Study on Hydrogen Removal Efficiency of Hydrogen Recombiners in Nuclear Power Plant Radioactive Waste Gas Treatment System

Jiayi Lin[1], Xiaojie Zhang[1], Tongwei Feng[1], Baihua Jiang[1,*]
[1]China Nuclear Power Engineering Co., Ltd., Beijing 100840, China
Corresponding author: Baihua Jiang (E-mail: BaihuaJcnpe@163.com)

ABSTRACT Aiming at the hydrogen removal efficiency issue of hydrogen-oxygen recombiners in the radioactive waste gas treatment system of nuclear power plants, this study conducted screening of three platinum-based catalysts and performed experiments on the effects of different process conditions on hydrogen removal performance. The results showed that catalyst 1# exhibited good catalytic activity and stability during continuous operation. After irradiation and the introduction of impurity iodine, the minimum full conversion temperature increased by 2–5 °C, and the activity recovery rate after heating and reduction regeneration reached over 90%. Under reaction conditions at 50 °C, 100 °C, and 150 °C, the limiting space velocity could reach 8000 h^{-1}, 52000 h^{-1}, and 96000 h^{-1}, respectively. The comprehensive screening identified a Pt composite catalyst, which demonstrated excellent low-temperature ignition activity at room temperature and could resist the impact of impurities and water vapor in the gas on the catalytic effect. For the inlet gas hydrogen concentration in the range of 1% to 3%, the outlet hydrogen concentration could be below 3000 ppm under experimental conditions, meeting the requirements for long-term operation of the waste gas treatment system in nuclear power plants.

INDEX TERMS hydrogen recombination; radioactive exhaust gas; hydrogen removal; nuclear power plant

I. INTRODUCTION

During the operation and maintenance of a nuclear power plant, the waste gas treatment system is a critical line of defense for controlling the release of radioactive materials into the environment. This system primarily collects and treats radioactive hydrogen-containing exhaust gases discharged from the Reactor Coolant System (RCS), the reactor coolant storage tank, and various other storage tanks. The typical characteristics of these exhaust gases include a certain concentration of hydrogen (usually derived from the radiolysis of water), a small amount of radioactive nuclides (mainly inert gases krypton and xenon, as well as trace amounts of iodine), and water vapor. Although hydrogen is not a radioactive substance, it is highly chemically active. If directly discharged or accidentally accumulated within the treatment system, it can easily trigger an explosion when encountering an open flame or static spark. This not only risks damaging the integrity of the waste gas treatment system but may also lead to a large-scale release of radioactive materials, causing a serious environmental incident.

To fundamentally eliminate explosive components from exhaust gases, hydrogen recombiners are used as hydrogen-removal equipment in nuclear power plant exhaust gas treatment process systems. The principle involves using supported noble metal catalysts (such as platinum/palladium on alumina) under specific temperature conditions to promote a flameless catalytic reaction between hydrogen and added oxygen, generating high-temperature water vapor [1]. After condensation and water removal, the remaining radioactive gases enter an activated carbon retention decay bed to achieve compliant emissions. Therefore, the operational stability and recombination efficiency of the hydrogen recombiner not only directly determine the safety of the exhaust gas treatment system, but also indirectly affect the load level of subsequent radioactive gas treatment units and the final environmental emissions.

However, the operating conditions of hydrogen-oxygen recombiners in the context of nuclear power plant flue gas treatment are relatively complex and show significant differences compared to general industrial hydrogen removal equipment [2]. On one hand, the hydrogen concentration in the flue gas to be treated fluctuates over a wide range, potentially varying from trace hydrogen levels during normal unit operation to hydrogen-rich conditions (>90%) before reactor shutdown or during degassing operations [3]. This

imposes strict requirements on the catalyst's ignition characteristics and dynamic response capability across a broad concentration range. On the other hand, the small amount of radioactive iodine carried in the flue gas may lead to catalyst poisoning, surface coverage, or sintering deactivation over long-term operation, thereby reducing the reaction activity and even triggering the risk of "hydrogen breakthrough." In addition, the coupled effects among system operating pressure, gas space velocity, preheating temperature, and water vapor content directly influence the heat and mass transfer efficiency of the recombiner bed. If heat removal is unbalanced, it may cause catalyst structural damage or system over-temperature interlock shutdown.

Based on current research, the performance evaluation of nuclear-grade hydrogen-oxygen recombiners is mostly focused on the verification of macroscopic efficiency under standard operating conditions. There is still a lack of systematic and in-depth experimental data supporting the understanding of microscopic deactivation mechanisms and dynamic response characteristics under the synergistic effects of varying concentration fluctuations, interference from impurity components, and long-term operational aging [4]–[6]. In particular, the validation of the applicability of domestically produced catalysts in real simulated exhaust gas environments has become a technical bottleneck that restricts the autonomous design of the equipment and its reliable long-term operation.

Based on this, this paper conducts bench-scale experimental research on hydrogen-oxygen recombiners based on the actual process requirements of a nuclear power waste gas treatment system. By building a simulation test loop, the study focuses on investigating the effects of stepwise changes in inlet hydrogen concentration, the introduction of impurity components, and different space velocity conditions on the ignition characteristics, conversion efficiency, and bed temperature distribution of the recombiner, aiming to reveal the disturbance mechanisms of characteristic components in nuclear power waste gas on the catalytic recombination process. The research results are expected to provide theoretical basis and experimental references for the engineering design optimization, operation procedure formulation, and catalyst life assessment of nuclear-grade hydrogen-oxygen recombiners.

II. EXPERIMENT SETUP AND METHODS

A. TEST SYSTEM

The process flow of the hydrogen-oxygen recombination reaction test system is shown in Figure 1. The system is mainly divided into three parts, including the gas supply unit, the gas preheating and catalytic recombination unit, and the gas concentration measurement and regulation unit.

(1) Air supply unit

The raw gas, consisting of nitrogen, hydrogen, air, and water vapor mixed in a certain ratio, is formulated into a raw gas with the required hydrogen and oxygen concentrations, primarily using nitrogen as the medium. The raw gas passes through a gas dryer to maintain a certain level of moisture in the gas.

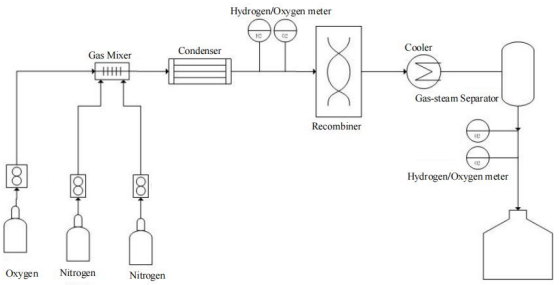


Fig. 1. Hydrogen-Oxygen Reformation Reaction Experimental System

(2) Gas preheating and catalytic composite unit

Hydrogen and oxygen in the raw gas undergo a chemical reaction under the action of a catalyst to produce water. The recombiner is equipped with a heating device, which accelerates the start-up speed of the hydrogen-oxygen recombination reaction by heating the catalyst, while reducing the condensation of water vapor on the catalyst surface. A thermometer is set to measure the temperatures of the inlet and outlet gases of the recombiner as well as the catalyst. The gas after the recombination reaction is discharged after cooling and dehumidification.

(3) Hydrogen-oxygen concentration measurement loop and hydrogen-oxygen concentration control device

The hydrogen and oxygen concentrations are monitored upstream of the combustor and at the exhaust emission outlet to evaluate the combustion efficiency. The oxygen injection amount can be automatically adjusted based on the hydrogen and oxygen concentrations upstream of the hydrogen-oxygen combustion device and the hydrogen-to-oxygen stoichiometric ratio setting, to ensure complete gas reaction.

B. EXPERIMENTAL PLAN

1) CATALYST OPTIMIZATION TEST

The catalytic activity, radiation resistance, iodine poisoning resistance, and regenerability of three platinum-based catalysts are evaluated using a catalyst reactor. The catalytic activity of the hydrogen recombiner is expressed by the lowest total conversion temperature (TLc) parameter; the lower the TLc, the better the activity. TLc is defined as the minimum reaction temperature required to reduce the residual hydrogen concentration in the exhaust gas to approximately 50 ppm [7]–[9].

The quartz sand buffer is filled at both ends of the catalyst reactor, and the catalyst is filled in the central isothermal zone, as shown in Figure 3. The reaction tube specification is 9×1.5 mm, and the length of the isothermal zone of the reactor is a$\geq$75 mm.

(1) Catalytic activity

First, nitrogen gas is introduced into the reactor at a space velocity of 10000 h^{-1} $\pm$ 100 h^{-1}. When the temperature is increased to 180°C at a rate of 2°C/min, the feed gas is

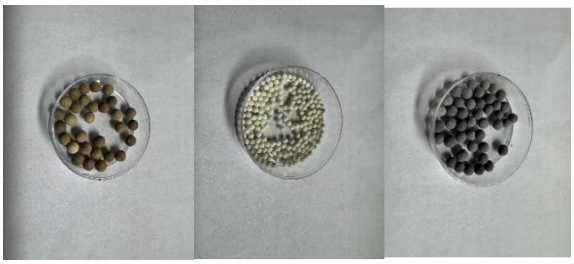


Fig. 2. Three platinum-based catalysts

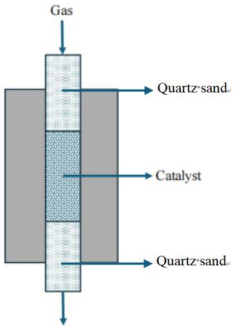


Fig. 3. Schematic diagram of the catalyst reactor

switched, with a hydrogen-to-oxygen ratio of 2:1, and the space velocity is maintained at 10000 h^{-1} ± 100 h^{-1} for 2 hours. Then, the temperature is gradually decreased in steps of 5°C. After each set temperature is reached, it is stabilized for 0.5 hours, and the hydrogen volume fraction at the outlet is analyzed using a trace hydrogen detector every 10 minutes. When the hydrogen volume fraction at the outlet exceeds 50 ppm for three consecutive measurements, the corresponding temperature is recorded. Starting from this temperature, the temperature is slowly increased in 1°C increments, and when the hydrogen volume fraction at the outlet drops to approximately 50 ppm, this temperature is defined as the minimum full conversion temperature. By determining the minimum temperature required for complete hydrogen conversion, the activity of various candidate catalysts is compared [10].

(2) Stability

Based on the lowest complete conversion temperature as the initial condition, three catalysts were operated continuously for 10 hours, and the hydrogen content in the exhaust gas was analyzed to evaluate their long-term operational stability.

(3) Radiation resistance

The TLc changes before and after irradiation (1000 Gy) were measured for the three catalysts separately [11].

(4) Iodine poisoning resistance

HI with a concentration of 100 ppm is introduced into the feed gas, and the change in TLc before and after the introduction of the iodine-containing gas is measured [12], [13].

(5) Renewability

The catalyst was soaked in water for 0.5 hours, followed by baking for 2 hours in an air atmosphere at 100°C. The TLc changes before and after the water soaking and drying treatment were tested [14], [15].

(6) Limit airspeed

The space velocity value at which the hydrogen concentration in the exhaust gas reaches 1000 ppm within a temperature range of 50°C-150°C.

Fig. 4. The test rig of the catalyst reactor

2) EFFECT OF PROCESS CONDITIONS ON THE RECOMBINATION OF HYDROGEN AND OXYGEN

Using the test rig, process condition verification tests were conducted on the selected catalyst, with a gas flow rate of 100 Nm^3/h and a space velocity controlled at 12000 h^{-1}, to investigate the effects of initial reaction temperature and humidity on the hydrogen-oxygen recombination reaction.

III. CATALYST EXPERIMENTAL RESULTS AND DISCUSSION

A. CATALYTIC ACTIVITY

The specific surface area of the three samples was analyzed using the Quantachrome Autosorb IQ3 fully automatic specific surface and porosity analyzer, with a test temperature of 77.35 K and nitrogen as the adsorbate. The analysis results showed that the specific surface areas of samples 1#, 2#, and 3# were 193.947, 93.887, and 175.219 m^2/g, respectively. Sample 1# has a larger specific surface area compared to the other two samples.

Figure 6 shows the test process data of the three catalysts, with the TLc values of samples 1#, 2#, and 3# being 65°C, 81°C, and 74°C, respectively. Sample 1# has a lower TLC value compared to the other two samples, indicating that it possesses the highest catalytic activity.Taken together, the lower TLc and larger specific surface area of catalyst 1# indicate that its accessible catalytic surface can sustain hydrogen oxidation with lower thermal input. The outlet hydrogen concentration decreased continuously as the temperature approached the complete-conversion threshold rather than changing irregularly, which also suggests a stable transition from kinetically limited conversion to near-complete reaction. Consequently, catalyst selection should not rely on surface area alone; the TLc response provides the more direct criterion because it reflects the combined

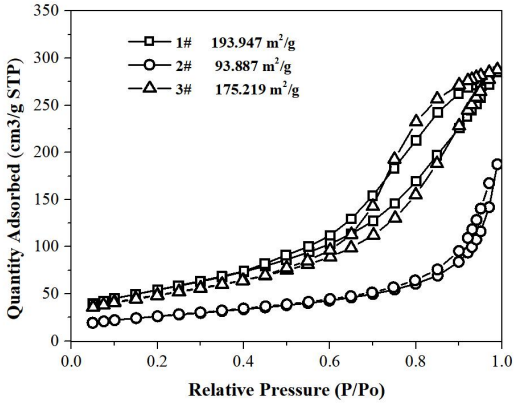


Fig. 5. Three sample specific surface area test curves

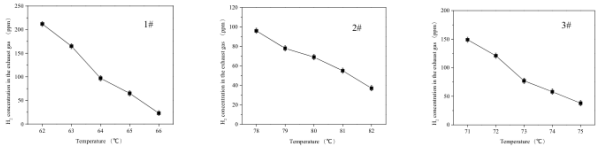


Fig. 6. Results of Catalytic Activity Test

effects of active-site availability, gas diffusion, and intrinsic reaction activity under the same feed conditions.

B. STABILITY

The punctual compressive strength of three samples was analyzed using a particle strength tester. The analysis results showed that the punctual compressive strengths of samples 1#, 2#, and 3# were 150, 27, and 71 N, respectively. Sample 1# exhibited a significantly higher punctual compressive strength compared to the other two samples.

Figure 7 shows the continuous operation data of the three catalysts. The analysis results indicate that the hydrogen content of catalysts 1# and 3# shows a decreasing trend during the last five hours of testing, while catalyst 2# exhibits greater fluctuations.

The stability results further distinguish catalyst 1# from the other candidates. Its high mechanical strength reduces the likelihood of particle fracture, fines generation, and local channeling during prolonged gas circulation. At the same time, the gradual decrease in outlet hydrogen concentration during the latter stage of the test indicates that catalytic performance did not deteriorate with operating time. In contrast, the larger fluctuation observed for catalyst 2# implies a less stable reaction state. These characteristics are important for maintaining a uniform pressure drop and predictable conversion efficiency in long-term waste-gas treatment.

C. RADIATION RESISTANCE

Figure 8 shows the results of the full conversion temperature measurements for the three catalysts after irradiation. The

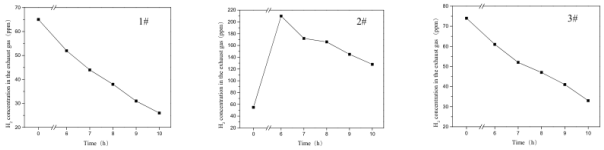


Fig. 7. Results of Catalytic Activity Stability Test

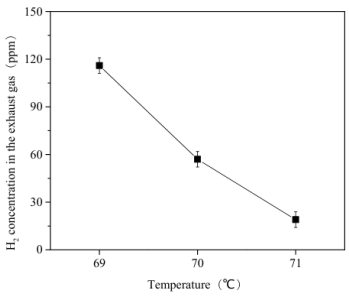


Fig. 8. Performance Test of Catalyst 1 against Iodine Poisoning

TLc values of catalysts 1#, 2#, and 3# after irradiation are 67°C, 86°C, and 77°C, respectively. Catalyst 1# exhibits the smallest change in TLc before and after irradiation compared to catalysts 2# and 3#, demonstrating the strongest irradiation resistance.The limited increase in TLc after irradiation shows that the catalytic surface of catalyst 1# retained most of its low-temperature activity after receiving the specified dose. Because the three samples were tested under the same irradiation and reaction conditions, the smaller temperature shift provides a direct comparative measure of radiation tolerance. This behavior supports the use of catalyst 1# in radioactive exhaust environments where accumulated radiation exposure may gradually alter the support structure or the state of the noble-metal active phase.

D. IODINE POISONING RESISTANCE

An anti-iodine poisoning performance test was conducted on catalyst No. 1. The test results showed that after introducing HI, the TLc of catalyst No. 1 increased from 65°C, to 70°C, indicating that iodine impurities in the gas have a certain impact on the reaction performance [16]. However, since the gas temperature generally rises above this temperature after the exothermic reaction, the hydrogen consumption requirements can still be met.The 5°C increase in TLc indicates that iodine-containing species inhibited part of the available catalytic surface but did not cause complete loss of activity. The remaining conversion capability at 70°C is particularly relevant because the recombination reaction is exothermic and the bed temperature rises after ignition. Therefore, once the reaction has been initiated, the operating temperature can compensate for the moderate activity loss caused by the tested iodine concentration. Nevertheless, the observed shift also confirms that iodine exposure should be considered in catalyst aging assessment and maintenance planning.

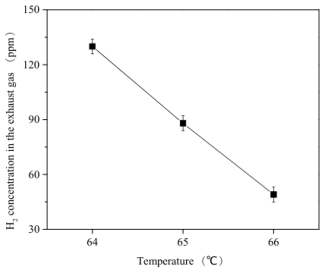


Fig. 9. Regenerability Test of Catalyst 1

E. RENEWABLE PERFORMANCE

As shown in Figure 9, the TLc of catalyst 1# after water soaking and drying treatment is 66°C, indicating strong regenerability.The TLc after regeneration was only slightly higher than the initial value, showing that the activity loss associated with water soaking was largely reversible under the applied drying treatment. This result suggests that temporary moisture coverage did not permanently destroy the active sites or the catalyst support. From an operational perspective, controlled heating and drying can therefore serve as a practical recovery measure after water ingress or shutdown conditions, provided that the catalyst is not subjected to prolonged liquid-water accumulation.

F. LIMIT AIRSPEED

The experimental results show that the 1# catalyst achieves maximum space velocity values of 8000 h^{-1}, 52000 h^{-1}, and 96000 h^{-1} at reaction temperatures of 50 °C, 100°C, and 150°C , respectively, as shown in Figure 10.The strong increase in allowable space velocity with reaction temperature reflects the combined influence of reaction kinetics and gas-solid contact. At 50°C, the lower reaction rate requires a longer residence time to maintain the outlet hydrogen concentration within the specified limit. As the temperature rises, hydrogen and oxygen are consumed more rapidly, allowing the same catalyst volume to process a substantially larger gas flow. The results therefore define an operational relationship between bed temperature and treatment capacity: low-temperature start-up requires conservative flow control, whereas stable high-temperature operation provides a greater throughput margin without changing the catalyst loading.

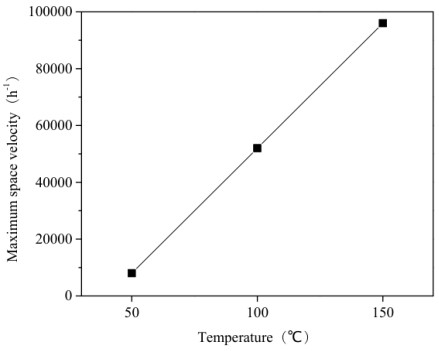
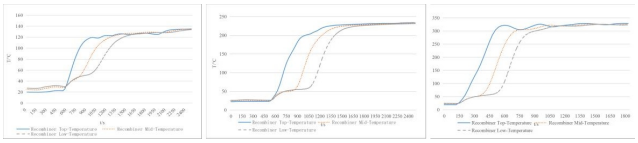


Fig. 10. Regenerability Test of Catalyst 1



(a) (b) (c)

Fig. 11. Effect of Initial Reaction Temperature on Dehydrogenation Performance

IV. HYDROGEN RECOMBINERS EXPERIMENTAL RESULTS AND DISCUSSION

The temperature values for the initial reaction temperature exploration experiments are all based on room temperature. Without activating the heater on the test rig, hydrogen and oxygen were introduced to observe whether the temperature of the catalyst inside the hydrogen-oxygen recombiner could increase. Exploration experiments were conducted with hydrogen concentrations of 1%, 2%, and 3%. It was determined that, at room temperature (20-25°C), hydrogen-oxygen recombination reactions can occur under different hydrogen concentrations [17], [18].

Starting from an ambient temperature of 20°C, hydrogen and oxygen are introduced to carry out a hydrogen-oxygen recombination reaction. Initially, the temperature at the lower part of the hydrogen-oxygen recombiner rises more rapidly, indicating that at low temperatures, hydrogen and oxygen require more contact time with the catalyst to react. As the catalyst temperature increases, hydrogen and oxygen preferentially react with the catalyst at the upper part, causing the upper catalyst temperature to rise more rapidly [19], [20]. Ultimately, the gas carries heat, and the temperatures of the catalyst in the upper, middle, and lower sections tend to become uniform.

For initial hydrogen concentrations with volume concentrations of 1%, 2%, and 3%, the temperature rise of the catalyst reaches 210°C, 230°C, and 310°C, respectively, when the complete recombination reaction finally stabilizes.

(a) 1% hydrogen concentration
(b) 2% hydrogen concentration
(c) 3% hydrogen concentration

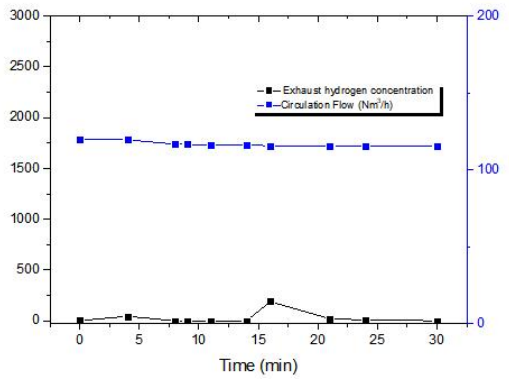


Fig. 12. Effect of Circulation flow on Hydrogen Removal Efficiency

A. EFFECT OF CIRCULATION FLOW ON HYDROGEN REMOVAL EFFICIENCY

Experiments with hydrogen concentrations of 1% and 3% were conducted at a circulation flow rate of 120 Nm^3/h. Increasing the circulation flow rate essentially increases the volume space velocity of the catalyst reaction, from 12000 h^{-1} to 14400 h^{-1} [21]. During the operation of the test rig, the temperature of the gas passing through the fan gradually increases, and under the condition of maximum fan power output, the flow rate exhibits certain fluctuations.

As can be seen from Figure 12, when the hydrogen concentration is 1%, there is no significant correlation between the operating flow rate (100-120 Nm^3/h) and the hydrogen concentration in the exhaust gas, meaning that the operating flow rate has no effect on the hydrogen-oxygen recombination efficiency.

B. EFFECT OF HUMIDITY ON HYDROGEN REMOVAL EFFICIENCY

The hydrogen-oxygen recombination reaction will produce water. Although heat exchangers and traps are installed in the system, a small amount of water vapor still enters the intake through circulation [22]. As the operating time increases, the humidity in the reaction gas will gradually increase. Under conditions with sufficient oxygen and when the temperature of the hydrogen-oxygen recombination reaction exceeds 100 °C, water vapor will not remain on the surface of the catalyst. The hydrogen concentration in the exhaust gas after hydrogen-oxygen recombination is less than 3000 ppm, and is basically maintained below 1000 ppm. Therefore, humidity has no significant effect on the efficiency of hydrogen-oxygen recombination.

C. EFFECT OF HIGH PRESSURE ON HYDROGEN REMOVAL EFFICIENCY

Exploration tests on the hydrogen-oxygen composite efficiency were carried out under a high pressure of 300 kPa for different hydrogen concentrations (1%, 2%, and 3%).

When the operating pressure is 300 kPa, the hydrogen concentration is 1%, the hydrogen-to-oxygen ratio is less than 2, and the catalyst temperature is above 110°C, the hydrogen concentration in the tail gas is basically close to 0, indicating that under the condition of ensuring sufficient oxygen and a certain temperature, the hydrogen-oxygen composite efficiency is close to 100%. When the hydrogen concentration is 2% and the hydrogen-to-oxygen ratio is less than 2, the hydrogen concentration in the tail gas is basically close to 0, indicating that the hydrogen-oxygen recombination efficiency is close to 100% when the oxygen supply is sufficient. When the hydrogen concentration is 3%, due to the high hydrogen concentration, the heat released by the reaction causes the catalyst temperature to approach 300°C. When both the temperature and the hydrogen-to-oxygen ratio meet the requirements, the hydrogen-oxygen recombination efficiency is close to 100%.

Therefore, under conditions of sufficient oxygen, with different hydrogen concentrations (1%, 2%, 3%), the hydrogen content in the exhaust gas remained below 3000 ppm, and even reached 0 ppm. The results are close to those obtained at a low pressure of 20 kPa.g, indicating that the operating pressure has no significant effect on the efficiency of hydrogen-oxygen recombination [22].

In summary, after a period of stable operation of this experiment, the hydrogen concentration was controlled at 1.5–2%, and the oxygen concentration at 2–3%. The system operated continuously for 2 hours, during which the hydrogen concentration in the exhaust gas remained below 3000 ppm, indicating a good hydrogen-oxygen recombination efficiency.

V. CONCLUSIONS

(1) After comparing the catalytic activity, continuous operation stability, and radiation resistance performance of the three candidate catalysts, catalyst No. 1 was selected as the optimal choice. By evaluating the iodine poisoning resistance and regenerability of the selected catalyst No. 1, it was proven that catalyst No. 1 can be applied in the bench test of the hydrogen-oxygen recombiner.

(2) The selected catalyst has good activity and can initiate the hydrogen-oxygen recombiner reaction at 20°C, allowing the electrical heater upstream of the recombiner to be optimized and potentially eliminated in the engineering design.

(3) The test showed the operating flow rate has no effect on the hydrogen-oxygen recombination efficiency. The other hands, the hydrogen-oxygen recombination reaction process produces water vapor, increasing the humidity within the system. Since the catalyst surface temperature exceeds 100°C during the reaction, the water vapor does not remain on the catalyst surface. Therefore, the effect of humidity on hydrogen-oxygen recombination efficiency during operation can be neglected. However, the impact of residual water vapor in the equipment on the catalyst after the reaction must be considered.

(4) Under the operating pressure of 300 kPa.g, with a hydrogen concentration of 1.5~2% and air as the oxidant, the equipment operated stably for 2 hours, and the hydrogen concentration in the exhaust gas remained below

3000 ppm throughout, indicating good hydrogen-oxygen recombination efficiency and stable catalyst performance; at the same time, during the 2-hour operation, the catalyst temperature remained around 200°C due to the heat released from the hydrogen-oxygen recombination reaction, while the hydrogen-oxygen recombination efficiency did not change, indicating good thermal stability of the catalyst under normal operating conditions.

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