

Enhanced ambient ammonia photosynthesis using nanosheets with light-switchable oxygen vacancies

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The issue to achieve efficient nitrogen (N₂) reduction to ammonia (NH₃) has posed a significant challenge for decades as the inert N \equiv N bond could be hardly broken because of the extremely large bond energy of 940.95 kJ mol⁻¹. To date, the industrial fixation of N₂ to NH₃ is monopolized by the energy-intensive Haber-Bosch process (673-873 K and 15-25 MPa), which unsustainably employs <u>natural gas</u> to make the hydrogen (H₂) feedstock with enormous energy consumption from fossil fuels, leading to a large amount of carbon dioxide (CO₂) emission. In this context, photocatalytic N₂ reduction is regarded as a sustainable alternative way for NH₃ synthesis from N₂ and water under ambient conditions.

However, the efficiency of most traditional photocatalysts is still far from satisfactory mainly due to the hard bond dissociation of the inert N_2 , which results from the weak binding of N_2 to the catalytic material and further inefficient electron transfer from photocatalyst into the antibonding orbitals of N_2 . In order to promote efficiency of N_2 photofixation, introducing the electron-donating centers as the catalytic activation sites for optimizing the N_2 adsorption properties and improving the photoexcited charge transport in the catalysts is a promising strategy.

Oxygen vacancy (OV) represents the most widely and prevalent studied type of surface defect for N_2 fixation. On one hand, OV can be facilely created for its relatively low formation energy; on the other hand, OV can assist photocatalysts to gain exciting N_2 fixation photoactivity by virtue of its superiority in N_2 capture and activation. Therefore, a



semiconductor with sufficient OVs may be favorable to improve their N₂ fixation performance. Transition metal (TM) doping is another widely investigated effective method to improve the photoactivity of N_2 fixation, because the TM species possess the advantageous ability of binding (and even functionalizing) with inert N₂ at low temperatures due to their empty and occupied d-orbitals, which can achieve the $TM-N_2$ interaction via "acceptance-donation" of electrons. Mo, as a critical element of the catalytic center in mysterious Mo-dependent nitrogenase, has attracted a lot of attention for the N₂ fixation. To this end, OVs-rich and Mo-doped materials would be ideal candidates for N₂ photofixation. In addition, layered bismuth oxybromide (BiOBr) materials have attracted numerous attentions because of their suitable band gaps and unique layer structures. For BiOBr-based semiconductors, such as Bi_3O_4Br and Bi_5O_7Br , it has been revealed that OV with sufficient localized electrons on their surface facilitates the capture and activation of inert N_2 molecules.

Recently, a research team led by Prof. Yi-Jun Xu from Fuzhou University, China reported that the introduction of OVs and Mo dopant into Bi_5O_7Br nanosheets can remarkably improve the photoactivity of N_2 fixation. The modified photocatalysts have showed the optimized conduction band position, the enhanced light absorption, the improved N_2 adsorption and charge carrier separation, which jointly contribute to the elevating N_2 fixation photoactivities. This work provides a promising approach to design photocatalysts with light-switchable OVs for N_2 reduction to NH_3 under mild conditions, highlighting the wide application scope of nanostructured BiOBr-based photocatalysts as effective N_2 fixation systems. The results were published in *Chinese Journal of Catalysis*.

More information: Xue Chen et al, Enhanced ambient ammonia photosynthesis by Mo-doped Bi5O7Br nanosheets with light-switchable oxygen vacancies, *Chinese Journal of Catalysis* (2021). <u>DOI:</u>



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