Low-temperature catalytic hydrogen recombiner based on hydrophobic catalysts

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Abstract

Currently, it is extremely promising to electricity generation increase by nuclear power plants. However, during its operation, any nuclear facility, including nuclear power plants, is a source of radioactive contamination, including the radioactive isotope of hydrogen - tritium. The solution of the technological flows detritization problem of nuclear industry enterprises is carried out by the most effective methods for the hydrogen isotopes separation. Due to the progress in the development of hydrophobic catalysts at the end of the 20th century, the chemical separation isotope exchange in the water-hydrogen system is considered the most effective separation method, characterized by high separation coefficients. In world practice, this process has found practical application in the water-hydrogen system, combined with the electrolyze (Catalytic exchange combine electrolyze – CECE-process).

The CECE-process is used for tritium isotopic purification from heavy and light water, and the processing of heavy water waste. To realize all the advantages of the CECE process, a device for the safe oxidation of hydrogen, which will ensure a complete transfer of the hydrogen stream to the liquid water stream, is required as the top node for the reversal of flows. The optimal solution is a low-temperature catalytic recombiner based on a hydrophobic catalyst for the hydrogen oxidation. The current lack of the necessary hydrophobic heat-resistant catalyst in Russia, and, consequently, the safe and reliable device, largely limits and complicates the use of the CECE process.

From all of the above, it follows that the creation of a reliable and highly efficient hydrogen oxidation reactor based on a domestic hydrophobic catalyst will significantly improve and simplify the top-node detritization plants and will ensure the introduction of advanced technologies in the field of hydrogen isotope separation.

The present **research work main goal** was the development of a Russian hydrophobic catalyst with higher heat-resistivity for low-temperature hydrogen combustion in stoichiometric (upper-) mixture with oxygen in recombiner using as the central element top flow reversal node for chemical hydrogen isotope separation during CECE-process.

Main **tasks** were:

1. Determination of the main laws, operational characteristics and operating parameters of a low-temperature catalytic converter using the domestic catalyst based on polymer carrier brand PXTY-3CM as the comparison sample.

2. Formulation of requirements for the basic properties of a hydrogen oxidation catalyst used in a low temperature recombiner.

3. Development of a method for preparing a hydrophobic heat-resistant platinum catalyst for the oxidation of hydrogen based on γ-Al_2O_3, modified to give its surface hydrophobic properties.

4. Establishment of the relationship between the synthesis conditions, including the modification method and type of modifier, and the characteristics of the prepared catalysts.

5. Testing the prepared catalyst in the process of low-temperature oxidation of hydrogen in a recombiner to determine the correlations between the properties of the catalyst and the efficiency of the oxidation process.

6. Development of a technological scheme, installation, and commissioning of a low-temperature converter for burning hydrogen in a stoichiometric mixture with oxygen based on a

hydrophobic catalyst as a part of the upper unit for flow reversal for hydrogen isotope separation units in a water-hydrogen system.

Scientific novelty:

1. An original methodology has been developed for the preparation of a new effective platinum hydrophobic heat-resistant catalyst for the hydrogen oxidation based on an inorganic carrier.

2. The optimal conditions for the synthesis of a hydrophobic platinum catalyst for the hydrogen oxidation with an active metal concentration of 0.2-0.5 wt. % based on the inorganic carrier γ-Al_2O_3 with a heat resistance of at least 623 K.

3. Correlations between the synthesis conditions, the type and concentration of the modifier, and the main physicochemical and catalytic properties of the synthesized inorganic-based hydrogen oxidation catalyst were established including determining the optimal synthesis parameters.

4. An original methodology has been developed for conducting accelerated “start-stop” stress testing of a hydrogen oxidation catalyst in a stoichiometric mixture with oxygen in low-temperature recombiner to determine the stability of its characteristics.

The practical significance of the work

1. A hydrophobic inorganic-based Pt/Al_2O_3 catalyst (modified) was developed with increased heat resistance of at least 623 K for the process of stoichiometric hydrogen oxidation in a low-temperature recombiner (operating temperature of the oxidation process is not more than 353 K) with direct contact of the coolant and catalyst.

2. The basic laws governing the operation of a low-temperature catalytic converter based on hydrophobic hydrogen oxidation catalysts (polymer Russian brand catalyst and the developed inorganic catalyst of the Pt/Al_2O_3-type (modified) in a wide range of hydrogen concentration in an oxygen (air) stream are determined.

3. A method of accelerated “start-stop” stress testing of a hydrogen oxidation catalyst in a stoichiometric mixture with oxygen is proposed to determine the stability of its characteristics during operation in laboratory conditions and relatively short times.

4. The developed catalyst was tested in a low-temperature converter as a WUOP of a hydrogen isotope separation unit.

Defense Provisions

1. The method of the catalyst inorganic carrier modification for the hydrogen oxidation in order to give its surface hydrophobic properties, characterized by a contact angle of 110-150^{O}.

2. The results of physico-chemical studies of samples of a hydrophobic catalyst for the hydrogen oxidation on an inorganic basis with increased heat resistance, including the main correlations between the composition of the samples and their properties.

3. The results of kinetic experiments and the calculation of the oxidation reaction parameters of low hydrogen concentrations in the air stream at temperature range 293-363 K, as well as their dependence on the samples composition.

4. The study results of the process of low-temperature catalytic hydrogen oxidation in a stoichiometric mixture with oxygen in a recombiner based on the developed catalyst, including an accelerated start-stop stress test of the catalyst properties stability and the launch of the top flow reversal unit as part of a hydrogen isotope separation facility in the “water-hydrogen” system.