錯体重合法をベースに合成した $Cu_2Sn_xGe_{1-x}S_3$ 粉末による光電気化学的な水素生成

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Photoelectrochemical hydrogen production by $Cu_2Sn_xGe_{1-x}S_3$ powder synthesized based on polymerized complex method ([1]Faculty of Engineering, Shinshu University, [2]Institute of Multidisciplinary Research for Advanced Materials, Tohoku University, [3]The University of Tokyo) ○Yusuke Ooka,[1] Sota Shiga,[1] Yosuke Kageshima,[1] Hiromu Kumagai,[2] Katsuya Teshima,[1] Kazunari Domen,[3] Hiromasa Nishikiori[1]

We have previously reported $Cu_2Sn_xGe_{1-x}S_3$ (CTGS) powder as a photoelectrode material that can drive photoelectrochemical hydrogen production reactions with relatively high efficiency using long wavelength light up to the near-infrared region. However, CTGS particles synthesized by the conventional solid-state reaction (SSR) had a wide particle size distribution and ragged shapes. Thus, there is room to improve the activity by improving the crystal quality.

In this study, we synthesized high-quality, fine-grained CTGS powder by sulfurizing the oxide precursor prepared by polymerized complex (PC) method. It was found that tuning the Sn/Ge ratio enabled the control of absorption edge wavelength as well as the improvement of the photoelectrochemical performances (Fig. 1a). In addition, CTGS synthesized based on the PC method showed the superior activity to that synthesized by the SSR (Fig. 1b). In the presentation, the characterizations of the synthesized CTGS particles will be also discussed in detail.

Keywords: Photocatalysts, Photocathodes, Hydrogen Evolution, Sulfide, Composition

近赤外領域までの長波長光を利用し、比較的高効率に光電気化学的な水素生成反応を駆動可能な光電極材料として、これまでに $Cu_2Sn_xGe_{1-x}S_3$(CTGS) 粉末を報告している[1]。しかし従来の固相法で合成した CTGS は粒度分布が広く不定形であるため、結晶品質改善による活性向上の余地が残る。

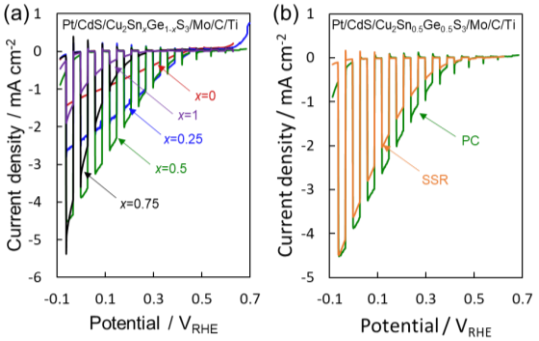

Fig. 1 (a) Current-potential curves for the particulate CTGS photocathodes with various Sn/Ge composition. (b) Current-potential curves for CTGS photocathodes synthesized by SSR or PC method.

本研究では、錯体重合法により合成した酸化物前駆体を硫化することで、高品質・微細な CTGS 粉末の合成を試みた。Sn/Ge 比を制御することで吸収端波長を制御可能であり、光電気化学特性を向上可能であることを見出した(Fig. 1a)。また、錯体重合法ベースで合成した CTGS は、既往の固相法で合成した CTGS よりも高い光電流値を示した(Fig. 1b)。発表では、合成した CTGS 粉末のキャラクタリゼーションについても詳細に議論する。

1) Y. Kageshima, *et. al.*, *J. Am. Chem. Soc.* **2021**, *143*, 5698–5708