学 位 論 文 内 容 の 要 旨

博士の専攻分野の名称　博士（総合化学）　氏名　ZHANG Meiqi

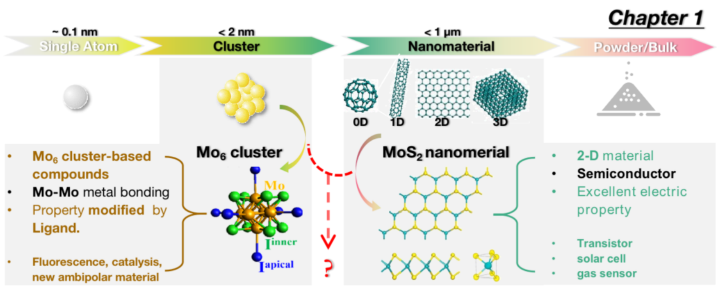
学 位 論 文 題 名

Exploration and development of novel functional MoS_2–based nanomaterials.

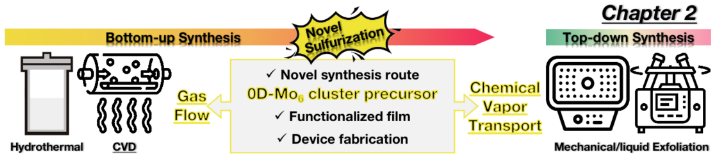
（新規機能性 MoS_2 系ナノ材料の探索と開発）

Transition metal dichalcogenides (TMDs), represented by molybdenum disulfide (MoS_2) are attracting much attention from researchers and have a rapid development in recent years. The MoS_2 nanomaterial has shown great application potential in the fields of optoelectronics, new energy, biosensing, and catalytical (including thermal catalysis, electrocatalysis, and photocatalysis). It is also expected to be applied as next-generation semiconductor materials in the industry application like transistors because of its unique tunable electronic structure, high carrier mobility.

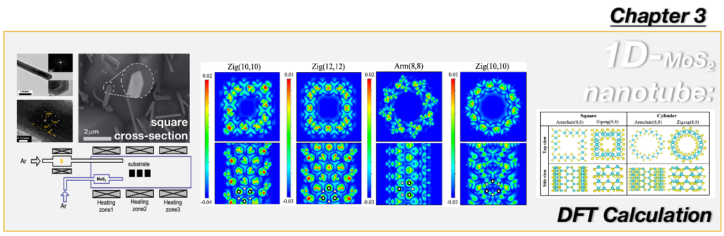
With the development of synthesis, controlled growth methods, and defect engineering, researchers have devoted themselves to the synthesis of functional MoS_2 nanomaterials based on various dimensions (0-3D). The high-performance defect-rich MoS_2, dominated by sulfur vacancies, as well as Mo-rich environments, have been extensively studied, which are attributed to the under-coordinated Mo atoms playing a great role as catalytic sites in the defect area.



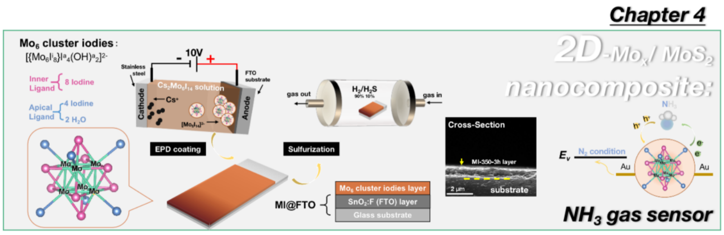
In this thesis, I focus on the novel functional MoS_2-based nanomaterials with various dimensions, which have an under-coordinated Mo atom or Mo cluster core. This thesis consists of six chapters: **chapter 1** presents the current status of MoS_2 nanomaterial and the significance of this research; **chapters 2-5** describe the results of this research; and **chapter 6** summarizes general conclusions of my research and future prospects.



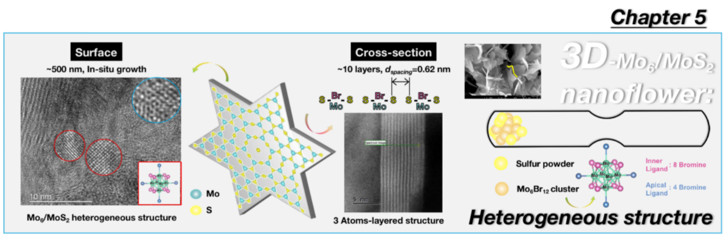
In the chapter 2, the novel synthesis strategy to generate MoS_2 by sulfurized 0D-Mo_6 cluster powder as a precursor have been explored. Traditional MoS_2 synthesis methods (e.g., chemical vapor deposition, hydrothermal synthesis, etc.) have been extensively investigated, and the use of catalysts to grow MoS_2 has also started to attract the attention of researchers. It has been reported that the growth of 1D-MoS_2 nanowires with rectangular cross-sections by using FeO nanoparticles as catalysts. This chapter firstly reviews and introduces these synthetic methods and their products, and then boldly explores and establishes a pioneering system for the sulfurization of 0D-Mo_6 cluster powder as a precursor to generate MoS_2 nanomaterial by gas flow method and chemical vapor transport reaction. The experimental results prove that this sulfurization strategy is indeed feasible.



In the chapter 3, a computational study on 1D-MoS_2 nanotubes with square cross sections is elucidate the growth mechanism and properties of experimentally synthesized nanotubes which is introduced in Chapter 2. The results show that these square nanotubes have strain energies lower than the traditional cylindrical ones in the small-diameter region. They also show a much smaller band gap and higher surface energies compared with the cylindrical ones. Zigzag and armchair chirality shows qualitatively different electronic structures about the localization of the edge states because of the under-coordinated Mo atoms located at the edges.



In the chapter 4, the Mo cluster-MoS_2 nanoparticle-built 2D thin film was creatively synthesized through the one-step sulfurization process, which is the development of the H_2/H_2S gas flow method based on Chapter 2. The Mo_6 cluster iodides nanoparticle (MI) coated on the fluorine-doped tin oxide (FTO) glass substrate via the electrophoretic deposition method (i.e., MI@FTO) were used as a precursor to form a thin film nanocomposite. Detailed investigations of the structure, reaction mechanism, and NH_3 gas sensing performance were carried out. The Mo_x cluster-MoS_2 thin film nanocomposites ($1 \leqslant x \leqslant 6$) were characterized and measured as gas sensors for the first time. Results indicated that after the sulfurization process, the response of MI@FTO for NH_3 gas increased by three times while showing conversion from p-type semiconductor to n-type, which enriches their possibilities for future device applications.



In the chapter 5, the 3D Mo_6/MoS_2 nanoflower was creatively synthesized through the sulfurization process in a vacuum which is the development of the Chemical Vapor Transport Reactions base on the Chapter 2. The significant effect of the pressure of the reaction system in a vacuum tube on the final MoS_2 product was revealed for the first time, laying the foundation for future Mo_6 cluster sulfurization processes. The structure of this star-shaped nanosheet has been observed and analyzed in detail, and successfully demonstrated that it has a novel Mo_6 cluster/MoS_2 heterogeneous structure for the first time.

In the chapter 6, overall summary of this thesis was presented.