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Plasma-activated catalytic surface reaction for CH_4 and CO_2 conversion

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Pulsed dry methane reforming (DMR; $CH_4 + CO_2 = 2CO + 2H_2$) in dielectric barrier discharge (DBD) and 12wt.% Ni/Al_2O_3 catalyst hybrid reaction was investigated for the efficient conversion of greenhouse gas (CH_4, CO_2) into syngas (H_2, CO) at low temperature. CO_2 flow was kept constant, while CH_4 was introduced into the reactor at constant duration and interval with various CH_4/CO_2 ratios. Although solid carbon was deposited during the reforming reaction, carbon was almost fully removed by turning off CH_4 flow and applying CO_2-fed DBD (Boudouard reaction; $C + CO_2 = 2CO$). Pulsed transient analysis of reforming reaction revealed that CH_4 dehydrogenation and the subsequent reverse water gas shift reaction (r-wgs; $CO_2 + H_2 = CO + H_2O$) is sufficiently fast with and without DBD, producing syngas with H_2 selectivity of 70% and CO selectivity of 90%, respectively. We also found that the Reverse reaction ($CO + 3H_2 = CH_4 + H_2O$) is negligible when the catalyst temperature is higher than 550 °C. In contrast, CO_2 conversion and carbon removal reaction (Boudouard reaction), was promoted clearly by DBD hybridization. Radical injection is primarily important step. Besides, selective surface heating by DBD such as charge recombination on the catalysts is anticipated to promote carbon diffusion through Ni catalyst particles and subsequent oxidation by adsorbed CO_2. DBD and catalyst hybrid reaction enabled higher CH_4 and CO_2 conversion without having serious coking problem. We also employed optical emission spectroscopy (OES) for the better insight into gas phase- and surface-reaction induced by DBD. Transient analysis of reforming products are nicely correlated with CO Angstrom system and C_2 high pressure Swan system, implying the carbon diffusion through Ni nanoparticles is the rate-determining step and clearly promoted by DBD. More detailed information about research background, energy efficiency, and thermodynamic analysis of plasma catalysis will be presented.