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TECHNOLOGICAL ASPECTS OF PURIFICATION AND SEPARATION OF HYDROGEN-CONTAINING GASES OBTAINED BY GASIFICATION OF SOLID CARBON-CONTAINING MEDIA

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Abstract. In this paper, the subject of study is the processes of purification and separation of hydrogen-containing gases obtained by plasma-chemical gasification of solid carbon-containing media. The work aims to establish the regularities of the influence of technological parameters of hydrogen-containing gases obtained by the gasification of solid carbon-containing media on the specific surfaces of filter partitions, membranes, and adsorbents used for their purification and separation. The graph-analytical research method is used in this work, which consists in finding rational specific surfaces of the processes of purification and separation of hydrogen-containing gases based on the obtained graphical dependencies.

Analytical dependencies for calculating the specific surface areas of the filter partition, the membrane, and the adsorbent were established. These dependencies take into account the technological parameters of hydrogen-containing gases and the technological parameters of their separation to produce high-purity hydrogen.

Based on the theoretical calculation results, a graphical dependence of the specific surface area of the filter partition on the temperature of hydrogen-containing gases formed during the gasification of solid carbon-containing media and the concentration of solid particles in the gas was obtained. This dependence shows that an increase in the concentration of solid particles and an increase in the temperature of hydrogen-containing gases lead to an increase in the specific surface area of the filter partition. The graphical dependence of the specific surface area of the polyimide membrane on the operating pressure and hydrogen concentration in the permeate and the graphical dependence of the specific surface area of zeolite 13X on the initial hydrogen concentration in the initial gas mixture from 60% to 90% at a final concentration of 99.99% are presented.

The paper presents a schematic diagram of purification and separation of hydrogen-containing gases to produce high-purity hydrogen. According to the diagram, the hydrogen-containing gas obtained by the gasification of carbon-containing media is sent for purification from solid particles to a filtration system, from where it is supplied to membrane separation for concentration, and the concentrated hydrogen-containing gas is sent to adsorption separation, where it is separated to produce high-purity hydrogen.

Keywords: carbon-containing medium, hydrogen-containing gas, hydrogen, filtration, membrane separation, adsorption.

1. Introduction

The growing negative impact on the environment of processing products of carbon-containing raw materials of various origins is an urgent issue of our time due to the growing energy consumption by the global economy and, as a result, the release of a significant amount of carcinogenic, toxic and harmful compounds into the atmosphere. The public attention is primarily focused on the problem of environmental pollution caused by greenhouse gas emissions of CO_2 and CH_4, which are generated during the processing of carbon-containing raw materials, causing overall climate warming. According to the plan, by 2050, the global society is going to switch to carbon-free, hydrogen-based energy consumption technology.

In general, new technologies for processing solid carbon-containing raw materials, such as coal of various degrees of metamorphism, sludge, and ash, must meet the following requirements: they must not produce environmental pollution problems, be efficient for industrial use, and be competitive in the energy market.

One way to solve the energy and environmental problems is to process carbon-containing raw materials by plasma-chemical conversion to produce hydrogen-containing gas and inert residue or valuable substances and materials from it.

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Existing autothermal technologies for converting carbon-hydrogen containing raw materials into energy gas - Winkler, Lurgi, Koppers-Totzek, and others - are based on direct combustion of a part of these raw materials, which leads to contamination of the gas phase with combustion products. Contamination is the main disadvantage of autothermal technologies. A promising alternative to autothermal technologies is allothermal technologies of high-temperature transformations with the production of hydrogen-containing gases, which more fully meet the requirements of modern production and ecology [1–3].

The most promising among allothermal technologies is a steam-plasma conversion, in which water is used as an oxidizing agent, which reduces harmful environmental emissions. Hence, this technology most fully meets environmental requirements. Studies conducted at M.S. Poliakov Institute of Geotechnical Mechanics of the National Academy of Sciences of Ukraine (IGTM of the NAS of Ukraine) have shown that the steam plasma technology for converting solid carbon-containing medium (SCCM) produces hydrogen-containing gas with hydrogen (H_2) content of 55–60% and carbon monoxide (CO) content of 35–40%; the maximum yield of combustible components (H_2 + CO) reaches 93–97% while increasing the caloric content to 11.5–11.7 MJ/m^3 due to the increase of the hydrogen component during the decomposition of water into components. Therefore, the resulting hydrogen-containing gas can be a promising source of raw materials in technologies for producing pure hydrogen, in the production of methyl alcohol and synthetic fuels.

However, to ensure compliance with the standards [4], according to which industrial hydrogen must meet the standards given in Table 1, there is a need for the preliminary purification from solid particles and subsequent separation into components of hydrogen-containing gases obtained as a result of plasma-chemical conversion of the SCCM.

Table 1 – Physical and chemical parameters of industrial hydrogen

Indicator name	Norm for brand	
	A	B
1. Volume fraction of hydrogen in terms of dry gas, %, not less than	99.99	99.95
2. Total volume fraction of oxygen and nitrogen, %, not more than	0.01	0.05
3. Mass concentration of water vapor at 20 °C and 101.3 kPa (760 mm Hg), g/m^3, not more than		
a) in pipelines	0.5	0.5
b) in pressurized cylinders	0.2	0.2

Accordingly, the subject of this study is the processes of purification from solid particles and separation into components of hydrogen-containing gases obtained by plasma-chemical gasification of the SCCM; the purpose of this work is to establish the regularities of the influence of technological parameters of hydrogen-containing gases on the specific surfaces of phase separation used for their purification and separation processes.

2. Methods

The graph-analytical method of research is used in this paper, which consists in finding rational specific surfaces of the processes of hydrogen-containing gases purification and separation based on the obtained graphical dependencies.

3. Theoretical part

Due to the fact that raw materials with a particle size of no more than 200 μm are sent for thermal conversion of the SCCM to produce hydrogen-containing gases, there is a possibility of removal unreacted particles with the resulting gas flow. Since the gas flow is cooled to normal temperatures after the thermal conversion unit, all vapours contained in the gas mixture are condensed and separated from the gas flow. Thus, relatively dry hydrogen-containing gases with a possible content of solid particles of no more than 200 μm are sent for purification. Therefore, taking into account the initial parameters of hydrogen-containing gases, the most rational method of purification from solid particles is filtration and, accordingly, the use of filters for their purification from mechanical impurities.

According to [5], various types of materials can be used as filter materials for the purification of hydrogen-containing gases containing solid particles. However, given that after cooling, the temperature of hydrogen-containing gases can reach 150 °C, it is appropriate to use materials with high heat resistance, such as nitron, lavsan, and fiberglass, where the permissible temperature is 120 °C, 130 °C, and 240 °C, respectively. Since different materials have different resistance and cost, further selection of the material for filter partition should be based on a technical and economic calculation.

Let's establish a dependence for determining the specific area of the filter partition, i.e., the surface area per unit flow rate of the gas mixture that is separated from solid particles, and determine the effect of technological parameters of the gas mixture on the specific area of the filter partition.

Specific surface area of the filter partition ($f_{sp}^{f \cdot p}$, m^2/(m^3/s)) is determined by the formula, using the dependence of the surface area of the filter partition, given in [6, 7]:

$$f_{sp}^{f \cdot p} = \frac{F^{f \cdot p}}{V} = \frac{1}{q}, \qquad (1)$$

where $F^{f \cdot p}$ – surface area of the filter partition, m^2; V – volumetric flow rate of the gas mixture, m^3/s; q – specific gas load during filtration, m^3/(m^2·s).

The specific gas load during filtration is determined by the formula [6, 7]:

$$q = q_{st} \cdot C_1 \cdot C_2 \cdot C_3 \cdot C_4 \cdot C_5, \qquad (2)$$

where q_{st} – standard gas load, depending on the type of dust, m^3/(m^2·s); C_1 –

coefficient that takes into account the peculiarity of filter element regeneration; C_2 – coefficient that takes into account the effect of dust concentration on the specific gas load; C_3 – coefficient that takes into account the influence of the dispersed composition of dust in the gas; C_4 – coefficient that takes into account the effect of the temperature of the gas to be cleaned; C_5 – coefficient that takes into account the requirements for gas purification quality.

Substituting formula (2) into (1), we obtain:

$$f_{sp}^{f \cdot p} = \frac{1}{q_{st} \cdot C_1 \cdot C_2 \cdot C_3 \cdot C_4 \cdot C_5}, \qquad (3)$$

After purification from solid particles, the next step in obtaining high-purity hydrogen from hydrogen-containing gases is their separation into components based on membrane and absorption separation processes.

The membrane units of Membrane Technology and Research, Inc. (MTR) [8] are designed to separate H_2 from synthesis gas and to regulate the H_2:CO ratio. In a typical system, hydrogen passes through the membrane forming a flow of purified hydrogen permeate and a hydrogen-depleted synthesis gas flow with an adjusted ratio. This ratio control makes it possible to produce high-purity CO for processes that require CO as a raw material. The developed units allow for efficient recovery of synthesis gas and hydrogen, achieving a recovery rate of 90% to 98% of the initial CO and producing hydrogen with a purity of 85% to 95% (vol.).

At present, other companies also produce membrane plants for gas separation: CanGas [9], Evonik [10], Air Liquide Advanced Separations (ALaS) [11], GENERON [12], which use a hollow membrane consisting of porous polymer fibers coated with a separation layer. The porous fiber has a complex asymmetrical structure, with the polymer density increasing towards the outer surface of the fiber. Gases at high pressures (up to 6.5 MPa) can be separated by using porous support layers with an asymmetrical structure. The thickness of the gas separation fiber layer does not exceed 0.1 µm, which ensures high relative gas permeability of the polymer membrane. The current level of technology makes it possible to produce polymers with high selectivity for various gases and, therefore, capable of providing a high degree of purity of gaseous products. A modern membrane module used in membrane gas separation technology consists of a removable membrane cartridge and housing. The density of fiber packing in the cartridge is estimated at about 500–700 square meters per cubic meter of the cartridge, which minimizes the size of gas separation units.

The analysis of membrane technologies for the separation of hydrogen-containing gases shows that their usage makes it possible to achieve a high degree of hydrogen recovery (up to 90%), and membrane plants are compact. However, these technologies are characterized by high operating costs due to the need to create high pressures (10 atm or more) and the impossibility of obtaining high-purity hydrogen (no higher than 60–70%) in single-stage plants due to the limited selectivity of membranes. In this regard, to obtain high-concentration hydrogen using membrane technologies, it is necessary to use a multi-stage (cascade) plant, which significantly increases the overall size of the plant. The cascade membrane unit should consist of roll membrane apparatus or hollow fiber membrane apparatus containing polymeric membranes, the choice of which depends on the composition of the hydrogen-containing gas, the degree of extraction and purity of the hydrogen produced.

Recently, short-cycle adsorption (SCA) units have been widely used in industry [13–20] to separate gas mixtures to obtain hydrogen, nitrogen, oxygen, methane, ethylene, and other components with high concentrations (from 95% to 99.9999%). This technology makes it possible to separate almost any gas.

The SCA technology is based on gas absorption by adsorbent using a pressure function. In the adsorption process, a mixture of gases is fed into the adsorber at increased pressure and ambient temperature. In this process, the easily adsorbed components of the mixture are absorbed, while the weakly adsorbed or non-adsorbed components pass through the apparatus, which separates the gas mixture. The adsorbent absorbs the gas to a state of equilibrium between adsorption and desorption, after which the adsorbent must be regenerated, i.e., the absorbed components must be removed from the adsorbent surface. This can be done either by increasing the temperature or by reducing the pressure. Typically, short-cycle adsorption uses heatless regeneration by reducing the pressure. For today, three methods of organizing a cyclic non-heating adsorption separation process have become widespread: pressure method - Pressure Swing Adsorbrion (PSA), vacuum method - Vacuum Swing Adsorption (VSA) and mixed method - Vacuum Pressure Swing Adsorption (VPSA).

An analysis of industrial adsorption technologies for the separation of hydrogen-containing gases shows that these technologies make it possible to produce high-purity hydrogen (up to 99.99%) using zeolites in the form of molecular sieves. However, adsorption technologies are characterized by a low degree of recovery - for single-stage plants no more than 60–70%; hydrogen-containing gases containing at least 50–60% hydrogen are subject to separation; a significant volume of the required adsorbent and, accordingly, the bulkiness of the equipment; the frequency of the process (for single-stage plants) and the need to increase the gas pressure from 5 atm. Therefore, a multi-stage plant is required to produce hydrogen of high concentration and with a high degree of recovery from hydrogen-containing gases obtained by thermal conversion of carbon-containing raw materials, which significantly increases the bulkiness of the equipment and operating costs.

Papers [21–25] show that the use of hybrid membrane-adsorption systems in the separation of hydrogen-containing gases obtained during the gasification of carbon-containing raw materials makes it possible to avoid the disadvantages of adsorption separation technologies characterized by a low degree of hydrogen recovery and the disadvantages of membrane technologies characterized by low purity of the hydrogen produced. When using membrane-adsorption systems, a high degree of hydrogen recovery (up to 90%) and hydrogen purity (up to 99.9%) are achieved due to their preliminary concentration in membrane plants and subsequent separation in adsorption plants.

Metal membranes based on palladium and its alloys can be used for hydrogen concentration. However, these membranes are characterized by high cost, irreversible “poisoning” of palladium, and the need to carry out the separation process at high temperatures from 673 K to 900 K. Therefore, polymeric membranes, which are cheaper than palladium membranes, such as polyimide, polysulfone, cellulose acetate, with H_2/CO separation factors of 41.0, 74.0, and 40.0, respectively, have been used for industrial-scale hydrogen separation [26, 27]. At the same time, these polymeric membranes have increased heat resistance and can be used to separate hydrogen-containing gases at temperatures up to 150 °C.

Let us consider the influence of the chemical composition of the gas to be separated on the membrane separation processes. Since the hydrogen-containing gas obtained during the gasification of the SCCM can contain such components as H2O, H_2O, H_2, CO_2, O_2, H_2S, N_2, CO, CH_4, C_3H_8, their kinetic diameters are, respectively, 2.65, 2.89, 3.3, 3.46, 3.6, 3.64, 3.76, 3.8, 4.3 (10^{-10} m or Å) (Fig. 1). That is, the pore size of the membrane partitions should be less than 3.3 Å, which makes it possible to concentrate hydrogen-containing gas, i.e., to increase the hydrogen concentration by removing other components in membrane systems.

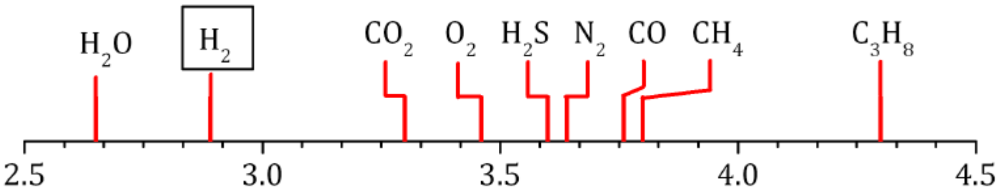


Figure 1 – Dependence of the kinetic diameter (relative size) on the type of gas

The pore size of polymeric membranes [29, 30] such as polyimide, polysulfone, and cellulose acetate depends on their structure and manufacturing method, but only polyimide can be produced with pore sizes less than 3 Å, so polyimide membranes are more effective for separating gas mixtures, including hydrogen production.

Let us establish a dependence for determining the specific area of the membrane, i.e., the surface area of the membrane per unit of flow rate of the gas mixture separated by the membrane process, and determine the effect of the technological parameters of the gas mixture on the specific area of the membrane. The specific area of the membrane is found from the dependence on the definition of the membrane surface (F^m, m^2) [29, 30]:

$$F^m = \frac{Q_{s.g}}{P \cdot \Delta p}, \tag{4}$$

where $Q_{s.g}$ – volumetric flow rate of the separated gas, m^3/s; P – membrane permeability, m^3/(m^2·s·Pa); Δp – difference in partial pressure of the gas passing through the membrane, Pa:

$$\Delta p = p_{in} - p_{out} = y_{in} \cdot P_{in} - y_{out} \cdot P_{out}, \tag{5}$$

where y_{in} and y_{out} – mole fractions of the separated gas at the inlet and outlet, respectively; P_{in} and P_{out} – total inlet and outlet pressure, respectively, Pa;

The volumetric flow rate of the separated gas is calculated from the total flow rate of the gas mixture (Q_{total}):

$$Q_{s.g} = Q_{total} \cdot x_{s.g}, \tag{6}$$

where $x_{s.g}$ – mass concentration of the gas to be separated in the initial gas mixture, kg/kg.

Let's find the specific surface of the membrane (f_{sp}^{m}, m^2/(m^3·s)), substituting the formulas (5) and (6) in (4):

$$f_{num}^{м} = \frac{F^{m}}{Q_{total}} = \frac{x_{s.g}}{P \cdot (y_{in} \cdot P_{in} - y_{out} \cdot P_{out})}. \tag{7}$$

The next stage of separation of hydrogen-containing gases after their concentration to 50–60% by membrane processes is adsorption processes, which allows to obtain high-purity hydrogen.

For the separation of hydrogen-containing gases, synthetic zeolites [31] of grades KA, NaA, CaA, NaX, and CaX are most widely used. The first letter of the zeolite brand corresponds to the form of the cation that compensates for the lattice charge (K+, Na+, Ca+), and the second letter indicates the type of crystal lattice. The choice of zeolite grade depends on the composition of the hydrogen-containing gas and the purity of the produced hydrogen.

In the process of hydrogen purification using zeolites, hydrogen-containing gas (synthesis gas) containing impurities is passed through a zeolite layer. Hydrogen molecules, as the smallest, pass freely through the pores, and impurity molecules (CO and others) are retained on the surface of the adsorbent due to the combination of smaller molecule size, lower polarity and weaker interaction with the adsorbent surface, which makes hydrogen more mobile and promotes its rapid diffusion through the pores of the adsorbent compared to carbon monoxide. Thus, zeolites act as molecular sieves in the separation of hydrogen-containing gases.

Taking into account the recommendations of [32, 33], we establish a dependence for determining the specific area of the adsorbent, i.e., the surface area per unit flow rate of the gas mixture separated by the adsorption process, and determine the effect of the technological parameters of the gas mixture on the specific area of the adsorbent.

The specific area of the adsorbent (f_{sp}^{ads}, m^2/(m^3·s)) is found from the dependence:

$$f_{sp}^{ads} = \frac{F^{ads}}{Q_{ads}}, \tag{8}$$

where F^{ads} – total surface area of the adsorbent, m^2; Q_{ads} – volumetric flow rate of the gas for adsorption, m^3/s.

Total surface area of the adsorbent:

$$F^{ads} = m^{ads} \cdot S^{ads}, \tag{9}$$

where m^{ads} – adsorbent mass, kg; S^{ads} – surface area per unit mass of adsorbent, m^2/kg.

Adsorbent mass:

$$m^{ads} = \frac{N_{CO}^{ads}}{q_{CO}}, \tag{10}$$

where N_{CO}^{ads} – the amount of absorbed CO, mole; q_{CO} – equilibrium adsorption capacity for CO, mole/kg.

The amount of absorbed CO:

$$N_{CO}^{ads} = n_{CO}^{in} - n_{CO}^{out}, \tag{11}$$

where n_{CO}^{in}, n_{CO}^{out} – the amount of CO contained in the gas flow at the inlet and outlet, respectively, mole:

$$n_{CO}^{in} = n^{in} \cdot C_{CO}^{out}, \tag{12}$$

$$n_{CO}^{out} = n^{in} \cdot C_{CO}^{out}, \tag{13}$$

where n^{in} – the amount of gas flow going to adsorption, mole; C_{CO}^{in}, C_{CO}^{out} – CO concentration in the gas flow at the inlet and outlet, respectively, mole/ mole.

The amount of gas flow going to adsorption is determined by the equation of ideal state:

$$n^{in} = \frac{Q_{ads} \cdot \tau_{ads}}{R \cdot T} \cdot P, \tag{14}$$

where τ_{ads} – time of the adsorption process, s; P – absolute pressure, Pa; R –

universal gas constant, R=8.314 J/(mole·K); T – absolute temperature, *K*.

Substituting formulas (9)–(14) into formula (8), we obtain:

$$f_{sp}^{ads}=\frac{\dfrac{Q_{ads}\cdot\tau_{ads}}{R\cdot T\cdot q_{CO}}\cdot P(C_{CO}^{in}-C_{CO}^{out})\cdot S^{ads}}{Q_{ads}}=\frac{P(C_{CO}^{in}-C_{CO}^{out})\cdot S^{ads}\cdot\tau_{ads}}{R\cdot T\cdot q_{CO}}, \qquad (15)$$

Since

$$C_{CO}=100-C_{H_2}, \qquad (16)$$

where C_{H_2} – H_2 concentration in the gas flow, mole/mole.

Then equation (15) has the form:

$$f_{sp}^{ads}=\frac{P(C_{H_2}^{out}-C_{H_2}^{in})\cdot S^{ads}\cdot\tau_{ads}}{R\cdot T\cdot q_{CO}}. \qquad (17)$$

4. Results and discussion

Based on formula (3), we obtained a graphical dependence of the specific surface area of the glass fiber filter membrane on the temperature of hydrogen-containing gases obtained during the gasification of the SCCM and the concentration of solid particles in the gas, which is shown in Fig. 2.

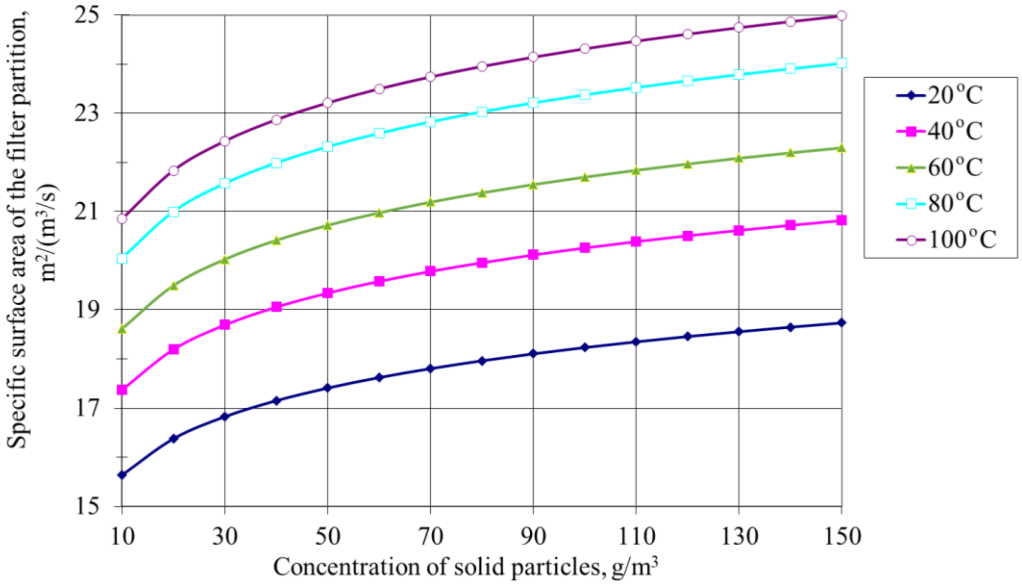


Figure 2 – Dependence of the specific surface area of the filter partition from glass fiber on the temperature of hydrogen-containing gases obtained during the gasification of the SCCM and the concentration of solid particles in the gas

The analysis of the results shows that an increase in the concentration of solid particles in hydrogen-containing gases from 10 g/m^3 to 150 g/m^3 leads to an increase in the specific surface area of the filter partition. Thus, at a temperature of hydrogen-containing gases of 20 °C, the specific surface area of the filter partition increases from 15.6 m^2/(m^3·s) to 18.7 m^2/(m^3·s). An increase in the temperature of hydrogen-containing gases from 20 °C to 100 °C also leads to an increase in the specific surface area, and at a concentration of solid particles in hydrogen-containing gases of 150 g/m^3, the specific surface area of the filter partition increases from 18.7 m^2/(m^3·s) to 24.9 m^2/(m^3·s).

Thus, the specific surface area of the filter partition increases with the increase in the concentration of solid particles and the temperature of hydrogen-containing gases, which leads to a corresponding increase in the area of the filter partition with an increase in the flow rate of hydrogen-containing gas. That is, the choice of the filter partition area should be made at the maximum values of the concentration of solid particles and the temperature of hydrogen-containing gases.

On the basis of dependence (7) and taking into account that the supply pressure or vacuum pressure (permeate pressure) from 10 MPa to 50 MPa is used in industry to separate the H_2/CO system [51, 52], a graphical dependence of the specific surface area of a polyimide membrane on the operating pressure and hydrogen concentration in the permeate was obtained (Fig. 3).

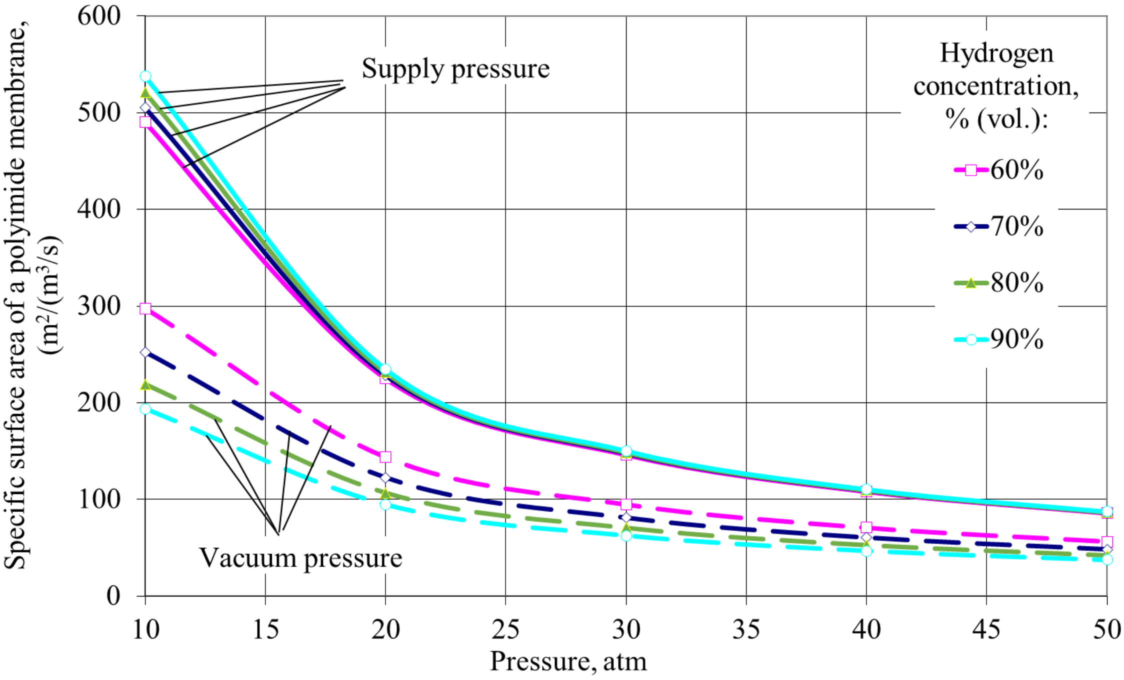


Figure 3 – Dependence of the specific surface area of a polyimide membrane on the operating pressure and hydrogen concentration in the permeate

The analysis of the results shows that an increase in the operating feed pressure from 10 MPa to 50 MPa leads to a decrease in the specific surface area of the membrane from 490 m^2/(m^3·s) to 86 m^2/(m^3·s), while changing the hydrogen concentration from 60% to 90% does not significantly affect the specific surface area of the membrane, since at the feed pressure the change in the final concentration does not lead to a change in the driving force of the process.

An increase in the operating vacuum pressure from 10 MPa to 50 MPa at a final concentration of 60% leads to a decrease in the specific surface area of the membrane from 298 $m^2/(m^3 \cdot s)$ to 56 $m^2/(m^3 \cdot s)$, and at a final concentration of 90% - from 194 $m^2/(m^3 \cdot s)$ to 37 $m^2/(m^3 \cdot s)$. At the same time, changing the hydrogen concentration from 60% to 90% significantly affects the value of the specific surface area of the membrane, so at a pressure of 10 MPa, the specific surface area decreases from 298 $m^2/(m^3 \cdot s)$ to 194 $m^2/(m^3 \cdot s)$, which indicates an effect on the driving force of the process at the operating vacuum pressure.

Thus, at the supply pressure, the specific surface area of the membrane decreases with increasing operating pressure and the change in the final concentration does not significantly affect its value; and at the vacuum pressure, the specific surface area of the membrane decreases both with increasing operating pressure and with increasing final concentration. That is, the choice of the operating pressure of the membrane process of syngas separation should be based on further technical and economic calculations.

On the basis of dependence (17), a graphical dependence of the specific surface area of zeolite 13X on the hydrogen concentration in the initial gas mixture was obtained, taking into account that after membrane separation, hydrogen-containing gas concentrated from 60% to 90% is fed for adsorption, and after separation by adsorption, hydrogen with a purity of 99.99% is obtained (Fig. 4).

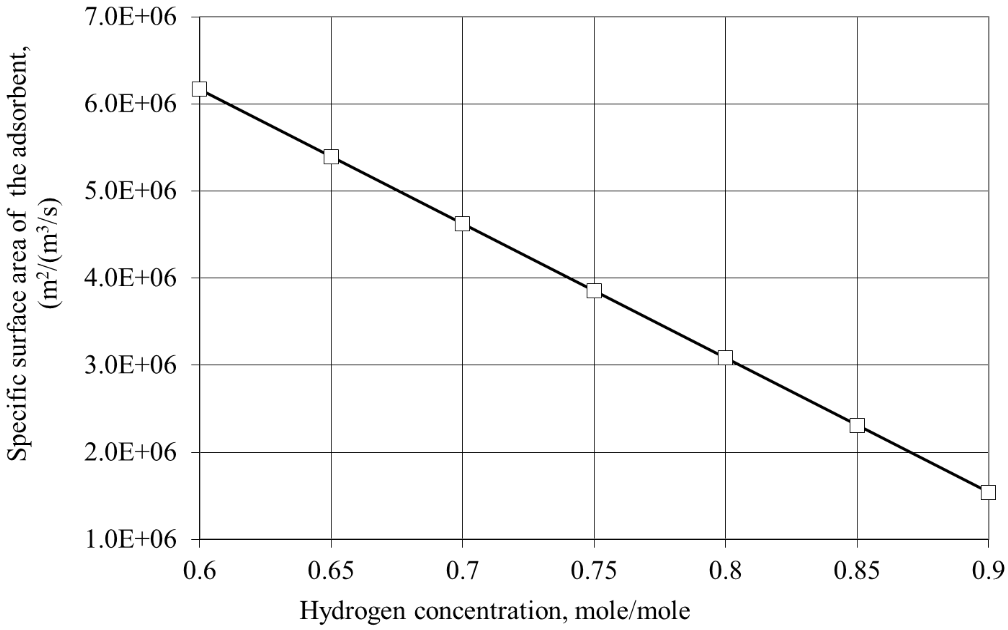
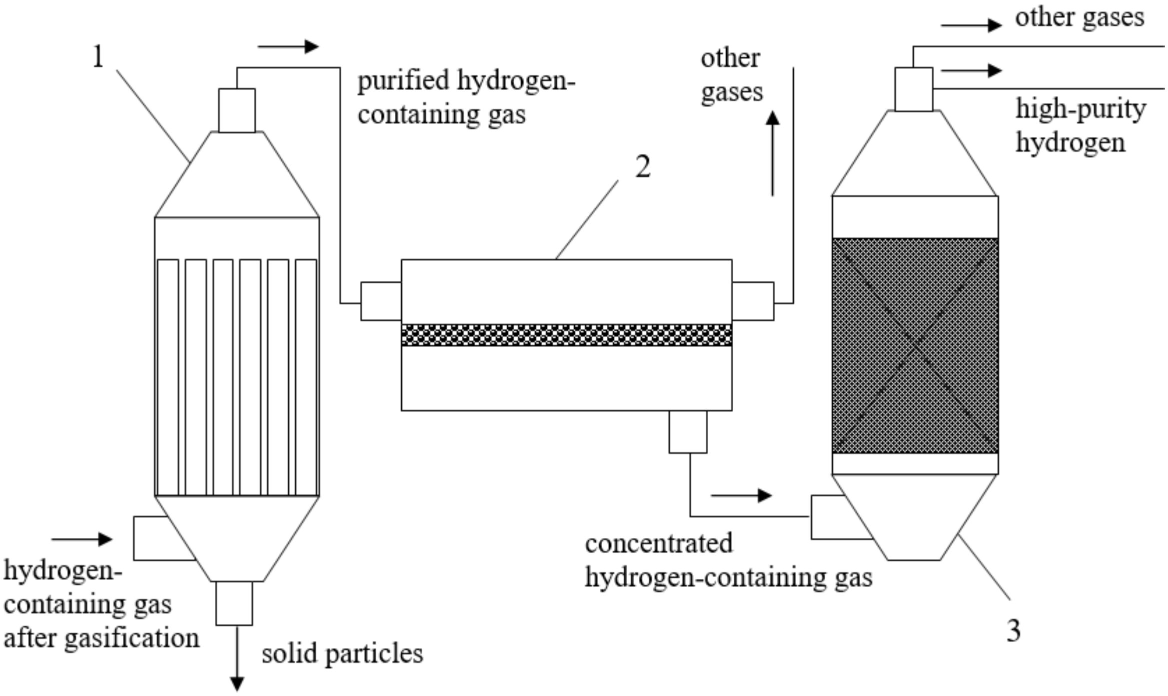


Figure 4 – Dependence of the specific surface area of zeolite 13X on the hydrogen concentration in the initial gas mixture

The analysis of the results shows that an increase in the hydrogen concentration in the initial gas mixture from 60% to 90% leads to a decrease in the specific surface area of zeolite 13X from $6.17 \cdot 10^6$ $m^2/(m^3 \cdot s)$ to $1.54 \cdot 10^6$ $m^2/(m^3 \cdot s)$, which indicates a decrease in the amount of absorbed carbon monoxide with a decrease in its initial concentration.

Based on the literature data and the obtained dependencies, a schematic diagram of purification and separation of hydrogen-containing gases to obtain high-purity hydrogen up to 99.99% was developed, which is shown in Fig. 5. According to this scheme, the hydrogen-containing gas obtained during the gasification of the SCCM is going to purification from solid particles to the filtration system 1, from where it is fed to the membrane separation 2 to concentrate it from 60% to 90%, then the concentrated hydrogen-containing gas is going to the adsorption separation 3, where it is separated to produce high-purity hydrogen with a concentration of up to 99.99%.



1 – filtration system; 2 – membrane separation; 3 – adsorption separation

Figure 5 – Schematic diagram of purification and separation of hydrogen-containing gases to produce high-purity hydrogen

5. Conclusions

1. Analytical dependencies for calculating the specific surface area of the filter partition, the specific surface area of the membrane, and the specific surface area of the adsorbent were obtained, which take into account the technological parameters of hydrogen-containing gases and the technological parameters of their separation to produce high-purity hydrogen.

2. A graphical dependence of the specific surface area of the filter partition on the temperature of hydrogen-containing gases formed during the gasification of solid carbon-containing media and the concentration of solid particles in the gas was obtained. The graphical dependence of the specific surface area of the polyimide

membrane on the operating pressure and hydrogen concentration in the permeate and the graphical dependence of the specific surface area of zeolite 13X on the initial hydrogen concentration in the initial gas mixture from 60% to 90% at a final concentration of 99.99% are presented.

3. A schematic diagram of purification and separation of hydrogen-containing gases to produce high-purity hydrogen with a concentration of up to 99.99% is presented. The diagram includes a solid particles purification system, a system for preconcentrating hydrogen-containing gas to 60–90% by membrane separation, and a system for producing high-purity hydrogen by adsorption separation.

Conflict of interest

Authors state no conflict of interest.

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ТЕХНОЛОГІЧНІ АСПЕКТИ ОЧИЩЕННЯ ТА РОЗДІЛЕННЯ ВОДНЕВМІСНИХ ГАЗІВ, ОТРИМАНИХ ПРИ ГАЗИФІКАЦІЇ ТВЕРДИХ ВУГЛЕЦЕВМІСНИХ СЕРЕДОВИЩ

Шевченко В., Опарін С., Давидов С., Гупало О.

Анотація. В наведеній роботі предметом дослідження є процеси очищення та розділення водневмісних газів, отриманих при плазмохімічній газифікації твердих вуглецевмісних середовищ. Мета роботи полягає у встановленні закономірностей впливу технологічних параметрів водневмісних газів, отриманих при газифікації твердих вуглецевмісних середовищ, на питомі поверхні фільтрувальних перегородок, мембран та адсорбентів, що використовуються для процесів їх очищення та розділення. В роботі використаний графоаналітичний метод дослідження, який полягає у пошуку раціональних питомих поверхонь процесів очистки та розділення водневмісних газів, основуючись на отриманих графічних залежностях.

Встановлено аналітичні залежності з розрахунку питомої площі поверхні фільтрувальної перегородки, питомої поверхні мембрани та питомої поверхні адсорбенту, які враховують технологічні параметри водневмісних газів та технологічні параметри їх розділення з отриманням водню високої чистоти.

На основі теоретичних результатів розрахунку отримано графічну залежність питомої площі поверхні фільтрувальної перегородки від температури водневмісних газів, утворених при газифікації твердих вуглецевмісних середовищ, та концентрації твердих часток в газі. Дана залежність показує, що збільшення концентрації твердих часток та зростання температури водневмісних газів призводить до підвищення питомої площі поверхні фільтрувальної перегородки. Наведено графічну залежність питомої поверхні мембрани, виготовленої з полііміду, від робочого тиску та концентрації водню в пермеаті та графічну залежність питомої поверхні цеоліту 13X від початкової концентрації водню в вихідній газовій суміші від 60% до 90% при кінцевій концентрації 99,99%.

Наведено принципову схему очищення та розділення водневмісних газів з отриманням водню високої чистоти, згідно якої водневмісний газ, отриманий при газифікації вуглецевмісних середовищ, направляється на очищення від твердих часток в систему фільтрації, звідки подається на мембранне розділення з метою його концентрування, а сконцентрований водневмісний газ направляється на адсорбційне розділення, де відбувається його поділ з отриманням водню високої чистоти.

Ключові слова: вуглецевмісне середовище, водневмісний газ, водень, фільтрування, мембранне розділення, адсорбція.