Development of $BaTaO_2N$ photocatalysts with distinctive carrier dynamics for one-step excitation overall water splitting

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Keywords: $BaTaO_2N$; photocatalysts; overall water splitting; cocatalyst; carrier dynamics

$BaTaO_2N$ (BTON) is an attractive perovskite oxynitride material and visible-light responsive photocatalyst, owing to its suitable band structures for hydrogen and oxygen evolution reactions. To date, several strategies have been employed to produce BTON with considerably less defects such as flux-assisted approach.[1-2] However, the one-step excitation overall water splitting (OE-OWS) utilizing BTON as the photocatalysts remains elusive to achieve. Herein, we report BTON that demonstrates OE-OWS and unravel its characteristics suitable for simultaneous production of H_2 and O_2 in pure water condition and under visible light irradiation.

The BTON was synthesized using amorphous Ta_2O_5 as a Ta precursor and blended with $BaCO_3$ as a Ba precursor, followed by nitridation under NH_3 gas. With suitable hydrogen evolution reaction (HER) and oxygen evolution reaction (OER) cocatalysts, the BTON split water to H_2 and O_2 at the nearly stoichiometric ratio (2.3 and 1.1 μmol/h for H_2 and O_2, respectively), suggesting successful observation of OWS utilizing BTON as the photocatalysts. The carrier dynamics of BTON synthesized from amorphous Ta_2O_5 was investigated by transient absorption spectroscopy to reveal its distinctive characteristics compared to other BTONs nitrided using the commercially available Ta_2O_5 precursors and with the addition of fluxes. Under 470 nm laser pulse excitation, the mid-IR absorption spectra of photocarriers in BTON exhibited monotonic increase behavior towards lower probe energies, indicating notable free carrier absorption characteristics, as depicted in Figure 1. This absorption feature has not been observed previously for BTON synthesized using conventional Ta_2O_5 precursor.[1] Detailed results will be presented at the conference.

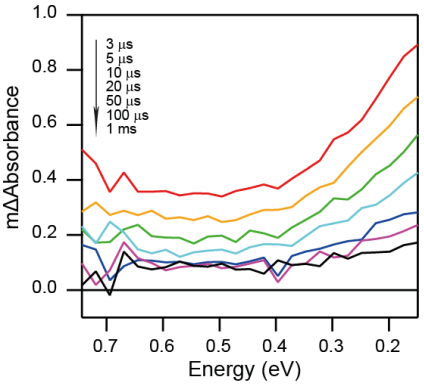

Figure 1. Transient absorption spectra of BTON BTON nitrided at 1173 K for 5 h under 470 nm laser pulses excitation. Measurement condition: room temperature and in N_2 (20 Torr).

References:

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