

Specifics of high-temperature sodium coolant purification technology in fast reactors for hydrogen production and other innovative applications

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Abstract

In creating a large-scale atomic-hydrogen power industry, the resolution of technological issues associated with high temperatures in reactor plants (900 °C) and large hydrogen concentrations intended as long-term resources takes on particular importance. The paper considers technological aspects of removing impurities from high-temperature sodium used as a coolant in the high-temperature fast reactor (BN-HT) 600 MW (th.) intended for the production of hydrogen as well as other innovative applications. The authors examine the behavior of impurities in the BN-HT circuits associated with the mass transfer intensification at high temperatures (Arrhenius law) in different operating modes. Special attention is given to sodium purification from hydrogen, tritium and corrosion products in the BN-HT. Sodium purification from hydrogen and tritium by their evacuation through vanadium or niobium membranes will make it possible to develop compact highly-efficient sodium purification systems. It has been shown that sodium purification from tritium to concentrations providing the maximum permissible concentration of the produced hydrogen (3.6 Bq/l according to NRB-99/2009) specifies more stringent requirements to the hydrogen removal system, i.e., the permeability index of the secondary tritium removal system should exceed 140 kg/s. Provided that a BN-HN-type reactor meets these conditions, the bulk of tritium (98%) will be accumulated in the compact sodium purification system of the secondary circuit, 0.6% ($\sim 4 \cdot 10^4$ Bq/s), will be released into the environment and 1.3% will enter the product (hydrogen). The intensity of corrosion products (CPs) coming into sodium is determined by the corrosion rate of structural materials: at a high temperature level, a significant amount of corrosion products flows into sodium. The performed calculations showed that, for the primary BN-HT circuit, the amount of corrosion products formed at the oxygen concentration in sodium of 1 mln^{-1} exceeds 900 kg/yr with fuel element claddings made of EP-912-VD steel and 464 kg/yr with molybdenum alloy claddings. For the secondary circuit, the amount of corrosion products totals 263 kg/yr for each loop. Taking into account the high-temperature experiments which demonstrated high efficiency of retaining corrosion product suspensions by the strainers located in the low-temperature area, it is proposed to cool sodium to the required temperature alongside the corrosion products retention on the mass transfer surfaces, including strainers. It is shown that, by using 30% of the power required to produce hydrogen with 50% efficiency, the BN-HT is capable of producing about $0.6 \cdot 10^6 \text{ m}^3$ of hydrogen per 24 hours which is sufficient for modern large-scale enterprises processing medium-grade crude oil or the implementation of other technologies.

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Introduction

Conceptual studies on the appearance of a high-temperature sodium-cooled fast power reactor (BN-HT) to create a large-scale atomic-hydrogen power industry have shown that developing of such a reactor is a real technical challenge [1]. In this context, a particularly important task is resolving technological issues associated with high tempera-

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tures in reactor plants and large hydrogen concentrations intended as long-term resources, including: developing a sodium coolant technology to be used at high temperatures and large hydrogen concentrations intended as long-term resources and ensuring corrosion stability of heat- and radiation-resistant high-temperature structural materials at the oxygen concentration in the sodium coolant of 1 mln^{-1} . Specific assessments were carried out for the BN-HT 600 MW (th.). The problems of removing impurities from high-temperature sodium were considered; special attention was given to the problem of sodium purification from hydrogen, tritium and corrosion products (CPs), the source intensity of which in the primary circuit of a high-temperature reactor plant intended for the hydrogen production increases by orders of magnitude.

Behavior of impurities in the BN-HT circuits in different operating modes

The heat removal by means of the coolant used in the reactor plant is accompanied by its interaction with impurities available in the liquid-metal system and their negative impact on the structural materials. The direction of these processes is determined by the difference of chemical potentials [2].

In a real nonisothermal system ($dT/dx \neq 0$), for its chemical potential, this relation is valid

$$\frac{\partial \mu}{\partial x} = \frac{dT}{dx} \left[\left(\frac{d\mu_0(T)}{dT} + R \ln \left(\frac{C}{C_{\text{sat}}} \right) \right) \cdot \frac{1}{dC/dx} + RT \frac{C_1}{C_{\text{sat}}} \left(C_{\text{sat}} - C \frac{dC_{\text{sat}}/dx}{dC/dx} \right) \right] \frac{dC}{dx} = B \frac{dC}{dx} \quad (1)$$

if, for the real system,

$$j = -\alpha \cdot d\mu/dx = -aB\mu \cdot dC/dx = -D \cdot dC/dx. \quad (2)$$

From (1) and (2), it is clear that the transfer of components in the nonisothermal fixed system, in addition to the coefficient α characterizing the diffusion properties of the medium, depends on a number of factors, including the impurity solubility, chemical potential temperature dependence (physical–chemical characteristics of the system) and temperature gradient which have a dominant role. When the fluid is moving, the hydrodynamic characteristics will also contribute to the mass transfer processes.

Using the temperature dependence of the constants characterizing the heat and mass transfer (Arrhenius formula),

$$k = k_0 \exp \{-E/(RT)\}, \quad (3)$$

(k is the constant characterizing the process; k_0 is the constant factor; E is the activation energy; R is the universal gas constant ($R = 8.31 \text{ J/(mole K)}$); T is the absolute temperature, K), it becomes clear that, when specific liquid metal system processes are considered, e.g., diffusion, permeability, solubility, absorption rate, gas pressure equilibrium, etc., their values will increase in the transition to high temperatures.

An increase in the constant for the above-mentioned processes is determined by their activation energy and temperature rise. But $T_2/T_1 \leq 2$, and the activation energy varies from

hundreds to tens of thousands of J/(mole K) and it is virtually temperature-independent for such a characteristic as the hydrogen pressure equilibrium over sodium. The highest values are characteristic of diffusion, permeability in solids and gas absorption rate kinetics. For the solubility of impurities, the activation energy is at least orders of magnitude lower than that of the diffusion processes. It should be noted that, for the same process, the activation energy of different materials may vary by several times and, in some cases, by an order of magnitude.

The qualitative analysis of the behavior of impurities in the liquid metal BN-HT systems in different operating modes shows that the coolant purification during the sodium receipt from transport containers, commissioning works and in standby modes can be done in cold traps (CTs). In modes of the sodium receipt from transport containers and commissioning works, conventional CT connection schemes can be used. In standby modes, if they are implemented after the stabilization of nominal conditions and operation under normal conditions, the inevitable appearance of radioactivity in the coolant should be considered.

In high-temperature systems with carbon concentrations of tens of mln^{-1} , the carbon thermodynamic activity due to the high sodium solubility as compared to NPPs of the BN-600 type increases by orders of magnitude. Therefore, in order to avoid carburization of structural materials, it may become necessary to remove carbon from hydrogen by the hot trap before switching to the nominal operating parameters.

Purification in nominal and standby modes required a special analysis since the intensity of the sources of hydrogen, tritium and corrosion products increases by orders of magnitude.

Sodium purification from hydrogen and tritium in high-temperature nuclear power plants

The specific behavior of hydrogen, tritium and cesium and their removal are considered in [3,4]; therefore, we shall focus only on the main results obtained for the BN-HT 600 MW.

With increasing hydrogen flows from the tertiary circuit into the secondary one by two–three orders of magnitude as compared to the hydrogen source in NPPs with the BN-600 reactor, it is possible to create compact purification systems with the required performance at hydrogen concentrations of tens of mln^{-1} , as the purification system (PS) performance in a first approximation is proportional to the hydrogen concentration in sodium. In this connection, sodium purification from hydrogen and tritium should be performed not by a cold trap (CT) but by their evacuation through the vanadium or niobium membranes. The combination of these two factors will create a highly compact system for removing hydrogen from sodium.

Sodium purification from tritium to concentrations providing its maximum permissible concentration (MPC) of 3.6 Bq/l in the produced hydrogen specifies more stringent requirements to the hydrogen removal system, i.e., its performance (permeability, and hence the size) should be increased.

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