Active Learning–Driven Design of g-C_3N_4 Photocatalysts for Solar CO_2 Reduction

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The photocatalytic reduction of CO_2 using solar energy is a promising strategy for sustainable fuel production, yet the rational design of efficient photocatalysts remains computationally expensive due to the large configurational space of materials, dopants, defects, and adsorption geometries. In this work, we investigate CO_2 reduction on graphitic carbon nitride (g-C_3N_4), a visible-light-responsive semiconductor, using Density Functional Theory calculations combined with an active learning framework. Pristine, doped, and defect-engineered g-C_3N_4 surfaces are systematically explored to evaluate electronic structure, band alignment, CO_2 adsorption energies, charge transfer, and molecular activation, particularly O–C–O bending. A Gaussian Process regression model is trained on an initial set of first-principles results and iteratively selects new candidate systems based on uncertainty and expected improvement criteria. This active learning strategy efficiently identifies the most informative calculations, reducing the total computational cost while maintaining high predictive accuracy. The results reveal clear correlations between dopant-induced electronic states near the conduction band and enhanced CO_2 activation, indicating favorable conditions for solar-driven reduction reactions. Overall, this study demonstrates that active learning significantly accelerates the discovery and optimization of g-C_3N_4-based photocatalysts, providing a data-efficient and scalable approach for artificial-intelligence-guided materials design in renewable energy applications relevant to future low-carbon energy technologies worldwide.