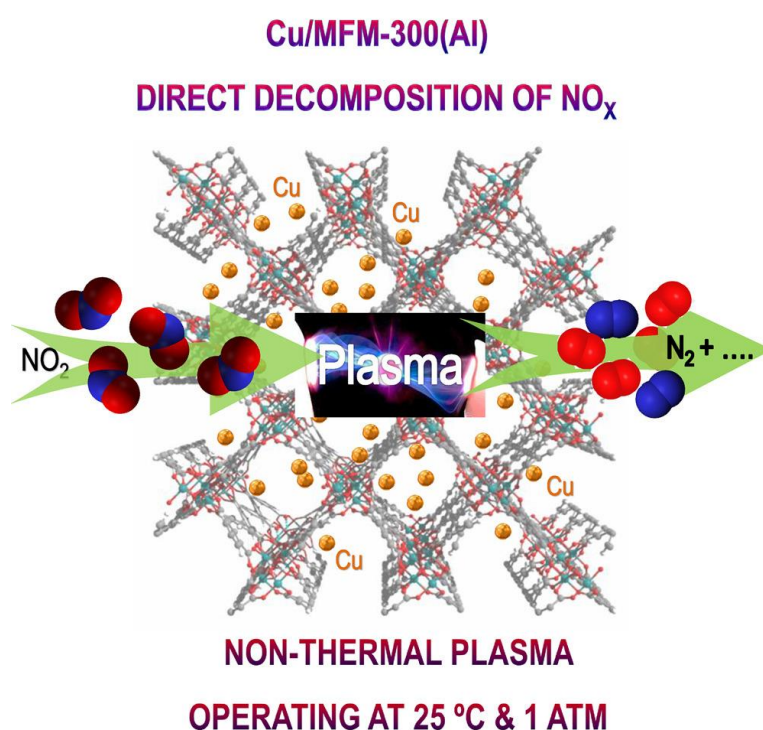


Catalytic decomposition of NO₂ over a copper-decorated metal-organic framework by non-thermal plasma

Nitrogen oxide (NO_x) causes significant impacts on the environment and human health, and nitrogen dioxide (NO₂) is the most toxic and prevalent form of NO_x in the atmosphere. Its removal and catalytic degradation into non-harmful species (*i.e.*, N₂) are thus important challenges. A great deal of effort has been devoted to developing catalysts for deNO_x processes with a focus on NO reduction from exhaust gases. The ammonia-assisted selective catalytic reduction (NH₃-SCR) over Cu-exchanged zeolites is by far the most effective method to reduce NO_x to N₂ and H₂O. However, this process has several inherent limitations, notably the high operating temperature (typically 250-550 °C), high running cost for reductants, use of corrosive and toxic NH₃, and potential release of NH₃ into atmosphere. In contrast, the catalytic degradation of NO₂ for domestic environments is poorly studied. The development of new efficient catalysts and catalytic processes to enable the direct decomposition of NO₂ at room temperature and without the use of reducing agents has, therefore, attracted considerable attention.

Porous metal-organic framework (MOF) materials show promise for the highly selective adsorption and separation of NO₂ at room temperature with the high porosity and tailored-to-design pore functionality. However, the application of MOF-based catalysts in deNO_x processes has been rarely explored, primarily due to the limited stability of MOFs against highly corrosive NO₂ and NH₃ at elevated temperatures. Non-thermal plasma (NTP) activation can promote deNO_x processes at room temperature by generating highly reactive species, especially vibrationally and electronically excited states of molecules (*e.g.*, N₂, O₂, NO), N and O atoms, radicals, and electrons with a typical electron temperature of 10⁴ °C. NTP activation in MOF-based catalysts has been shown to enhance performance in catalytic reactions, with the structure and porosity of the MOF being preserved. There are thus powerful drivers for the development of efficient deNO_x systems that can operate at room temperature and avoid the use of toxic reductants.

In this study here, a new process combining a robust MOF-based catalyst and NTP-activation has been developed for one-through conversion of NO₂ into N₂ without use of any external reducing agent at room temperature. The rigid and robust open structure of MFM-300(Al) offers an excellent platform to embed uniformly dispersed Cu nanoparticles of diameters of *ca.* 1 nm using a simple incipient wetness impregnation method. Cu/MFM-300(Al) shows simultaneously high NO₂ conversion and high N₂ selectivity, as well as an excellent long-term stability under the NTP activation at 25 °C and 1.0 bar. The high catalytic activity of Cu/MFM-300(Al) is attributed to the



unique formation of $\text{Cu}^{2+}\cdots\text{NO}$ nitrosylic adducts, which facilitates the dissociation of NO to improve the yield of N_2 . Compared with conventional thermal-based catalysis, NTP activation can effectively preserve the structure and porosity of MOF-based catalysts. Coupled with emerging stable MOFs showing ultra-high and selective NO_2 adsorption, we are now seeking to design new MOF-based catalysts to drive future development of efficient reductant-free de NO_x processes to mitigate air pollutants.

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