

*Regular Paper***Optical Floating Zone Crystal Growth of Rare-Earth Tantalates $RE_8Ta_2O_{17}$ (RE = Gd, Y, and Lu)****Yueshen ZHOU^{1,2}, Dongsheng YUAN^{1,*} and Kiyoshi SHIMAMURA^{1,2,*}**¹*National Institute for Materials Science, Tsukuba 305-0044, Japan.*²*Graduate School of Advanced Science and Engineering, Waseda University, Shinjuku, Tokyo 169-8555, Japan.*

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Abstract

The bulk crystal growth of rare-earth tantalates $RE_8Ta_2O_{17}$ (RE = Gd, Y, and Lu) was first achieved using the optical floating zone method. The crystals are colorless and exhibit a high transparency with an absorption edge at ~ 265 nm. They are characterized in fluorite-type structure $Fm-3m$, and their compositions were found shifted from the nominal RE/Ta ratio of one, being smaller as the ionic radius of RE increases. Under light excitation of 230 nm, all the $RE_8Ta_2O_{17}$ samples emit at ~ 280 nm originating from Ta-O charge transfer. However, undoped $RE_8Ta_2O_{17}$ series only emit a weak luminescence due to the strong self-absorption of their UV emissions.

Keywords: Floating zone technique, Rare-Earth Tantalates, $RE_8Ta_2O_{17}$, Crystal

1. Introduction

Rare-earth tantalates exhibit promising potentials across various applications, including refractories, scintillators, laser hosts, and oxygen ion conductors [1-3]. They are characterized by high melting points due to the refractory nature of RE_2O_3 and Ta_2O_5 , particularly for compounds with large RE_2O_3 ratio, thus their single-crystal growth is not easy by the conventional Czochralski and Bridgeman methods. Consequently, most of the studies so far are focused on ceramics [4-9].

Among this family, Gd_2O_3 , Y_2O_3 , and Lu_2O_3 based binary tantalates attract the most attention as hosts for laser and luminescence. Yokogawa et al. studied the phase relations within the Gd_2O_3 - Ta_2O_5 and Y_2O_3 - Ta_2O_5 systems [4,5]. By solid-state reactions and thermal analyses, they confirmed the congruently melting compounds $RETaO_4$ (RE = Gd, Y) at approximately 50% RE_2O_3 . Furthermore, a fluorite-type cubic phase (F phase, space group $Fm-3m$) was found at around 80% RE_2O_3 , being stable above 1500°C but decomposing into RE_2O_3 & RE_3TaO_7 during cooling [4,5]. This F phase exhibited disordered cation and anion sublattices [10]. In a similar vein, Xing et al. explored different

Lu_2O_3 - Ta_2O_5 compounds through solid-state reactions, constructing a phase diagram spanning 0-100 mol% Lu_2O_3 [6]. Unlike in the Gd-Ta and Y-Ta systems, the melt with a molar ratio of 1:1 did not yield a single-phase $LuTaO_4$ but with a coexistence of Lu_3TaO_7 . A single cubic phase Lu_3TaO_7 was also identified, melting congruently within the range of 70-75 mol% Lu_2O_3 . But for the composition $\sim 80\%$ Lu_2O_3 , the F phase became unstable at high temperatures exceeding 1500°C [6].

Considering the relatively high density of $RE_8Ta_2O_{17}$ and the possible luminescence from Ta-O charge transfer centers (commonly observed in tantalates) [7, 11], its single crystal growth is highly demanded in order to evaluate the optical properties as intrinsic scintillators.

Herein, we used the optical floating zone method to grow the cubic phase $RE_8Ta_2O_{17}$. The crystals were characterized in terms of crystal structure, chemical composition, optical transmittance, and photoluminescence properties.

2. Experimental method

The growth of $RE_8Ta_2O_{17}$ (RE = Gd, Y, and Lu) transparent crystals was conducted in Ar atmosphere using an optical floating-zone furnace (FZ-T-12000-X-I-VPM-NS-PC, Crystal Systems

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