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Electrification of CO_2 Conversion Processes by Fluidized-bed Plasma

The emerging electricity-driven plasma catalysis demonstrates the key advantages of overall flexibility, encompassing feed gas, green energy source and operation, which align precisely the core requirement for global CO_2 capture and utilization. However, unlike thermal catalysis where simply a catalyst plays the main role, the promising plasma catalysis is much more complex: besides surface catalytic analysis, the rarely studied plasma characteristics and dynamic plasma–catalyst interaction are equally important, posing the key obstacles to its development. Herein, we aim to maximize the plasma catalytic synergism (PSC) for efficient CO_2 valorization. We firstly focus on the plasma–catalyst interaction behaviors, comparatively analyzing the properties of both plasma and catalyst bed in each representative system, and conclude the fluidized-bed dielectric barrier discharge (FB-DBD) reactor is the ideal candidate. Thereafter, in CO_2-to-CO conversion via Langmuir-Hinshelwood (L-H) mechanism using CO_2–CH_4 reforming as a model, we proposed FB-DBD and experimentally verified its superiorities over packed-bed DBD (PB-DBD) in PCS. We analyzed the forward catalysis behavior, heat transfer, discharge properties and reverse reaction; the forward CO_2 conversion was selectively promoted to reach the thermal equilibrium. In addition, the CO_2 was further co-reacted with more reactive H_2 via the representative Eley-Rideal (E-R) pathway in FB-DBD to produce CO, by which the CO_2 conversion well beyond the thermal equilibrium. Meantime, we obtained the criterion of DBD for maximizing the PCS: low electron energy and high electron concentration by increasing streamer amount, instead of streamer size. Furthermore, in CO-to-X conversion (X = carbon as a model) via the typical Mars-van Krevelen (MvK) mechanism, we verified the vibrational excitation of CO by FB-DBD and in situ reduction of Fe_3O_4 by H_2 jointly contributed to an efficient CO conversion to the unique spiral type carbon nanofibers.