§5. Development of Advanced Catalyst for Oxidation of Tritium and Quantification of Mass Transfer Coefficient

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Recovery of tritium released into the working area of fusion power plants or tritium-handling facilities is a important technique to establish the safety of the fusion technology. It is also necessary to develop a technique that enables accurate monitoring of environmental tritium around the facility where tritium is handled. The catalytic oxidation and adsorption is the most conventional and reliable method for removing tritium that is accidentally released into the working area of these facilities. The catalytic oxidation and adsorption is also a key process for monitoring of environmental tritium, which is used to capture tritium in the atmosphere. The catalysts used for these purposes need to posses high catalytic performance for low temperature combustion of tritium and tiritated methane as well as selectivity in catalytic reactions. Up to now, the authors have worked on the development of such catalysts. However, if the actual process is considered, there are further demands for catalyst and catalytic process. For example, if accidental tritium releases take place, large amounts of air should be processed by the air cleanup system. Therefore, the air cleanup system needs to be designed to be able to deal with the air with high volumetric velocity. Other than this, compactness and simplicity, efficient heating and endurance in long term use and repeated use are also required. With this background, the purpose of this study is to reexamine required catalytic performance from wider perspective and to develop advanced catalysts in a scientific way. This time, the authors tested the applicability of honeycomb catalysts, which is considered to be effective for the treatment of high throughput process gas and efficient heating, for catalytic oxidation of tritium tritiated methane. Furthermore, the selection of advanced materials such as perovskite type of catalysts, carbon nanotubes and carbon fibers were performed, and their fabrication methods were investigated as well.

In the experiments, a reactor made of quartz was used. The temperature of the reactor was changed in the range of ambient to 673 K. The argon gas containing hydrogen and methane of 0.1 % was introduced to the reactor. The concentration of hydrogen and methane at inlet and outlet stream of the reactor was measured with a gas chromatograph. The flow rates were controlled with conventional mass flow controller. The catalytic activity of honeycomb catalysts deposited with platinum and palladium was investigated using the experimental apparatus described above.

The honeycomb catalysts possess favorable characteristics in comparison with conventional pellet type of catalysts used in the packed bed reactor. If the honeycomb is made of metals, the honeycomb catalysts possess higher heat conductivity. Furthermore, there is a possibility that the catalyst itself can be directly heated by applying currents. The pressure drop along the honeycomb catalysts is smaller than packed bed catalysts, as well. For this reason, the catalysts used to deal with the exhaust gas of automotive are of honeycomb type. In practice, the pressure drop along the honeycomb catalyst was calculated by assuming actual cell sizes. The pressure drop along the axial direction in the packed bed catalyst reactor was also estimated using empirical equations. The results of these estimations indicate that the pressure drop can be substantially decreased when the packed bed catalyst is replaced with the honeycomb catalysts in the air cleanup system.

The effect of the cell density of the honeycomb and the amounts of deposited noble metals on the catalytic activity was examined. In this investigation, cordierite and 20%Cr-5%Al-Fe metal honeycombs were used. In these honeycomb substrates, platinum or palladium was deposited. The experimental results on catalytic activity of these catalysts suggest that increasing the cell density and increasing amounts of deposited noble metal improve their catalytic activity. It was also found that the catalytic performance of these honeycomb catalysts are comparable or better than those of conventional alumina based noble metal catalyst, which are used in the form of packed bed, if the honeycomb catalysts have appropriately high cell density and appropriate amounts of noble metals are deposited. Since it was proven that the metal honeycomb catalysts also possess higher catalytic performance, it should be worthwhile to investigate such metal honeycombs as can be heated by direct application of current.

With regard to more advanced catalysts, the fabrication of perovskite type of catalysts was tested using sold-solid reactions. The analysis with x-ray diffraction indicates that the prepared catalysts reproduce the diffraction pattern of perovskites reported in other literatures. The perovskite types of catalysts prepared in this way were now under tests of catalytic performance. In addition to this, the investigation of fabrication process of perovskite types of catalysts with higher catalytic performance is also being performed. The catalytic activity of these catalysts will be examined near future. As more advanced carbon materials, carbon nanotube and carbon fiber were examined in terms of suitable characteristics for catalysts and fabrication methods, and the selection of the materials was conducted. In the next term, these materials will be obtained, and the method of noble metal deposition on the carbon materials will be performed. Furthermore, the preparation of catalysts and investigation of the activity of those catalysts will be performed.