New LGT crystal for Ultra-Stable Resonators

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Among the crystals belonging to the langasite family, the Langatate ($La_3Ga_{5.5}Ta_{0.5}O_{14}$ also called LGT) seems to be the best candidate to substitute to the quartz crystal for acoustic waves applications and frequency output devices. But, its capability to substitute to quartz depends on the crystalline quality of the production of the harvested “boule”. Indeed, the crystal quality (its homogeneity and its atomic structure as well as the amount of chemical impurities) can be very different from one supplier to another. So, to compare and analyze the strategic interest of this crystal, we start a sufficiently researched program, supported by the French DGA, to enable us to address the parameters of the growth process, implemented on “Cristal Innov” platform.

The Langatate is an incongruent material grown from a ternary melt of La_2O_3 – Ga_2O_3 and Ta_2O_5 by the Czochralski technique along <00.1> or <2-1.0> in iridium crucible and slightly oxidizing atmosphere. Furthermore, a post-growth annealing performed under vacuum or in air at different temperatures will determine the color of the boule, which varies from colorless to orange or pale-green color. So, we attach to analyze the stoechiometry (i.e. the amount of the majors La, Ga, Ta) but also the influence of different annealing processes performed in the pulling machine immediately after growth and in other oven into different atmospheres. We do not forget that the annealing has become necessary to remove the stress induced by the growth.
We analyze the impurities levels and we shall determine also the fine structure variation in the material (cell-parameters, inter-atomic distances, angles…) in terms of the Ga/Ta substitution rate and chemical composition. For that, two complementary experimental techniques will be used: single crystal X-ray diffraction and Electron Probe Micro Analysis.

After analyzing previous crystal coming from different suppliers and new ones from “Cristal Innov”, we perform resonators working at about 40 MHz (on its 3rd or 5th overtone) allowing estimation of the quality of a small volume of crystal. We compare also their Q.f product values with those of quartz AT- or SC-cut resonators working at the same frequency. For this manufacturing, we have to determine the best process to achieve thin resonators (thickness around 100 µm) with polished surfaces and deposited electrodes. We note that the grinding and polishing processes are deeply different than those of quartz resonators.

Finally, the use of LGT crystals in frequency and time applications is discussed and some perspectives are presented as conclusions.