Journal of Crystal Growth 395 (2014) 80–89

Contents lists available at ScienceDirect

Journal of Crystal Growth

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

Vertical Bridgman growth of sapphire—Seed crystal shapes and seeding characteristics

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ARTICLE INFO

Article history:
Received 17 January 2014
Received in revised form
11 February 2014
Accepted 6 March 2014
Communicated by R.S. Feigelson
Available online 15 March 2014

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

ABSTRACT

The growth of sapphire by the traditional vertical Bridgman (VB) method was studied by using various shapes of seed crystals and tungsten (W) crucibles shaped to match the seeds. Approximately 2-in. diameter, *c*-axis sapphire single crystals were reproducibly grown from three kinds of seed: thin, tapered and full diameter. Factors relating seed type to single-crystal growth are discussed, including the reproducibility of seeding processes, and the generation and elimination of low-angle grain boundaries (LAGBs). What was learned facilitated the subsequent growth of large-diameter, 3-, 4- and 6-in., *c*-axis single-crystal sapphires from full-diameter seeds.

1. Introduction

Methods of growing sapphire for the fabrication of GaN-based LED devices have recently been attracting considerable attention. Well known techniques for growing large and high-quality sapphire crystals include the Kyropoulos (KP) [1,2], heat exchange (HE) [2–4], edge-defined, film-fed growth (EFG) [2,5–7], and Czochralski (CZ) methods [2,8]. All these methods have been studied extensively and compared in detail [2,9–13].

We have investigated sapphire crystal growth using the traditional vertical Bridgman (VB) method, in which the crystals are grown in a crucible with rotation and translation in a hot zone with an appropriate temperature distribution. We first demonstrated with our VB growth of *c*-axis, 3-in.-diameter sapphire that grown crystals could be easily and nondestructively released from a W crucible and that the crucibles could be reused many times [14]. We also showed that the easy crystal release was due to a gap between the crucible inner wall and the grown crystal formed by differential thermal contraction during cooling to room temperature [14]. Our experimental demonstrations and theoretical confirmations of crucible reusability have received favorable attention for their practical value in the industrial production of large sapphire. The reuse of crucibles has been a very important consideration in the HE and VB methods, in which the alumina melt is solidified in the crucible under crystal shape control.

However, we have not yet presented detailed information about the size and shape of seed crystals, the seeding process with precise temperature control, and the single-crystal growth process after seeding. Reproducible seeding with single-crystal growth was very difficult when we could observe neither the seed crystal nor the growth interface during the seeding process in either the HE or the VB method. In the HE method, a seed crystal placed at the bottom of the crucible is kept from melting by a cooling flow of He [3]. This allows seeding that reproducibly yields single-crystal growth, with the seed crystal remaining unmelted. However, in the VB method, with no special technique to prevent the melting of the seed crystal, it is very important to implement precise temperature measurement and control in the vicinity of the crucible bottom during seeding. It was well known that the difficulty of reproducible seeding for single-crystal growth can be strongly dependent on the shape and size of the seed crystal. For the traditional VB method, however, there are a few reports [14,15] on the shape and size of seed crystals, and on the details of the seeding process when they are used.

In this paper, we first propose a technique for precise temperature measurement at the crucible bottom in the VB growth furnace and verify its utility in providing reproducible seeding. Then, we

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http://dx.doi.org/10.1016/j.jcrysgro.2014.03.007

examine three representative kinds of seed crystals: thin, tapered and full-diameter seeds, used to grow *c*-axis single-crystal sapphire in the traditional VB growth technique. We discuss their differences with respect to reproducible seeding for single-crystal growth, and the generation and elimination of low-angle grain boundaries (LAGBs) [9] in their seeding processes.

2. Experimental

Fig. 1 shows a diagram of the VB furnace used in our experiment. The temperature distribution as measured with no crucible present is shown at the right side in the figure. The W crucible is mounted on a crucible shaft that can rotate and also translate vertically. The graphite heater is powered by a radio-frequency coil positioned towards the outer periphery of the carbon-felt heat shield. An argon atmosphere is maintained in the airtight chamber at just over 100 kPa.

Fig. 2 shows the shapes of the seeds and crucibles used in these experiments. The thin seed, the tapered seed and the full-diameter seed are shown in Fig. 2(a)–(c). The inside of the crucibles in which the sapphire crystals were grown were tapered by a few degrees to make it easy to release the crystal from the crucible [14].

The VB growth processes with these seed crystals and crucibles are as follows. Charging process: the seed and raw materials are placed in the crucible. Melting process: the crucible is elevated into the high-temperature zone and all raw materials are melted. Seeding process: a part of the seed is melted during the slow elevation of the crucible into the high-temperature zone, where the temperature gradient is about 10 °C/cm. Growth process: the crucible is lowered at 3 mm/h. Cooling process: the crucible is cooled to room temperature as the heating power is reduced to zero.

The VB furnace shown in Fig. 1 was used in the basic examination of seeding and growth of crystals with 2-in. diameter main bodies. However, crystals with diameters greater than 3 in. were grown by using another large-sized resistance-heating furnace described in Ref. [14].

The crystals grown were cut and both sides mirror-polished as experimental specimens. Green-laser scattering was used to observe the profile of the seeding interface. A crossed polarizer was used to evaluate the LAGBs. X-ray topography was also used to evaluate the LAGBs, the internal residual stress and the dislocation distribution.

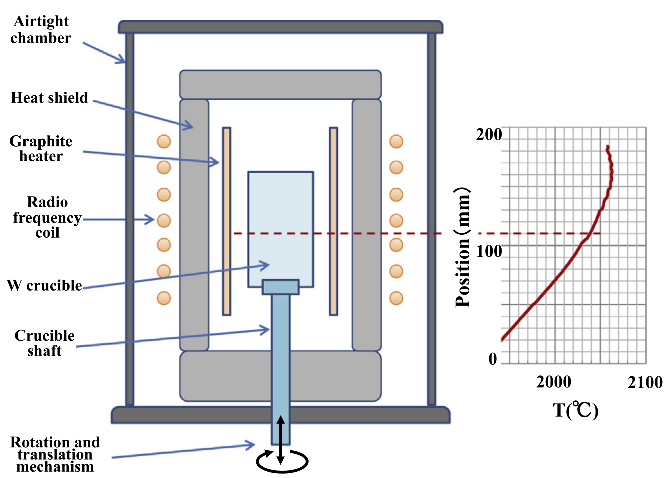

Fig. 1. VB furnace with the temperature distribution as measured without crucible.

3. Results and discussion

3.1. Temperature measurement and seeding process

It was very important to achieve satisfactory reproducibility in the seeding process, requiring precise measurement and control of the temperature near the crucible bottom. This was so because the present VB method provides no mechanism to prevent the melting

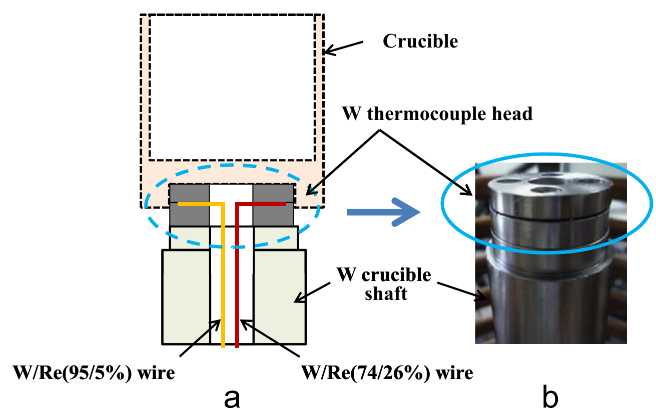

Fig. 3. Schematic drawing of the constructed thermocouple (a) and photograph of the thermocouple used in our experiment (b).

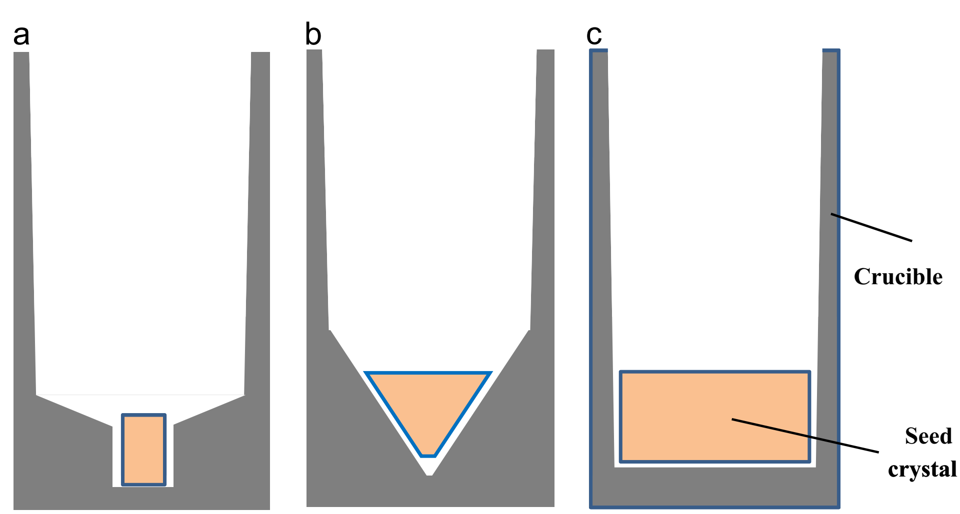

Fig. 2. Shapes of seeds and crucibles.