

NITROGEN FIXATION AS NO_x ENABLED BY A THREE-LEVEL COUPLED ROTATING ELECTRODES AIR PLASMA COMBINED WITH NANO-SIZED TiO₂

X. LEI¹, X. LU¹, S. JIN¹, Z. LI¹

¹State Key Lab of Advanced Electromagnetic Engineering and Technology, School of Electronic and Electrical Engineering, Huazhong University of Science and Technology, Wuhan, Hubei, People's Republic of China

A novel three-level coupled rotating electrodes air plasma with nano-sized TiO₂ photocatalysts is developed for evaluation of nitrogen fixation. Factors influencing NO_x concentration and energy cost, including gas flow rates, N₂ fractions, relative humidity levels, blade numbers at each rotating electrode and rotating speed, are examined. Gas flow rates prove to have no effect on the rotational temperature of N₂ 337.1 nm and the emission intensities of N₂⁺ and N₂, but specific energy input (SEI) and species' residence time can be shorter with higher air flow rates, resulting in lower NO_x concentration and energy cost. The addition of H₂O also has a positive effect on both NO_x concentration and energy cost. Optical emission spectrum (OES) shows that air + H₂O plasma has stronger 336 nm (NH) and 309 nm (OH) emission lines than air plasma, indicating that NH and OH are the key species in NO_x enhancement. Final results show that the exceptional synergistic effect between TiO₂ and three-level coupled rotating electrodes air plasma significantly increases the NO_x concentration by 68.32% (from 4952 to 8335 ppm) and reduces the energy cost by 40.55% (from 2.91 to 1.73 MJ mol⁻¹) at gas flow rate of 12 l min⁻¹ and relative humidity level of 12%, which beats the ideal thermodynamic energy limit ~2.5 MJ mol⁻¹ for the thermal gas-phase process. A possible mechanism for enhanced NO_x production with TiO₂ is discussed: Highly energetic electrons in plasma contribute to the formations of the electron-hole pairs and oxygen vacancy (V_o) on the TiO₂ catalyst surface, which may facilitate the dissociative adsorption of O₂ molecules to form superoxide radical groups (like O₂⁻) and H₂O molecules to form surface hydroxyl groups (like OH·), and thus, improving energy efficiency.