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Title: Development of Heteroanionic Metal oxide Nanostructures for Solar and Mechanical Energy Harvesting

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**Abstract:** Water scarcity and the energy crisis are two interrelated global challenges that pose significant threats to environmental sustainability and human well-being. Water scarcity arises from the mismanagement of water resources, exacerbated by industrialization. Concurrently, the energy crisis stems from heavy reliance on fossil fuels, which not only deplete finite resources but also contribute to greenhouse gas emissions and climate change. Emerging technologies like photocatalysis and piezocatalysis offer promising solutions by harnessing inexhaustible and green renewable energy sources. Photocatalysis utilizes solar energy to drive chemical reactions for clean fuel production, water pollutant removal, and fine chemical synthesis. In parallel, piezocatalysis transforms mechanical energy into chemical energy. However, to fully unlock their potential, the development of high-efficiency and stable catalysts is urgently needed. Metal hetero-anionic oxides stand out in this endeavor, displaying remarkable visible light absorption capabilities attributed to their elevated valence bands, a consequence of multiple anions. In sharp contrast to conventional metal oxides, constrained by wide band gaps that hinder visible light capture, these materials possess the unique ability to harness visible light, offering a promising path to a more sustainable future. Moreover, their robust ferroelectric properties enable efficient mechanical energy conservation, ushering in an era where both photocatalysis and piezocatalysis can coexist and flourish. The research conducted in this thesis represents a groundbreaking endeavor, focusing on the sustainable synthesis of a novel category of hetero-anionic metal oxide, specifically the Sillen Aurivillius phase. This unique material holds immense promise for applications in wastewater treatment, energy harvesting, and organic transformation. What truly distinguishes this research is the remarkable discovery of the self-activation property inherent to the Sillen Aurivillius phase. Catalysis has long grappled with the challenge of catalyst degradation over time, often neglecting the crucial aspect of catalyst stability during catalytic processes. In this context, the development of an efficient and self-activating catalyst stands as a critical achievement. This breakthrough not only addresses the long-standing challenges in catalysis but also paves the way for a new era of sustainable and robust catalytic processes. In the first part of this thesis, the development of a sustainable method to synthesize the layered Sillen Aurivillius (SA) phase under ambient conditions and the assessment of its exceptional photocatalytic capabilities is described. The work focused on addressing the longstanding challenge posed by Rhodamine B (RhB), a stubborn and toxic textile dye that has plagued researchers for decades. The current work uncovered the potential of producing Rhodamine 110 (Rh110), an overlooked intermediate in RhB degradation. The approach utilizes RhB and the SA phase as catalysts to efficiently produce pure Rh110 under sunlight. Notably, this catalyst not only maintains stability but also exhibits self-activation during successive cycles, a standout trait among photocatalysts. The production of Rh110 was further improved by increasing perovskite layers in the SA phase. In the second part, the SA phase's piezocatalytic activity was explored for the first time, specifically in piezocatalytic water splitting. Through high-pressure synchrotron X-ray diffraction and theoretical studies, we gained insights into the catalyst's crystal structure and electronic properties under piezocatalytic conditions. Lastly, the synergistic effects in piezo-photocatalytic wastewater treatment using real-world mimicking pollutant feedstock were demonstrated and the origin of the same was illustrated.

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