Study on methane decomposition in plasma
プラズマ中のメタン分解に関する研究

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In the fuel cycle system of a fusion reactor, tritium is extracted from exhaust gas and reused. Plasma decomposition is one of techniques for extracting hydrogen from hydrocarbon. In order to evaluate a direct decomposition of methane using helium RF plasma, a flow-type plasma reactor utilizing capacitively coupled plasma was developed and direct decomposition of methane was investigated varying supplied RF power and total gas pressure. The overall decomposition rate of methane in helium plasma increased proportionally to the supplied RF power but decreased with increasing total pressure.

1. Introduction
Impurities in the exhaust gas from a vacuum chamber of a fusion reactor are removed in a fuel lean up system. Tritium contained in impurities gases is extracted in a tritium recovery system. Some amounts of hydrocarbon are included in the exhaust gas, when graphite materials are used in a part of plasma facing components. Especially, a considerable amount of hydrocarbon will be released in a discharge-cleaning to aim tritium recovery from plasma facing wall. In tritium handling facility, tritiated hydrocarbon is generally oxidized in a catalytic bed and recovered in an adsorption bed as water vapor. However, it is preferable that tritium is collected as molecular, considering it is reused as a fuel. Direct decomposition methods of methane with gettering materials [1-3] and Ni catalyst [4] have been investigated. In these ways, a regeneration process using hydrogen or oxygen is needed because decomposition performance deteriorates by carbon deposits. Moreover, the heating for the improvement of decomposition rate causes permeation loss of tritium. Direct decomposition by plasma is one of candidate techniques for tritium recovery from hydrocarbon. It is considered that the deterioration of decomposition performance of plasma by carbon deposits is small because the decomposition reaction is caused by the collision of hydrocarbon with electron in plasma. Therefore, it is expected that hydrocarbon is decomposed continuously for a long time without a removal process of carbon deposits. In the present study, decomposition behavior of methane in helium RF plasma was investigated varying supplied RF power and gas pressure.

2. Experimental
A flow-type plasma reactor utilizing capacitively coupled plasma was used for observing a direct decomposition behavior of methane. Fig.1 shows a schematic diagram of the experimental apparatus. Details of the size of the plasma reactor and the experimental procedure have been mentioned in [5]. The observation of components in the outlet gas was performed using a quadrupole mass spectrometer. The decomposition reaction progresses stepwise by the collision of methane and electron in plasma. When the decomposition rate is assumed to depend on first-order in methane concentration and the reverse reaction can be ignored, the following equation is given:

\[ Q \frac{dC_{CH_4}}{dv} = -k_{decomp} C_{CH_4} \]  

where \( Q \) is the volumetric flow rate in plasma \([\text{m}^3/\text{s}]\), \( C_{CH_4} \) is the methane concentration \([\text{mol}/\text{m}^3]\), \( v \) is the micro-volume in plasma \([\text{m}^3]\), \( k_{decomp} \) is the overall reaction rate constant \([1/\text{s}]\). When the total volume of plasma is represented as \( V \) \([\text{m}^3]\), the following equation is obtained by integration of eq.(1).

![Fig.1 A schematic diagram of the experimental apparatus.](image-url)
\[
\ln \frac{C_{CH_4,\text{out}}}{C_{CH_4,\text{in}}} = -k_{\text{decomp}} V \frac{1}{Q}
\]  

(2)

3. Results and discussions

Fig. 2 and Fig. 3 show the volumetric flow rate dependence on the decomposition rate at 150 Pa and 870 Pa. The logarithm natural of methane decomposition rate decreases linearly with increasing the inverse of the volumetric flow rate. This tendency was also observed in the experiment at 370 Pa. It was confirmed that the decomposition rate has large pressure dependence. The overall decomposition rates were obtained from the gradient of each line. These are compared in Fig. 4, where the values have been reported in [5] were also shown. The decomposition rate is clearly proportional to the supplied RF power. On the other hand, it decreases with increasing total pressure. These phenomena are speculated to be related to the electron density and energy in plasma. The electron density at around port 2 was measured by Langmuir probe method. It was found that the electron density linearly increases with increasing the supplied RF power. The present authors considered that a decomposition rate decreases depending on total pressure since an electron loses energy by the collision with neutral particles when the pressure is high.

4. Summary

A flow-type plasma reactor utilizing capacitively coupled plasma was developed and direct decomposition of methane was investigated varying supplied RF power and gas pressure. The overall decomposition rate of methane in helium plasma increased proportionally to the supplied RF power but decreased with increasing total pressure.

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