

Photocatalytic performance of $\text{Li}_2\text{LaTa}_2\text{O}_6\text{N}$ layered oxynitride for H_2 evolution and CO_2 reduction

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Depletion of fossil fuels and serious environmental problems has grown our interest toward renewable energy sources. For example, production of H_2 by water splitting and reduction of CO_2 into useful carbon compounds using exhaustible sunlight are both attractive. Semiconductor photocatalyst is one of the most promising materials to induce chemical reactions by using light energy. Among semiconductor materials, layered compounds are especially fascinating due to its unique photocatalytic properties derived from the layered structure as have demonstrated so far [1]. However, most of layered compounds are active only under UV light irradiation, and thus it is desirable to develop a new layered material that works under visible light. In this work, we present a synthesis of layered oxynitride $\text{Li}_2\text{LaTa}_2\text{O}_6\text{N}$ and the photocatalytic properties for H_2 evolution and CO_2 reduction under visible light.

$\text{Li}_2\text{LaTa}_2\text{O}_6\text{N}$ was obtained via ammonia nitridation of a precursor oxide prepared by polymerized complex method at 1173 K. XRD pattern of the resulting material is shown in Fig. 1A. Although a very small diffraction peak attributable to LaTaON_2 was detected, the diffraction pattern was almost identical to that of $\text{Li}_2\text{LaTa}_2\text{O}_6\text{N}$ [2], indicating successful synthesis of the compound. In addition, obtained $\text{Li}_2\text{LaTa}_2\text{O}_6\text{N}$ exhibited a visible light absorption in diffuse reflectance spectrum (DRS, Fig. 1B), with a band gap energy of approximately 2.5 eV that was estimated from the absorption edge of the spectrum.

First, H_2 evolution activity of $\text{Li}_2\text{LaTa}_2\text{O}_6\text{N}$ was examined from methanol aqueous solution with the aid of Pt as H_2 evolution co-catalyst. H_2 was evolved under visible light irradiation. However, the evolution rate was gradually

decreased over irradiation. DRS measurement revealed that the absorption edge was shifted to shorter wavelength region after the reaction, indicating elimination of nitrogen from lattice and instability of the compound in aqueous environment.

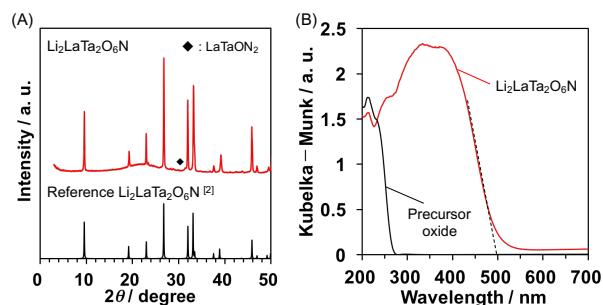


Fig. 1 (A) XRD and (B) DRS of $\text{Li}_2\text{LaTa}_2\text{O}_6\text{N}$.

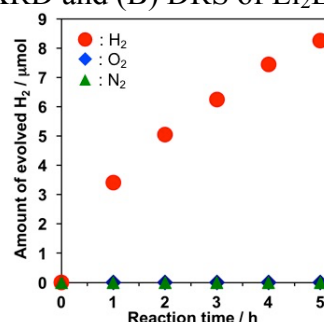


Fig. 2 Time course of H_2 evolution over Pt/ $\text{Li}_2\text{LaTa}_2\text{O}_6\text{N}$ under visible light ($\lambda \geq 400$ nm).

$\text{Li}_2\text{LaTa}_2\text{O}_6\text{N}$ was also tested for CO_2 reduction combined with a Ru-Ru binuclear complex in acetonitrile solution containing triethanolamine as an electron donor. After 15 h irradiation, 120 μmol of formic acid was detected as CO_2 reduction product. The turnover (the number of produced formic acid to the adsorbed complex) exceeded 100, confirming catalytic cycle. The activity was relatively high compared to similar CO_2 reduction systems previously reported using CaTaO_2N and Y-Ta oxynitride [3, 4].

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