INVESTIGATION OF SOME FACTORS INFLUENCE ON THE WORK OF EXTRA PURE HYDROGEN GENERATOR

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The developed models of ultra-pure hydrogen generators had been designed, produced and tested. The influence of membrane temperature and form, position in the flame, installation of additional screens etc. were investigated. The special device, so called hydrogen comprемator (accumulator-compressor), was designed, produced and investigated. Its function is to accumulate low pressure pure hydrogen flow (pressure is less than 1 Torr) from generator and to convert it to high hydrogen pressure of about 15 atm in the comprеmator volume. The combined work of hydrogen generator and comprеmators was investigated. Hydrogen generator and comprеmator were tested in the regime of pure hydrogen puffing in plasma device DSM-1, providing the required parameters during experiment on the examine of metal erosion. The examines had shown capacity for work of both hydrogen generator and comprеmator in the conditions of real plasma physical experiments.

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INTRODUCTION

Earlier the method and laboratory model of the ultra-pure hydrogen generator were suggested for hydrogen production, when ultra-pure hydrogen (purity is better than 99.999 vol. %) is generated in only one technological process [1, 2]. On the realization of this technology the process of pure hydrogen production could be carried out at the same time with hydrocarbon materials utilization (combustion), e.g., during heating of water. The process of hydrogen generation is realized with help of diffusion-catalytic membrane which is placed in the flame of combustion and separates the volume of hydrocarbon combustion from the volume for pure hydrogen accumulation. Only hydrogen can pass through the diffusion-catalytic palladium (its alloys) and nickel membranes due to its high hydrogen diffusivity in these metals, providing ultra high purity of more than 99.999 vol. % [3].

The proposed method could be used in hydrogen energy, chemical, electronics and fuel cells industries, electro-chemical generators, gas chromatography, in proton accelerators, under control thermonuclear fusion problem solution in many plasma devices, etc. But to realize this it was needed to investigate in detail the influence of some factors, such as membrane form and position in the flame, installation of additional screens, to develop construction of diffusion-catalytic membrane module and hydrogen generator, to solve the problem of accumulation and compression of low pressure pure hydrogen flow from generator, etc.

1. EXPERIMENTAL AND RESULTS

The developed construction of membrane module is presented in Fig. 1. It includes Pd-membrane of 6x0.25x190 mm dimensions. Ni-tubes with the same dimensions were also used. Unlike the earlier used membrane module the side of the membrane tube plunged into the flame of hydrocarbon combustion is hermetically argon welded and valve has been installed between the other side of membrane tube and the volume of hydrogen flow measurement.

Fig. 1. Developed diffusion-catalytic membrane module

This membrane module had shown high reliability and maintainability. Then on its base two models of ultra-pure hydrogen generators HG-1 (hydrogen productivity is about 3…10 l/hour with the use of Pd-membrane) and HG-2 (hydrogen productivity is about 7…15 l/hour using two Pd membranes) had been designed, produced and investigated.

The ultra-pure hydrogen generator HG-1 is presented in Fig. 2.

Fig. 2. General view of hydrogen generator HG-1

The generator diffusion module was made on the base of WhisperLite Universal Stove model #06630 (USA) and includes one cylindrical or semi-tore Pd diffusion membrane (we will get productivity about 1 l/hour, if Ni membrane to use instead of Pd). The generator is connected to butane/propane balloon. The generator construction allows gasoline using, too. Dimensions of generator are 400x400x250 mm. Thermal power – 3…3.6 kW, in dependence on fuel consumption

(0.2…0.25 l/h). In parallel with hydrogen generation water heating could be performed. The measurements of hydrogen flow through Pd and Ni membranes were carried out with the method of constant pressure, similar as described earlier [4]. Productivity in our case means hydrogen amount generated per time unit. We used such units of productivity – normal cm^3 of hydrogen per second (Ncm^3/s) or normal liters of hydrogen per hour (l/hour). This value is equal to hydrogen flow rate Q Ncm^3/s from diffusion-catalytic membrane. If to divide Q on the area of membrane surface it will be the specific hydrogen flow $Ncm^3/s.cm^2$ or specific productivity q. The scheme of the hydrogen productivity measurement is shown in Fig. 3.

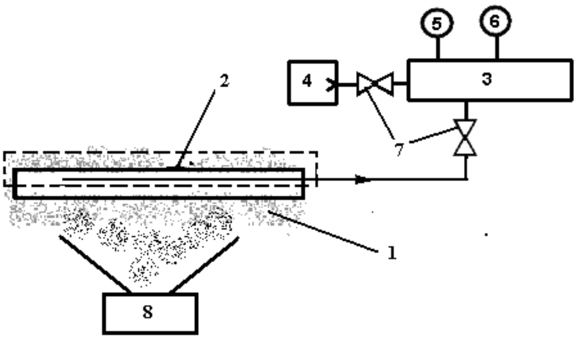

Fig. 3. Scheme of productivity measurements: 1 – flame; 2 – diffusion-catalytic membrane; 3 – vacuum chamber; 4 – pump; 5 – vacuum-sensing device; 6 – mass-spectrometer; 7 – valves; 8 – gas/gasoline stove

Under generator work, first, the membrane 2 internal volume, the measurement vacuum chamber 3, hydrogen conductors and valves were pumped to a pressure of about $5x10^{-6}$ Torr. This pressure is measured by vacuum-sensing device 5 and it is the initial pressure P_0. Then the gas/gasoline stove 8 is ignited and the palladium tube 2 is heated to 700…800ºC. The temperature to which the palladium tube 2 is heated depends on the location of the palladium tube 2 within the influence of the flame or on the flame intensity (Fig. 4, green rhomb points). The temperature was controlled by a chromel-Copel thermocouple. Hydrogen, formed on the Pd-membrane surface faced to the flame, diffuses according to Fick's law through membrane and desorbs to the internal volume of membrane. As soon as hydrogen desorbs, the pressure P in the vacuum chamber 3 increases. When the temperature of the palladium tube 2 is 800ºC, the measured pressure in vacuum chamber 3 is 0.15 Torr. The hydrogen flow rate to vacuum chamber $Q = 1.3 (P-P_0)\cdot S$, where P_0 (Torr) is an initial pressure, P(Torr) – measured a final pressure, S (l/s) is the pumping speed. In our case S= 5 l/s so hydrogen flow $Q \approx 1\ N\cdot cm^3/s$ (or ~3.6 l/hour) passes from the membrane to the vacuum volume 3.

Then the hot (uncooled) screen made of flat stainless steel (Fig. 5, and dotted line in Fig. 3) was installed over the membrane. At the palladium tube 2 temperature of 800 degrees C, the measured hydrogen flow to vacuum chamber 3 was ≈5 l/h instead of about 3.5 l/h without screen (see Fig. 4). So, the result of additional screen installation is the essential enhance of device productivity. If cold screen (volume with water) was installed the hydrogen productivity was the same as for hot screen. So, the thermal energy generated by flame can be used not only for membrane heating but for parallel technological process, i.e., for water heating or boiling. Note, that the presence of the diffusion-catalytic membrane in the flame did not noticeably influence on the water heating (boiling) parameters. Time required for heating up to 100ºC increased on less than 10 %.

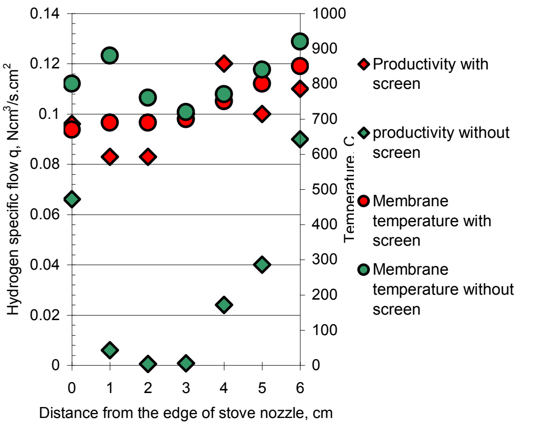

Fig. 4. Specific hydrogen productivity vs membrane position in the flame

Fig. 5. Working hydrogen generator HG-1 with hot screen

Two forms (shape) of screens were investigated: flat (plane) 20x3 cm dimensions and semi-cylindrical with radius R = 1.2 cm and length l=20 cm. For both the effect of productivity increasing was observed (Fig. 6) and it is the same for both screens.

The influence of form of the diffusion-catalytic membranes on hydrogen productivity (hydrogen flow) was investigated for semi-tore and cylindrical membranes with the same dimensions. Note, in first case the circular disk type stainless steel screen was used, including the variant when screen was the bottom of volume for water heating/boiling. The large tore radius R has to be less than flame dimension. And the screen diameter has to be rather more than large tore radius. In the second case the flat right-angled stainless steel screen was used as in Fig. 5 is shown.

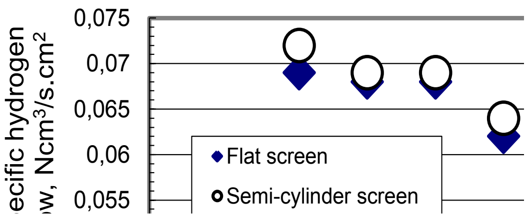

Fig. 6. The influence of screen shape on hydrogen productivity

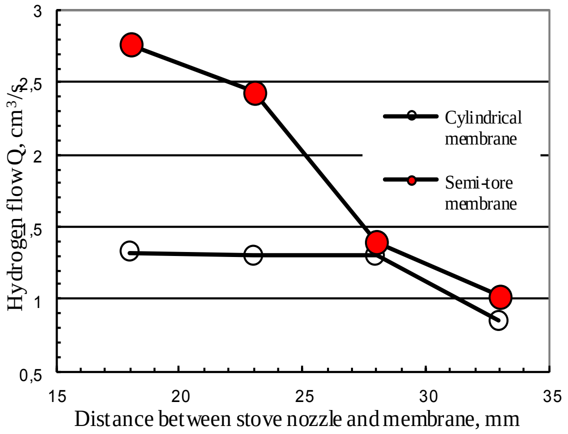

Fig. 7. Hydrogen flow dependence on the distance of Pd diffusion-catalytic membrane from the stove nozzle

The tests have shown that hydrogen productivity of semi-tore type membrane is in about half as much again more than that for usual cylindrical membrane. It is seen in Fig. 7 this effect is observed on the optimal distance 18…23 mm between membrane and flame source. For longer distances productivity is the same for both membrane forms. Note, that for long length membranes it is possible to make membrane in form of spiral of Archimedes or part of such spiral.

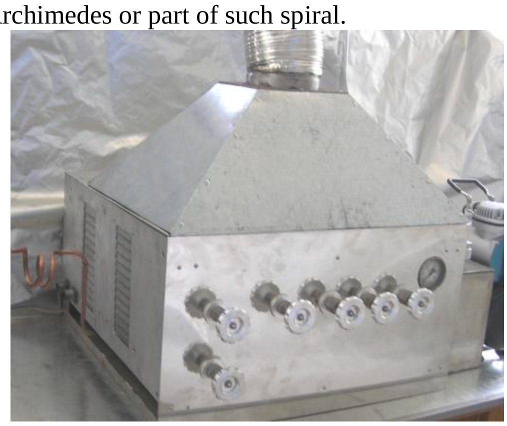

Fig. 8. General view of hydrogen generator HG-2

The pure hydrogen generator HG-2 with productivity 7…15 l/hour had been created (Fig. 8). The generator was made on the base of WhisperLite Universal Stove, too, and includes two semi-tore Pd diffusion membranes (Ni membranes were used, too.). The generator was connected to butane/propane balloon or to gasoline source. Dimensions of generator are 550x550x300 mm. Thermal power is 3…3.6 kW, similar as for HG-1 generator. The generator is completed with two hydrogenompremators (compressor-accumulator). This special device has been created and investigated to convert the resulting low pressure (less than 1 Torr) hydrogen flow from generator to high pressure (up to 15 atm.) This allows to use this system for hydrogen feeding through the autonomic system inlet.

The hydrogen compremator includes high pressure box with two connections for pumping and getter inputting, electric heater and shield (Fig. 9). The installed special filter prevents hit of powder into pumping system. Hydrogen compressor dimensions are: diameter 80 mm, length 350 mm. The internal volume of compremator chamber allows to place there about 1000 g of Zr-Ni getter.

Fig. 9. Compremator chamber with heater and shield

Batch of Zr-Ni powder was produced in the IPTTMT NSC KIPT. It was previously tested in the special stand GAS [5] to determine the main characteristics: hydrogen sorption rate at low pressures, capacity, optimal sorption/desorption temperatures, etc. The average size of powder particles was 0.2…0.8 mm. About 500 g of Zr-Ni getter had been placed into compremator to provide hydrogen sorption. After getter activation procedure and saturation with hydrogen flow from generator during 25 hours at the room temperature, such compremator provided about 5 atm pressure after hydrogen desorption at the temperature of 500ºC. This allows use pure hydrogen generator for hydrogen puffing in numerous vacuum devices with autonomic systems for hydrogen inlet, high hydrogen pressure is required in which.

The heater capacity is 1.5 kW. The heater provides the compremator heating to temperature of 700…750ºC during 7…10 min (with shield). Power consumption in the regime of hydrogen accumulation is 0 W, and it is about 1…1.5 kW in the regime of hydrogen desorption.

The joint operation of hydrogen generator with compremator (hydrogen compressor-accumulator) was investigated. The combined system (hydrogen generator and compressor) works in the following way. At first, valves V_1-V_6 (Fig. 10) are opened and system is pumped to pressure of about $5\cdot10^{-6}$ Torr. Then valves V_1-V_2 are shut off and the flame of gas/gasoline combustion is ignited. Hydrogen diffuses through membranes and is puffed into vacuum system to measure hydrogen generation productivity (rate of hydrogen flow through membrane). Then valves V_2-V_6 are shut off and valve V_1 is opened. Hydrogen is trapped by getter in compremator 4. This process is carried out at the room temperature of compremator. After full getter saturation valve V_1 is shut off. The gas/gasoline stove is switched off to cut off the hydrogen flow through membranes. Then the compremator heater switched on to heat getter to the temperature of 450…500°C. Due to hydrogen desorption process the pressure in compressor increases up to 5…15 atm. (in dependence of saturation time, temperature, getter properties, etc.). In our experiment the times of saturation were 25/250 hours and maximum pressures were 5/15 atm.

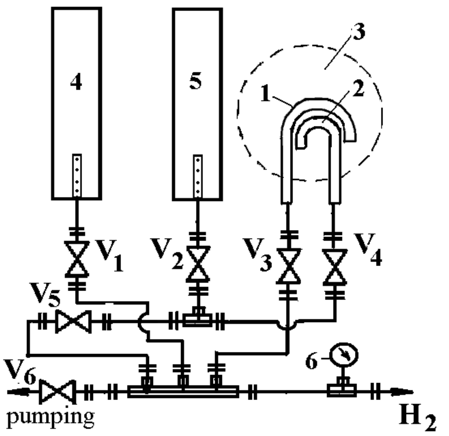

Fig. 10. The scheme of the hydrogen generator HG-2: 1, 2 – Pd membranes; 3 – gas/gasoline stove; 4, 5 – compremators; 6 – manometer; V_1 and V_2 – valves for compremators; V_3 – change over valve; V_4 and V_5 – valves for diffusion membranes; V_6 – valve for pumping

Various work regimes of the generator are possible: the direct generated hydrogen puffing into vacuum systems (one or two working membranes), regimes of hydrogen accumulation in compremators (one or two compremator is working) and high pressure hydrogen using, demounting of one compremators and its using without of generator, etc.

It is known, that pure hydrogen is widely used for research experiments in the frame of the program of thermonuclear fusion devices creation: erosion behavior of materials, hydrogen trapping and desorption phenomena, etc. In the Institute of Plasma Physics of the NSC KIPT (Kharkov, Ukraine) the investigations of the erosion behavior of various materials used in thermonuclear plasma devices were carried out early and are carried out now on the DSM-1 device [5]. Two regimes are usually used: plasmas of mirror Penning discharges and magnetron type discharges regime.

To test the work of hydrogen generator and compremator in the real experimental conditions they were installed in the DSM-1 device instead of hydrogen balloon. In this work the regime of mirror Penning discharges was used. Steady state mirror discharges were ignited in magnetic field of 0.05 T under work gas (it was hydrogen from compremator or generator) pressure about 0.2 Pa. The discharge voltage was 1 keV, discharge current change in the range of 2 mA. Hydrogen flow in the DSM's vacuum chamber was provided by puffing of desorbed hydrogen from compremator during its heating up to 500ºC at the working forevacuum pump (5 l/s) and turbo molecular pump (500 l/s). Such regime was supported during 20 minutes that was enough to measure the erosion rate of material.

In the next experiment hydrogen generator was used for hydrogen puffing in DSM-1 device. The initial pressure in the DSM vacuum chamber was $5x10^{-6}$ Torr and it is provided by working forevacuum and turbo molecular pumps. After switch on the hydrogen generator the work pressure of $1…2x10^{-3}$ Torr was obtained in the DSM chamber and it was supported during the experiment (~20 min). The estimated hydrogen productivity of the generator was 1…2 N· cm^3/s (3.5…7 N·l/hour) in dependence on the flame intensity. The plasma parameters in this experiment was the similar to above mentioned for experiment with hydrogen compremator.

So, the examines had shown capacity of both hydrogen generator and compremator for work in the conditions of real physical experiments.

2. DISCUSSION

Note, that the effect of enhancing of pure hydrogen generation productivity due to screen installation above the diffusion-catalytic membrane was not known and evidence before the beginning experiments with screen. Its initial function was to protect membrane from mechanical impacts to prevent its damage because the membrane material has no sufficient mechanical strength. As the screen is made of higher mechanical strength and heatproof material than membrane tube material, the probability to fracture thin membrane is essentially decreased and device reliability increases. On the other hand such screen could, e.g., to decrease the amount of generated hydrogen (productivity) due to partial isolation of diffusion-catalytic membrane from the flame flows. There are not exact information on the real reasons of such increasing of productivity. The process of pure hydrogen production from the flame is very complete and dependents on many factors, such as temperature, work gas concentration (pressure), physical-chemical reaction rates, membrane placing in the flame, form and structure of flame, etc. Such possible simple mechanism can be suggested to explain this effect: The flame intensity could be different below the membrane and above the membrane. On the top of membrane tube flame intensity could be lower and it could lead to decreasing of hydrogen flow through this part of membrane. Note, that at the same time the membrane temperature can be similar for both places due to high metal thermal conductivity. In Fig. 4 you can see that the membrane temperature with screen even some lower than for membrane without screen. The screen installation above the membrane could increase of flame flows to top of membrane. Moreover, due to screen the flame intensity could be more uniform along the membrane. If that's the case, the concentration of work gases near the membrane surface increases and hydrogen flow (productivity) through membrane also increases. Of course, forming of flame flows to top of membrane and along tube could be dependent on the screen form and its distance from membrane.

Some words on the screen dimensions. The exact radius of semi-cylindrical screen is not important, but it have to be not rather small in order to prevent screening of tube surface from the flame. In our case it is enough screen radius about two tube diameters (about 12 mm). But, e.g., if we will use the diffusion-catalytic membrane which will be consisting of more than one tube, the variant of large flat screen could be more preferable.

So for numerous modifications of hydrogen generator the screen shape or dimensions or distances from stove could be different. The optimal configuration provides from the one hand the protection of membrane and on the other hand, as had shown our experiments the essential increase of hydrogen productivity.

CONCLUSIONS

Two models of pure hydrogen generators HG-1 (hydrogen productivity is about 5…10 l/hour with the use of Pd-membrane) and HG-2 (hydrogen productivity is about 7…15 l/hour with two Pd membranes) had been designed, produced and were tested. The optimal forms of diffusion catalytic membranes were choused. The installation of the additional screen above the diffusion-catalytic membrane leads to essential increasing of hydrogen productivity up to two times. It could be explained as the result of the flame intensity increase on the membrane top part and membrane temperature stabilization.

Hydrogen compremator (accumulator-compressor) was designed, produced and tested. It is the device with the function to accumulate low pressure pure hydrogen flow (pressure is about 0.5 Torr and lower) generated in generator and then to obtain in compremator volume high hydrogen pressure of about 15 atm. Generator HG-2 had been completed with two such compremators. The combined work of hydrogen generator and compremator was investigated. Both HG-1 and compremator was tested in the regime of the work on plasma device DSM-1, providing the required parameters. The examine had shown capacity for work of both hydrogen generator and compremator in the conditions of real physical experiments.

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ИССЛЕДОВАНИЕ ВЛИЯНИЯ РЯДА ФАКТОРОВ НА РАБОТУ ГЕНЕРАТОРА ОСОБО ЧИСТОГО ВОДОРОДА

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Разработаны, изготовлены и испытаны усовершенствованные модели генераторов особо чистого водорода. Изучено влияние на производительность генераторов таких факторов, как температура и форма диффузионно-каталитических мембран, их положение в пламени, установка дополнительных экранов и др. Специальное устройство, так называемый компрематор (аккумулятор-компрессор водорода), был изготовлен и испытан. Его функцией является накопление водорода низкого давления (ниже 1 Торр), поступающего из генератора, и конвертация его в водород высокого давления (около 15 атм) в объёме компрематора. Исследована совместная работа генератора водорода и компрематора. Генератор и компрематор испытаны в режиме напуска чистого водорода в плазменную установку ДСМ-1, обеспечивая необходимые параметры во время экспериментов по изучению эрозии металлов. Испытания показали хорошую работоспособность генератора и компрематора в условиях реального физического эксперимента.

ДОСЛІДЖЕННЯ ВПЛИВУ РЯДУ ЧИННИКІВ НА РОБОТУ ГЕНЕРАТОРА ОСОБЛИВО ЧИСТОГО ВОДНЮ

Г.П. Глазунов, О.Л. Конотопський, Д.М. Виноградов, М.М. Бондаренко, С.М. Мазніченко, О.П. Свинаренко

Розроблено, виготовлено і випробувано вдосконалені моделі генераторів особливо чистого водню. Вивчено вплив на продуктивність генераторів таких чинників, як температура і форма дифузійно-каталітичних мембран, їх положення в полум'ї, установка додаткових екранів і ін. Спеціальний пристрій, так званий компрематор (акумулятор-компресор водню), був виготовлений і випробуваний. Його функцією є накопичення водню низького тиску (нижче 1 Торр), що поступає з генератора, і конвертація його у водень високого тиску (близько 15 атм) в об'ємі компрематора. Досліджена спільна робота генератора водню і компрематора. Генератор і компрематор випробувані в режимі напуску чистого водню в плазмову установку ДСМ-1, забезпечуючи необхідні параметри під час експериментів з вивчення ерозії металів. Випробування показали хорошу працездатність генератора і компрематора в умовах реального фізичного експерименту.