ABSTRACT

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Title: Design and Synthesis of Porous Materials for Visible Light-Driven Carbon Dioxide Reduction: A Pathway to Sustainable Energy Solutions.

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Carbon dioxide (CO_2) reduction has emerged as a vital approach to addressing two pressing global challenges: climate change and sustainable resource management. As a major greenhouse gas, CO_2 has significantly contributed to global warming, primarily due to rapid industrialization and urban expansion over the past century. To mitigate its environmental impact, CO_2 can be treated not as waste but as a valuable raw material. Among the emerging technologies, photocatalytic CO_2 reduction stands out for its ability to convert CO_2 into valuable products such as fuels and chemicals by harnessing renewable, abundant, and inexpensive solar energy. This strategy not only helps reduce greenhouse gas emissions but also supports the development of a circular carbon economy, while simultaneously addressing the global energy crisis by enabling the production of value-added fuels and chemicals from CO_2. This scientific approach embraces the concept of transforming waste into valuable resources, paving the way toward a cleaner and more sustainable future.

To address these challenges, in this thesis, we focus on the design and development of a series of porous polymers including covalent triazine-based frameworks (CTFs), metal-organic frameworks (MOFs), and conjugated microporous polymers (CMPs) with diverse backbone architectures. These porous materials have been further employed as solid supports for the incorporation of redox-active metal centres and metal coordination complexes through various post-synthetic modification strategies. This approach aims to develop robust and efficient photocatalysts for sustainable CO_2 reduction into value-added chemicals under visible light irradiation. The core findings and detailed discussions are presented across four main chapters, namely **Chapters** 2, 3, 4, and 5. In **Chapter 2**, single-atom catalysts were successfully synthesized by anchoring Co^{2+} ions into a covalent triazine framework (CTF-TPE). The prepared catalysts are proved to be a potential catalyst for CO_2 to CO conversion. **Chapter 3** presents the fabrication of Zn (II)- and Co (II)-based imidazolate frameworks—IFP-1(Zn), IFP-5(Co), and a bimetallic IFP-1(Zn/Co)—achieved via partial substitution of Zn with Co, all demonstrating promising photocatalytic CO_2 reduction to CO activity. In **Chapter 4**, a Ru (II) bis-terpyridine complex was integrated into a Zr-based NDI-linked MOF via solvent-assisted ligand exchange, yielding Zr-NDI@Ru-tpy, which enabled selective CO production under visible light. Finally, **Chapter 5** describes the design of two metal–salen-based conjugated microporous polymers, salen-CMP@Co and salen-CMP@Zn, for syngas generation. Notably, salen-CMP@Co exhibited superior activity, and the CO/H_2 ratio could be precisely controlled by tuning the acetonitrile–water composition. Overall, this thesis offers valuable insights and contributes significantly to the advancement of photocatalytic CO_2 conversion for sustainable energy and environmental solutions.

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