6th Asia-Pacific Conference on Plasma Physics, 9-14 Oct, 2022, Remote e-conference

N_2 fixation, CO_2 conversion, and CH_4 valorization in atmospheric pulsed plasmas

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Energy-efficient and environmentally benign alternatives for small molecules (N_2, CO_2 and CH_4) conversion are urgently needed, especially the green-chemistry, electricity-driven processes using renewable energy, to near the ultimate goal of zero carbon emissions. The paper introduces a potentially ground-breaking approach to use renewable energy in three major industrial reactions: 1) N_2 fixation, 2) CO_2 conversion and 3) CH_4 valorization, which have drastically lower carbon footprint (up to over 90% with respect to current ones). The high-energy electrons and/or gas fast-heating effect of low-temperature plasma will lower the reaction barrier and/or promote the reaction equilibrium under near ambient temperature and pressure.

1) N_2 fixation: The indirect approach involving plasma nitrogen oxidation ($N_2 \rightarrow NO_x$) in pulsed spark discharge[1] and further catalytic reduction to ammonia ($NO_x \rightarrow NH_3$) have been proposed to approach the zero-carbon emissions target[2], with new insights into free-radical-chain reactions by plasma kinetics modeling and optical diagnosis (as shown in figure 1). The approach to convert N_2/H_2O directly to NH_3 in pulsed plasma-catalytic reactor is also an alternative way for Haber–Bosch process[3].

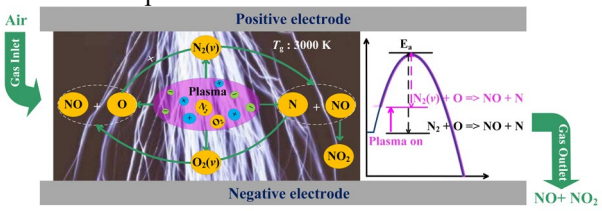

Figure 1. Sustainable plasma-driven N_2 oxidation for NO_x formation by a nanosecond pulsed spark discharge

2) CO_2 conversion: The dry reforming of CH_4 to syngas in atmospheric pulsed dielectric barrier discharge plasmas are optimized by pulse parameters (pulse rising time, width and frequency)[4] and reactor structures (electrodes, controlled temperature and packing materials) [5], as shown in figure 2. The methanation of CO_2 has also been implemented in atmospheric pulsed dielectric barrier discharge plasma coupling with structured catalyst and its reaction mechanism is speculated by a plasma kinetics model[6].

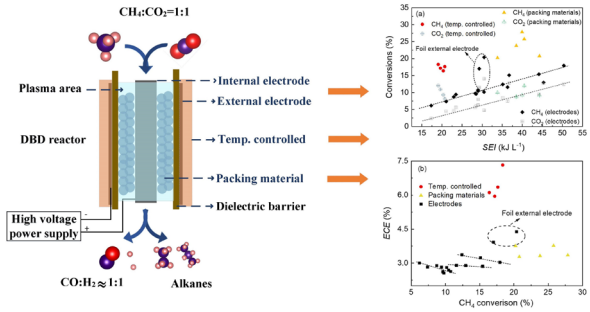

Figure 2. The dry reforming of CH_4 to syngas in atmospheric pulsed DBD plasmas

3) CH_4 valorization: We acquire a potential result that the maximum CH_4 conversion of 90% and H_2 yield of 38% could be achieved by the microsecond pulsed spark discharge with a desirable energy efficiency of 44%[7], and the possible energy and chemical pathways are shown in figure 3. On the other hand, the dissociation CH_4 for H/CH_3 radicals by the pulsed dielectric barrier discharge plasma show the possibility for plasma-catalytic CH_4 directly to fine chemicals[8].

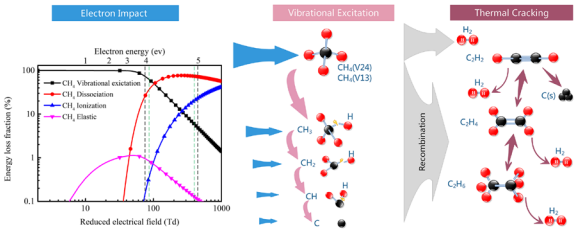

Figure 3. The schematic diagram of possible energy and chemical pathways in pulsed CH_4 spark discharge

References

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Note: Abstract should be in (full) double-columned one page.