§12. Hydrogen and Methane Oxidation Performance of Honeycomb Hybrid Catalyst for a Tritium Removal System

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Necessity of honeycomb catalysts
It had been examined about the performance of oxidizing catalysts for hydrogen and methane containing gases considering tritium contaminated exhaust air treatment system. The conventional tritium removal technique is oxidation of tritiated hydrogen and hydrocarbon to water by packed type catalytic oxidation reactors, followed by adsorption process on a molecular sieve bed. The usual catalysts tested were ceramic particles on whose microscopic surfaces precious metals like platinum (Pt) or palladium (Pd) were adsorbed. If the tritium removal system is applied for safety issue of a tritium treatment in a large nuclear fusion facility, the processing gas volume and throughput will be large with increasing flow rate of processed gas, and the packed bed of oxidation reactor is considered to have a large pressure drop. In order to reduce pressure drop in the oxidation reactor and mechanical load applied on a blower, we have proposed to apply for a honeycomb type catalyst, which exhibits a low pressure drop, made of a metal or cordierite ceramic. This is because the honeycomb type has large open space. These honeycomb type catalysts impregnated with platinum or palladium had an adequate performance to oxide hydrogen and methane in air.

Property of Platinum and Palladium catalysts
In our previous study it was found that a platinum (Pt) catalyst exhibited higher oxidation reaction rate of hydrogen than that of a palladium (Pd) one, to the other hand a Pd catalyst exhibited higher oxidation of methane than that of a Pt catalyst. Therefore, to make use of each characteristic, we tried to impregnate with both Pt and Pd on a honeycomb type porous ceramics of which composition is CaO-Al$_2$O$_3$, SiO$_2$, and TiO$_2$.

Experimental methods
The shape of a honeycomb ceramics used was 20 mm in a diameter and 50 mm in a length and the cell density was 200 cell/inch$^2$. The catalysts were experimentally made by impregnating mixed solution of Pt and Pd or impregnating a solution of Pt after impregnating a solution of Pd. The ratio of each catalyst was consisted of 1:1 and total amount of precious metals was 2 g/L. The evaluations of oxidation performance were carried out by using a flow-type reactor. The catalytic reactor column was heated up stepwisely to 400 $^\circ$C, and each appropriate temperature it was hold about 1 hour. The feed processing gas was a mixture of H$_2$, CH$_4$, O$_2$ and N$_2$ in the ratio of 0.1:0.1:20:8.79 (vol%).

Experimental results
Experimental results for hydrogen gas oxidation are shown in Fig. 1. In the figure the oxidizing rate coefficients of simple 2 g/L of the Pt catalyst and the Pd catalyst are compared. The catalysts of Pt 1 g/L and Pd 1 g/L impregnating with mixed solution or individual solution, and the catalyst impregnating simple 2 g/L Pt solution show almost the same reaction rates comparing the Pd catalyst. Also the experimental results for methane gas oxidation are shown in Fig.2. Comparing the oxidizing reaction rates of these catalysts, it was summarized as follows. The best is impregnating mixed solution of Pt and Pd, or simple Pd solution, and the next was impregnating individual Pt and Pd solution.

As the results, the synergy effect on oxidation performance would be exhibited on the honeycomb hybrid catalyst which was impregnated with a mixed solution of Pt and Pd. The hybrid catalyst would be applicable for a safety system of future nuclear fusion facility. The present hybrid catalyst can also be applied to collect tritiated gases individually hydrogen and methane gases for the measurement of tritium in the atmospheric air around the fusion facility.

Fig.1 Hydrogen gas oxidizing rate coefficients.

Fig.2 Methane gas oxidizing rate coefficients.