

NON-THERMAL PLASMA PROCESSING FOR THE CONVERSION OF HYDROCARBONS

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Abstract. Non-thermal, atmospheric pressure plasma processing has been utilised for the conversion of methane and carbon dioxide into syngas, higher hydrocarbons (C₂-C₄) and H₂O at a lower reduced energy density than has previously been shown. The synergistic combination of BaTiO₃ packed-bed dielectric barrier discharge (DBD) with zeolite or alumina catalysts at low applied power (~1 W) has been investigated. Of the catalysts tested, zeolite NaY produced the most promising CH₄ and CO₂ conversions of 29 % and 13 %, respectively. Plasma parameters including feed gas composition and total flow rate have been varied in order to gain a better understanding of the factors which govern the overall conversion of methane into specific product channels.

1. INTRODUCTION

Methane is the second most important gas responsible for the greenhouse effect, having a global warming potential 21 times greater than that of CO₂. Methane is extensively found as natural gas, coal mine and oil bed emissions, landfill site emissions and sustainably-produced biogas. Landfill gas and biogas are produced with significant quantities of CO₂. Consequently, CO₂ reforming of CH₄ (dry reforming) is a particularly attractive area for research, as it contributes to the reduction of two greenhouse gases and the development of clean, sustainable fuels such as hydrogen and methanol. Although it is known that plasma-catalysis has a synergistic effect on dry reforming, a comprehensive understanding of the reaction mechanisms is presently lacking. Various zeolite and transition or noble metal catalysts have been investigated using DBD [1, 2] and corona [3] discharges. These studies have shown that plasma-catalysis can significantly reduce operating temperature compared with conventional reforming processes, modify the selectivity of products and eliminate poisoning effects of the catalyst.

2. EXPERIMENTAL SECTION

The experimental set-up is illustrated in Figure 1. The DBD reactor consists of a 226 mm long Pyrex tube of 25 mm internal diameter containing two cylindrical electrodes placed 30 mm apart and connected by a 10 kV, AC power supply. The area between the electrodes is packed with barium titanate (BaTiO₃) beads of 3.5 mm diameter. The discharge volume of the reactor is 4.63 cm³.

Zeolite NaY was supplied by Sigma-Aldrich. Zeolites NaA (pore size = 4 Å) and CaA (pore size = 5 Å) were supplied by Fluka Chemika. 0.5 % Ni/γ-Al₂O₃ was prepared as described by Harling [4]. All catalysts were used in powdered form as supplied and were calcined by heating to 873 K for 3 hours prior to use. The catalysts (~ 250 mg) were packed in the DBD reactor occupying the spacing between the BaTiO₃ beads.

CH₄ and CO₂ (BOC Gases) flows were varied between 100-300 mL/min and controlled by mass flow controllers (MKS flow control systems). All experiments used a gas operating pressure of 1 bar. The system was purged for one hour prior to each experiment to allow stabilisation of gas mixtures.

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