Investigations in a substantiation of high-temperature nuclear energy technology with fast-neutron reactor cooled by sodium for manufacture of hydrogen and other innovative applications

A.P. Sorokin, A.A. Trufanov, A.P. Ivanov, F.A. Kozlov, A.A. Kamaev,
V.V. Alekseev, A.V. Morozov A.V.

State Scientific Center of the Russian Federation – Institute for Physics and Power Engineering named after A.I. Leypunsky», Obninsk, Russia

E-mail contact of main author: sorokin@ippe.ru

Abstract. It is shown, that there is a principal possibility to provide demanded parameters of a high-temperature fast reactor with the sodium coolant for production of a considerable quantity of hydrogen, for example, on the basis of one of thermochemical cycles or on the basis of high-temperature electrolysis with high factor of thermal use of the electric power. Demands to system of clearing from hydrogen on the basis of a principal new method – pumping out through special membranes are formulated: the coefficient of permeability of system of clearing of the secondary circuit from tritium should exceed 140 kg/s. Taking into account high temperature experiments in which high efficiency of deduction of suspended matters of products of corrosion on the filters installed in low-temperature zone is shown, it is offered to use a principle of work of a cold trap: to chill sodium to necessary temperature with simultaneous deduction of products of corrosion on mass transfer surfaces, including filters.

Key Words: high-temperature fast reactor, production of hydrogen, sodium coolant.

1. Introduction

Nuclear power generation is not an alternative or a competitor within the general strategy of development of the fuel and power generation complex of the country, but, instead, it offers additional potential of preservation of efficiency of available fuel resources during extended periods of time and possibility to enhance reliability and safety of power supply becoming “the source of the source” of power and other resources. Several alternative strategies of development of nuclear power generation are currently under discussion [1, 2]. One of the main requirements to future nuclear power generation technology – its large scale – implies enhanced level of safety of all its elements including reactor facilities and application of technologies of closed nuclear fuel cycle [3, 4]. Development of innovative fast reactors with stressful temperature and dose rate loads with application of sodium as a coolant constitutes important direction of formation of the new technological platform [5, 6].

The most significant problem determining future development of environmentally clean power generation is the inclusion of hydrogen in the fuel cycle. Hydrogen is a very attractive element as the replacement of oil and gas although by itself it is not a source but, instead, a carrier of energy. It is anticipated that the need in hydrogen production will be sharply increased in the nearest future. Currently the main method of hydrogen production is the methane reforming with steam. However, from the viewpoint of long-term perspective of large-scale hydrogen production, the above method is not viable since it requires consumption of non-renewable hydrocarbon resources and is accompanied with emission of greenhouse gases in the environment. That is why alternative methods of production of hydrogen with application of water splitting methods using thermal chemistry or electrolysis processes requiring high-temperature source of heat for enhancement of efficiency of the above processes are investigated [7, 8].

Due to the application of such coolants as gases and liquid metals (sodium, lead) Generation IV nuclear reactors can serve as such sources of high-temperature heat [9–12]. Coolant temperature at the outlet of the core of such reactors can reach 900 – 950°C. This is a new class of nuclear reactors designed for both ensuring electric power generation with enhanced thermal efficiency (50%), and, as well, for the supporting the technological processes during hydrogen production, coal gasification and liquefaction, advanced oil cracking, conversion of biomass into liquid fuel, in chemical industry, metallurgy, etc.

Conceptual studies on the selection of the general outlook of high-temperature sodium-cooled fast power reactor (BN-VT) for large-scale nuclear and hydrogen power generation [13] demonstrated that designing such nuclear reactor is a realistic task. Addressing the issues of purely technological nature associated with high levels of temperature in the reactor facility (RF) becomes the first priority including the development of sodium coolant technology at elevated temperatures and high hydrogen concentrations during extended lifecycle, application of heat resistant and radiation resistant structural materials, ensuring corrosion resistance of such materials at oxygen concentrations present in sodium coolant at the level of 0.1 ppm becomes the first priority [14]. Discussion of complex (not only neutronics, but thermal hydraulics and technological) studies for substantiation of conceptual design and safety of 600-MW-th BN-VT high-temperature sodium-cooled reactor for production of hydrogen is the objective of the present paper.

2. Composition and technical characteristics of the BN-VT reactor facility

BN-VT reactor facility. Composition of BN-VT RF (*FIG. 1*) includes sodium-cooled fast reactor, three cooling loops of the emergency heat removal system, three sets of equipment of secondary cooling loops for transfer of high-potential heat from the reactor to hydrogen producing chemical installations or to gas-turbine plant intended for supplying electricity to chemical equipment. Each of the loops contains intermediate heat exchanger incorporated in the reactor vessel, centrifugal pump and pipeline for removal and re-introduction of sodium in the reactor.

Existing requirements on the safety and financial performance of reactors of new generations were taken into consideration in outlining the general configuration of the reactor under study. Innovative ideology of fast reactor on the basis of achievements and success of sodium-cooled fast reactor technology is further developed in it. BN-600 reactor successfully operated already during more than 30 years was chosen as the basis for the design of the reactor under investigation. Main technical characteristics of BN-VT RF are presented in Table 1.

Pool type configuration of main equipment of the primary (radioactive) cooling loop inside the reactor vessel consisting of the pressure vessel and secondary containment, which facilitates ensuring high level of safety and allows removing the boxes for arrangement of auxiliary systems of the first cooling loop. Pressure vessel of the reactor intended for installation of in-reactor equipment, sodium and argon of the primary cooling loop and arrangement of sodium circulation represents vertical cylindrical tank equipped with cone-shaped reactor vessel lid and elliptical bottom with support plate. From the inner side the reactor vessel is in contact with sodium with exception of its upper part (lid) working in contact with argon gas blanket of the reactor, and from the outer side with argon contained within the safety gap between the internal pressure vessel and outer safety containment.

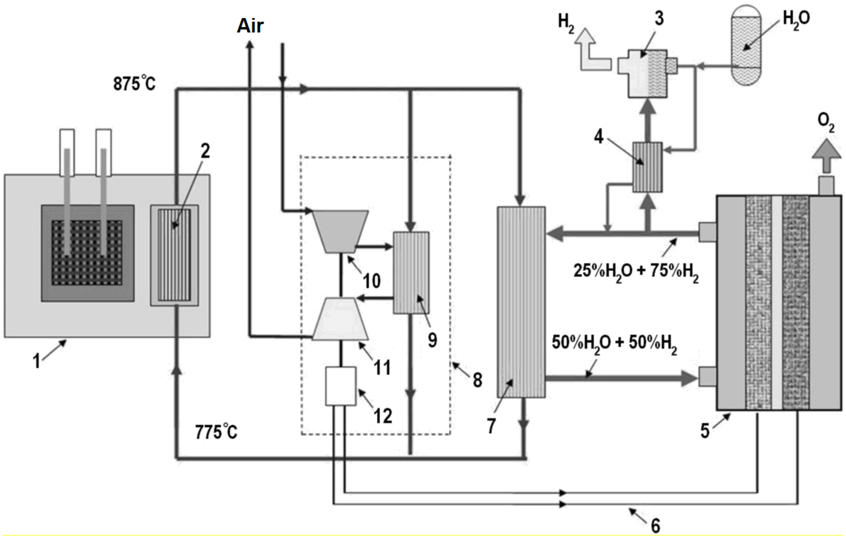

FIG. 1. Schematic layout of reactor facility for production of electricity and hydrogen on the basis of technology of solid oxide water electrolysis: 1 – fast reactor; 2 – intermediate heat exchanger; 3 – hydrogen separator; 4 – heat exchanger; 5 – solid oxide electrolysis cell; 6 – electrical power supply to the electrolysis cell; 7 – steam generator; 8 – gas turbine plant; 9 – heat exchanger; 10 – compressor; 11 – turbine; 12 – electrical generator

TABLE 1. MAIN TECHNICAL CHARACTERISTICS OF BN-VT REACTOR

Name of technical parameter	**Parameter value**
Rated thermal power	600 MW
Number of heat removal loops	3
Coolant temperature – core inlet – core outlet – intermediate heat exchanger inlet – intermediate heat exchanger inlet	 800°C 900°C 775°C 875°C
Sodium flow rate through one intermediate heat exchanger Absolute coolant pressure at the core inlet	1379 kg/s ≤ 1.0 MPa
Excess pressure within reactor gas volume	0.054 MPa

Reactor core, intermediate heat exchangers, main circulation pump of the first cooling loop, emergency cooling heat exchangers, electrochemical hydrogen sensor, electrochemical oxygen and carbon sensor, pipeline for coolant supply core, gas compensation and overflow pipes and fuel cladding leak detection system are arranged inside the vessel of such nuclear reactor adapted for generation of heat. Because of their large dimensions cold traps (CT) are removed outside the reactor vessel.

Nuclear reactor characteristics. Based on the degree of preparedness it is intended to preserve as the initial step the existing reactor design and use uranium oxide fuel with changing only the temperature level. The main objective during this phase is to reveal the bottlenecks from the viewpoint of already well developed design leaving the issue of selection of structural materials open. Existing high culture of BN reactor design and technical solutions tested during many years of reactor operation should facilitate practical implementation of the reactor facility [15]. During subsequent phases it is possible depending on the obtained results to address the possibility of use of other fuel compositions: MOX-fuel, mixed uranium-plutonium nitride fuel inside container type fuel rod, thorium fuel cycle and other potentially promising solutions requiring practical substantiation.

Design of fuel assemblies, reactor core configuration and fuel loading map are similar for the BN-VT reactor to those for BN-600 reactor [16]. BN-VT reactor core consists of a set of fuel assemblies (FAs), CPS rods, neutron sources, boron control and steel control systems arranged in the reactor core according to triangular lattice with average spacing equal to 98.35 mm. Reactor core consists of 369 uranium loaded FAs with three types of enrichment, 27 CPS rods and two neutron sources. Along the radius the core is divided into three zones with different fuel enrichment. FAs contain sections of the top and bottom axial blankets consisting of pellets of depleted or natural uranium dioxide arranged inside the cladding common with fuel pellets. Assemblies of the radial blanket are arranged surrounding the reactor core.

It can be expected based on the correlation between power capacities of BN-600 reactor and BN-VT reactor under design that with reduction of thermal power from 1470 to 600 MW (by ~ 2.5 imes) time interval between reactor core reloading can be increased from 140 days to one year – 330 days. Available efficiency of the reactor core shim system is expected to be sufficient with adequate safety margin for burn-up compensation, while increased temperature reactivity effect (isothermal reactor core heating from the reloading temperature to the coolant inlet temperature for the rated reactor power) can be compensated. Remaining reactivity effects should not significantly change. Characteristics of the reactor power unit are presented in Table 2.

TABLE 2. MAIN CHARACTERISTICS OF BN-VT REACTOR POWER UNIT

Characteristic	**Value**
Power (thermal)	600 MW
Nuclear fuel	UO_2
Core dimensions (D×H) according to reactor vessel	3900×1300 mm
Reflector thickness	200 mm
Flat-to-flat dimension and wall thickness of hexagonal FA jacket	96×2 mm
Number of fuel rods in the FA	127
Material of FA jackets, fuel cladding and spacer wiring	EP-912-VD
Fuel cladding diameter and wall thickness (d×δ)	6.9×0.4 mm
Cross-section dimension of spacer wiring – for 91 central fuel rods – for 36 peripheral fuel rods	 ∅1.05 mm 6×1.3 mm
Fuel pellet sizes (bush) – outer diameter – internal diameter	 ∅5.9 mm ∅1.7 mm
Reactor core height	1030 mm

Heights of axial blankets – top – bottom	300 mm 350 mm0.6×1.3
Gas void height	617 mm
Total FA length	3500 mm
Time between reloading operations	330 days
Reload temperature	230°C
Maximum fuel cladding temperature	1025°C
Total temperature reactivity effect (230°C → T_{in}) (230°C → 368°C) /(230°C → 800°C), %Δ*K*/*K*	–1.431
Total reactor power reactivity effect ($T_{in} \rightarrow N_{rat.}$), %Δ*K*/*K*	–0.452

Some safety issues. Specific feature of reactor operation within the hydrogen production complex is the need to take into consideration the probability of hydrogen penetration inside the reactor core along the coolant circuit. The implemented calculation studies demonstrated (Table 3) that hydrogen penetration within the limits of permissible allowances produces practically no effects on the neutronics characteristics of the reactor and on safety parameters of the reactor.

TABLE 3. VARIATION OF REACTOR REACTIVITY VERSUS CONCENTRATION OF HYDROGEN IN THE COOLANT

Hydrogen concentration in the coolant	**Reactivity gain introduced by hydrogen present in sodium composition**
0 pcm	0.000 %Δ*C*/*C*
50 pcm	0.0081 %Δ*C*/*C*
100 pcm	0.014 %Δ*C*/*C*
150 pcm	0.022 %Δ*C*/*C*
200 pcm	0.027 %Δ*C*/*C*
250 pcm	0.032 %Δ*C*/*C*

As a result of materials research the possibility of using at elevated concentrations of hydrogen in sodium and oxygen concentrations less than 2 million, one of a number of construction materials (molybdenum, niobium, steel EI-847, ES-912-HP, EI-732) with sodium temperatures up to 750°C.

Higher temperature level increases the probability of sodium boiling. Removal of sodium causes insignificant negative void reactivity effect explained by the use of uranium fuel. Thus, the requirement of significant increase of pressure in the primary cooling loop becomes not necessary. In order to organize closed fuel cycle there exist the possibility of examination of the use of uranium-thorium fuel cycle with close characteristics of reactivity effects. Key problem for high-temperature reactors is the fuel pin stability. The situation is somewhat mitigated for the proposed RF design due to the selection of low thermal load on the fuel pins. Maximum fuel burn up can be additionally reduced.

3. Structural materials

Selection of high-temperature material for particular reactor conditions is the most complex task from the viewpoint of reactor design. Alloys with high heat resistance and corrosion resistant in contact with sodium coolant at temperatures from 900 to 1200°C and radiation resistant up to 100 dpa are required as the material for fuel cladding. Results of studies of corrosion of structural materials are presented in Refs. [16 – 19]. Molybdenum and niobium alloys possessing attractive production properties and high temperature resistance along with corrosion resistance in contact with sodium coolant can be examined as such alloys.

Alloys of the basis of molybdenum can serve as the most suitable structural materials, but, however, their presence results in noticeable increase of neutron absorption which requires adjustment of fuel enrichment. According to preliminary estimations, increase of fuel enrichment in correspondence with maximum molybdenum concentration will not result, taking into account the available significant reactivity margin, in the violation of reactor safety requirements in the process of normal operation and during emergency situations. The problem of use of molybdenum-based structural material can be resolved by adjustment of fuel isotopic composition.

EP-912-VD steel was examined as possible optional structural material. This alloy with standard denomination Х15Н35В10Б (developed by the FSUE VIAM and the IPPE) is one of the promising structural materials for applications to work in contact with sodium coolant in air atmosphere at temperatures of 900 – 950°C. High short-term and long-term strength of the alloy is combined with high plasticity and viscosity characteristics at temperatures up to 950°C and at hot deformation temperature, stability of structure and of mechanical properties, good corrosion resistance in sodium coolant, as well as with high oxidation resistance at high temperatures. Argon electric-arc welding of sheets with thicknesses up to 12 mm is recommended to be performed using welding wire of ХН60ВТ, 06Х15Н60М15 and Х15И35В12 brands ensuring high resistance of metal welds against formation of hot cracks. Absence of molybdenum in the composition of steel is the important characteristic (Table 4).

TABLE 4. CHEMICAL COMPOSITION OF HIGH-NICKEL STAINLESS STEEL EP-912-VD [17]

C	**Si**	**Mn**	**S**	**P**	**W**	**Ni**	**Nb**	**Fe**
0.03	0.32	0.06	0.005	0.005	9.13	35.97	0.93	Rem.

Heat resistant 07Х15Н30В5М2 (CHS81) chromium-nickel austenitic steel developed by the «Prometey» Central Research Institute of Structural Materials (Table 5) is the alternative structural material. This steel is recommended for operation at temperature equal to 900 – 950°C. Studies of strength characteristics, corrosion resistance in sodium coolant and thermal stability performed at the «Prometey» Central Research Institute of Structural Materials demonstrated that the steel in question possesses the complex of physical, mechanical and technological properties required for its use in high-temperature nuclear reactors.

Comparison of reactivity gain introduced in the reactor by structural materials made of the above steels is shown in Table 6. Structural materials of BN-600 reactor core (cold-worked CHS-68 steel) introduce in the reactor core reactivity equal to – $2{,}218 \cdot 10^{-2}$ ($\Delta C/C$). This difference can be compensated for by using the reactor CPS. Therefore, preference can be given to CHS-81 steel although the final choice can be made after implementation of comprehensive studies of different structural materials as applicable to high-temperature nuclear reactor.

TABLE 5. COMPOSITION OF CHS81 STAINLESS STEEL [18]

C	**Si**	**Mn**	**S**	**P**	**W**	**Cr**
≤ 0.07	≤ 0.2	0.8 – 1.2	≤ 0.01	≤ 0.015	4.5 – 5.5	14.0 – 17.0

Ni	**Mo**	**Ti**	**Al**	**Other**	**Standart**
29.0 – 31.0	0.8 – 2.2	≤ 0.06	≤ 0.12	Cu ≤ 0.08 N ≤ 0.03 Fe ≤ rem. Y ≤ 0.05	TU14-1-3970-85 TU14-1-4244-87

TABLE 6. CONTRIBUTION OF CHEMICAL ELEMENTS IN THE COMPOSITION OF REACTOR CORE STRUCTURAL MATERIALS IN THE EFFECTIVE NEUTRON MULTIPLICATION FACTOR, C_{ef}

Chemical element	**EP-912-VD**		**CHS-81**	
	$\Delta K/K$	Nuclide composition	$\Delta K/K$	Nuclide composition
Fe	$-1.08 \cdot 10^{-2}$	25.9%	$-8.78 \cdot 10^{-3}$	22.6%
Cr			$-3.35 \cdot 10^{-3}$	8.6%
Ni	$-1.67 \cdot 10^{-2}$	39.9%	$-1.40 \cdot 10^{-2}$	36.0%
Mo			$-3.12 \cdot 10^{-3}$	8.0%
W	$-1.43 \cdot 10^{-2}$	34.3%	$-8.42 \cdot 10^{-3}$	21.7%
Mn			$-1.16 \cdot 10^{-3}$	3.0%

4. Sodium technology

Behavior of impurities in the BN-VT circuits at different operation regimes. Heat removal using a coolant in the reactor system is followed by its interaction with impurities and their negative effects on the materials of construction of the liquid metal present in the system. The direction of these processes is determined by the difference in chemical potentials [19].

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

$$k = k_0 \exp\left(-\frac{E}{RT}\right),$$

(the k – constant characterizing the process; k_0 of – constant factor, the E – activation energy, the R – universal gas constant (R = 8.31 J/(mol·K)); T – the absolute temperature, K), it is clear that the examining specific system liquid metal processes, such as diffusion, permeability, solubility, rate of absorption, the equilibrium pressure of the gases, the transition to the high temperatures of their importance will increase.

The increase of the specific constants for the above processes is determined by the activation energy and increasing temperature. But the $T_2/T_1 \leq 2$, and the activation energy varies from hundreds to tens of thousands of J/mol·K, and for such characteristics as the equilibrium pressure of hydrogen over sodium it is virtually independent from temperature. The highest values E are characteristic of diffusion processes, permeability in solids and for the kinetics of gas absorption rate. For the solubility of impurities in the activation energy of the order of and lower than the activation energy for diffusion processes. It should be noted that the same process for the activation energy of different materials may vary several times in some cases by an order.

Qualitative analysis of the behavior of impurities in the liquid metal BN-VT systems in various regimes of operation shows that the cleaning of the coolant sodium impurities in reception regimes of transport containers, the start - up and the parking regimes can be made of cold trap (CT). The sodium reception regimes of transport capacities and the start - up can be used to connect conventional circuits CT. In the parking regime, if they are implemented after the regime for nominal parameters and operation at nominal parameters, should be considered the inevitable appearance of radioactivity in the coolant.

In systems with high concentrations of carbon tens ppm, its thermodynamic activity due to the high solubility compared to sodium NPP BN-600 increases by orders of magnitude. Therefore, in order to avoid carbonization of construction materials, you may need to clean carbon from the hot trap before entering the nominal parameters.

Cleaning operation regimes at the nominal parameters and parking require special analysis, since the intensity of the source of hydrogen, tritium, corrosion products increases by orders of magnitude.

Cleaning the sodium-hydrogen and tritium in the high-temperature nuclear power plant. The peculiarity of hydrogen behavior of tritium and cesium and cleaning them is considered in [20, 21]. Therefore, we focus only on the main results obtained for the BN-VT 600 MW.

With increasing hydrogen flow from the third circuit in the second to two-three orders of magnitude as compared to the hydrogen source in nuclear power plants with a BN-600 creation of compact purification systems (PS) with the required performance is possible with hydrogen concentrations of tens of millions in minus first degree, as performance PS in a first approximation proportional to the hydrogen concentration of the sodium. Thus sodium purification of hydrogen and tritium should not produce a cold trap (CT) and their evacuation through the membranes of vanadium or niobium. The combination of these two factors will create a highly compact sodium hydrogen purification system.

Cleaning up sodium from tritium concentrations in the produced hydrogen provide its maximum permissible concentration (MPC) of 3.6 Bq/l, imposes more stringent requirements for the removal of the hydrogen system: its performance (permeability, and hence the size) should be increased. Under these conditions for NPP BN-VT bulk of tritium, 98% will be battery-Rovan in compact PS of the second circuit of sodium, 0.6% (~ $4{\cdot}10^4$ Bq/s) will go to the environment, and 1.3% – in the manufactured product. If released in compact PS ~ $4{\cdot}10^4$ Bq/s requirements ensure proper environmental conditions, set forth in [22], it can easily be satisfied using the methods that are widely used today in the nuclear industry.

As a large mass of tritium accumulated in compact PS, when the large-scale use of nuclear power question about the future of hydrogen requires special consideration.

The behavior of corrosion products in installations with sodium coolant. Corrosion products in the operation of facilities are constantly coming in sodium. Numerous studies of corrosion of structural materials published in monographs [23, 24]. Research in this area continues to see the last 20 years. For example, [25 – 26].

Results of evaluation of the PC source intensity in the BN-VT circuits are shown in Table 7. It should be noted that the non-isothermal system in the corrosion rate in high temperature zone should not depend on the temperature difference between hot and cold zones contour, with its decrease in the high temperature zone, it should decrease. Evaluations conducted by us, it is assumed that the number of corrosion products the sodium entering the reduced 6-fold.

TABLE 7. INTENSITY CORROSION PRODUCTS REVENUE IN THE SODIUM FIRST AND SECOND BN-VT CIRCUITS WHEN OPERATING AT THE NOMINAL PARAMETERS, kg/year

Primary circuit			**Secondary circuit*⁾**	
Homogeneous system	Heterogeneous system		Homogeneous system	
All equipment – EP-912-HP	Fuel element – Molybdenum (Alloys)	Intermediate heat exchanger – EP-912-HP	Intermediate heat exchanger – EP-912-HP	Pipeline – EP-912-HP
900	Negligible	464	662	914
*⁾ on six loops of the second circuit in sodium of each loop of the second circuit fed 263 kg/year				

Sodium purification system from corrosion products (SPSCP). It is known that the low effectiveness of CT in sodium clean modern nuclear power plants from the corrosion products. However, special experiments have shown that the mesh filter installed behind the heat exchanger (at this°exchanger in which the sodium temperature is decreased with 750 (temperature the sodium washed of impurity source) to 420, with corrosion products retention rates for estimates similar to unity, and the proportion of impurities deposited on the surface of the heat transfer tube ~ 3% of the number found by the corrosion products in the filter. Given these results, the development SPSCP principle of operation has been selected CL: sodium cooled to the desired temperature, followed by retention of suspensions of corrosion products on the strainers.

5. Conclusion

The results of neutron-physical and thermal research reactor facility BN-VT 600 MW (th) have shown that there is a fundamental possibility of building on the existing structure of the reactor BN-600 to provide the required parameters of high-temperature fast reactor for the production of large amounts of hydrogen with a high coefficient of thermal use and high efficiency of electricity production, while meeting the safety requirements.

Use the schematic production of electricity and hydrogen on the basis of technology solid oxide electrolysis of water, taking into account the use of a fundamentally different of the method of purification – removal of hydrogen and tritium from sodium evacuating them through a special membrane. It is shown that the efficiency of such a system is ~ 40%, and the volume-produced in the hydrogen is $2.8{\cdot}10^4$ l/s (under normal conditions).

According to the proposed method of calculation the results by the mass transfer of hydrogen and tritium in sodium circuit reactor facility BN-VT 600 MW (th) were obtained. The real danger of tritium in the finished product arises after combustion of hydrogen in the atmosphere.

Therefore, when calculating cleaning system parameters and operation of hydrogen and tritium concentrations in sodium secondary circuit was assumed that the maximum allowable tritium concentration in the produced hydrogen should not exceed 3.26 Bq/l. Cleaning the sodium of tritium concentration to providing hydrogen in the produced maximum permissible concentration equal to 3.26 Bq/l, charged an additional requirements for purification from hydrogen system: permeability coefficient of the second circuit cleaning system for tritium should exceed 140 kg/s.

Taking into account the results of available high temperature tests (maximum temperature 900°C), which shows a high retention efficiency suspensions PC to filters (retention factor close to unity), set in the low temperature zone, it is proposed the development PS of corrosion products use the principle of cold trap: sodium cooled to the desired temperature while retaining the mass transfer on the surfaces of the PC, including filters.

BN-VT with a thermal capacity of 600 MW using 30% power for the production of hydrogen with an efficiency of 50% would produce about $0.6 \cdot 10^6$ m^3 of hydrogen per day, enough for a modern large-scale enterprises, processing crude oil of medium quality and other technologies.

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