Melt Homogenization Improvement By Optimizing the Rotation Profile

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Abstract

During the last decades, mid-IR (3-12µm) laser sources have attracted a particularly attention due to their potential applications in different fields like infrared counter-measures (e.g: missile jamming) and remote chemical sensing (e.g: LIDAR). As transparency of common nonlinear oxide crystals is limited to about 4µm, many researches have been focused on non-oxide like chalcopyrite compounds because of their wide transparency in the mid-IR range and their high nonlinearity. Thus, compounds like ZnGeP2 (ZGP) and AgGaS2 (AGS) have been developed and are currently among the most used for the laser frequency conversion in this range.
For several years, Onera has been working on the development of these strategic materials. Currently, ZGP, AGS and also AGGS (AgGaGeS4, Figure 1) single crystals have been successfully obtained. However, these materials have intrinsic drawbacks (multiphonons absorptions, low laser damage threshold), which limit their efficiency in some applications. Thus, research for better materials have been undertaken in order to develop new crystals allowing to reach higher power and to extend the transparency range beyond 9µm.
To process those crystals, there are three main steps: chemical synthesis, crystal growth and annealing. The present paper is focused on the crystal growth which is performed in a Bridgman-Stockbarger system: a compound is translated from a zone above the melting temperature to a zone below where it is crystallized. One of the main problems we encounter is the melt inhomogeneity inside the crucible during the growth. Indeed, the real composition at the crystallization point can vary more or less around the stoichiometric proportion. This can affect the crystal quality and it is important to minimize it.
The goal of this work is to model, using COMSOL Multiphysics® software, the influence of the simplest parameter we have access to: the rotation.
A new system controlling crucible's rotation was added on the furnace to permit cyclic crucible accelerations. Thus, using the Heat Transfer in Fluids and Heat Transfer in Solids interfaces, we modeled the thermal environment, the rotation profile and their influence on the melt convections (Figure 2). Then, at a defined time (once the temperature is stabilized, instant t' on the Figure 2), a species distribution inhomogeneity (Transport of Diluted Species interface) is imposed. Then, we observe the time necessary to restore the homogeneity. Different rotation profiles were tried to optimize this time: no rotation, constant rotation, cyclic rotation.
First, phase transition (liquid to solid) is not considered, to improve calculation and reduce

computing's time (Figure 3). Very different behaviors were observed depending on the profile. Optimization is in progress. Then, the phase transition will be considered at the crystallization temperature isotherm to observe the influence of the rotation profile on the crystallization interface's shape.

Figures used in the abstract

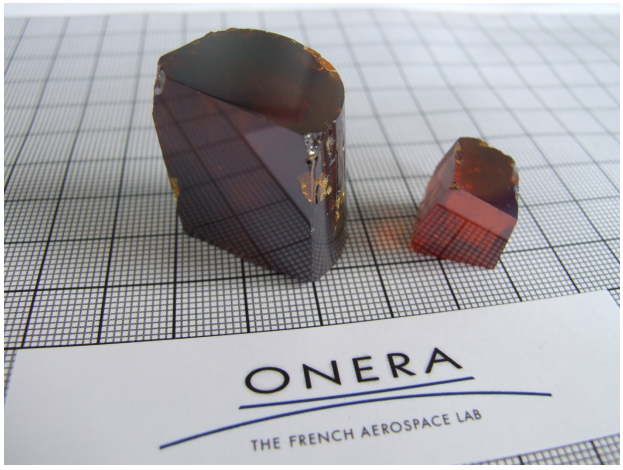

Figure 1: AgGaGeS4 ingots obtained with the Bridgman technique.

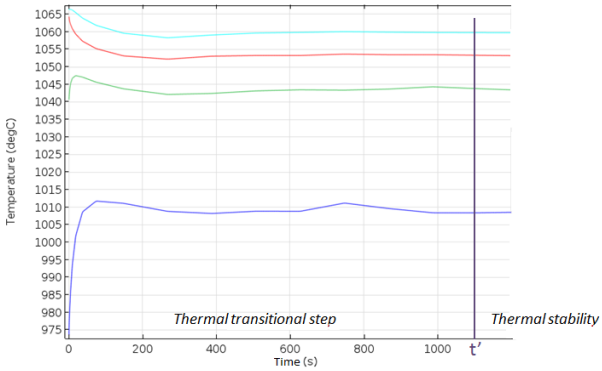

Figure 2: Thermal evolution in the crucible: once t=t', the system is in a thermal stability stage.

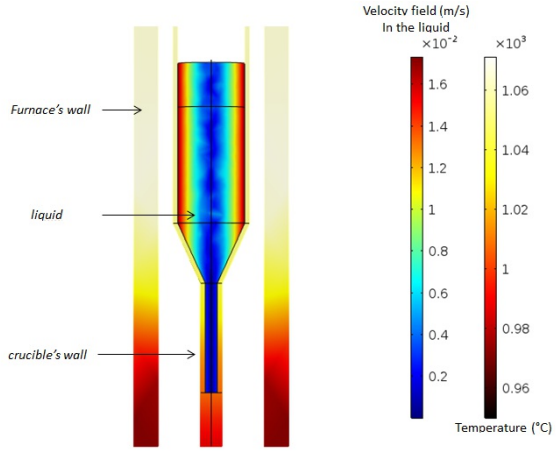

Figure 3: Results at t>t' for a constant rotation of 12 rpm (showing velocity field in the liquid in m/s, and the temperature on the walls of the furnace and the crucible in °C).