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Growth & characterization of Langasite crystals for SAW device applications

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Abstract

LGS crystal which melts congruently at 1470 ^{0}C has been grown by Czochralski method. The langasite crystal of length about 1cm grown along its Z-axis was cut in X and Y directions , polished and subjected to various characterization studies. Phase and structure of the grown crystal was confirmed by Powder XRD measurement. FTIR spectrum was carried out to confirm the functional groups present in the grown crystals. The optical behavior was studied by UV–vis-NIR analysis. Electrical properties such as Dielectric constant, Resistivity, Conductivity and Piezoelectric coefficient have also been studied.

Keywords:*Langasite, Czocharlski technique, Piezoelectric ,SAW device,*

Introduction

Piezoelectric materials play a very important role in the modern industry. The most widely spread products of piezoelectric engineering is radio frequency (RF) control elements (resonators) and selection (filters)[1, 2]. The main material used in piezoelectric engineering is quartz which is having highly stable frequency-temperature characteristics and is the reason for its application in resonator production. One of the piezoelectric materials with high $K^2_{emc}$is lithium tantalate, enabling to design wide band filters. However, due to the low frequency-temperature stability and the low quality factor (Q) of resonators made of this material the application of these filters is limited. Langasite(LGS) crystal belongs to piezoelectric materials with the value of electromechanical coupling coefficient intermediate between quartz crystal and that of lithium tantalate. LGS has attracted increasing attention because of its excellent properties in applications of surface acoustic wave, bulk acoustic wave, and sensors fields[3,4]. An important feature of LGS is that it undergoes no phase transitions up to its melting point, which stimulates the development of langasite- based piezoelectric sensors[5,6]. In the present work LGS crystal has been grown by Czochralski method and subjected to XRD, FTIR, UV and Electrical characterization.

II. Synthesis

The LGS polycrystalline material synthesis was carried as per the following reaction

$$3\ La_2O_3 + 5\ Ga_2O_3 + 2\ SiO_2 \text{ -------}\rightarrow 2\ La_3Ga_5SiO_{14}$$

The synthesis of LGS has been carried out by solid state sintering method from precursors of 99.99% pure La_2O_3, Ga_2O_3 and SiO_2 weighed according to stoichiometry ratio and then well mixed in a planetary mill for 6 hours. To compensate the loss of Ga by volatisation at high temperature excess amount of Ga was taken. Mixtures were then pressed at 10 kgf/cm^2 to pellets and sintered for 12 h at 1100^{0}C.The polycrystalline LGS pellet is shown in Figure 1.

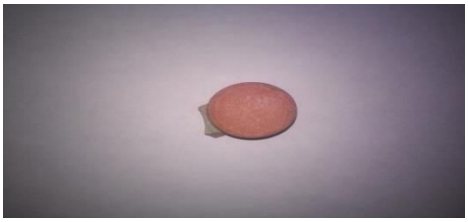

Fig.1.The prepared Polycrystalline LGS pellet

Growth of Langasite crystal

Langasite crystal was grown along its *Z*-axis by the conventional Czochralski technique. Initially polycrystalline LGS pellets were charged into Pt/10%Rh crucibles of 50 mm diameter and 50 mm height. Pellets of LGS weighing 150 g which is much less than the volume of the crucible were taken in the crucible in order to avoid overflow of material from the crucibles when the pellets are molten. The crucible was then placed on a crucible holder which can be moved vertically along the furnace axis, thus allowing an appropriate positioning of the crucible within the temperature gradient of the furnace. The basic process was to melt the pellets completely and then to cool it to observe the exact crystallization temperature of the material for the present setup. Charge was heated to 1600 ^{0}C , held at that temperature for 3 h for complete melting and then cooled down slowly to find the solidification temperature. After several runs the crystallization temperature was found to be 1480°C, and a polycrystalline seed crystal (Figure 10) was introduced at this temperature so as to initiate the growth process by pulling and simultaneous rotation of the seed rod. A pulling rate of 0.5-3 mm/h and rotation rate of 5-40 rpm were maintained during growth process. The as grown single crystal of size 38x22x9 mm is shown in figure 2a.The X and Y cut crystal are shown in fig.2b.

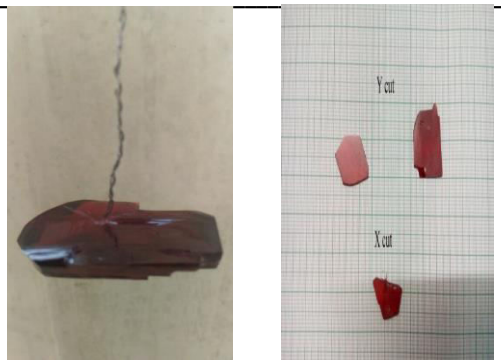

Fig. 2. Photograph of grown LGS crystal

IV. Characterization

i) X-ray diffraction study

Powder X-ray diffraction analysis of the polycrystalline LGS prepared by solid state reaction method and LGS crystal was carried out using PANalytical-X-ray diffractometer in the range 10 - 70° to confirm the structure and phase. The Kα radiations from a copper target (λ = 1.5406 Å) was used.The Powder-XRD spectrum of polycrystalline LGS and LGS crystal are shown in Figure 3. From the Powder-XRD spectrum, the formation of LGS phase and structure was confirmed. The calculated cell parameter values are a=8.1624 , c=5.093. The diffraction pattern well agreed with the data of standard for LGS (JCPDS No: 72-2249).

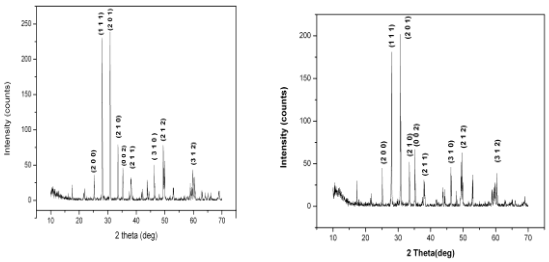

Fig.3. The Powder-XRD spectrum of polycrystalline LGS crystal (a) and Grown LGS crystal (b)

UV-Vis-NIR absorption spectrum

The absorption spectrum of Langasite crystal was recorded using Lambda 35 UV Winlab Spectrometer. The UV spectrum was recorded between 400 to 1100 nm. The recorded absorption spectra of Langasite crystal is shown in Figure 4. The absorption edge was observed in the range 500 - 600 nm.

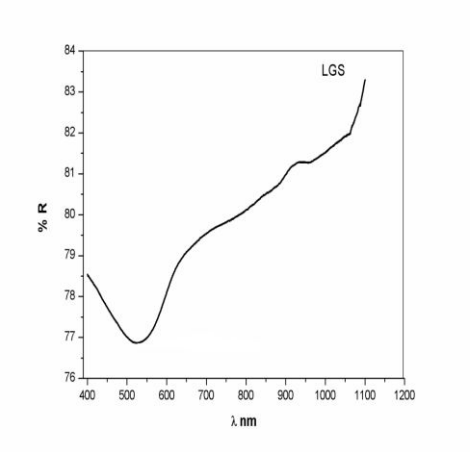

Fig.4 UV reflected spectrum of LGS crystal

FTIR Analysis

Figure 5 shows the From the FTIR spectra of the sample shown in figure 5 the functional groups are analyzed taking into account the molecular structure of the material. The peak at 450 $cm^{-1}$is assigned to the O-La-O stretch. The absorption bands in the region of 575 cm^{-1} are due to O-Si-O stretching mode. Stretching at 628 $cm^{-1}$and 735 $cm^{-1}$are the evidence for the existence of Ga-O group. The band at 675 cm^{-1}signifies the La-O stretching vibration. The Si-O stretching vibration obtained at 779 and 879 cm^{-1}.

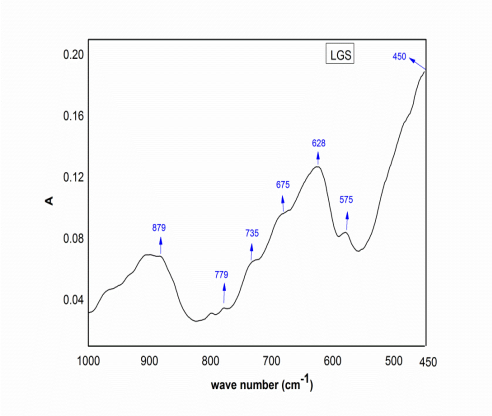

Fig.5 FTIR recorded spectrum of LGS crystal

Dielectric studies

Frequency dependence of dielectric constant for Langasite crystal in the frequency range 100 Hz to 3 MHz at 313 Kis shown in figure 6. It is observed that dielectric constant is high at low frequency which is due to the contribution of various polarization[7]. At high frequencies, only electronic polarization with large relaxation time exists and all other polarizations cease[8]. Hence the net dielectric constant decreases as frequency increases. The dielectric constant for the Langasite crystal at 1MHz was calculated found to be 8.87.

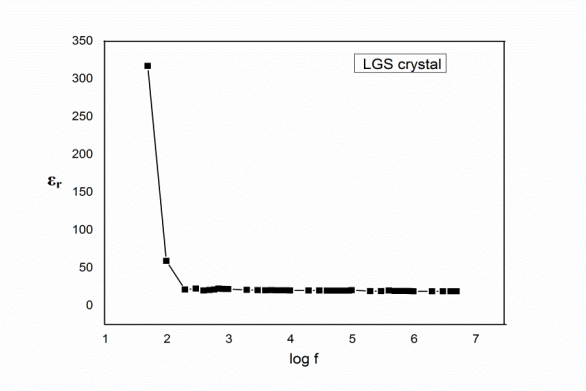

Fig.6 Frequency dependence of dielectric constant for LGS crystal at 313 K

Tangent loss (tanδ) found to have higher value at low frequency and decreases with increasing frequency, illustrating the relaxation process. The higher dielectric loss that occurs at lower frequency may be due to an accumulation of free charge. The polar ionization which occurs due to the charge accumulation of decreases, leads to a decrease in the value of the dielectric loss.

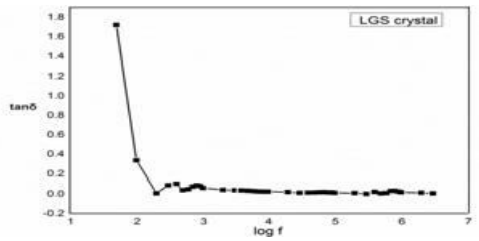

Fig.7 Frequency dependence of dielectric loss for LGS crystal at 313 K

AC conductivity

Figure 8 shows the frequency dependence of AC conductivity (σ_{ac}) at 313 K for Langasite crystal. The AC electrical conductivity (σ_{ac}) of the LGS crystal was calculated from the following equation,

$$\sigma_{ac} = \omega \varepsilon_r \varepsilon_0 \tan\delta$$

where ω (=2πf) is the angular frequency, f is the applied frequency. From fig.8, it is clear that the conductivity is independent of frequency at low frequency region whereas above the characteristic frequency the conductivity increases with increase in frequency. The high conductivity at higher frequencies confirms the short-range intrawell hopping of charge carriers between localized states[9].

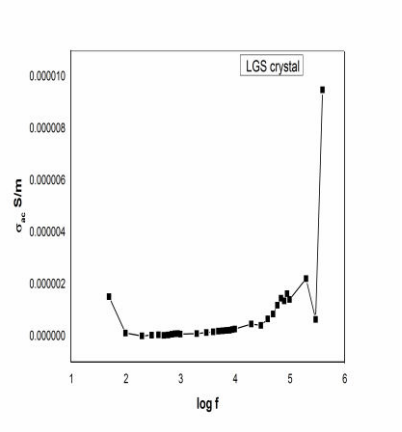

Fig.8 Frequency dependence of AC conductivity for LGS crystal at 313 K.

Piezoelectric studies

The piezoelectric coefficient (d_{33}) was measured for the grown Langasite crystal using APC USA make YE2730A model d33 meter. The Piezoelectric coefficient of Langasite crystal was found to be 5 pC/N.

Conclusion

Langasite single crystal of length 1cm was grown by Czochralski technique . The structure of LGS was confirmed by powder XRD. UV-Vis –NIR absorption spectrum shows absorption edge at 525 nm. The various functional groups present in the crystal were confirmed by FTIR analysis. The dielectric measurements show that the dielectric constant and dielectric loss decreases with increase in frequency. The AC conductivity values found to have higher values at high frequency. Piezoelectric coefficient of the crystal was found to be 5 pC/N.

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