Journal of Crystal Growth (xxxx) xxxx–xxxx

Contents lists available at ScienceDirect

Journal of Crystal Growth

journal homepage: www.elsevier.com/locate/jcrysgro

Improvement of dopant distribution in radial direction of single crystals grown by micro-pulling-down method

Yuui Yokota[a,*], Tetsuo Kudo[b], Valery Chani[b], Yuji Ohashi[b], Shunsuke Kurosawa[a], Kei Kamada[a,c], Zhong Zeng[d], Yoshiyuki Kawazoe[a], Akira Yoshikawa[a,b,c]

[a] New Industry Creation Hatchery Center (NICHe), Tohoku University, Japan
[b] Institute for Materials Research, Tohoku University, Japan
[c] C&A Corporation, Japan
[d] Department of Engineering Mechanics, College of Aerospace Engineering, Chongqing University, China

ARTICLE INFO

Communicated by Francois Dupret

Keywords:
A1. Segregation
A2. Growth from melt
A2. Single crystal growth
B1. Oxides
B3. Scintillators
B3. Solid state lasers

ABSTRACT

Two iridium crucibles with one and five capillaries at a nozzle were developed for the growth of Ce-doped $Y_3Al_5O_{12}$ [Ce:YAG] single crystals by the micro-pulling-down [μ-PD] method. The purpose of the study was to examine the effect of the capillary number on the Ce distribution in the radial direction of the crystals. The crystals grown from 2 mol% and 5 mol% Ce-doped YAG melts were then cut perpendicular to the growth direction and polished to produce specimens suitable for the measurement of Ce distribution. The Ce:YAG crystals grown using the crucible with one capillary [Ce:YAG(1 C)] had greater Ce content in the central portion of the crystal when compared to its peripheral parts. On the other hand, in the case of the Ce:YAG crystal grown using the crucible with five capillaries [Ce:YAG(5 C)], the increased Ce concentration was detected in five specimen sections positioned just below the capillaries. The results indicated that increase of the number of capillaries is effective practice that results in an improvement of the radial homogeneity of the single crystals grown by the μ-PD method.

1. Introduction

The micro-pulling-down [μ-PD] technique is one of the methods of single crystal growth from the melt. It is widely used for materials research because it is relatively simple and exceptionally fast when compared with conventional growth methods [1–5]. Fig. 1(a) presents a schematic diagram of the typical μ-PD system developed for the growth of oxide single crystals using a radio-frequency [RF] induction heating. In such μ-PD configuration, a metal crucible plays the dual role of the melt container and the heater due to its placement into the high-frequency electromagnetic field produced by the RF inductor. The bottom of the crucible is equipped with a nozzle (or die) that is produced with one or several capillary channels. These channels are responsible for the continuous delivery of the melt from the crucible container to the bottom of the nozzle as illustrated in Fig. 1(b). At the beginning of the growth, when the starting materials are completely melted, the seed is displaced in the upward direction to establish its physical contact with the melt sited on the bottom of the nozzle. This event corresponds to growth initiation. Thereafter, the growth is progressed by pulling-down the melt and its solidified part (crystal) in the downward direction. A relatively high temperature gradient in the vicinity of the liquid-solid interface and the small size (cross-section) of the grown crystal allow application of growth rates that are much larger than those used in conventional methods such as Czochralski [Cz], Vertical Bridgman [VB], and Floating Zone [FZ] techniques.

Many optical and scintillating materials are composed of a host (matrix) substance that contains one or several doping ions as (an) emission center(s). Generally, the dopant concentration affects optical and scintillation properties of the material [6–8]. Therefore, inhomogeneity of the dopant distribution in the single crystals causes degradation of the optical and scintillation properties and uncertainty of the material parameters and material performance. Experimental and simulation results on the dopant distribution in the single crystals grown by the μ-PD method were discussed in the past [9–13]. The results indicated that the dopant concentration in the central parts (regarding cross-sectional cuts) was larger than that in the peripheral parts when the segregation coefficient, k, of the dopant exceeded unity ($k > 1$). Oppositely, the dopant content in the center of the crystal was lower than that around the periphery in the case of $k < 1$. Thus, the

* Correspondence to: 2-1-1, Katahira, Aoba-ku, Sendai, Miyagi 980–8577, Japan.
E-mail address: yokota@imr.tohoku.ac.jp (Y. Yokota).

http://dx.doi.org/10.1016/j.jcrysgro.2016.11.119

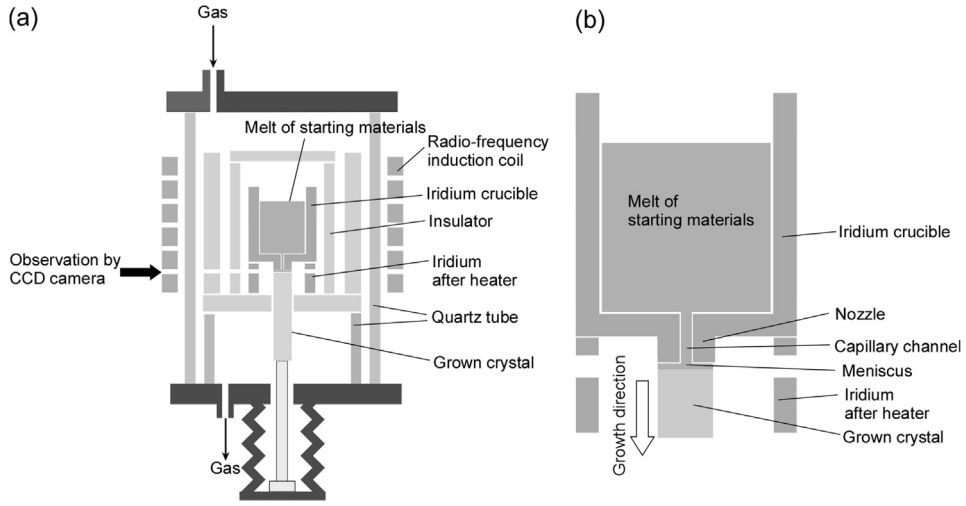

Fig. 1. (a) Schematic diagram of the μ-PD system used for the growth of oxide crystals and (b) details of the μ-PD crucible setup.

inhomogeneity of the dopant distribution in the single crystals grown by the μ-PD method is an important issue that should be examined in details. It is assumed that improvement of the uniformity of the μ-PD crystals may result in an improvement of their optical and scintillation properties.

Based on the above considerations, we simulated the dopant distribution in the radial direction of single crystals grown by the μ-PD method using the parameters of Ce-doped $Y_3Al_5O_{12}$ [Ce:YAG] together with crucibles with one and five capillaries in the nozzle to improve the homogeneity of the dopant distribution [14].

2. Simulation model and results

The dopant distribution in the radial direction of Ce:YAG single crystals grown by the μ-PD method using crucibles with one and five capillaries was simulated numerically. The computational model only includes the capillary-channel and the meniscus zone. Details of the simulation model was described in [14]. The equilibrium segregation coefficient of the Ce ion in a YAG single crystal is much less than one [12] and the Ce ion retains preferentially in the melt rather than the crystal during the crystal growth. In the simulation, the melt is taken as an incompressible Newtonian fluid with constant viscosity and density. The results of the simulations demonstrated that the application of crucibles with a multi-channel capillaries design could improve the dopant distribution in the radial direction. In such conditions, the Ce concentration at the spots corresponding to the positions of the capillaries could be larger at several locations as compared to the unique location in the Ce:YAG crystals grown using the crucible with a single capillary channel. In this way, the overall uniformity of the crystal could be improved. For experimental examination of the above conclusions, we developed two Ir crucibles with one and five capillary channels, and Ce:YAG single crystals were grown by the μ-PD method using these crucibles to understand the effects of the capillary number on the dopant distribution.

3. Experimental

Ce:YAG single crystals were grown by the μ-PD method using two types (1 C and 5 C) of Ir crucibles with a square-shaped nozzle at the bottom (Fig. 2). The 1 C crucible had one capillary channel positioned at the center of the bottom of the nozzle as shown in Fig. 2(a). Alternatively, there were five capillaries at the center and on the diagonal lines of the bottom of the nozzle of the 5 C crucible as illustrated in Fig. 2(b). The diameters of their capillary channels were ϕ0.4 mm and so the total cross section of the five capillary channels

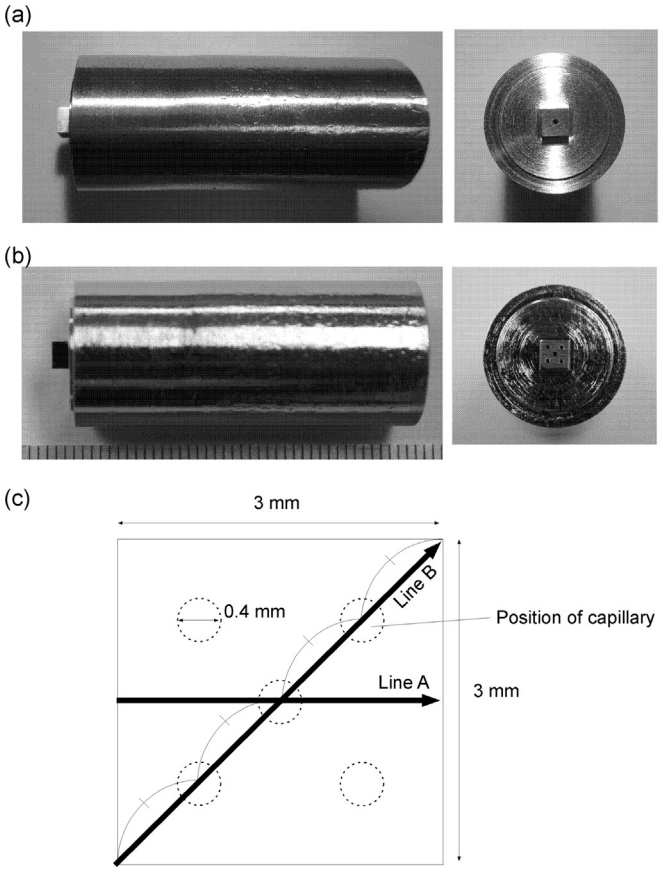

Fig. 2. View of the Ir crucibles produced with (a) one and (b) five capillary channels at the nozzle. (c) The chemical composition analysis of the polished specimens cut perpendicular to the growth axes was performed along lines A and B.

was five times larger than the cross-section of the one capillary channel. The capillaries in the 5 C crucible were placed diagonally at equal distances as illustrated in Fig. 2(c).

Starting materials were prepared from α-Al_2O_3, Y_2O_3, and CeO_2 powders (> 4N purity). The powders were mixed at nominal compositions of $(Y_{1-x}Ce_x)_3Al_5O_{12}$ with x=0.02 and 0.05, and the mixed powders were sintered at 1100 °C for 12 h in air. The as-sintered substances were loaded into the crucibles and Ce:YAG single crystals were grown according to the diagram shown in Fig. 1. Details of the single crystal growth by the μ-PD method are described in [3,4].