INTERNATIONAL JOURNAL OF HYDROGEN ENERGY 41 (2016) 3320–3325

Available online at www.sciencedirect.com
ScienceDirect
journal homepage: www.elsevier.com/locate/he

Review Article

On the problem of efficient production of hydrogen reducing gases for metallurgy utilizing nuclear energy☆

V. Klimova, V. Pakhaluev, S. Shcheklein*

Ural Federal University, Department of Nuclear Power Plants and Renewables, 19 Mira str., Ekaterinburg 620002, Russia

ARTICLE INFO

Article history:
Received 20 September 2015
Accepted 20 September 2015
Available online 19 January 2016

Keywords:
Hydrogen economy
High temperature gas-cooled nuclear reactor
Methane conversion
Synthetic gas
Reducing environment

ABSTRACT

One of the promising ways to reduce the energy consumption of metal production is the use of direct reduction technologies. The use of high capacity heat from high temperature gas-cooled nuclear reactors (HTGR) for the production of reducing gases from methane feedstock is a rational solution to the problems of fossil resources saving, reducing of power intensity of synthesis process and drastic lowering of foul gases release.

The application of radial flow scheme in the nuclear reactor core and in the methane converter allows to create the installations with minimal dimensions and lower hydraulic losses in comparison with currently existing units. It was conducted the investigation of gas flow and heat exchange with the elements of pebble bed in the case of radial flow, which allowed to receive the design equations for such an apparatus. The results of calculation of main thermohydraulic features of a prospective methane converter with radial flow of gaseous reaction mixture using these equations are given.

Introduction

Nowadays we can observe active development of atomic energy as an electricity source. But the share of electricity in total energy resources consumption is about 20%. The rest 80% of oil, natural gas, coal and other energy resources is expended for housing and transport needs and energy supply of various industrial technological processes. Burning of organic fuel in energotechnological processes inevitably leads to the generation of anthropogenic greenhouse gases. Developing of large-scale nuclear hydrogen energy can supplant expensive hydrocarbonic fuel from the energy sector and save oil and natural gas for the branches of industry where it is more difficult to replace them [1–5]. It also allows to reduce drastically the foul gases release.

We can regard a high temperature gas-cooled nuclear reactor (HTGR) as an energy source for the industrial

☆ This paper is the English version of the paper reviewed and published in Russian in International Scientific Journal for Alternative Energy and Ecology “ISJAEE” issue 154, number 14, date 18.07.2014.

* *Corresponding author*. Tel.: +7 902 442 15 47; fax: +7 343 375 95 08.
E-mail address: s.e.shcheklein@urfu.ru (S. Shcheklein).

http://dx.doi.org/10.1016/j.ijhydene.2015.12.148

processes because it allows to receive high-grade heat of 800–1000 °C. Pebble bed HTGR has additional benefit because this type of nuclear reactor does not need to stop its operation for refueling. The heat production in HTGR is realized by cooling of the reactor core consisting of spherical fuel elements with helium moving in the space between the fuel elements. The main disadvantage of HTGR consists in the following. It is demanded to spent large amount of energy to pump the coolant through several meters of pebble bed. Therefore, the auxiliaries of such plants are unattractively large. The application of radial gas flow allows to receive a compact apparatus with lower hydraulic losses in comparison with traditional axial flow [6].

Iron and steel industry has the second position after power engineering in the rank of consumers of fuel resources. One of the promising ways to reduce the energy consumption of metal production is the use of technologies of direct reduction of metals, especially iron, from the ore. Direct reduced iron (DRI) is produced from oxides by destructing chemical bounds without melting. The result of direct iron reducing is the highly metalized material with a complex of features (composition, structure, size etc.) which depends on the applied technology. The main DRI consumer is electrometallurgy, but it can also be processed in open-hearth and converter furnaces instead of scrap metal. Blast-furnace process is entirely excluded in this type of production [7].

That is why raw material that is achieved in the direct iron reducing process allows to reduce the negative impact of metallurgy on the environment, including the reduction of the carbon dioxide emission into atmosphere. The methods of metal production without blast-furnaces and in the first place the direct reduction method are widespread all over the world. It is connected first of all with increasing need for pure metal.

There are two factors that limit the development of direct reduction methods. The first one is the need of rich ore to maximize productivity. The second is the need of natural gas or other reducing gases and a sufficient quantity of energy resources to run the process effectively.

DRI-technology with hydrogen-containing gases

One of the most effective ways of the direct reducing of iron from the ore is the application of hydrogen-containing gases. The technologies of iron direct reducing from oxides (the so-called DRI and HBI technologies) are used actively worldwide. These methods consume up to 400 m^3 of natural gas per 1 ton of metallic pellets produced [7].

Today we can observe the trend of iron production profitability reduction for the conditions of the rise in prices of the organic fuel.

In the DRI process the enriched iron ore is reduced by the certain gas mixture under the high temperature conditions. Iron ore is usually processed in the form of pellets that contain about 70% of iron by weight. The reduction process can be run in shaft furnaces or rotating pipe furnaces in the atmosphere of reducing gases such as carbon monoxide and hydrogen [7].

Let's examine the process of DRI production on the example of Midrex process. Midrexcompany is the recognized leader of the DRI facilities market which works in this sphere since 1969.

Enriched iron ore in the form of pellets or lumps is charged into Midrex shaft furnace. The typical raw material for the process has the following content:

- Fe – 67%
- SiO_2+Al_2O_3 – 3%
- S – 0.008%
- TiO_2 – 0.15%
- P – 0.03%.

The overall reduction reactions are

$$Fe_XO_Y + CO = Fe + CO_2 \tag{1}$$

$$Fe_XO_Y + H_2 = Fe + H_2O \tag{2}$$

The product of the Midrex process contains 90 … 94% Fe.

The process of iron reducing from oxides runs by sequential transition from higher to lower oxides. It can be schematically described as

$$Fe_2O_3 \rightarrow Fe_3O_4 \rightarrow FeO \rightarrow Fe;\ \text{temperature higher than } 570\,^\circ C$$

or

$$Fe_2O_3 \rightarrow Fe_3O_4 \rightarrow Fe;\ \text{temperature lower than } 570\,^\circ C$$

Hydrogen has lower reducing ability than carbon monoxide if the reaction temperature is lower than 810 °C, but in the temperature range higher than 810 °C it becomes much more stronger deoxidizer [3].

The rate of the oxides reducing process increases significantly if pure hydrogen is used as a reducer. Due to technical and economic difficulties of hydrogen production the sphere of its application is limited and it is used for the production of metallic powders and the creation of nitric and hydrogenous atmosphere for the processes of thermochemical treatment of metals.

The large-scale production of metal pellets is connected with the reducing processes based on synthetic gas (mixture of CO and H_2). Synthetic gas can be produced in a steam methane converter or Midrex reformer from natural gas. It also can be obtained from coal or coking gas. The reducing gas composition affects the quantity of the iron produced. The greatest iron output was achieved when H_2 to CO ratio in the reducing gas fed into the shaft furnace for the reducing process was equal to 1.

The reaction of steam methane conversion

$$CH_4 + H_2O = CO + 3H_2 - 206\ \text{kJ/mol}, \tag{3}$$

$$CO + H_2O = CO_2 + H_2 + 41\ \text{kJ/mol} \tag{4}$$

in view of high endothermicity is run in pipe furnaces with large amount of upright pipes filled with dispersed catalyst and heated by gas burners from the outer side. To heat the steam and methane mixture and to supply the heat of endothermic reaction, large amount of gas is consumed, up to 50% of total amount.

The production of hydrogen by electrolysis for the purposes of metallurgy is limited by high energy consumption of the electrolysis process. Electrolysis energy consumption is 5 times higher than the inherent energy capacity of the received hydrogen. The other problem is high level of CO_2 emission due to the contribution of thermal power plants producing electricity.

The production of hydrogen and hydrogen containing gases in the process of methane conversion has better efficiency and ecological features than electrolysis. For example, the method energy consumption exceeds hydrogen energy capacity in 1.14 times.

Application of HTGR heat for the production of hydrogen containing gases

A nuclear facility for energotechnological purposes consists of a reactor circulation circuit which includes HTGR with helium cooling, gas blowers and other systems, and a technological circuit including chemical reactors. To ensure the facility safety and reliability nuclear and technological circuits are separated with intermediate helium to helium heat exchanger. The nuclear reactor helium outlet temperature is 950 … 1000 °C. The intermediate heat exchanger helium outlet temperature is 900 … 950 °C, that is sufficient for the steam methane conversion reaction [8].

A schematic diagram of the facility is shown in Fig. 1.

Helium of the first circuit heated in HTGR (1) is fed into the intermediate heat exchanger (2), where it transfers heat to the helium of the second circuit. The helium of the second circuit is directed into the pipes of the methane converter (5) where it supplies the endothermic reaction with heat. After the converter, helium with lower temperature arrives in the steam generator (6) to produce steam. Helium circulation is provided by gas blowers (3) and (4).

Features of radial gas flow in the plants with pebble bed

HTGR reactor core with radial gas flow consists of two coaxial cylindrical collectors of greater and lesser radius with walls perforated for the gas passage with spherical fuel elements between them. To calculate the thermohydraulic parameters of such plants we have to take into consideration the peculiarities of radial flow which are connected with velocity gradient in the radial direction. In the case of accelerated flow through the pebble bed the so-called relaminarization effect is observed. In the case of moderated flow we can observe gas turbulization. Both phenomena affect hydrodynamic and heat-exchange characteristics of the process.

The authors have investigated the occurrence of relaminarization and turbulization effects in the case of radial gas flow in the pebble bed [6,8,9]. We have chosen the intensity of static pressure pulsations as a characteristic of relaminarization effect presence. This parameter is connected with vortexes at the spherical elements surface that create pressure pulse at the separation moment. Relative energy of pressure pulsations was measured as

$$\eta = \frac{2\sqrt{\overline{p}^2}}{\rho U_r^2} \quad (5)$$

where $\sqrt{\overline{p}^2}$ is mean-square level of static pressure pulsations on the sphere surface, ρ – gas density, U_r – local flow velocity on the assumption of pebble bed absence.

The results of η measurement for different Reynolds numbers are shown in Fig. 2. For radial flow η lays in the span of 0.95 (nearby outer collector, low Re_r) and 0.1–0.25 (nearby inner collector, $Re_r > 10^3$). It is connected not only with gas rate, but also with gas acceleration in radial direction and relaminarization effect. For axial flow η almost does not depend on the flow velocity in the different points in flow direction.

The authors have carried out experiments on the facility described in Ref. [6], and the analysis of the results allowed us to deduce several equations that can be used to calculate thermohydraulic characteristics of the plants with radial gas flow in pebble bed.

It was obtained an equation to calculate Nusselt number $Nu_r = \alpha_r d_s/\lambda$ taking into account the peculiarities of radial flow:

$$Nu_r = 0.36\frac{(1-\varepsilon)^{0.33}}{\varepsilon}\chi_r^{-1.6}\cdot Re_r^{0.62}, \quad (6)$$

where α_r is local heat-transfer coefficient, W/(m² K); λ – gas heat conductivity factor, W/(m·K); d_s – spherical element diameter, m; ε – pebble bed porosity.

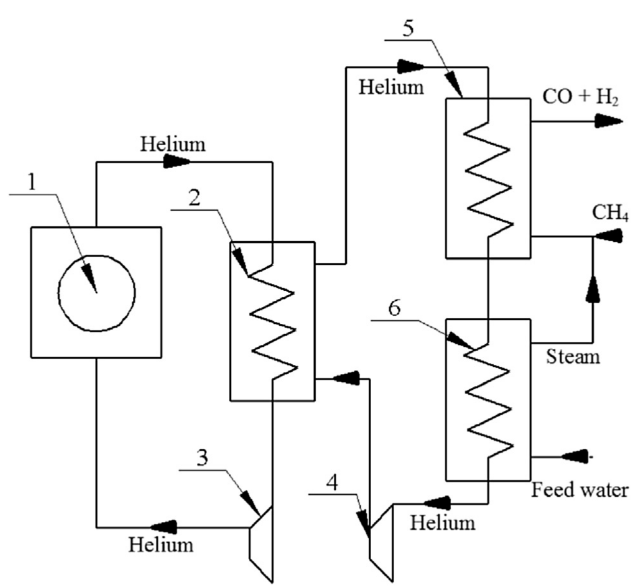

Fig. 1 – Energotechnological facility schematic diagram.

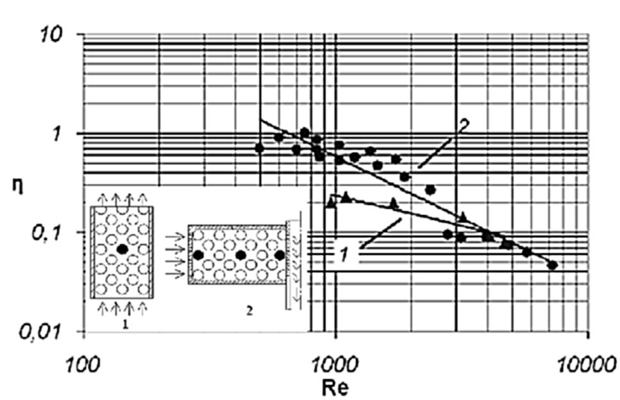

Fig. 2 – Dependency of static pressure pulse intensity on Reynolds number: 1 – axial flow, 2 – radial flow.

Reynolds number for radial gas flow is determined as

$$Re_r = \left(\frac{G}{2\pi H}\right) \cdot \frac{d_s}{r \cdot \nu}, \quad (7)$$

where G is gas volume flow rate, m^3/s; H – pebble bed height, m; ν – dynamic viscosity coefficient, m^2/s; r – radial coordinate of spherical element, m.

The remaining parameter, χ_r, includes the influence of gas flow local acceleration:

$$\chi_r = 4\left({r}/{r_{in}} + {r_{in}}/{r} + 2 \right)^{-1}, \quad (8)$$

where r_{in} is the radius of inner collector.

Eq (6) turns into the formula of heat transfer coefficient for axial gas flow in the pebble bed in case of low relative curvature of the collectors. The relaminarization effect is observed for the outer and inner collectors' ratio r_o/r_{in} >1.15 [6].

Hydraulic resistance for the pebble bed can be described by the Darcy–Weisbach equation:

$$\Delta p = \xi_r \cdot (8\pi^2 d_s)^{-1} \cdot \rho \cdot \left(\frac{G}{H}\right)^2 \cdot \left(\frac{1}{r_{in}} - \frac{1}{r_o}\right), \quad (9)$$

where ρ is gas density; ξ_r – hydraulic resistance coefficient for radial flow.

It was experimentally achieved the dependency of hydraulic resistance coefficient from main regime and design parameters:

$$\xi_r = \frac{108}{\varepsilon^4} \cdot Re_{\bar{r}}^{-0,79} \cdot \chi \ \text{ for } ds = 15 \text{ and } 22 \text{ mm and } 0.4 \le \varepsilon \le 0.44; \quad (10)$$

$$\xi_r = \frac{43,6}{\varepsilon^4} \cdot Re_{\bar{r}}^{-0,35} \cdot \chi \ \text{ for } d_s = 7 \text{ mm and } \varepsilon = 0.31. \quad (11)$$

In eqns (10) and (11) parameter χ establishes linkage between the hydraulic resistance coefficients for radial flow ξ_r and axial flow ξ:

$$\xi_r = \xi \cdot \chi; \ \chi = 4\left({r_o}/{r_{in}} + {r_{in}}/{r_o} + 2 \right)^{-1}. \quad (12)$$

Parameter χ includes the influence of collectors' relative curvature. In the case of insignificant curvature r_o/r_{in} <1.15 the values of hydraulic resistance coefficients for radial and axial flows are approximately the same.

Reynolds number in the eqns (10) and (11) is calculated with averaged velocity

$$U_{\bar{r}} = \frac{G}{2\pi \bar{r} H}, \quad (13)$$

where $\bar{r} = 0.5(r_o + r_{in})$.

The empirical equations achieved reveal the variation of hydraulic resistance coefficient in the range of outer and inner collectors ratio $3.5 \le r_o/r_{in} \le 7.1$ and Reynolds number in the pebble bed average cross-section $Re_{\bar{r}} = 3 \cdot 10^2 \ \ldots \ 5 \cdot 10^4$. Maximum mean square error was 13% for spherical elements with diameter 15 or 22 mm and 22% for $d_s = 7$ mm because of poor hydrodynamic characteristics of little static pressure receivers.

Formulas for Δp and Nur possess quite simple structure and reflect design and regime parameters of plants with radial flow in pebble bed and can be used for these plants' design. The relaminarization effect appearance is accompanied by decrease of hydraulic resistance and heat transfer coefficient in radial direction in comparison with axial flow in pebble bed.

High temperature gas-cooled reactor with radial gas flow

The equations given above were used to perform estimation of thermohydraulic parameters of HTGR reactor core. The calculation input data are given it Table 1.

The calculation results are given in Fig. 3. The calculation was performed for taken values of inner collector radius $r_{in} = 0.4 \ \ldots \ 0.2$ m Fig. 3 shows the outer collector radius r_o, reactor core height H, pressure losses Δp and hydraulic resistance coefficient ξ_r. The value of $r_{in} = 0.8$ m was taken for further calculation, the results of it are given in Table 2.

The data of Table 2 gives the opportunity to compare the calculation results for the primary circuit of nuclear plant with radial flow and axial flow HTGR of the same capacity. Pressure drop in the radial flow reactor core is given considering the losses for perforation of inner and outer collectors.

Table 2 shows that application of radial flow diagram allows to save up to 60% of gas blower power consumption. The core with axial flow diagram has fuel rating $q_v^{ax} = (0.75 \ldots 0.8) q_v^{rad}$, for the same height $H_{ax} = H_{rad}$. This fact allows us to conclude that radial flow HTGR has more compact design. On the other side, calculation shows that hydraulic resistance of axial flow HTGR is approximately 7.5 times higher due to higher flow velocities in the pebble bed space.

Methane converter with radial flow of reacting gases

Secondary circuit of the plant shown in Fig. 1 includes a methane converter for synthetic gas production. Synthetic gas produced at the plant can be transported through pipelines to the consumers which are represented not only by metal industry but also ammonia, methanol and synthetic fuel

Table 1 – HTGR calculation input data.

Parameter	Designation	Value
Heat power, W	Q_t	$330 \cdot 10^6$
Coolant pressure at core inlet, Pa	P_{in}	$5 \cdot 10^6$
Coolant temperature, °C		
Core inlet	T_{in}	500
Core outlet	T_o	950
Fuel elements		Spherical
Fuel element diameter, m	d_s	0.06
Fuel element core diameter, m	d_c	0.05
Fuel rating, W/m^3	q_v	$9.5 \cdot 10^6$
Core volume, m^3	V	44
Coolant mass flow, kg/s	G_m	141.2
Nusselt number for core inlet [10]	Nu_{in}	88.2
Reynolds number for core inlet	Re_{in}	2035

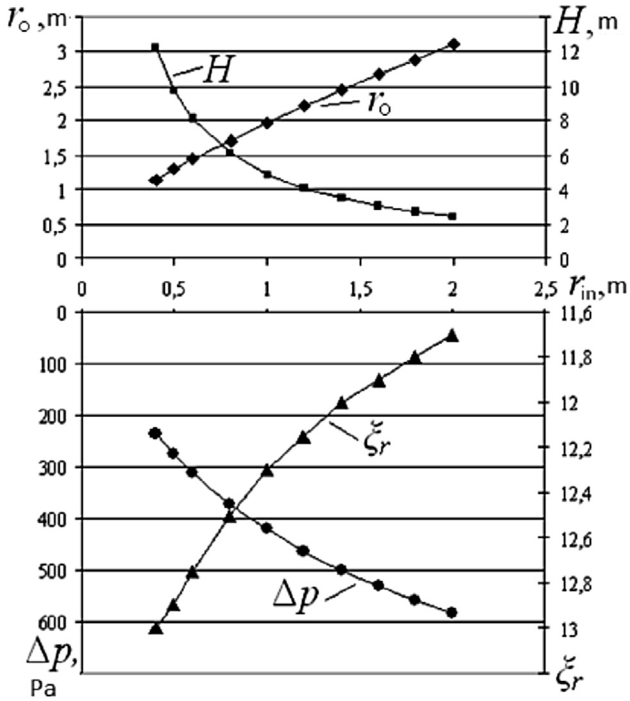

Fig. 3 – Reactor core calculation results.

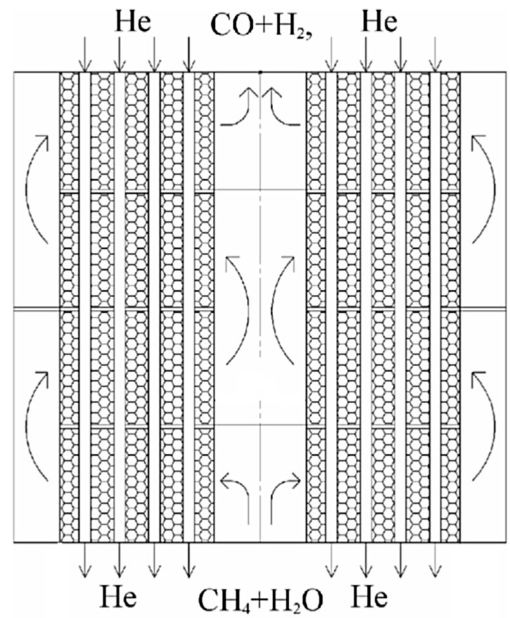

Fig. 4 – Steam methane converter schematic diagram.

producers. It is expedient to provide the methane converter heating through a system of pipes with the helium of the second circuit circulating inside. The catalyst granules fill the space between tubes of the converter. The reacting mixture of gases can flow in axial or radial direction relative to the apparatus axis.

Apparatuses with radial gas flow in the granular layer have substantial advantage in comparison with axial flow apparatuses because of significantly lower flow pressure loss in the layer. This leads not only to the productive capacity rise and energy saving but also to the reduction of specific quantity of metal due to better apparatus compactness [8].

To intensify the heat transfer process in the tube furnaces with catalyst layer the multipass circulation of reacting gases is usually applied. In this case a convergent-divergent regime is realized in radial flow apparatuses. The reacting gas flow is accelerated in one section and is decelerated in the following section.

The schematic diagram of a methane converter with radial flow is shown in Fig. 4. The catalyst granules are laid in the cavity between two coaxial cylindrical collectors of greater and lesser radius with walls perforated for the gas passage.

For design calculation it is necessary to consider the peculiarities of radial gas flow connected with the flow velocity variation from the entrance of distributing collector to the exit of receiving collector [6].

The heating of steam and methane mixture ($CH_4 + H_2O$) is implemented by a system of vertical pipes with helium heat transfer agent. The intensity of heat transfer from the helium inside the pipes to the reacting mixture can be sufficiently increased with the use of different methods of flow swirling inside the pipes and rational choice of radial flow mode.

Table 2 – HTGR primary circuit calculation results.

Parameter	Radial flow	Axial flow
Pressure drop in reactor core, Pa	505	7360
Primary circuit pressure drop, Pa	4000	10 860
Temperature before gas blower, °C	498.8	499.2
Gas blower power consumption, MW	0.413	1.12
Power transfer to the secondary circuit, MW	316.8	316.8

Table 3 – The results of thermohydraulic calculation of the methane converter.

Parameter	Methane rate (tons per hour)		
	500	1000	1500
Quantity of heat, MW			
Total	6.85	13.71	20.56
For gas mixture heating	0.93	1.87	2.80
For endothermic reaction	5.92	11.84	17.76
Helium rate, kg/s	17.6	35.2	52.8
Characteristics of heat exchange tubes			
Length, mm	3000		
Diameter × wall thickness, mm^2	40 × 2.5		
Spacing, mm	56		
Heat transfer coefficient, $W(m^2 K)$			
For heating zone	120.6	133.8	143.4
For reaction zone	155.5	168.8	178.4
Heat exchange surface, m^2	231.9	425.5	601.9
Converter diameter, m	1.62	2.10	2.45
Pressure loss for the reacting gas, kPa	3.79	12.5	24.6
Pressure loss for helium, kPa	3.17	3.71	4.14
Needed catalyst volume, m^3	0.24	0.48	0.72
Space between tubes, m^3	3.2	5.5	7.5

The calculation of main dimensions of the steam methane converter with radial gas ($CH_4 + H_2O$) flow was made with the use of experimental formula given in Refs. [6,8]. An experimental facility with granular layer filling the space between tubes was used to achieve data on the heat exchange between the gas inside the pipes and the gas moving in the granular layer.

The results of thermohydraulic calculation are given in Table 3.

Conclusion

The suggested scheme of synthetic gas production with high temperature gas-cooled reactor with radial gas flow in pebble bed is rather universal and can be used to supply the synthetic motor fuels, metallurgic and chemical industry with hydrogen-containing raw material.

The data of calculation show the advantages of plants with radial gas flow. HTGR with radial flow of coolant has more compact design and much lesser hydraulic resistance in comparison with axial flow due to lower velocities of gas in pebble bed.

Acknowledgements

This research is funded by the Russian Scientific Fund (Research Project 15-19-30001).

REFERENCES

[1] Ponomarev-Stepnoy NN, Stolyarevskii AYa, Pakhomov VP. Nuclear-hydrogen power engineering. Moscow: Energoizdat; 2008. p. 76–89.

[2] Stolyarevskij AYA, Khusnutdinov VA. Innovatsionnye tekhnologii atomno-vodorodnoj ehnergetiki v proekte «Bakcharskaya stal'». Mezhdunarodnyj nauchnyj zhurnal «Al'ternativnaya ehnergetika i ehkologiya» ISJAEE 2007;11(55):164–73. ISSN 1608-8298.

[3] Market Potential for non-electric applications of nuclear energy. Vienna: IAEA; 2002.

[4] Matsui K. Possible scenarios and its effects by non-power. Oarai, Japan: The Institute of Applied Energy, Kunitomi K., Japan Atomic Energy Agency; 16 April 2007.

[5] IAEA-Tecdoc-1085 Hydrogen as an energy carrier and its production by nuclear power. 1999.

[6] Klimova VA, Pakhaluev VM, Shcheklein SE. Numerical simulation and experimental investigations of hydrodynamics and heat transfer for radial gas flow in an apparatus with ball packing. Therm Eng 2011;4(V.58):325–30.

[7] Yaroshenko YuG, Gordon YaM, Hodorovskaya IYu. Energy efficient and resource-saving technologies in iron-and-steel industry. Ekaterinburg: UIPC; 2012. p. 526–76.

[8] Klimova VA, Pakhaluev VM. Energotechnological system of long-distance atomic heat supply with radial gas flow installations. Int Sci J Altern Energy Ecol 2012;3(107):26–30.

[9] Klimova V, Pakhaluev V, Shcheklein S. Production of reducing environment for metallurgy using nuclear energy. WIT transactions on ecology and the environment, vol. 190. WIT Press; 2014. ISSN 1743-3541.

[10] Popov SV. Static thermophysics of HTGR with pebble bed core. Atomno Vodorodnaya Energ I Tech 1982;4:126–9.