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3D large eddy simulation (LES) calculations and experiments of natural convection in a laterally-heated cylindrical enclosure for crystal growth

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ABSTRACT

The ammonothermal crystal growth technique is one of the most effective methods used for growing Gallium Nitride (GaN) crystals in terms of the final product quality and manufacturing efficiency. Popular applications of GaN crystals include light emitting diodes (LEDs) and high-frequency electronic devices. A laterally-heated cylindrical enclosure with a top to bottom temperature gradient is considered in this study to investigate the fluid mechanics and heat transfer in an ammonothermal crystal growth reactor. Three-dimensional (3D) large eddy simulations (LES) results of natural convection in a laterally heated cylindrical reactor, using the commercial computational fluid dynamics (CFD) software, ANSYS FLUENT, are presented in this paper for a Rayleigh number (Ra) of 8.8×10^6. The Ra is defined based on a length scale that is equal to the ratio of volume to the lateral area of the cylinder ($R/2$). In addition, an experiment of a geometrically- and dynamically-similar geometry is developed, and particle image velocimetry (PIV)-based flow visualizations are carried out for the purpose of validating the numerical model. Comparisons between experiments and numerical simulations showed that flow patterns were qualitatively similar, and Fourier transforms of velocity magnitudes at selected points in the domain matched reasonably well. An added interesting observation in the simulation was the existence of temperature inversion, which has potential implications on the choice of mineralizer (acidic/basic).

1. Introduction

Natural convection occurs due to the density differences resulting from concentration or temperature gradients. It is observed in several natural flows such as in the ocean and in the atmosphere, where a temperature gradient results in transport of nutrients and pollutants. The effectiveness of heat transfer due to natural convection also has widespread practical applications in energy storage, solar heating configurations, nuclear reactors and crystal growth industry. With regard to the crystal growth industry, Gallium nitride (GaN) crystals are manufactured via an ammonothermal process that utilizes natural convection in order to circulate the nutrients that create GaN in crystallized form in the reactor.

The ammonothermal crystal growth technique is similar to the hydrothermal technique, that is utilized to grow the quartz crystals, with an important distinction. The difference is the type of the solvent in the reactor, which is ammonia for ammonothermal, and water for hydrothermal techniques [1–3]. The two distinct thermal zones in an ammonothermal crystal growth reactor would create a buoyant force in the reactor. The pre-loaded GaN crystals in the nutrient basket are dissolved in the dissolution zone and, by virtue of natural convection, they are transported to the crystallization zone, where they deposit on the seeds. In the ammonothermal method, as GaN crystals are almost insoluble in pure ammonia, mineralizers (acidic or basic) are added to the autoclave. Other alternative crystal growing methods are hydride vapor phase epitaxy (HVPE), solution growth, sodium flux, and the stoichiometric melting methods [4,5].

Natural convection has been studied both experimentally and numerically by several researchers for the past few decades [6–26]. Numerical studies included both 2D and 3D, and rectangular and cylindrical enclosures with different boundary conditions. The thermal configuration has a very significant role in determining the characteristics of temperature and flow distribution in the reactor. They primarily include side-wall heating, and top and bottom wall

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heating with either constant temperature or heat flux. In addition, previous investigations have also looked at the effect of heat generation sources within the domain. The current study tries to introduce and analyze a relatively new thermal configuration, where the lower hot wall at a constant temperature in a cylindrical enclosure is separated from the upper colder wall, again at a constant temperature, by an insulated section, with no heat source.

With regard to natural convection specifically in a cylindrical enclosure, there have been quite a few 2D and 3D numerical and also experimental studies with a variety of thermal configurations [27–31]. For instance, Chakir et al. [32] performed a 2D numerical calculation of turbulent natural convection of mixed gases in a horizontal annulus. Several simulations of thermal configurations involving a constant heat flux and constant temperature on the inner cylinder were carried out, while the outer cylinder was maintained at a constant temperature. The heat transfer and fluid motion were analyzed for Rayleigh number (*Ra*) values ranging from 10^5 to 10^{10}, and numerical results were compared to experiments. Barhaghi [33], studied the structure of turbulent natural convection boundary layers, and the effect of the buoyancy on a mixed convection boundary layer using direct numerical simulations (DNS) and large eddy simulations (LES). Flow and turbulence parameters in all cases were studied, and the results were compared to experiments as well. More recently, Malik et al. [34], performed a CFD analysis of conjugated heat transfer within a bottom-heated cylindrical enclosure. The study investigated the influence of the inner cylinder material and the outer cylinder geometric configurations on the heat transfer mechanism within the enclosure. Kang et al. [35], studied experimentally the effects of the Prandtl number and the curvature on the natural convection in a cylindrical geometry. In order to study the effect of curvature, they measured the Nusselt numbers for *Ra* of 1.4×10^9 - 3.7×10^{12} at $Pr = 2094$. They showed a good agreement with laminar natural convection correlation data on the vertical plates.

Several investigations have also been performed about the role of the natural convection in a crystal growth process [5,36–42]. Experimental and numerical studies in a rectangular cuboid were carried out by Li et al. [37]. The upper half of the sidewalls was cooled uniformly, while the lower half of the sidewalls was heated. The *Ra*, based on the width of the chamber as the characteristic length, was 1.77×10^8. They showed a good agreement between the experimental data and the 3D numerical results, with respect to the fluid flow and thermal distributions. Li et al. [43] also carried out experiments and 3D numerical simulations of flow and heat transfer in a cylindrical hydrothermal reactor, again cooled on the upper-half and heated on the lower-half, separated by a baffle. Due to the fluid exchange at the baffle opening, an unsteady jet-like flow pattern in each of the chambers of the reactor was observed. They also concluded that the time-averaged flow profiles and temperature were axially symmetric. Erlekampf et al. [44], conducted numerical simulations of an ammonothermal autoclave. The effect(s) of various baffle shapes on the flow pattern, fluctuations of the temperature as well as flow velocities were studied. They came to the conclusion that 3D numerical simulations were necessary to accurately model the heat and mass transport in the ammonothermal autoclaves, rather than a 2D model. The current authors previously performed a 2D axisymmetric numerical study in a vertical cylinder with laterally-heated walls, using ANSYS FLUENT. A *Ra* criteria based on flow transition from fully laminar flow to transitional flow was introduced [45]. They considered the cylindrical volume of the reactor to its lateral area as the characteristic length in calculating the *Ra*.

There have been several experimental and numerical investigations with regard to ammonothermal-based GaN crystal growth [46–61]. D'Evelyn et al. [62] used high-pressure ammonothermal crystal growth to produce high-quality GaN single crystals. The pressures and temperatures were varied between 5 and 20 kbar and 600–1000 C, respectively. They managed to reduce the level of impurity in the GaN crystals and successfully demonstrated homoepitaxial laser diodes. They concluded that this technique could produce high-quality bulk GaN substrates. Bao et al. [63] studied different types of acidic mineralizers such as NH_4Cl, NH_4Br, NH_4I and NH_4F, for the ammonothermal GaN crystal growth method. It was concluded that NH_4F was a promising mineralizer, since it could afford a negative temperature gradient in supercritical ammonia at a temperature range of 550–650°C. Furthermore, it could increase the growth rate and quality of GaN crystals. Mirzaee et al. [64] presented an in-depth 2D axisymmetric numerical study for the ammonothermal GaN crystal growth technique in a Rene-41 autoclave, including fluid flow, heat transfer, and dissolution and crystallization rates. The operating temperature and pressure were 500°C and 1.5 bar, respectively. The top and bottom parts of the external sidewalls had a constant temperature of 525°C and 475°C, respectively. They proposed to create a gap between the nutrient basket and the sidewall in order to have a stronger flow in the nutrient basket. Although, the presence of the gap resulted in a better mixing and transportation of the nutrient particles, it adversely affected the crystal growth rate near the seeds. The proposed model (with the gap) showed a steady growth rate of 92μm per day.

There is still a need for detailed experimental observations and numerical analysis of natural convection in a cylindrical enclosure with laterally-heated boundary conditions. The current study presents 3D LES calculations of turbulent natural convection in a laterally-heated cylindrical enclosure at a *Ra* number of 8.8×10^6, $Pr = 5.85$, and an aspect ratio (H/D) of 5.17, using the commercial CFD software, ANSYS FLUENT. LES uses filtering operations in order to solve the filtered governing equations such that, the turbulent scales are separated. As a result, the large scales are resolved, while the sub-filter scales are modeled using subgrid-scale (SGS) models [65]. Such a methodology is superior to a Reynolds-averaged Navier-Stokes (RANS) approach, which models all scales. Also, LES is computationally cheaper than DNS, which resolves all the length scales. Over the last two decades, LES has demonstrated excellent comparisons with experiments for nonreacting flows. See the review paper by Pope [66] for comprehensive list of LES investigations.

Flow patterns and thermal maps are analyzed through contours of instantaneous velocity and temperature on selected planes. An experiment, with a similar setup as the numerical simulations, is also carried out, and flow patterns and velocities are compared to the computational results. In order to assess the effectiveness of the heating in terms of the consistency of the wall temperatures, which has direct implications on the correlation between experiments and computations, several thermocouples are placed on the inner wall of the reactor. For the numerical results validation purposes, the thermal boundary conditions (temperatures of the walls) are recorded and then directly extended to the numerical computations. For that purpose, Type-K surface thermocouples are installed on the inner wall of the enclosure. It was found during the experiments that these thermocouples recorded constant temperatures along the walls (variation of less than 0.3° C) both in the azimuthal and longitudinal directions, and in the cold and hot sections. Therefore, the simulations are conducted using constant wall temperatures germane to the experiment. This work attempts to provide more information about the flow patterns and the thermal distribution in a cylindrical crystal growth reactor.

The unique aspect of the current work is the usage of 3D LES in conjunction with experiments for investigating relatively large laterally-heated cylindrical reactors. It should be noted that the size