channel radius             | 30·10⁻³   | m      |
| Permeation area                  | 90.16·10⁻⁴ | m²     |
| Pressure                         | 0.1013·10⁵ | Pa     |
| Initial area concentration in the inner channel | 0.46 | ppbv |

HYDRA-IBRAE/LM calculation results of the permeation rates in comparison with experimental data and analytical solution are shown in Table 7. Results are presented for three different temperatures: 673 K, 823 K and 973 K.

**Table 7. Comparison of calculation results with experimental data and analytical solution.**

| Temperature, K | Calculation result, μCi/m²/h | Analytical solution, μCi/m²/h [1] | Experimental data, μCi/m²/h [2] |
|----------------|------------------------------|-----------------------------------|----------------------------------|
| 673            | 0.1138                       | 0.1202                            | 0.119±0.0595                     |
| 823            | 0.644                        | 0.683                             | –                                |
| 973            | 2.158                        | 2.298                             | 1.29±0.645                       |

As shown in Table 7, there is good agreement between calculation results and analytical solution. For lower temperatures a HYDRA-IBRAE/LM result is in the experimental accuracy limits, but for higher temperatures the results are dramatically different from experimental data. It can be explained by existence of the oxide film in the tubes in case of higher temperatures, but there is no such information in the experimental materials.

**6. Conclusion**

In the paper verification results of modelling of tritium transfer are presented. For this modelling the special module TRITIUM was developed in HYDRA-IBRAE/LM code. This module contains models describing the main physical processes connecting with tritium behavior: tritium transfer in pipes, tritium permeation through tube walls, tritium leakage into atmosphere and tritium accumulation in cold trap. All equation coefficients are computed in dependence of current status of a system, which makes the module useful and flexible tool for tritium transfer simulation.

The results of experiment simulation, comparative calculations and analytical tests are shown in the article. A good agreement between the experimental data and HYDRA-IBRAE/LM results has been obtained for temperatures about 673 K (there is no sufficient experimental data for other temperatures). An average relative error was no more 0.01% for analytical tests. These results...
demonstrate that HYDRA-IBRAE/LM allows modeling physical processes, connecting with tritium transfer in reactor loops.

References
[1] Ohashi H and Sherman S R *Tritium movement and accumulation in the NGNP system interface and hydrogen plant*, INL/EXT-07-12647.
[2] Yang L, Baugh A and Baldwin N L *Study of Tritium permeation through peach bottom steam generator tubes*, 1977.