Journal of Crystal Growth 401 (2014) 146–149

Contents lists available at ScienceDirect

Journal of Crystal Growth

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

Vertical Bridgman growth of sapphire crystals, with thin-neck formation process

K. Hoshikawa [a], T. Taishi [a], E. Ohba [a,b], C. Miyagawa [a,b], T. Kobayashi [b], J. Yanagisawa [b], M. Shinozuka [b]

[a] *Faculty of Engineering, Shinshu University, Japan*
[b] *Fujikoshi Machinery Corporation, Japan*

ARTICLE INFO

Available online 27 January 2014

Keywords:
A2. Bridgman technique
A2. Growth from melt
A2. Seed crystals
A2. Single crystal growth
B1. Sapphire
B3. Light emitting diodes

ABSTRACT

A new technique is proposed in the traditional vertical Bridgman growth of sapphire crystals, in which thin-neck formation follows the initial seeding. Low-angle grain boundaries generated at the periphery of the seeding interface were eliminated at the thin neck, and the *c*-axis sapphire crystals with main bodies free from low-angle grain boundaries were grown.

1. Introduction

Many techniques for growing sapphire crystals for the fabrication of GaN-based LED devices have been actively discussed in recent years. The Kyropoulos (KP) method [1,2], the heat exchange (HE) method [1,3], the edge-defined, film-fed growth (EFG) method [1,4], and the Czochralski (CZ) method [1,5] are well known and important techniques for growing large high-quality sapphire crystals.

We have applied the traditional vertical Bridgman (VB) technique to the *c*-axis sapphire crystal growth [6]. The one difference between the HE method [3] and the VB method [6] is in the seeding process. The seed crystal placed in the crucible bottom is kept from melting by a cooling flow of He gas in the former method. In the latter, the temperature and its gradient in the vicinity of the seed must be controlled with precision.

We investigated the growth of sapphire crystal using the VB method with various shapes of seeds and crucibles, aiming at single-crystal growth. We achieved acceptable reproducibility with a very low temperature gradient of about 10 °C/cm and produced *c*-axis sapphire single crystals of about 50 mm in diameter from all three kinds of seeds: full-diameter, thin, and tapered.

We have progressed to more advanced investigations to eliminate crystal defects, such as low-angle grain boundaries (LAGB), which may originate at the seeding interface. Now we can propose a new growth technique that uses the thin-neck formation process in a traditional VB growth procedure.

In this paper, we will introduce the new process in the VB technique and the growth of *c*-axis sapphire crystals free from LAGB.

2. Experimental

Fig. 1(a) is a diagram of the VB furnace used in our experiment. The temperature distribution as measured without a crucible present is also shown in Fig. 1(b). The crucible was mounted on a crucible shaft that can both rotate and translate vertically. An argon atmosphere was maintained in an airtight chamber at just over 100 kPa. The carbon heater was powered by a radio-frequency coil. A maximum temperature of about 2100 °C was maintained in the central portion of the carbon heater.

A schematic drawing of the newly proposed thin-neck formation process is shown in Fig. 2. A molybdenum (Mo) or tungsten (W) crucible with a seed well, a thin neck and a main body is prepared as shown in Fig. 2(a). The inner diameters of the seed well, the thin neck and the main body are respectively 10–30, 3, and about 50 mm. Fig. 2(a) also shows the charging processes of the raw materials and the seed crystal. The raw materials, blocks of sintered alpha-alumina with a density of about 3.5 g/cm^3, are put into the crucible from above as shown in Fig. 2(a). A *c*-axis seed crystal of 10, 20 or 30 mm diameter by 50 mm long is inserted into the seed well at the bottom of the crucible as shown in Fig. 2(a). All of the raw materials and part of the seed are melted by moving the crucible up into the high-temperature heating zone as shown in Fig. 2(b). Fig. 2

E-mail address: khoshi1@shinshu-u.ac.jp (K. Hoshikawa).

0022-0248/$ - see front matter © 2014 Published by Elsevier B.V.
http://dx.doi.org/10.1016/j.jcrysgro.2013.12.051

(c) shows the neck formation process. In this process, the thin-neck portion, with a diameter of 2 mm (smaller than the seed crystal diameter of 10–30 mm) is grown up to the main body by moving the crucible down. Fig. 2(d) shows the growth process of the main body. The downward speed of the crucible was 3 mm/h in all cases, and it rotated at a constant speed of 5 rpm. The heating power was decreased to zero at a constant rate, as all growth processes terminated and the furnace cooled to room temperature.

The crystals grown were cut and both sides were mirror-polished as experimental specimens. X-ray topography was used to evaluate the LAGB.

3. Results and discussion

3.1. Release of seeds and grown crystals from the crucible

Photographs of three typical crystals grown with the neck formation processes are shown in Fig. 3. The crystal in Fig. 3(a1) is the main body grown from the seed crystal with 10 mm diameter shown in Fig. 3(b1). The crystal in Fig. 3(a2) is the main body grown from the seed crystal with 20 mm diameter shown in Fig. 3(b2). The crystal in Fig. 3(a3) is the main body grown from the seed crystal with 30 mm diameter shown in Fig. 3(b3). Crystals (a1) and (a2) were grown in W crucibles and crystal (a3) was grown in a Mo crucible.

Sapphire crystals grown with a main-body diameter of about 50 mm were easily released from the Mo and W crucibles. It was confirmed as previously reported [6] that the size of the gap between the crystal periphery and the inner wall of the crucible is large enough to allow release due to the different thermal shrinkage of sapphire and the crucible material. No correlation was found between the difficulty of release and the conical shape as the diameter increased from the thin neck to the main body, as shown in Fig. 3(a1), (a2) and (a3). It was presumed that the crystals, together with the main body and seed, were consistently separated by an unintentional break at the thin-neck portion due to the different contraction of the sapphire crystal and crucible material.

It is important for the re-use of crucibles that the seed crystals are easily released from the seed wells. Seed crystals of 10 and 20 mm diameter were easily released from the W crucibles, as shown in Fig. 3(b1) and (b2), respectively. In contrast, seed crystals more than 30 mm in diameter could be released from the seed well of Mo crucibles, as shown in Fig. 3(b3), but not those with diameters less than 20 mm. It is considered that the seed release differences between Mo and W crucibles were due to their different contractions during cooling [6].

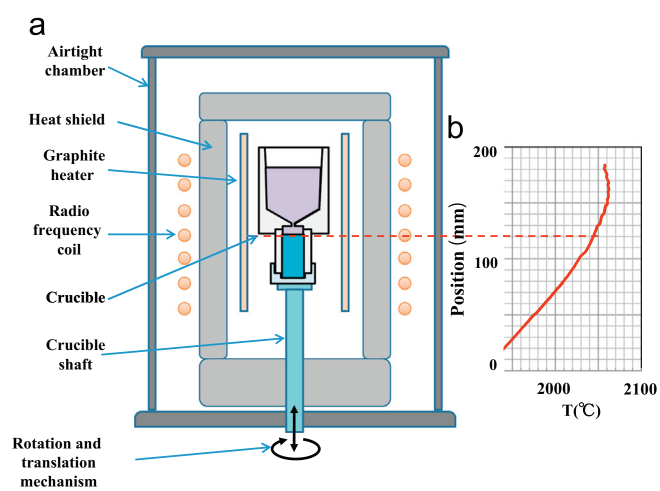

Fig. 1. Vertical Bridgman growth furnace used in thin-neck formation process (a) and temperature distribution as measured without crucible (b).

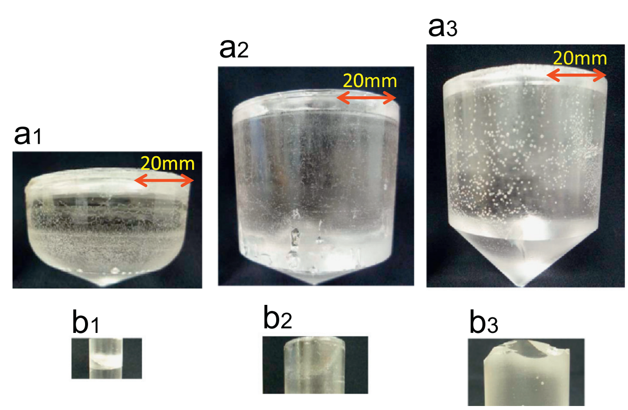

Fig. 3. Sapphire crystals grown by vertical Bridgman method with thin neck formation process. Crystals (a1) and (a2) were grown in W crucibles and crystal (a3) was grown in a Mo crucible. *c*-axis seeds with diameters of 10 mm (b1), 20 mm (b2) and 30 mm (b3) were used.

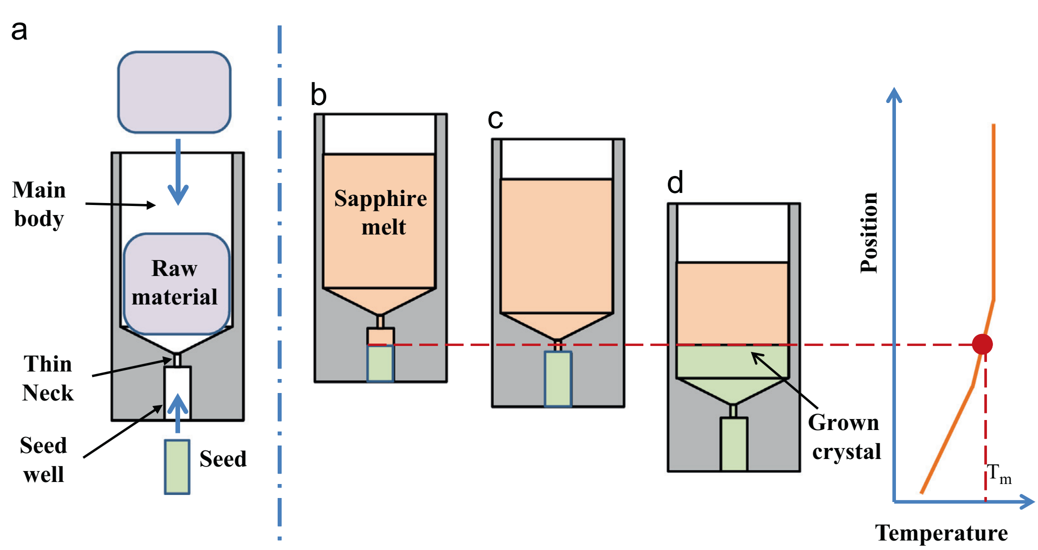

Fig. 2. Vertical Bridgman growth with thin neck formation process.