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The role of plasma and catalyst in methanation of CO_2 with plasma catalysis

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CO_2 recycling technology is one of the most important technologies for human beings. Methanation is a reaction in which methane is produced by hydrogenation of CO_2. Methane can be used as an energy carrier for about 1 billion times longer storage period and 1,000 times higher storage capacity than conventional batteries [1]. Other possible applications include the use of methane as a raw material for chemical products and in existing infrastructure. The reaction equation for CO_2 methanation, also known as the Sabatier reaction, is as follows,

$$CO_2 + 4H_2 \rightarrow CH_4 + 2H_2O;\ \Delta H = -165\ \text{kJ/mol}. \quad (1)$$

The reaction is generally accelerated by the use of a thermal catalyst. However, the exothermic reaction makes it difficult to control the temperature, and the catalyst is deactivated by overheating. In the case of the most common Ni/Al_2O_3 catalyst, the catalytic activity is halved in about 100 h [2]. The main causes of catalyst deactivation due to overheating are the increase in particle size due to sintering and the carbon coating on the catalyst surface due to coking. These phenomena strongly depend on temperature [3]. Therefore, it is important to realize the process at lower temperatures for long-term stable methanation.

To solve this problem, plasma catalysis is attracting attention. Plasma catalysis can reduce the process temperature [4]. This reason is still unclear, but it is due to some interactions between the plasma and the catalyst, such as the excitation of the feed gas and the activation of the catalyst by the plasma.

In our research, we have achieved a CO_2 conversion of 90% and a methane selectivity of 35% at room temperature by using a helicon plasma with high electron density and electron temperature [5]. By using low pressure, the effect of the reverse reaction was reduced, and the reaction rate constant for CO_2 conversion by electron impact was derived. The effect of the catalyst was also evaluated by placing catalysts on the surface of the electrode in the capacitively coupled plasma (CCP) reactor, and we found that Cu, which is generally not used in methanation reactions, showed high catalytic activity in plasma catalysis [6].

However, there is still much that remains unknown about the fundamentals of plasma-catalyzed methanation, such as the reaction process and suitable conditions for the methanation reaction. In this study, we report the role of plasma and catalyst in methanation of CO_2 with plasma catalysis.

The experiments were carried out in the CCP reactor. The plasma is generated between two Cu plates placed in parallel. Here, the Cu plates act as catalysts. The Cu plates have a diameter of 50 mm and the distance between the electrodes is 6 mm. A mixture of CO_2 and H_2 gas was supplied to the reactor. The gas flow rates of CO_2 and H_2 were 1 sccm and 6 sccm, respectively. The pressure was 750 Pa. The discharge power was 100 W. The gas composition was measured by quadrupole mass spectrometer (SRS QMS300). The catalysts are heated by ion bombardment, and the temperature was measured by a thermocouple.

Figure 1 shows the dependence of CO_2 conversion and CH_4 yield on catalyst temperature. Whereas the CO_2 conversion is almost independent of temperature, the CH_4 yield increases with temperature. This indicates that CO_2 is mainly decomposed in the gas phase, while CH_4 is mainly produced on the catalyst surface. In the case of atmospheric pressure plasma-catalyst combinations, which are common in studies of plasma catalysis, the temperature of the catalyst surface has a strong influence on the decomposition of CO_2 [4]. However, in the case of low-pressure plasma, the conversion of CO_2 is independent of temperature. This is because the contribution of the CO_2 formation reaction in the gas phase is large under atmospheric pressure. In other words, by using low pressure plasma, the contribution of the gas phase reaction and the catalyst surface reaction can be discussed separately. In the presentation, factors affecting CO_2 decomposition in the gas phase and active species contributing to methane production on the catalyst surface will be discussed in detail.

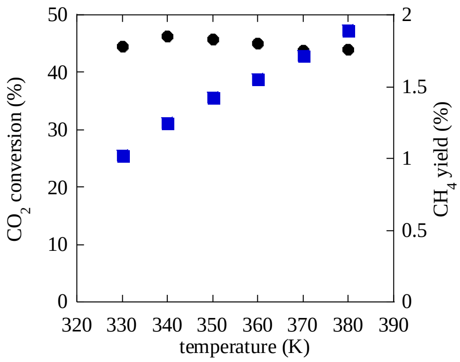

Figure 1. Dependence of CO_2 conversion and CH_4 yield on catalyst temperature.

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