

Treatment of Tritium in Off-gas by Catalytic Oxidation

Sung Paal Yim, Kwang-Rag Kim, Minsoo Lee, Seungwoo Paek, Hongsuk Chung, Do-Hee Ahn
Korea Atomic Energy Research Institute, 150 Deokjin-dong, Yuseong-gu, Daejeon, 305-353, Korea,
nspyim@kaeri.re.kr

1. Introduction

The gaseous tritium (HT, DT or T₂) is contained in off-gas from research reactors, nuclear power plants that use heavy water and tritium-handling facilities. This off-gas has been usually released to the environment without removal of tritium because the tritium concentration is low enough to satisfy the permissible release concentration specified in regulation. However, as the regulation tends to be strengthened for increasing the safety more and as the generation of off-gas containing relatively high concentration of tritium is also expected due to the expansion of the use of tritium in nuclear industry, it is recently being recognized that the treatment of off-gas containing tritium is important [1].

The tritium in off-gas is usually treated by two steps. It is first oxidized to tritiated water vapor by a catalyst and then the vapor is adsorbed in a molecular sieve drier. Conventionally the oxidation is carried out by using catalyst in a temperature range of 150~200°C, which is high to adsorb directly the tritiated water vapor in molecular sieve [2, 3]. And other approaches have been employed to oxidize the tritium in off-gas at ambient temperature [4].

In the experiments, as a preliminary study of the treatment of tritium in off-gas, the oxidation of hydrogen in air is investigated at room temperature by using two kinds of catalysts which are a precise metal catalyst supported on polymer and another precise metal catalyst supported on alumina.

2. Experiments

2.1. Catalyst

As a precise metal catalyst supported on polymer, the material with 1 wt.% of platinum impregnated on SDBC (Styrene-Divinybenzene Copolymer) was fabricated in the shape of about 4 mm pellet for the experiment. And as a precise metal catalyst supported alumina, a palladium catalyst with the palladium content of 0.5 wt% and the shape of 3.2 mm pellet was purchased from Aldrich Chemical and used after purging with dry nitrogen for 24 hrs.

2.2. Equipment and Procedure

The equipment was designed to observe the oxidation of hydrogen in air by catalyst. As is shown in Figure 1, it consists of compressed gas cylinders, mass flow controllers, a mixer, a catalytic reactor and a

hydrogen analyzer. The air stream with constant hydrogen concentration and constant flow rate is formed by the mass flow controllers and the mixer. It is introduced to the catalytic reactor packed with catalyst. Hydrogen in air is oxidized to form water vapor by catalyst through the reactor. The conversion ratio of hydrogen to water can be determined by measuring the hydrogen concentrations at the inlet and outlet of the reactor, respectively.

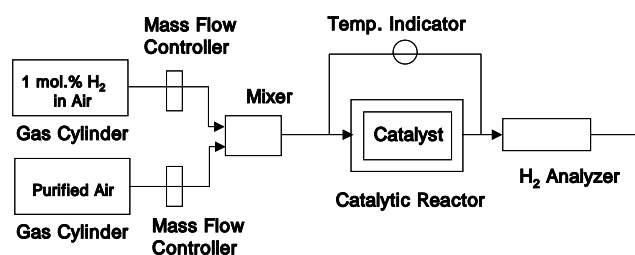


Figure 1. Flow diagram of experimental equipment system.

The air with the hydrogen concentration of 0.1 mol% was made at room temperature ($20 \pm 3^\circ\text{C}$) and introduced to the reactor. The reactor was also operated at room temperature. The flow rate was controlled in 24 L/hr. Two reactors which have the dimension of 3.8 cm in diameter by 50 cm height and 2.5 cm in diameter by 20 cm height were respectively used in the experiment. The hydrogen concentration of air was measured by means of a hydrogen analyzer (Fuji Electric Instrument Co., Ltd) with thermal conductivity detector. The quantity of catalysts packed in reactor was changed in the range of 30~90g.

3. Result and Discussion

In the catalytic reactor packed with the platinum catalysts supported on SDBC (1wt% Pt/SDBC catalyst), the change of the conversion ratio with time is shown in Figure 2. The conversion ratio is represented as follows.

$$f = 1 - C / C_0 \quad (1)$$

where f is the conversion ratio, C is the hydrogen concentration at the outlet and C_0 is the hydrogen concentration at the inlet. In the figure, it is shown that the conversion ratio decreases rapidly with time at the initial stage and it approaches constant value for later time. The conversion ratio after the time has elapsed enough depends on the quantity of catalyst in reactor and it increases with the quantity of catalyst. The

conversion ratio is about 0.9 when the 90g of catalyst was packed in the reactor

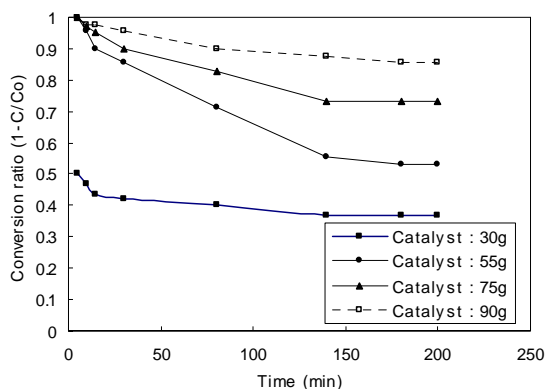


Figure 2. Changes of conversion ratio of 1 wt% Pt/SDBC catalyst.

Although no generally accepted mechanism on the reaction between hydrogen and oxygen on a metal surface exists in spite of extensive studies, based on the assumption of a pseudo-homogeneous first-order reaction, the apparent kinetic rate constant could be obtained by plotting the experimental data according to the following equation [5]:

$$-Q \ln(1 - f) = \frac{1}{k} \cdot V \quad (2)$$

where k [min^{-1}] is the apparent kinetic rate constant, Q [L/min] is the flow rate of the gas, and V [L] is the volume of the catalyst.

The plot to obtain the apparent kinetic rate constant of the 1 wt% Pt/SDBC catalyst is shown in Figure 3. The apparent kinetic rate constant of 1 wt% Pt/SDBC catalyst is 0.68 min^{-1} from the plot.

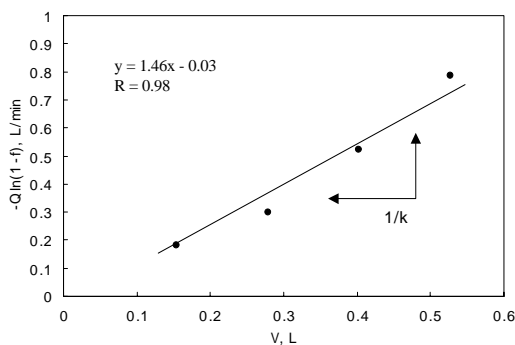


Figure 3. Plot of the experimental data to obtain the apparent kinetic rate constant of 1 wt% Pt/SDBC catalyst

For the palladium catalysts supported on alumina (0.5 wt% Pd/Alumina catalyst), the changes of the conversion ratio with time are shown in Figure 4. The conversion ratio reaches 0.9 when 80g of catalyst was packed in the reactor. It is similar to that of 1 wt% Pt/SDBC catalyst. However, the apparent kinetic rate

constant could not be obtained yet due to the lack of experimental data. The additional experiments are currently being performed to get the rate constant of 1 wt% Pt/SDBC catalyst.

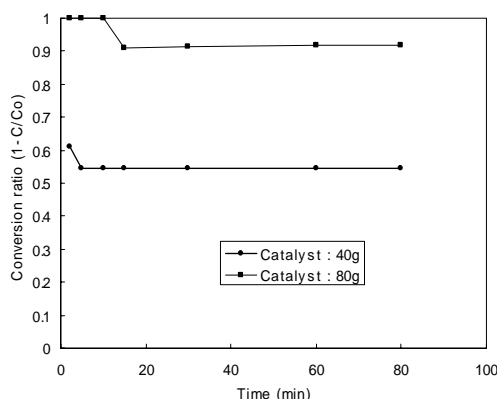


Figure 4. Changes of conversion ratio of 0.5 wt% Pd/Alumina catalyst.

4. Summary

In this study, it was confirmed that 1 wt% Pt/SDBC catalyst and 0.5 wt% Pd/Alumina have the capability to oxidize hydrogen to water vapor at room temperature. Based on the results, at present, the effects of the concentration of moisture and hydrogen in air and the flow rate to reactor are being investigated on the catalytic oxidation in more detail.

Acknowledgement

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