学 位 論 文 内 容 の 要 旨

博士の専攻分野の名称　　博士（工学）　　氏名　林　學毅

学 位 論 文 題 名

Multielement–Doped Tungstic Acids via Submerged Photosynthesis for Enhanced All-Solar Photoelectrochemical Responses

(全太陽光電気化学応答のためのマルチ元素タングステン酸の水中光合成)

The purpose of this dissertation is to achieve low-cost all-solar utilization by tungstic acid synthesized via submerged photosynthesis of crystallites method.

Solar energy utilization is an excellent strategy for renewable energy production. Since sunlight covers a vast area of the Earth's surface, enabling individual households scattered across various regions to independently generate solar power is more important than constructing large centralized solar power plants. One of the drawbacks of solar energy is the instability of sunlight exposure. Therefore, stable methods of solar energy storage are essential.

Photoelectrochemical energy storage materials, which allow both solar power generation and energy storage within the same material, offer a more efficient alternative compared to other storage methods and significantly reduce costs and physical storage space. However, photoelectrochemical energy storage materials are quite rare. Tungsten acids, as one of the candidates, face the challenge of poor absorption efficiency in the visible and infrared light regions —an issue commonly found in many solar energy materials. One potential solution to this problem is multielement doping. Nevertheless, existing multielement doping methods typically require high temperatures and involve complex or toxic precursors.

The submerged photosynthesis of crystallites method is a novel material synthesis technique. By utilizing light irradiation energy as an additional driving force for crystallization reactions, it enables the synthesis of nanoparticles in a simple aqueous solution under ambient temperature and pressure. However, there have been no reports of using this method for multi-element doping.

Briefly, to achieve low-cost solar energy storage, it is necessary to evaluate the feasibility of multi-element doping using the submerged photosynthesis of crystallites method and to analyze the photoelectrochemical storage performance of the resulting doped tungstic acids.

In Chapter 1, I will briefly review the status of solar energy utilization and storage, including various forms of solar energy storage, and an introduction to nanomaterials and their fabrication processes. Finally, I will explain the development and principles of the submerged photosynthesis of crystallites method and describe the purpose of this dissertation.

In Chapter 2, I first attempted to compare the tungstic acids generated under UV irradiation and in a dark environment. The results showed that, in terms of both particle size and crystallinity, the samples formed in the dark were significantly inferior to those formed under UV light. This preliminarily confirmed the necessity of UV irradiation in the synthesis process.
In Chapter 3, I introduced Mo elements into the reaction solution. It was found that varying Mo concentrations led to the formation of three different crystalline phases. Among them, the Mo–W nanocomposite crystalline phase, exhibited the best light absorption and electrochemical performance. This composition was subsequently used in Chapter 4.
In Chapter 4, I incorporated Cu, Fe, and Mn as doping elements into the solution. Electron microscopy analysis confirmed that the submerged photosynthesis of crystallites method can successfully synthesize multielement–doped tungstic acids. The absorbance analysis showed a 27 % improvement in solar light absorption efficiency. Doping significantly increased the photogenerated charge carriers. Combined with the planar defects induced by doping, the photogenerated capacitance increased by 18.7 times. These results indicate that multielement doping achieved through the submerged photosynthesis of crystallites method is an effective approach to enhance solar energy storage.
In Chapter 5, I provide a summary of the entire dissertation and point out that future studies should further explore in situ analysis techniques, crystal orientation control, and substrate-based growth in order to realize the vision of all-solar light utilization.
This dissertation has confirmed that multielement–doped tungstic acids can be synthesized by UV irradiation under room temperature. By adjusting the doping elements, it can achieve all-solar utilization.