LOW-TEMPERATURE CATALYTIC HYDROGEN-TO-WATER CONVERTER WITH DIRECT CONTACT OF THE REACTIONARY MIX AND THE COOLANT

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Introduction

At many plants of nuclear power industry there is a specific problem of safe conversion of hydrogen into water without any leak of the combustion product (water) into the environment. Such a problem arises every time when hydrogen contains appreciable amount of deuterium and, especially, radioactive isotope of hydrogen, tritium. In the first case the question is a loss of the expensive product (heavy water), in the second case it is protection of the environment from tritium radiation. Thus the quantities of hydrogen streams being converted depend on a specific aim and change from single burning of several cubic meters of hydrogen up to long-term (year and more) continuous conversion of hydrogen stream achieving hundred of cubic meters per hour. The last problem concerns the plants of hydrogen isotopes separation by a method of chemical isotope exchange between hydrogen and water. It is supposed, in particular, that such plants will be used with a view of isotope clearing heavy-water coolant of Canadian power reactors CANDU [1]. The problems like ones mentioned above arise at many Russian plants as well.

Conversion of hydrogen into water can be carried out by different methods. Those are the simple flame burning of hydrogen in the atmosphere of oxygen or air, the oxidation on copper oxide, the catalytic oxidation on various catalysts of molecular hydrogen activation. Each of those methods has the lacks: either a low level of the safety or loss of the conversion product or complication of the plant flow sheet related to the need of water catching.

In the present work it is reported about the development of the catalytic hydrogen converter where the hydrophobic catalyst allowing to carry out the oxidation reaction in the device with the direct contact of reactionary mix and cooling water is used. For the first time the idea of an opportunity of creating such a device has been stated in [2]. In the present work this idea has been advanced, modified and finished with concrete devices of different productivity [3].

Results and Discussion

The layout of the experiment and principle of the device operating is explained on the Fig. 1.

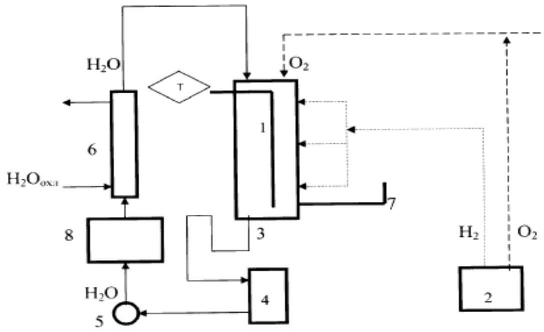

Fig. 1. The layout of the converter testing experiment.

Hydrogen is going to the reactor 1 from the electrolyzer 2. Water formed after hydrogen oxidation is flowing down into the reservoir 4 through hydraulic lock 3 and then moving into the reactor by the pump 5 preliminary being passed through the heat-exchanger 6 cooled by external water. The pipe 7 is intended for dumping of superfluous gases. Stoichiometric amount of oxygen to the device is supplied with the same electrolyzer 2 and superfluous one from an external source. In a series of experiments the thermostat 8 is used for heating of water circulated.

The catalytic converter is the cylindrical vessel made of stainless steel and supplied with three inputs of hydrogen distributed uniformly along the reactor. Inputs of hydrogen inside the device are ending with the rings made of metal tube centered along the reactor axis and provided with the apertures directed downwards. The reactor is filled with a mix of the catalysts RCTU-3SM prepared according to [4] and metal spiral-prismatic packing, volumetric ratio between them was varied. Along an axis of the reactor there is a pocket for the thermocouple that allows measuring the temperature in any cross-section of the reactor during its operating by moving the thermocouple. In the top cover of the device there are two pipes for input of oxygen and cooling water, and in its bottom part there are pipes for output of cooling water and not reacted gas.

During systematic researches of the pilot converter (internal diameter = 62 mm, height = =800 mm) designed for productivity G_{H2} = 0.30 m^3/h of H_2 (normal conditions here and further) per hour as a part of the plant shown in the Fig. 1 all parameters necessary for steady operating of the converter have been determined and optimized:

- size of the catalyst RCTU-3SM granules,
- a volumetric ratio between catalyst and hydrophilic packing in the converter,
- distribution of the catalyst density trough the length of the converter,
- flow rate of water circulating through the converter and the temperatures at the input and output of the converter.

As an example the parameters of operating the converter in one of modes are resulted in the Table 1: volumetric ratio between the catalyst and packing (stainless steel spiral-prismatic packing with the size of an element 2x2x0.2 mm) was 1:10, the size of catalyst granules was 0.8-1.0 mm, a flow rate of water and its temperature at the input of the converter were 22.5 l/h and T_1=25^{0}C correspondingly.

Table 1. Operating parameters of the pilot converter

№	1	2	3	4	6	7
G_{O2}, l/h	65	90	115	140	170	160
G_{H2}, l/h	50	100	150	200	300	300
T_2*, ^{0}C	27	36	41	50	64	64

*-Temperature of water at the output of the converter.

Let us note that at all operating modes the concentration of hydrogen at the output of superfluous oxygen from the converter (pipe 7 in the Fig.) did not exceed 4ppm, i.e. the achievable degree of its conversion into water was close to 100%. Besides that, in a mode of № 6 the long experiments have been carried out to determine the time resource of the catalyst. For this purpose its catalytic activity has been measured by an independent method (rate of the isotope exchange reaction between hydrogen and water vapor at given temperature) before loading it into the converter and after its work during 100 hours. Measurements have shown that activity of the catalyst has remained constant.

The results obtained have allowed creating the device of a larger productivity. Its sizes have made: internal diameter = 150 mm, height = 800 mm. It is loaded with the catalyst RCTU-3SM with the size of a granule 0.4-0.6 mm and packing with the size of an element 3x3x0.2 mm at their volumetric ratio 1:10 (a gradient of the catalyst loading along the height of the converter has been applied). The converter has been tested at flow rates of hydrogen up to 3.6 m^3/h. The results are listed in the Table 2.

Table 2. Results of testing the enlarged converter.

№	A flow rate of circulating water, l/h	G_{H2}, m^3/ч	G_{O2}, m^3/ч	T_1, ^{0}C	T_2, ^{0}C
1	240	0.56	0.40	53	56
2	240	1.12	0.66	55	60
3	240	1.72	0.92	56	61
4	290	1.72	0.92	55	61
5	330	2.30	1.43	48	56
6	330	3.00	2.00	48	59
7	350	3.60	2.40	55	67

At all operating modes of the converter the concentration of hydrogen in waste gas also was lower than 4ppm. We shall notice, however, that when operating at the last mode the periodic clicks were audible inside the converter that testifies that this mode is close to limiting for the converter of given sizes. We shall notice, that the increase in productivity of the device due to increase of its dimensions will demand the additional researches related to creation of an optimum temperatures profiles in a cross-section and height of the converter.

Thus, the work done has allowed creating the low-temperature catalytic converter of hydrogen with productivity not less than 3 m^3/h.

In summary we shall note that (if necessary) there is a principal opportunity of creating a source of low-potential heat (hot water with temperature up to 85-90 ^{0}C) on the basis of the converter developed.

References

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3. Decision about the patent application of the Russian Federation №2008135535 from 01.09.2008

4. The patent of the Russian Federation №2307708 from 31.01.2006.