Modeling of Convective Transport in Crystallization of Gallium Nitride by Basic Ammonothermal Method

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Gallium nitride (GaN) crystals of the best structural quality are grown by ammonothermal method [1]. Its main idea is to dissolve polycrystalline GaN (i.e., feedstock) in supercritical ammonia (NH_3) in one zone of an autoclave and transport it to the second zone where the solution is supersaturated and crystallization of GaN on native seeds takes place. An appropriate temperature gradient between these two zones enables the convective mass transport. Some mineralizers are added to NH_3 in order to enhance the solubility of GaN. Thus, the growth can be proceeded under different environment: basic or acidic. The type of environment is determined by the choice of mineralizers. Ammonobasic growth makes use of alkali metals or their amides as mineralizers, while in ammonoacidic growth halide compounds are present. Due to technological limitations (tightly closed reactors) and extreme conditions (chemically aggressive environment and high NH_3 pressure), information about processes in the reactors are not straightforwardly accessible in ammonothermal growth experiments. The autoclaves are like "black boxes" controlled by temperature measured outside of them. A detailed analysis of convective transport can lead to a deeper understanding of the ammonothermal crystal growth process and a further, faster and better development of this technology. Bigger than 2-inch GaN crystals of high structural quality and different type of conductivity (highly conductive and semi-insulating) are grown during an ammonothermal process and there is still room for improving their structural quality and uniformity of incorporated dopants. It is well-known that the homogeneity of the growth process depends on the uniformity of supersaturation. The latter, in turn, can be ensured by a homogenous and laminar convective flow of the reagents to the growth zone at proper and constant temperature–pressure conditions. A quantitative analysis of the convective transport at 3D approximation would allow to find the best growth conditions for a uniform crystallization of GaN. Therefore, in this work convective transport in NH_3-based supercritical solution during crystallization of GaN is examined. The Computational Fluid Dynamics (CFD) method was applied for modeling the processes inside autoclaves of large volume, thus with high values of Rayleigh and Grashof numbers, at difference of temperature between feedstock and growth zones of the order of a few hundred degrees. These conditions mean that flow is buoyancy-driven and can become turbulent. A time-dependent set of Navier-Stokes equations (continuity equation, momentum and energy conservation equations) was used to describe the density, pressure, velocity, and temperature of the moving fluid. The flow and the change in the mass transfer in time were analyzed. The solution of the mentioned equations allowed to determine the flow velocity and temperature during an ammonothermal crystal growth process. A model geometry was build basing on the documentation of a working autoclave. Boundary conditions were calculated basing on operating conditions of working autoclaves. ANSYS Academic engineering simulation software was applied. The geometry of the autoclave and the model mesh were created using tools from the ANSYS package. Experimental data were applied to verify the modeling results. The determined and analyzed morphology, structural quality and growth rate of ammonothermal GaN crystals were linked to a convective transport model.

[1] Kucharski R., Sochacki T., Lucznik B., Bockowski M.,Growth of bulk GaN crystals, J. Appl. Phys., 128 (5), 2020, 050902