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

Munakata, K., Okabe, Y. (Interdisciplinary Graduate School of Engineering Sciences Kyushu University),
Kawano, T., Asakura, Y., Uda, T.
Tanaka, M. (Nippon Kucho Service Co., Ltd.)

For the establishment of the safety of the fusion technology, recovery of tritium released into the working area of fusion power plants or tritium-handling facilities is quite important. 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 tritiated 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 intensively investigated the oxidation performance 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 and tritiated methane. Furthermore, the sorption of hydrogen into carbon nano-tube was investigated.

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, and it was found that the increase of content of catalyst metal and the increase of cell density raise the performance of the catalysts. Therefore, the honeycomb catalysts with the cell density of 400 CPSI were intensively examined. The experimental results obtained using the experimental apparatus were analyzed and reaction constants for oxidation of hydrogen and methane were quantified. The results of the analysis indicate that cordierite honeycomb catalysts deposited with platinum has the best performance for the oxidation of hydrogen. It is also suggested that the metal honeycomb catalyst deposited with platinum has the oxidation performance comparable to a conventional packed bed catalyst deposited with platinum for the oxidation of hydrogen. With regard to the oxidation of methane, it was found that the metal honeycomb catalyst deposited with palladium has the best oxidation performance. It is also found that the cordierite honeycomb catalyst deposited with palladium has the oxidation performance comparable to a conventional packed bed catalyst deposited with palladium for oxidation of methane. The quantification of reaction constants for oxidation of hydrogen and methane over the honeycomb catalysts enables the quantitative design of catalyst bed of larger scale for recovery of tritium and tritiated methane.

Furthermore, the sorption of hydrogen in carbon nano-tube was investigated. The packed bed reactor charged with particles of carbon nano-tube was used for this study. An argon gas containing hydrogen was introduced to the reactor at ambient temperature, and the concentration of hydrogen in the outlet stream of the reactor was measured. However, the experimental result indicates no appreciable sorption of hydrogen. Several researchers have reported the sorption of hydrogen into carbon nano-tubes. Thus, it is necessary to investigate the effect of temperature and carrier gas. Additionally, the effect of catalyst on the sorption of hydrogen needs to be investigated.