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High efficiency NO_x synthesis and regulation using dielectric barrier discharge in the needle array packed bed reactor

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Nitrogen, existing in amino acids, chlorophyll, and nucleic acid, is the necessary component of living organisms. Above 99% of the nitrogen is present in the atmosphere as N_2 which has the stable N≡N structure and electron configuration. As a result, the nitrogen fixation process is essential to use N_2 reasonably. The Haber-Bosch (H-B) process, which takes primary responsibility to global population survival and crop growth is an energy intensive chemical process. In detail, accounts for 1%-2% of the world's energy consumption and release 300 million tons of CO_2 per year. Considering the grim situation of world growing population and greenhouse gas emission, the new nitrogen fixation technology which can reduce the environmental pressure and energy crisis is necessary to be developed.

The plasma has been considered as a promising technology to develop nitrogen fixation effectively. Among different discharge types, the DBD, especially in the packed bed reactor (PBR) which can generate non-thermal plasma and play good synergy effect with catalyst, is widely used in NO_x synthesis process. However, comparing with other plasma types, achieving the reduction of the energy cost or the increase of the NO_x production in PBRs is always a challenge and a lot of research works has been carried out.

As can be concluded from previous works, most PBRs used for gas-phase NO_x synthesis are driven by alternating current (AC) power supply, and the PBRs driving by nanosecond power supply is rarely applicated in gas-phase NO_x synthesis.

To further improve the energy efficiency of gas-phase NO_x synthesis in PBRs, the optimization of plasma sources is an important path. In addition, much attention should also be paid to the innovations of electrode structure which can directly influence the utilization rate of raw material gases.

In this paper, a needle array packed bed reactor excited by nanosecond pulse voltage is employed to synthesize NO_x efficiently with α-Al_2O_3 and γ-Al_2O_3 packed materials. The pulse width, pulse rising time, pulse repetition rate, and oxygen content are adjusted to regulate the NO_x synthesis results, such as NO_x concentration, product selectivity, and energy cost. The discharge power, key reactive species, and the rate coefficient are calculated to provide clear insight into the NO_x synthesis pathways. It is found that the NO_x can be synthesized efficiently in this needle-tubes array packed bed reactor, the highest NO_x concentration of 1.12% and 0.97% are obtained in unfilled reactor and γ-Al_2O_3 packed bed reactor, respectively, which are nearly two times higher than the results reported in previous works under the same energy cost and the same type of discharge. In addition, increasing pulse width and pulse repetition rate can significantly enhance the NO_x concentrations because of the increased energy input and rate coefficient.

References

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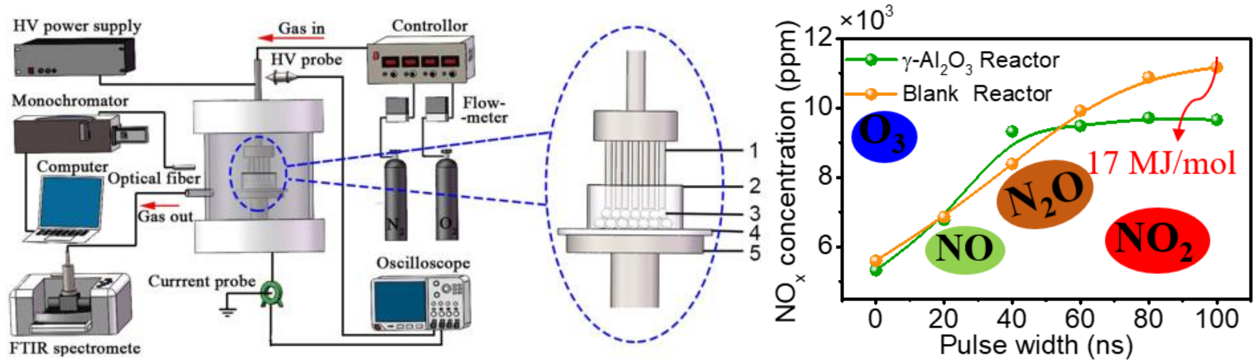

Figure 1. experiment setup and magnified view of the PBR (left) and the NO_x synthesis results under different pulse width in PBR (right)