Boosted Photoconversion of SF₆ over Defective ZnGa₂O₄ Nanosheets under Mild Conditions

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EXPERIMENTAL SECTION

Synthesis of the ZnGa₂O₄ nanosheets:

The described procedure involves the dissolution of 220 mg of zinc acetate dihydrate $(Zn(CH_3COO)_2 \cdot 2H_2O)$ in 20 mL of deionized water. After that, 512 mg of gallium nitrate $(Ga(NO_3)_3)$ is added to the solution, followed by stirring for 20 minutes. Finally, 10 mL of anhydrous ethylenediamine is added to the mixture. Following the 10-minute magnetic stirring, the liquid is transferred to a 50 mL Teflon lined hydrothermal autoclave. The autoclave is then sealed, and the contents are heated to 200 °C for a duration of 24 hours. Subsequently, the autoclave is removed from the heat source and left to cool naturally at room temperature. After the reaction in the hydrothermal autoclave, a white precipitate is obtained. This precipitate is washed several times with water and ethanol to remove any impurities that may have been formed during the reaction.

Synthesis of the defective ZnGa₂O₄ nanosheets:

After obtaining the white $ZnGa_2O_4$ nanosheets, they are subjected to further treatment. The nanosheets are heated at a temperature of 400 °C in a 5% H₂/Ar atmosphere for a duration of 1 hour. This specific atmosphere ensures a controlled environment for the heating process.

Characterization:

TEM images were performed with a HT7800 TEM with an acceleration voltage of 200 kV. HRTEM images were carried out on a Talos F200X G2 TEM/STEM with a spherical aberration corrector. ESR spectra were measured on an ESR spectrometer (JEOL JES-FA200) at room temperature. XRD patterns were obtained from a Philips X'Pert Pro Super diffractometer with Cu K α radiation ($\lambda = 1.54178$ Å). UV-vis diffuse reflectance spectra were measured on a Perkin Elmer Lambda 950 UV-vis-NIR spectrophotometer. XPS spectra were acquired on an ESCALAB MKII system with Al K α (*hv* = 1486.6 eV) as the excitation source. The binding energies obtained in the XPS spectral analysis were corrected for specimen charging by referencing C 1s to 284.8 eV. Raman spectra and Room temperature photoluminescence (PL) spectra were detected on a RenishawRM3000 Micro-Raman system. Synchrotron-radiation photoemission spectroscopy (SRPES) were executed at the National Synchrotron Radiation Laboratory (NSRL) in Hefei, China. BET surface area was acquired on automatic microporous gas adsorption analyzer system (Microtrac MRB 2023 M BELSORP). Micromeritics ChemiSorb 2720 with a thermal conductivity detector (TCD) were used to conduct temperature-programmed desorption (TPD) of the samples.

Photocatalytic SF₆ conversion measurements:

The photocatalytic SF₆ conversion measurements were conducted in a sealed off-line reactor (Perfectlight Limited, Beijing). In the photocatalytic SF₆ conversion process, 10 mg as-synthesized catalysts were initially dispersed in 2 mL deionized water and then spined dropped onto a quartz glass. After heating at 60 °C for 30 minutes, the as-synthesized catalysts were successfully deposited on the quartz glass (diameter: 3.6 cm

area: 10.2 cm²). After putting the quartz glass in the reaction cell as well as injecting 8 mL cetonitrile solution on the bottom, the reaction cell was vacuum-treated for three times, which was then pumped by high-purity SF₆ (99.99%) to reach an atmospheric pressure. While the temperatures of the reaction cell were controlled at 60°C by recirculating cooling water system during irradiation. The light irradiation comes from a CEL-HXF300 Xe lamp (Beijing China Education Au-light Co., Ltd.) with a standard AM 1.5G filter, outputting the light density of about 100 mW/cm². The instrument was initially added some acetonitrile solution, and then pumped and purged for three times, which was finally filled by high-purity SF₆ to reach an atmospheric pressure.



Figure S1. XRD patterns for the Vo-ZnGa₂O₄ and ZnGa₂O₄ nanosheets.



Figure S2. TEM image for the $ZnGa_2O_4$ nanosheets.



Figure S3. HRTEM image for the $ZnGa_2O_4$ nanosheets.



Figure S4. (a) Zn 2p and (b) Ga 2p XPS spectra a for the defective $ZnGa_2O_4$ nanosheets and the $ZnGa_2O_4$ nanosheets.



Figure S5. Schematics illustrating the electronic band structures for the Vo-ZnGa₂O₄ and ZnGa₂O₄ nanosheets.



Figure S6. TEM image for the Vo-ZnGa₂O₄ nanosheets after photocatalysis.



Figure S7. XRD patterns for the Vo-ZnGa₂O₄ nanosheets before and after photocatalysis, in which the pattern of "Before catalysis" was indexed to that of the Vo-ZnGa₂O₄ in Figure S1.



Figure S8. TEM image for the ZnGa₂O₄ nanosheets after photocatalysis.



Figure S9. XRD patterns for the $ZnGa_2O_4$ nanosheets before and after photocatalysis, in which the pattern of "Before catalysis" was indexed to that of the $ZnGa_2O_4$ nanosheets in Figure S1.