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論 文 要 旨

THESIS SUMMARY

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要旨（英文 800 語程度）
Thesis Summary (approx.800 English Words)

This thesis, entitled "Material Informatics Approaches to Explore the Potential of Ternary Metal Sulfide as the CO_2 Reduction Electrocatalyst," aims to make a significant contribution to the early-stage exploration of ternary metal sulfides as potential sustainable green energy materials for CO_2 reduction electrocatalysis. It introduces innovative empirical material informatics strategies to overcome the challenges associated with studying metal sulfides, providing a novel perspective on material design and optimization. In summary, this thesis consists of eight chapters: five main chapters and three supporting chapters. The discussion of material informatics in this thesis is constructed around two main topics: (1) material screening and (2) insight for material development. The first strategy aims to develop an alternative approach to exploring metal sulfides, while the second strategy aims to elucidate the CO_2 reduction reaction (CO_2RR) activity of ternary metal sulfides, specifically focusing on the role of sulfur.

Chapter 1, "Introduction," briefly introduces the problems of CO_2 emissions and the potential of electrochemical CO_2RR using metal sulfides as a sustainable renewable energy solution. It describes the obstacles of using single metal sulfides as electrocatalysts and emphasizes the importance of elevating the system to a bi-metallic (ternary metal sulfide) system. The chapter also highlights the current lack of discussion on ternary metal sulfides, underscoring the challenge of investigating trends and studying activity-determining factors.

Chapters 2 and 3 discuss empirical material informatics strategies for the early-stage screening of ternary metal sulfides.

Chapter 2, "Empirical Approach for Early-Stage Ternary Metal Sulfide Exploration," presents a developed burdenless material screening workflow. This methodology leverages experimental data and integrates density functional theory (DFT) calculations with machine learning (ML) without requiring high-throughput experimental tools. The goal is to clarify which properties should be prioritized to predict and obtain optimal electrocatalysts. This study employs high-dimensional regression ML models trained on a dataset of 18 samples, specifically targeting ternary metal sulfides selective for syngas carbon monoxide (CO) production. The CO selectivity trend of ternary metal sulfides is mapped using two low-cost computational properties: structure volume and DFT surface total energy. Additionally, insights into the challenges of describing ternary metal sulfides selective for hydrocarbons (CH) production are provided.

Chapter 3, "The Alternative Descriptor for Predicting CO-Selective Ternary Metal Sulfide," offers a more profound analysis based on the ML models from Chapter 2, aiming to provide straightforward guidance for early-stage CO-selective ternary metal sulfide screening. This chapter analyzes new, unseen ternary metal sulfide structures from open-source databases, emphasizing the importance of considering crystal structure beyond atomic composition in catalyst design. It identifies that ternary metal sulfides with hexagonal lattice systems containing two cations among Zn/In/Cd are optimal for CO-selective electrocatalysts.

Chapters 4 and 5 discuss empirical material informatics strategies for studying the dynamics of metal sulfides, focusing on the role of sulfur during electrochemical CO2RR-a significant experimental challenge.

Chapter 4, "Machine Learning Interatomic Potential (MLP) for Material Simulation," introduces a

machine learning technique for molecular dynamics (MD) simulation, the MLP method, advantageous for low-cost material simulation. This chapter provides an example of artificial neural network (ANN)-based MLP construction on a vast, multi-component open-source dataset and applies it to a specific MD application. It also addresses a strategy for low-cost MLP construction using transfer learning and subset selection, offering potential benefits for the broader community.

Chapter 5, "Dynamic Behavior Study of Ternary Metal Sulfide during Electrochemical CO_2RR," investigates the role of sulfur during electrochemical CO_2RR using the MLP method from Chapter 4. This chapter focuses on $ZnIn_2S_4$ due to its high selectivity for CO products. A database of $ZnIn_2S_4$ is empirically constructed based on various experimental results, including XRD, in situ XAFS, in situ FTIR, and XRF. A simulation benchmark with In_2S_3 is also conducted to clarify the advantages of the bi-metal system.

Chapter 6, "Scientific Contribution of This Work," outlines the scientific contributions of this study. This work offers a novel perspective on material design strategies, introducing an empirical approach that does not rely on high-throughput setups. It presents a generalizable and burdenless method for material exploration, with potential applicability across various research interests. Additionally, this study provides valuable insights into the flexibility of sulfur as a specific parameter for determining the CO_2RR activity of metal sulfides.

Chapter 7, "Conclusion," summarizes the overall conclusions.

Chapter 8, "Appendix A," serves as a supporting data for statements in the main body of the thesis.

Chapter 9, "Appendix B," is a specific extension of Chapter 4, details my research on the cost-effective construction of MLPs using the transfer learning method. This aspect of the research was set aside from the main work because applying transfer learning to metal sulfides is still not fully supported. Nevertheless, the findings from this work hold potential benefits for the broader community.

Overall, this study lays the groundwork for future research by demonstrating the practical application of these methodologies across various fields, from energy storage to environmental remediation. By bridging the gap between qualitative observations and quantitative analysis, this thesis also offers a comprehensive understanding of the factors influencing CO_2RR activity in metal sulfides, paving the way for the development of more efficient and selective catalytic systems.