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Repetitively nanosecond-pulse discharge: Non-thermal Plasma Generation and Energy Conversion

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Gas discharge is an interdisciplinary research field related to plasma physics, electrical engineering, fluid mechanics, and others. However, the classical Townsend theory (in 1903) and Streamer theory (in 1937) could not convincingly describe the fast breakdown process in nanosecond-pulse discharges (NPDs) at high pressure. This is due to that the high gradient electric field is close to or exceeds the spatial-temporal variation rate of traditional electron avalanche and streamer development.

Therefore, new strategies are needed to investigate NPDs, where new phenomena and characteristics appear. NPDs also provide more change to improve the performance of plasma in different applications [1].

Here, run-away electron (RAE) beam and X-ray with an energy up to tens of keV from NPDs are measured with time and energy resolution. It is found that RAE beam appears during the rising edge of high-voltage pulse before the breakdown happens. Based on this experimental observation, the fast breakdown theory guided by RAEs is proposed. This is verified by the Monte-Carol simulation that electrons with a high initial energy can induce the formation of diffuse discharge in large volume, while low energy electrons will result in filamentary discharge.

Generally, the breakdown process of NPDs develops in a form of fast ionization wave (FIW), i.e., the discharge propagation is driven by electron multiplication and charge separation under the strong electric field. The electric field during discharge propagation in dielectric barrier discharge and atmospheric pressure plasma jet is measured using electric field induced second harmonic method [2, 3]. It is found that FIW exists universally in NPDs, with typic feature of field enhancement at the FIW front region but weak electric field in the plasma channel after the FIW front. The memory effect of residual particles in repetitively NPDs also influences the FIW propagation and energy coupling processes.

Furthermore, repetitively NPDs, featured by high electric field, high-energy electrons and high activity particles, have been applied in the field of energy conversion, including nonoxidative conversion of CH_4, CO_2 conversion, and NH_3 synthesis.

It is found that the maximum CH_4 conversion 90% and H_2 yield 38% with energy conversion efficiency of 44% are achieved using pulsed spark discharges [4]. Hydrogen atom is an important intermedial species and its related energy transfer processes dominate in NPDs for CH_4 conversion [5]. The short pulse rise-time simultaneously promotes reactant conversions and energy efficiency when NPDs are used to drive the CH_4/CO_2 dry reforming [6].

The highly-adjustable NPDs and nickel foam (NF) based catalysts ($Ni\text{-}Fe_x\text{-}Al_{1-x}/NF$) synergistically enable the high-performance CO_2 methanation [7]. It is demonstrated that the electron-induced vibrational excitations CO(v) and Ni-Fe active phases (<10 nm) contribute to the low-temperature CO_2 hydrogenation. NH_3 yield rate reached 198.3 μmol cm^{-2} h^{-1} with the strategy that tandem coupling of plasma N_2 oxidation and electrochemical NH_3 synthesis, with Faradaic efficiency almost 100% [8].

In addition, the relationship between the optical emission spectroscopy and the relative concentration of gaseous products in plasma-enabled CH_4 conversion is investigated [9]. It is found that the intensity ratio of C_2 $A \rightarrow X$ and CH $A \rightarrow X$ can function as a good indicator of the relative concentration of saturated and unsaturated hydrocarbon products and gas temperature, over an extremely wide range of plasma parameters. The link between OES and gaseous products is bridged with the intermediate active species and is revealed by reaction kinetics model.

In summary, this work systematically introduces the plasma characteristic of NPDs and their applications in energy conversion, which open a new window toward a deeper understanding of gas discharge physics and a better performance in applications.

References

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