Title: Computational Investigation of AlCoCrFeNi High-Entropy Alloy as a Catalyst for CO_2 and CO Reduction Reactions

Nabila Tabassum[1], Yamini Sudha Sistla[*]
IIT Roorkee[1], Shiv Nadra Institution of Eminence, Greater Noida*

The increasing concentration of carbon dioxide (CO_2) and carbon monoxide (CO) in the atmosphere has become a major environmental concern, driving the demand for advanced catalytic materials capable of converting these harmful gases into useful products. High-entropy alloys (HEAs), characterized by their multi-principal element composition and unique physicochemical properties, have recently emerged as promising candidates for catalytic applications due to their exceptional structural stability, tunable electronic configuration, and synergistic elemental interactions. In this work, the catalytic potential of AlCoCrFeNi high-entropy alloy (HEA) toward CO_2 and CO reduction reactions is systematically investigated through computational approaches.

The primary objective of this study is to evaluate the adsorption, activation, and reduction behavior of CO_2 and CO molecules on the surface of AlCoCrFeNi HEA and to understand the role of its constituent elements in enhancing catalytic performance. Structural optimization of the HEA system is first performed to determine the thermodynamic stability and equilibrium lattice configuration. Surface slab models are then constructed to simulate the exposed catalytic facets, followed by adsorption studies of CO_2 and CO molecules at various active sites on the alloy surface.

Adsorption energy calculations are carried out to identify the most favorable adsorption configurations and determine the interaction strength between gas molecules and the HEA surface. Further analysis of molecular geometry, including bond elongation and bending angle variation, is employed to evaluate the activation of adsorbed species. Charge transfer mechanisms and electronic interactions between adsorbates and surface atoms are examined using charge density analysis and density of states (DOS) calculations to reveal the underlying catalytic mechanism.

The multicomponent nature of AlCoCrFeNi HEA is expected to provide diverse active sites and enhanced electronic coupling, promoting stronger adsorption and improved catalytic activity compared to conventional binary or ternary alloys. Transition metal constituents such as Fe, Co, and Ni are anticipated to contribute significantly to d-band center modulation, thereby facilitating adsorption and intermediate stabilization during reduction reactions.

The outcomes of this study will provide theoretical insights into the design of HEA-based catalytic materials and establish AlCoCrFeNi HEA as a potential multifunctional catalyst for sustainable carbon capture and conversion technologies. This research contributes to the growing field of computational catalyst design and offers a pathway toward developing efficient materials for environmental remediation and clean energy applications.

Keywords: High-Entropy Alloy; AlCoCrFeNi; CO_2 Reduction; CO Reduction; Catalysis; Density Functional Theory; Adsorption; Electronic Structure.