The problems of using a high-temperature sodium coolant in nuclear power plants for the production of hydrogen and other innovative applications

A.P. Sorokin, V.V. Alexeev, Ju.A. Kuzina, M.A. Konovalov
SSC RF – IPPE, Obninsk, Russia

sorokin@ippe.ru

Abstract. The intensity of the hydrogen sources arriving from the third contour of installation in second in comparison with the hydrogen sources on NPP BN-600 increases by two – three order at using of high-temperature nuclear power plants with the sodium coolant (HT-NPP) for drawing of hydrogen and other innovative applications (gasification and a liquefaction of coal, profound oil refining, transformation of biomass to liquid fuel, in the chemical industry, metallurgy, the food-processing industry etc.). For these conditions basic new technological solutions are offered. The main condition of their implementation is raise of hydrogen concentration in the sodium coolant on two – three order in comparison with the modern NPP, in a combination to hydrogen removal from sodium and its pumping out through membranes from vanadium or niobium. The researches with use diffusive model have shown possibility to expel a casium inflow in sodium through a leakproof shell of fuel rods if vary such parameters as a material of fuel rods shell, its thickness and maintenance time at design of fuel rods for high-temperature NPP. However maintenance of high-temperature NPP in the presence of casium in sodium is inevitable at loss of leakproof of a fuel rods shell. In these conditions for minimisation of casium diffusion in structural materials it is necessary to provide deep clearing of sodium from cesium.

1. Введение
The future of hydrogen power engineering will depend to a considerable degree on the efficiency of technological hydrogen production processes. As a rule, these processes use high-grade heat, the generation of which requires considerable energy resources. The same applies to efficient petroleum products refinery processes. Gas and petroleum products rather than renewable sources of energy are presently used for obtaining such heat.

However, this method does not hold much promise [1], because it involves additional depletion of resources, the natural reserves of which are far from being unlimited, and the possibility of using nuclear energy for these purposes is being discussed. A sodium-cooled fast-neutron nuclear reactor may become one of possible sources producing high-grade energy [2–4].

The using of traditional sodium technology, central to which is removal of hydrogen from sodium by means of cold traps only, does not hold promise for development of high-temperature nuclear power installations (NPPs): the intensity of hydrogen flows from the tertiary to the secondary coolant...
circuit in a high-temperature NPP increases by several tens of times as compared with that in the BN-600 reactor.

This is attributed to the following two main factors. First, this is the kinetics relating to the physicochemical processes in the liquid metal system: in accordance with the Arrhenius equation, the constants determining this kinetics are exponential functions of temperature (as the temperature increases by a few hundreds of degrees, the values of these constants grow by several orders of magnitude). And second, the pressure of hydrogen in the process systems of the third coolant circuit is essentially higher both in case of hydrogen production and in refinement of petroleum products than it is in the third coolant circuit of modern nuclear power plants (NPPs).

Of all admixtures that are contained in high-temperature liquid-metal systems, only hydrogen and cesium were analyzed in this study. It is shown that, if the intensity of hydrogen sources becomes several tens of times higher, new technologies for removing hydrogen from sodium must be developed, with a yield essentially higher than that implemented at modern NPPs. An increased concentration of hydrogen in sodium – 100 to 1000 times higher than in the sodium circulating in the secondary coolant circuit of modern NPPs – is the main condition for implementing such technology.

In this study, we estimated the possible ingress of cesium into sodium through the tight fuel rod cladding and showed that the sodium circulating in a high-pressure NPI must be purified from cesium to a deeper extent than it is done at the presently existing NPPs equipped with fast-neutron reactors, e.g. the BN-600 reactor.

2. Hydrogen in sodium of a high-temperature nuclear power plant

The requirements imposed on hydrogen removal systems (HRSs) from sodium are obvious: it is necessary to ensure the required throughput and capacity of an HRS with respect to sodium hydride and removal of hydrogen from the coolant to the preset concentrations. In so doing, both the HRS operational safety and its cost must be taken into consideration.

The change of hydrogen concentration in the sodium circulating in the secondary coolant circuit with the preset operating parameters of a high-temperature NPP and with the known design solutions can be calculated from the balance equation. For the secondary coolant circuit this equation has the following form Figure 1:

\[ J_{3/2} = J_2 + J_{2/AM} + J_{1/AM} \]  

Figure 1. Scheme of hydrogen mass transfer in the loops of a high-temperature NPP.
The hydrogen flow rates through surfaces of the sodium circuits were estimated using the following equation:

\[ J = K_P \left( P_2^{0.5} - P_1^{0.5} \right) S / \delta, \]  \tag{2}

where \( J \) is the hydrogen flow rate, kg/s; \( K_P \) is the permeability coefficient for hydrogen through a structural material (one of the major parameters of hydrogen mass transfer in circuits of a high-temperature NPP), kg·Pa\(^{0.5}\)·ppm·s\(^{-1}\); \( P_1 \) and \( P_2 \) is the partial pressures of hydrogen on either side of a surface, Pa; \( S \) is the surface area, m\(^2\); and \( \delta \) is its thickness, m.

Considering, that at concentration of hydrogen in sodium smaller concentration of saturation, concentration of hydrogen \( (C_{H_2}, \text{kg/kg}) \) is connected with a hydrogen equilibrium pressure in sodium \( (P_{H_2}, \text{Pa}) \) by expression [5]:

\[ C_{H_2} = K_S \sqrt{P_{H_2}}, \]  \tag{3}

where \( K_S \) is Sieverts constant equal to 4.34·10\(^{-7}\), Pa\(^{0.5}\).

For practical calculations of hydrogen flows in a concrete design of the technological equipment at the chosen maintenance parameters of high-temperature NPP the equations were used:

the hydrogen flow rate from the tertiary to the secondary coolant circuit

\[ J_{3/2} = K_{3/2} \left( P_3^{0.5} \cdot K_S - C_2 \right), \]  \tag{4}

the hydrogen flow rate from the secondary to the primary coolant circuit

\[ J_{2/1} = K_{2/1} (C_2 - C_1), \]  \tag{5}

the hydrogen flow rate from the secondary coolant circuit in the ambient medium

\[ J_{2/AM} = K_{2/AM} C_2, \]  \tag{6}

the hydrogen flow rate from the primary coolant circuit in the ambient medium

\[ J_{1/AM} = K_{1/AM} C_1, \]  \tag{7}

the hydrogen flow rate from the secondary coolant circuit to the its HRS

\[ J_2 = G_{CT2} \beta (C_2 - C'), \]  \tag{8}

the hydrogen flow rate from primary coolant circuit to the its HRS

\[ J_1 = G_{CT1} \beta (C_1 - C'), \]  \tag{9}

where \( K_{3/2}, K_{2/1}, K_{2/AM}, K_{1/AM} \) are coefficients of permeability which were determined taking into account the operating parameters of the high-temperature NPP and the adopted design solutions and \( K_{i,j} = K_i^{(i,j)} \cdot (S_1^{(i,j)}) / \sigma^{(i,j)} \); \( C_1 \) and \( C_2 \) are the hydrogen concentrations in the primary and secondary coolant circuits, kg/kg; \( G_{CT1}, G_{CT2} \) are flow rates through cold traps of primary and secondary coolant circuits; \( \beta \) is factor of hydrogen retention; \( C' \) is concentration of saturation for hydrogen at an outlet temperature from a cold trap, kg/kg.

The value of \( C' \) was calculated using the expression [6]:

\[ C'(T) = 10^{0.467 - 3023/T}. \]  \tag{10}

The value of \( K_{3/2}, K_{2/1}, K_{2/AM}, K_{1/AM} \) was calculated using the expression which defined \( K_P \) for 12Kh18N10T steel [7]:

\[ J = K_P \left( P_2^{0.5} - P_1^{0.5} \right) S / \delta, \]  \tag{2}

where \( J \) is the hydrogen flow rate, kg/s; \( K_P \) is the permeability coefficient for hydrogen through a structural material (one of the major parameters of hydrogen mass transfer in circuits of a high-temperature NPP), kg·Pa\(^{0.5}\)·ppm·s\(^{-1}\); \( P_1 \) and \( P_2 \) is the partial pressures of hydrogen on either side of a surface, Pa; \( S \) is the surface area, m\(^2\); and \( \delta \) is its thickness, m.
\[ K_p(T) = e^{-20.3-8833/T}. \]  

3. Determining the initial parameters for calculating hydrogen mass transfer

For estimating the surface areas of the primary coolant circuit we used the data of [2]. The surface area of the secondary coolant circuit was evaluated proceeding from the supposed layout of components, namely, taking into account that the process facilities are arranged at a distance of 500 m from the reactor plant and are connected with each loop of the secondary coolant circuit by forward and return pipelines made of Grade 12Kh18N10T steel with a standard size of 325×12 mm.

The following initial data of the high-temperature NPP were used in the calculations are presented in Table 1.

Table 1. Parameters of the high-temperature NPPs.

| Parameters | BN-600 | Phenix | High-temperature NPP |
|------------|--------|--------|----------------------|
| Reactor heat rating, MW | 1470 | 563 | 600 |
| Number of secondary circuit loops, pcs. | 3 | 3 | 6 |
| Number of intermediate heat exchangers (IHE), pcs. | 6 | 6 | 6 |
| Number of steam generators or technological heat exchangers (THE), pcs. | 8 section per one loop | 3 | 6 THE |
| Surface areas, m\(^2\): | | | |
| IHE (one module) | 1590 | 457 | 577 |
| SG or THE (per one loop) | 4968 | 717 | 1000 |
| Reactor vessel | 764 | 202 | 150 |
| Pipelines and equipment of secondary circuit (external surfaces) | 2055 | 671 | 1050 |
| Wall thickness, mm: | | | |
| IHE | 1.4 | 1 | 1.5 |
| SG or THE | 2.5 | 2/3, 6/4 | 5.0 |
| Reactor vessel | 30 | 25 | 30 |
| Secondary circuit pipelines | 13 | 6/7 | 12 |
| Temperature, °C: | | | |
| IHE | 535–365 | 560–395 | 900–800 |
| SG or THE | 510–315 | 550–343 | 875–800 |
| Reactor vessel (average value) | 450 | 480 | 850 |
| Pipelines and equipment of secondary circuit: | | | |
| – hot pipelines | 510 | 550 | 875 |
| – cold pipelines | 315 | 343 | 800 |

Results of calculation of the permeability coefficients are presented in table 2.

In view of uncertainty in the selection of structural material and adopted design solutions, the calculation results are tentative in nature.

Table 2. Hydrogen permeability coefficients through surfaces of the technological equipment in high-temperature NPP (for six loops).

| Coefficient, kg·s\(^{-1}\) | Value |
|--------------------------|--------|
| \( K_{3/2} \)            | 3.2    |
| \( K_{2/1} \)            | 1.6    |
4. Hydrogen pressure in the tertiary coolant circuit

It is shown in monograph [1] that high-temperature electrolysis, thermochemical decomposition, and hybrid cycles are the most efficient cycles for obtaining hydrogen. High levels of temperature are required for implementing these processes; as regards hydrogen pressure at different stages of the process, no detailed information is given on this matter in [1], and it is only pointed out that the pressure of all components may be around a few atmospheres. In [8], it is indicated that the hydrogen pressure in the tertiary coolant circuit at the process heat exchanger outlet may reach a few atmospheres, which is two or three orders of magnitude higher than in the tertiary coolant circuit of the BN-600 reactor, the level of which is, according to our assessments, around 500 Pa. In refining petroleum products using hydraulic cracking and reforming, the hydrogen pressure is equal to several tens of atmospheres [9]. Therefore, in estimating the effect the hydrogen pressure in the tertiary circuit has on the hydrogen flow rate in the secondary circuit, we used a parametric approach. The calculation results are given in Table 3.

Table 3. Hydrogen flow rates from the tertiary circuit to the secondary circuit and from the secondary circuit to the ambient medium vs. the hydrogen pressure in the tertiary circuit and its concentration in the secondary circuit (for six loops).

| Hydrogen pressure in the tertiary circuit $P_{H_2}^{III}$, MPa | Hydrogen flow rate, kg/s | $C_{H_2}^{III}=0.2$ | $C_{H_2}^{III}=100$ |
|-------------------------------------------------------------|--------------------------|---------------------|---------------------|
|                                                             | $J_{3/2} \cdot 10^3$ | $J_{2/AM} \cdot 10^3$ | $J_{3/2} \cdot 10^3$ | $J_{2/AM} \cdot 10^3$ |
| 0.1                                                        | 4.3                     | 1.5                 | 1.2                 | 0.7                 |
| 0.4                                                        | 8.7                     | 1.5                 | 5.5                 | 0.7                 |
| 0.9                                                        | 13.0                    | 1.5                 | 9.9                 | 0.7                 |

It can be shown that the hydrogen flow rate from the primary circuit into the ambient medium calculated from the expression

$$J_{2/1} = J_{1/AM_1} = K_{1/AM_1} + \frac{K_{2/1}}{K_{1/AM_1} + K_{2/1}}$$  \hspace{1cm} (12)

is a few orders of magnitude smaller than it is from the secondary circuit into the ambient medium; this is why it is not included in Table 3.

5. Calculating the parameters of the HRS for secondary circuit sodium

In using CTs as the main component of the hydrogen removal system from sodium, it was assumed that they operate in the traditional mode; i.e., that the temperature at the trap outlet is 120°C, and the working concentration of hydrogen in sodium was specified in a parametric way. With such mode of operation, the HRSs used in the secondary circuit should accumulate the entire hydrogen arriving from the tertiary circuit except for the leaks escaped from the primary and secondary circuits into the ambient medium, i.e.,

$$J_2 = J_{3/2} + J_{2/AM} + J_{1/AM}.$$ \hspace{1cm} (13)

It follows from (8) that the flow rate through all cold traps in the secondary circuit is given by
\[ G_{CT2} = \frac{J_2}{\beta(C_2 - C')} \]  

(14)

There are two approaches to determining the CT volume.

- **The first approach** is based on the time for which sodium must dwell in the CT at the specified flow rate through it. For the presently recommended CT design, the coolant dwelling time in the CT \( \tau_{dw} \) is taken equal to 900 s, hence

\[ V_{CT} = G_{CT2} \tau_{dw}, \]  

(15)

- According to the second approach, the CT volume is determined taking into account the specified CT life time \( \tau_{dw} \) with the high-temperature NPP operating at the nominal load and the cold trap capacity with respect to sodium hydride \( C^* \) (it is taken that \( C^* = 10 \text{ vol} \% \) and \( \tau_{res} = 1 \text{ year} \))

\[ V_{CT} = 100 J_2 \frac{\tau_{res} M_{NaH}}{C^* M_H \rho_{NaH}}, \]  

(16)

where \( M_{NaH} \) and \( M_H \) are the molecular masses of sodium hydride and hydrogen, kg/mol, and \( \rho_{NaH} \) is the sodium hydride density, kg/m\(^3\).

Using the data given in Table 3, we calculated the sodium flow rates through the hydrogen removal system as functions of hydrogen pressure in the tertiary circuit and working concentration of hydrogen in the secondary circuit sodium. The calculation results are given in Table 4.

**Table 4.** Sodium flow rate through the CTs in one secondary circuit loop, kg/s, and their volume, m\(^3\), vs. the hydrogen pressure in the tertiary circuit and working concentration of hydrogen in the secondary circuit sodium

| Hydrogen pressure in the tertiary circuit \( P_{HIII}, \text{mPa} \) | Sodium flow rate through the CTs in one secondary circuit loop and their volume | \( C'_{HII} = 0.2 \) | \( C'_{HII} = 100 \) |
|---|---|---|---|
| 0.1 | 360/340* | 0.07/26 |
| 0.4 | 730/670 | 0.8/280 |
| 0.9 | 1100/1000 | 1.5/540 |

* – The numbers in the numerator indicate the flow rate through the HRS CTs, and those in the denominator indicate the cold trap volume. The CT volume was calculated using (15) and (16), and the larger one of these two values was taken.

With the hydrogen concentration in the secondary coolant circuit corresponding to operation of modern NPPs at the nominal parameters (0.1–0.2 ppm) [10], and with the hydrogen pressure in the tertiary coolant circuit equal to 0.1 MPa, the sodium flow rate required for maintaining the hydrogen concentration at a level of 0.2 ppm must be no less than 1400 m\(^3\)/h, and the CT volume must be no less than 340 m\(^3\).

Technically, such a system can be constructed. For example, in case of using cold traps similar to those used in the BN-600 reactor, the system will consist of approximately a hundred of such traps. In our opinion, such a solution is unacceptable not only from economic, but even from aesthetic considerations!

Clearly, in order to reduce the required volume of CTs and flow rates through them, it is necessary to reduce hydrogen flow rates to a great extent (by several tens of times) by using low-permeable materials.
Assessments show that in order to obtain working concentrations at a level of around 0.2 ppm, the permeability must be reduced by a factor of 100. The data of [11] testify that for temperatures around 800°C such problem has not been solved as yet.

Hydrogen flow rates from the tertiary to secondary circuit can be limited or reduced by using special design solutions, but this will entail essential complication of the process equipment.

The necessary quantity of CTs can be reduced by using efficient regeneration methods that would allow a CT to be used several times [12–14].

In this case, approximately 2500 kg of hydrogen will be released from the cold trap per year, which will contain around $1.2 \times 10^{14}$ Bq of tritium. In our opinion, it is advisable not to discharge this hydrogen into the environment, but to utilize using more advanced technologies of the future hydrogen power engineering [1]. In so doing, it should be borne in mind that since the obtained hydrogen contains tritium, its long-term storage for no less than 80 years will be required, after which its use in the industry will become possible because the content of tritium in it will have become by that time lower than the maximum admissible concentration.

Since a considerable amount of tritium may diffuse together with hydrogen into the environment through the secondary circuit pipelines and equipment, it is also advisable to collect this hydrogen and tritium in dedicated systems. This can be implemented by enclosing the pipelines in special casings.

The results presented in table 3, testify to possibility of clearing of sodium from hydrogen pumping out through materials with high permeability. In the capacity of such materials membranes from a steel, nickel, niobium and vanadium have been observed. At estimations was considered, that level of temperatures at which these HRSs should be maintained, it is equal 800°C, and a thickness of the membrane – 0.3 mm.

The hydrogen flow rate, $m^3/s$ through a membrane was estimated using of the equation (2) which at $P_2 \gg P_1$ has following equation:

$$J = K_p P_2^{0.5} \frac{S}{\delta}.$$  \hspace{1cm} (17)

The data on the coefficients of hydrogen permeability through a steel, nickel, vanadium and niobium are presented on figure 2 and to table 5.

![Figure 2. Coefficient of hydrogen permeability through different membrane materials vs. temperature.](image)
Table 5. Hydrogen permeability coefficients for membranes made of different materials.

| Material        | Coefficients $A$ and $B$ in equation (18) | Temperature range, °C | Source |
|-----------------|------------------------------------------|-----------------------|--------|
| Grade           |                                          |                       |        |
| 12Kh18N10T steel| 1.90                                     | 400–900               | [18]   |
| Nickel          | 3.40                                     | 350–550               | [19]   |
| Niobium         | 2.75                                     | 100–300               | [20]   |
| Vanadium        | 0.326                                    | 100–300               | [20]   |

The coefficient of hydrogen permeability through different membrane materials is determined from the formula

$$\log K_p = -A - B/T.$$  \hspace{2cm} (18)

The membrane surface area $S$ was calculated from the formula

$$S = \frac{J_2 RT}{M_H} \cdot \frac{K_S}{C_2} \cdot \frac{\delta}{K_p},$$  \hspace{2cm} (19)

where $J_2$ is quantity of hydrogen which HRS should remove from secondary circuit, kg/s; $R$ is the gas constant equal to 8.31 J/(K mol), and $\delta$ is a thickness of the membrane wall, m.

The calculation results are given in Table 6, including the necessary surface area and the number of tubes for the hydrogen removal system designed as a device made of 1 m – long 0.3 mm – thick tubes with a diameter of 10 mm operating at a temperature of 800°C.

Table 6. Necessary membrane surface area and number of tubes in the system for removing hydrogen from sodium by vacuumizing for different materials and with different hydrogen concentrations in the secondary circuit and $J_2$ (at the hydrogen pressure in the tertiary coolant circuit equal to 0.1 MPa per a secondary circuit loop.

| Membrane material | $C_{H_2} = 0.2$ | $C_{H_2} = 100$ |
|-------------------|-----------------|-----------------|
|                   | $S$, m$^2$      | Number of tubes in the HRS | $S$, m$^2$ | Number of tubes in the HRS |
| Nickel            | 3·10$^5$        | 10$^7$          | 74         | 2400 |
| Steel             | 1.1·10$^5$      | 3.6·10$^6$      | 28         | 900  |
| Niobium           | 2.7·10$^3$      | 8.6·10$^4$      | 0.7        | 22   |
| Vanadium          | 7·10$^2$        | 2.2·10$^4$      | 0.2        | 6    |
| $J_2$, kg/s       | 7.2·10$^5$      | 9·10$^6$        |            |      |

The data presented in Table 6 testify that it is possible to construct systems for removing hydrogen from sodium by evacuating it through membranes made of materials having high permeability for hydrogen. Niobium and vanadium were considered as such materials.

For estimating whether it is possible to operate a high-temperature NPP at the nominal parameters with high concentrations of hydrogen in the sodium taking the BN-600 reactor as an example, we analyzed the effect of hydrogen on the reactor nuclear-physical characteristics, on the NPP safety, on the corrosion of structural materials, on the possibility of implementing the technological processes,
and on the thermal-hydraulic characteristics. It was shown that the concentration of hydrogen in sodium up to 150 ppm does not have an essential adverse effect on the nuclear-physical characteristics.

In analyzing the effect of hydrogen on the safety of a high-temperature NPP it was supposed that the equilibrium pressure of hydrogen above sodium should not exceed 0.1 MPa. If the hydrogen pressure is higher than this level, then in case of an emergency situation caused by loss of tightness of the gas pocket, the pressure of gas above the sodium will drop to atmospheric pressure, and the sodium will become "foamed" due to release of gaseous hydrogen.

The concentration of hydrogen in sodium at which the equilibrium pressure of hydrogen above sodium at the nominal parameters will not exceed 0.1 MPa calculated using the data of Banus [18] is equal to 108–114 ppm. Hence, for safety reasons, the hydrogen concentration must be lower than the above-mentioned values.

The effect of hydrogen on the corrosion of structural materials and on the change of their properties in a wide range of hydrogen concentrations has not been studied. It should be expected that with oxygen concentration in sodium equal to a few ppm or less than 1 ppm (such concentration of oxygen is required in using refractory materials) a growth of hydrogen concentration in sodium will not have a negative effect on the corrosion characteristics of coolant. Hence, operation of a high-temperature NPP with increased concentration of hydrogen in sodium is possible with respect to this indicator (the effect of hydrogen on the corrosion of structural materials). It should be pointed out that the use of structural materials at high pressures of hydrogen in the technological processes that are supposed to be implemented in high-temperature NPPs has presently been mastered in the industry. This also testifies that it is possible to operate high-temperature NPP systems with sodium coolant at increased concentrations of hydrogen in sodium.

From technological considerations, it is dangerous if a crystalline phase (suspensions of sodium hydride) appears in sodium in cooling down the installation (a transition to the operating conditions in which the sodium temperature is decreased to 250°C). To avoid this process, it is necessary that the hydrogen concentration in sodium did not exceed its solubility at the same temperature. The hydrogen solubility in sodium as a function of temperature is described by formula (10). It can be seen that the solubility of hydrogen in sodium does not exceed 5 and 15 ppm at temperatures equal to 250 and 300°C, respectively. Hence, for this condition to be satisfied, the admissible concentrations of hydrogen in sodium must be limited to the above-mentioned values. However, if a need arises to operate at higher concentrations, this limitation can be removed provided that a correct combination is maintained between the temperature drop rate and extent of hydrogen removal from sodium under cooling down condition.

6. Diffusion of cesium from under the fuel rod into the coolant and structural materials of a high-temperature nuclear power plant

Specialists of the Institute for Physics and Power Engineering named after A.I. Leypunsky (IPPE) showed [19] that stainless steel that had been in contact with sodium for a long period of time (120 000 h at a temperature of up to 430°C) contains cesium, which diffused into it to a depth of up to 100 μm. As was pointed out above, the physicochemical processes become more intense as the temperature increases. In this connection, and taking into account the fact that cesium had diffused into steel to a depth of 100 μm, the question arose about possible diffusion of cesium into sodium through the tight fuel rod cladding during operation of a high-temperature NPP at the nominal parameters and about possible "impregnation" of its materials with cesium in the course of their being for a long time in contact with sodium that contains cesium.

1 These assessments were carried out by the employees of the Institute for Nuclear Reactors and Thermal Physics A.P. Ivanov and V.Yu. Stogov.
Since the fuel rod cladding thickness to the curvature radius ratio has a small value, the fuel rod cladding can be regarded as a flat wall. In view of this, the following relations are valid:

\[
C(x, t) = C_1 + \left( C_2 - C_1 \right) \frac{x}{x_0} + \frac{2}{\pi} \sum_{n=1}^{\infty} \frac{C_2 \cos(n\pi) - C_1}{n} \sin \left( \frac{n\pi x}{x_0} \right) e^{-\left( \frac{n\pi x}{x_0} \right)^2 \frac{t}{\tau}} ,
\]

(20)

\[
J_{Cs}(t) = -D \frac{\partial C}{\partial x} \bigg|_{x=x_0} = \frac{D C_1}{x_0} \left( 1 + \sum_{n=1}^{\infty} \cos \left( n\pi \right) e^{-\left( \frac{n\pi x}{x_0} \right)^2 \frac{t}{\tau}} \right) ,
\]

(21)

where \( x \) varies from 0 to \( x_0 \), \( \tau \) is time, \( s \); \( C_1 \) is the concentration of cesium on the cladding surface on the fuel side, \( \text{kg/m}^3 \); \( C_2 \) is the concentration of cesium at the cladding – coolant boundary, \( \text{kg/m}^3 \); and \( D \) is the coefficient of cesium diffusion in the fuel rod cladding, \( \text{m}^2/\text{s} \), which is an exponential function of temperature (Figure 3).

The distribution of cesium concentration over the fuel rod cladding thickness \( C(x, t) \) was obtained, and the cesium flow rate into the coolant \( J_{Cs}(t) \) through the tight fuel rod cladding at a temperature of 1000°C was estimated [21].

It can be seen from Figures 4 and 5 that for the assumed fuel rod service time (equal to 2.5 years), cesium will diffuse into the steel cladding from the fuel – fuel rod cladding boundary to the fuel rod – coolant boundary (the fuel rod cladding thickness is 0.3 mm) and to a depth of 0.1 mm from the fuel – fuel rod cladding boundary in case of using a molybdenum cladding. The flow rate of cesium at the fuel rod – coolant boundary reaches the value of \( 2 \times 10^{-16} \text{ kg/(m}^2\text{s}) \) for the cladding made of steel, and for the cladding made of molybdenum its flow rate will be negligibly small. This means that release of cesium may take place through the 0.3 mm – thick tight fuel rod cladding made of steel during the fuel rod service life, whereas the ingress of cesium into sodium through the 0.3 mm – thick tight fuel rod cladding made of molybdenum can be neglected. It follows from (20) and (21) that the ratio of stationary flow rates of cesium through the steel and molybdenum claddings is equal to the ratio of diffusion coefficients through them: \( (1.88 \times 10^{-16}/9.2 \times 10^{-18}) = 20 \), and the time taken for the process to
achieve steady-state parameters is equal to the inverse quantity; i.e., this time for the molybdenum cladding is a factor of 20 longer than it is for the steel cladding.

**Figure 4.** Distribution of cesium concentration over the 0.3 mm thick fuel rod cladding in molybdenum (1, 2) and steel (3, 4) for different intervals of time (1, 3 – 1 year and 2, 4 – 2.5 years).

The ingress of cesium into sodium through the tight fuel rod cladding can be reduced by varying the parameters of the complex $D \tau / x_0^2$. For example, if we increase the steel cladding thickness by a factor of 2, the ingress of cesium into sodium through it can be neglected.

If a loss of fuel rod cladding tightness occurs, operation of the high-temperature NPP with certain content of cesium in the sodium will be unavoidable. In view of this, diffusion of cesium into the structural materials should be minimized in order to improve the radiation environment. To this end, the sodium must be subjected to deep purification from cesium to the minimal concentrations by means of the methods used in NPPs, such as BOR-60, BN-600, and EBR-II.
It should be pointed out that in calculating the solubility of hydrogen in sodium and permeability of hydrogen through structural materials as functions of temperature, we had to extrapolate the data presented in the literature from the temperature region 300-400°C to the interval of temperatures typical for high-temperature NPPs. But since such extrapolation has not hitherto been substantiated experimentally, numerically, or theoretically, it should be emphasized once again that the presented data are tentative in nature.

7. Conclusion

It has been shown from the results of the performed investigations that in case of using high-temperature NPPs (and other innovating technologies) for obtaining hydrogen, the sources of hydrogen entering from the tertiary to secondary circuit have intensity two to three orders of magnitude higher than the sources of hydrogen at an NPP equipped with a BN-600 reactor. Under such conditions, with using the traditional technology for operation of a sodium-cooled NPP, and with the need to maintain the working hydrogen concentration equal to 0.1–0.2 ppm, bulky metal- and energy-intensive systems for removing hydrogen from sodium will have to be developed and operated in the case of using both cold traps and other proposed innovative technical solutions, including those based on evacuating hydrogen from sodium through materials with high permeability with respect to hydrogen.

The proposed innovative sodium treatment technology in case of hydrogen concentrations at a level of several tens of ppm taken in combination with removing hydrogen from sodium by evacuating it through membranes made of vanadium or niobium makes it possible to develop high-temperature compact and high-efficient systems for removing hydrogen from sodium.

A preliminary study aimed at analyzing the effect of high hydrogen concentrations in sodium on the nuclear-physical characteristics of the reactor, on its safety, on the corrosion of structural materials, on the implementation of technological processes, and on maintenance of the required thermal-hydraulic parameters has shown that it is in principle possible to use such technology in high-temperature NPPs.

The possible ingress of cesium into sodium through the tight fuel rod cladding has been estimated using the diffusion model. It has been shown from this analysis that ingress of cesium into sodium through the tight cladding can be excluded by varying such parameters as cladding material, its thickness, and time of operation.

For securing better radiation environment, measures must be taken for purifying sodium from cesium to the minimal concentrations by applying the methods used, for example, in the BOR-60, BN-600, and EBR-II reactors.

References

[1] Shpil‘rain E.E., Malyshenko S.P., and Kuleshov G.G. *An Introduction to Hydrogen Power Engineering*. Ed. by V.A. Legasov. – Moscow: Energoatomizdat, 1984 [in Russian].

[2] Kalyakin S.G., Kozlov F.A., Sorokin A.P., Alexeev V.V., Konovalov M.A. Investigations in substantiation of high-temperature nuclear energy technology for a fast neutron reactor with a sodium coolant for hydrogen production. *J. Proceedings of universities. Nuclear energy*, 2016. No. 3. P. 104–115 [in Russian].

[3] Adamov E.O., Dzhalavyan A.V., Lopatkin A.V., Molokanov N.A., Muravev E.V., Orlov V.V., Kalyakin S. G., Rachkov V.I., Troyanov V.M., Avrorin E.N., Ivanov V.B., and Aleksakhin R.M. The conceptual statements in the strategy for development of the Russian nuclear power industry in the outlook up to 2100. *Atomic Energy*, 2012. V. 112. No. 6. P. 319–330 [in Russian].

[4] Rachkov V.I., Tjurin A.V., Usanov V.I., Voshchinin A.P. *Efficiency of nuclear energy technology: system criteria and development directions*. – Moscow: TCNIIAAtominform, 2008 [in Russian].

[5] Kozlov F.A., Volchkov L.G., Kuznetsov E.K. and Matyukhin V.V. *Liquid-Metal Coolants of
Nuclear Power Installations: Purification from Impurities and Their Monitoring. Ed. by F.A. Kozlov – Moscow: Energoatomizdat, 1983 [in Russian].

[6] Wittingham A.C. An equilibrium and kinetic study of the liquid sodium-hydrogen reaction and its relevance to sodium-water leak detection in LMFBR systems. *Journal of Nuclear Materials*, 1976. V. 60. P. 119–131.

[7] Geld P.V., Ryabov R.A. *Hydrogen in metals and alloys* – Moscow: Metallurgiya, 1974 [in Russian].

[8] Morozov A.V. and Sorokin A.P. *Methods for obtaining hydrogen and prospects of using a high-temperature sodium-cooled fast-neutron reactor for its production*. Proceedings of the International Seminar "High-Temperature Projects", Kalpakkam, India, November 14–15, 2011.

[9] Plate N.L. and Sivinskas E.V. *Chemical Principles of Monomer Technology: A Handbook* – Moscow: IAPC "Nauka/Interperiodika", 2002 [in Russian].

[10] Chechetkin Yu.V., Kizin V.D. and Polyakov V.I. *Radiation Safety of an NPP Equipped with a Sodium-Cooled Fast-Neutron Reactor* – Moscow: Energoatomizdat, 1983 [in Russian].

[11] Karpov D.A., Kislov I.F., Mazaev S.N., Moiseev V.Ya., Shabanov A.P., Dubinin G.V., Dubrovskii V.A. and Khazov I.A. *Studying the possibility of using protective coatings for reducing hydrogen penetration through the structural materials* – URL: [http://www.rvs.itssoft.ru:8000/article/sart.html?id=239&conf_id=4](http://www.rvs.itssoft.ru:8000/article/sart.html?id=239&conf_id=4).

[12] Kozlov F.A. and Volchkov L.G. *A method for regenerating the cold traps of impurities in alkali metals* – USSR Inventor's Certificate No. 429468, Byul. Otkr., Izobr., No. 19 (1974).

[13] Kozlov F.A. and Volchkov L.G. A new method for regenerating the traps for purifying alkali metal coolants. *Atomic Energy*, 1975. V. 39. No. 4. P. 310.

[14] Kozlov F.A., Volchkov L.G., Tonov B.I., Nalimov Yu.P. and Likharev V.A. *A method for regenerating the cold traps of impurities in alkali metal coolants* – USSR Inventor's Certificate No. 473221, Byul. Otkr., Izobr., No. 21 (1975).

[15] Eshbakh G., Gross U., Llayapcoeprep K. and Shullien S. *Hydrogen diffusion and penetration through steel / Sorption Processes in Vacuum*. Ed. by K.N. Myznikov – Moscow: Atomizdat, 1966 [in Russian].

[16] Courouau J.-L., Labatut D. *Hydrogen and tritium distribution in FBRs. Modeling and cross-over calculations in a reference case, for the PHENIX and BN-600 reactors*. Note Technique No. 97036 dated 25.06.97. Cadarache, 1997. P. 17–20.

[17] Belovodskii L.F., Gaevoi V.K. and Grishmanovskii V.I. *Tritium* – Moscow: Energoatomizdat, 1985 [in Russian].

[18] Fromm E. and Gebkhart E. *Gases and Carbon in Metals* – Moscow: Metallurgiya, 1980 [in Russian].

[19] Lastov A.I., Pavlinchuk E.A., Konovalov E.U., Sheinker I.G., Efimov I.A. and Mezentsev A.N. Distribution of radionuclides over the primary circuit pipeline wall thickness in a BR-10 reactor. *Atomic Energy*, 1986. V. 60. No. 4. P. 262–264.

[20] Kozlov F.A. and Konovalov M.A. *Assessment of cesium release through the steel claddings in a high-temperature sodium coolant*. Proceedings of the Scientific-Technical Conference "Thermal Physics – 2012" – Obninsk: SSC RF – IPPE, 2012. P. 430–437.

[21] Matzke H.J., Linker G. Study of the diffusion of cesium in stainless steel using ion beams. *J. Nucl. Mater.*, 1977. V. 64. No. 1–2. P. 130–138.

[22] Isaichev V.I., Larikov L.N. and Chernaya L.F. About the diffusion mechanism of group IV elements in Mo and W. *Ukr. Fiz. Zh.*, 1980. V. 25. No. 4. P. 591–598.