

Nitrided Layered Ruddlesden–Popper Phase KLaTiO_4 and $\text{K}_2\text{La}_2\text{Ti}_3\text{O}_{10}$ Crystals: Photocatalytic Water Oxidation Activity and Fabrication of Their Nanosheets

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Ultrathin nanosheets, with infinite planar lengths and atomic or molecular thickness, presenting intriguing physical, chemical and electronic properties compared to their bulk counterparts are extensively regarded as an emerging class of the thinnest advanced functional nanomaterials for various applications. A wide variety of layered d^0 transition metal (Ti^{4+} , Nb^{5+} , Ta^{5+} , Mo^{6+} , and W^{6+}) oxides have been exfoliated into their elementary host layers. In addition to the Ruddlesden–Popper (RP) and Aurivillius (AV) phase oxide nanosheets, the Dion–Jacobson (DJ) phase oxide nanosheets were also fabricated by exfoliation, and an enhancement in their photocatalytic activity was researched. In this work, we have synthesized RP phase KLaTiO_4 and $\text{K}_2\text{La}_2\text{Ti}_3\text{O}_{10}$ crystals by solid-state reaction and studied their thermal stability under an NH_3 flow at 800 °C for 3, 5, 7, and 10 h to obtain their phase-pure oxynitride counterparts. With the maximum nitridation time of 10 h, the nitrided KLaTiO_4 and $\text{K}_2\text{La}_2\text{Ti}_3\text{O}_{10}$ crystals maintained the crystal structures of the oxide precursors and showed the longest absorption edges at about 540 and 600 nm, respectively. We have also investigated the effect of protonation on photocatalytic water oxidation activity of the pure and nitrided KLaTiO_4 and the pure, nitrided, and re-oxidized $\text{K}_2\text{La}_2\text{Ti}_3\text{O}_{10}$ crystals without cocatalyst under visible light. The $\text{HLaTi}(\text{O},\text{N})_4$ and $\text{H}_2\text{La}_2\text{Ti}_3(\text{O},\text{N})_{10}$ crystals exhibited higher photocatalytic activity compared to other samples. The nanosheets of the pure and nitrided KLaTiO_4 and the pure, nitrided, and re-oxidized $\text{K}_2\text{La}_2\text{Ti}_3\text{O}_{10}$ crystals were fabricated through the protonation-intercalation-exfoliation processes as well as by sonication. As shown in Figure 1, thin nanosheets of the nitrided KLaTiO_4 crystals can easily be dispersed in water.

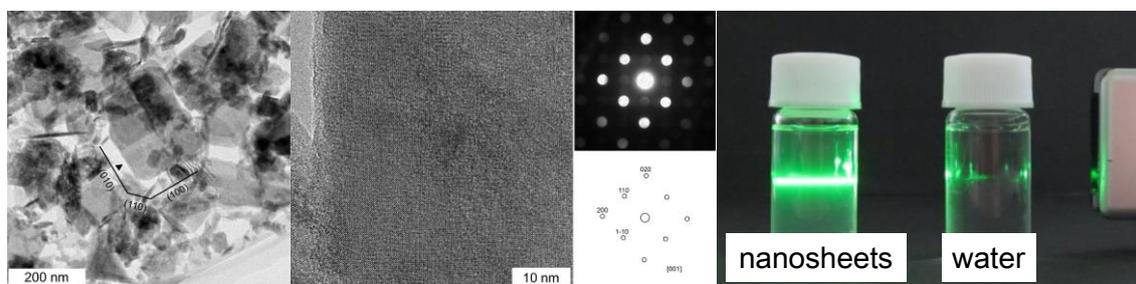


Figure 1. TEM and HRTEM images, SAED pattern and the colloidal suspension of the fabricated oxynitride $[\text{LaTi}(\text{O},\text{N})_4]$ nanosheets.