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Title:	Integrated Approaches in Nanomaterials Design: From Metal- Free Photocatalysts to Metal Oxo Clusters for Enhanced Catalytic Performance.
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Abstract:	Visible light-driven hydrogen (H ₂) evolution from water (H ₂ O) has triggered an extensive demand for high-abundant, low-cost semiconducting photocatalysts towards sustainable energy harvesting. Even though graphitic carbon nitride (g-C ₃ N ₄) heralds great promise as a metal-free inexpensive photocatalyst, its efficiency has been limited due to low visible light absorption, stacked layers-driven restricted mass transfer, and poor charge- transfer dynamics. Although many attempts have been made to exfoliate the layers and tune the photophysics but finally end up with either complex synthetic steps or very low yield. Herein, we have decorated the surface of g-C ₃ N ₄ by carbon nanotube (CNT) by two composite formation strategies. The composite made under hydrothermal conditions and calcination technique showed around 6 and 3 times higher H ₂ evolution rates respectively under visible light irradiation compared to the bare g-C ₃ N ₄ . The enhanced photocatalytic hydrogen production can be attributed to facile charge transfer through the network of CNT spread on the photoactive surface of g-C ₃ N ₄ . This work highlights the potential of metal-free composites to realize highly efficient heterogeneous photocatalytic systems for solar hydrogen production. II Synthesis of various metal oxo clusters, including those of Zr and Hf using fatty acids as stabilizing ligands commonly employed in oxide nanocrystal stabilization. Our focus was on the M ₆ O ₄ (OH) ₄ (OOCR) ₁₂ type clusters, recognized as pivotal in materials science. We successfully synthesized different types of Zr, Hf, and Ti clusters. Exploring the catalytic potential of zirconium and hafnium derivatives in esterification, we discovered the superiority of oxo clusters over traditional forms like MOFs and nanocrystals. Specifically, comparing UiO-66 MOF and ZrO ₂ nanocrystals with Zr ₁₂ clusters revealed the remarkable catalytic efficiency of clusters, attributed to their enhanced surface-to- volume ratio. Moreover, our analysis, including Pair Distribution Function studies, affirmed the structural integrity of the clusters, reinforcing their efficacy as catalysts in homogeneous, zirconium/hafnium-mediated esterification reactions. These findings provide crucial insights into the active catalytic species and pave the way for advanced applications in catalysis and materials science
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