New Approach of Interdigitated Transducers Engineering for High-Temperature Surface Acoustic Wave Sensors

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***Abstract*—The need of Surface Acoustic Wave (SAW) sensors operating at high-temperature (T ⩾ 600 ° C) is increasing. Generally, such SAW sensors are based on langasite (LGS) family crystals, with noble metal as interdigital transducers (IDTs), and oxide/oxynitride as a cladding layer. In addition, a buffer layer is systematically used to promote the noble metal adhesion. At high temperatures, material instabilities lead to several issues related mainly to interdiffusion and agglomeration and consequently in frequency instability and irreproducibility. In this work, after having tested several buffer layers (metals or oxides), the platinum grown directly on LGS surface showed the best stability at high temperature. The effect of surface quality of the LGS crystal and Pt thickness on the agglomeration of Pt was studied, as well. The final optimized stack consisted only of 3 materials: LGS, Pt IDTs, and SiO_2 cladding layer, and it did not show any degradation at 800 °C. The frequency of such SAW devices remained stable in the limits errors over 16 days.**

Keywords— SAW, sensor, high temperature, fabrication

I. Introduction

Surface Acoustic Wave (SAW) devices are used in a wide range of applications: sensors (pressure, temperature, humidity, chemicals…), oscillators, and radio frequency filters, usually operating at ambient conditions. The need of SAW sensors operating at high-temperature (T ⩾ 600° C) is increasing. Several studies have investigated the possibility to use SAW technology in harsh environments (corrosive and high temperature). Generally, such SAW sensors are based on langasite (LGS) family crystals, with noble metal as interdigital transducers (IDTs), and oxide/oxinitride as a cladding layer. LGS family crystals are piezoelectric at high temperature and offer plenty of possibilities with respect to the composition as long as the crystals keeps following the characteristic structure $A_3BC_3D_2O_{14}$ structure, where, A, B, C, and D, indicate cations site, respectively: a twisted Thomson cube (8 oxygen ions coordinated), an octahedron (6 oxygen ions coordinated), and a tetrahedron for both C and D (4 oxygen ions coordinated). At high temperatures, SAW device instabilities are mainly related to material interdiffusion and noble metal agglomeration. In addition, the systematic use of a buffer layer (noble metal adhesion promoter), which also gets oxidized and/or diffuses, decreases (even more) the device stability. Many of the reports focus on triggering the composition of the stack in order to increase high-temperature stability of the devices [1]–[7]. However, the more materials are added to the stack, the higher probability of material interaction/interdiffusion. Indeed, using a buffer layer, coupled with co-evaporated Pt-alloys/oxide composite playing the role of IDTs, and a cladding layer offer to the materials the possibility to migrate more easely through grain boundaries (e.g. energetic interfaces) and to react with other stack materials or with oxygen (diffused from the atmosphere or from oxides in the stack). This leads to dewetting process, continuous material degradation, electrode conductivity drop, frequency instability, and finally to device failure. Previous works dealing with high-temperature SAW sensors have built a strong database on materials reactivity. However, a robust material solution is still needed to enable the SAW frequency stability and reproducibility at high working temperature.

Obviously, usual approach faces a dilemma: adding stabilizers of agglomeration/dewetting creates other kind of instabilities (reactivity and interdiffusion). In this paper, different approach to stabilise SAW device structure at high-temperatures is proposed by limiting the number of materials. Several buffer layers (metals or oxides) and direct Pt growth have been tested. The improvement of LGS/Pt interface quality has also been part of the study by means of LGS surface treatment. Finally, SiO_2 was used as a cladding layer for the Pt IDTs and the stability of the SAW devices has been studied.

II. Process and Fabrication Details

For the studies of stack stability at high temperatures, different metal and oxide layers were deposited by means of magnetron sputtering. The typical annealing of these material heterostructures was performed in air at 800°C for 48h. The material surfaces/interfaces were studied by using focused ion beam (FIB) cross-section cut, scanning electron microscopy (SEM) and atomic force microscopy (AFM). The SAW resonators with operational frequency of 248 kHz were fabricated by using standard lift-off process of UV lithography. The thickness of the Pt electrodes, deposited directly on LGS surface by evaporation, was 300 nm. The SiO_2 cladding layer with thickness of 50 nm was deposited by sputtering. The resonators were characterized by admittance measurements, usinga ROHDE and SCHWARZ Vector Network Analyzer (VNA).

III. Results and Discussion

A. Pt/LGS interface and adhesion layer

First of all, several adhesion layers have been tested in LGS/Pt interface, which follows the common direction of former studies. Ti metal being widely coupled with Pt, it was the first-choice material. In total accordance with other works

[8], [9], the high affinity between Ti and O makes Ti diffusing across Pt layer leading in the formation of Ti oxide. Fig. 1 shows the critical diffusion of the Ti layer which is completely scattered in Pt layer and converted into oxide after annealing process.

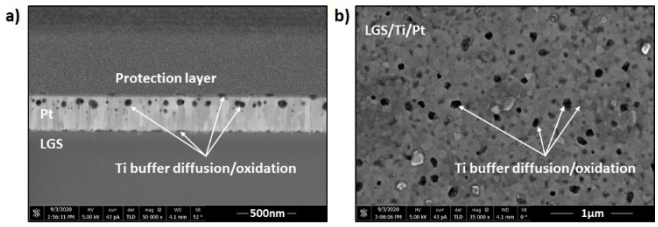

Fig. 1 SEM cross-section images of LGS/Ti/Pt (continuous film) stack after annealing 48h at 800°C: cross-section (a) and surface (b).

Device fabrication using other metals (Zr, Hf, Ta, Y, Nb, Cr) has been deeply investigated in literature, as well [3], [8], [10]. Interdiffusion usually causes conductance drop or even open circuit due to dewetting and consequently device failure. These transition metals oxidize easily at high-temperature: either by oxygen diffusing from the atmosphere or from the substrate, or by metal diffusion itself to the surface.

Metal buffer layers being unusable, corresponding oxide have been considered instead of the pure metal with the idea that if the metal is already oxidized, it will not migrate anymore in the stack. In this work, TiO_2, $LaNiO_3$, and ZnO interface layers have been studied. $LaNiO_3$ has been found to react with LGS at 800 °C as can be seen from the degradated surface in SEM image (Fig. 2 c). No interdiffusion was observed between ZnO and Pt but a strong dewetting process of Pt was present on ZnO surface after annealing (Fig. 2.b). In the case of TiO_2 interface layer, the annealed Pt surface presented big crystals, initiation of dewetting on the crystal boundaries, and very few spots related to interdiffusion (Fig. 2.a). Ababneh *et al. (2015)* [9] have shown a high stability of TiO_2. Thus, this residual TiO_2-Pt interdiffusion might be caused by the oxygen nonstoichiometry in TiO_2, which could introduce the migration of partially oxidized Ti.

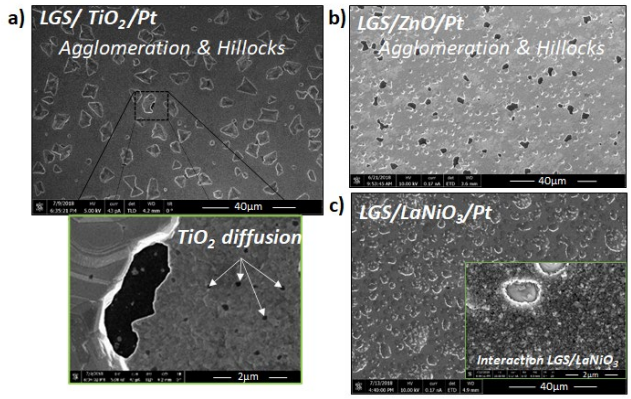

Fig. 2 SEM surface images of LGS/TiO_2/Pt with a zoom of the black-doted area on the bottom (a), LGS/ZnO/Pt (b), LGS/$LaNiO_3$/Pt with inset of a zoom showing the appearance of crystallites with different composition (c).

To study further the influence of oxide surface energy on Pt agglomeration, Pt has been directly deposited on several other oxide crystals ($SrTiO_3$, different Sapphire orientations, $LaAlO_3$, $(LaAlO_3)_{0.3}(Sr_2TaAlO_6)_{0.7}$) and the adherence was good and the very slow agglomeration appeared due to higher Pt surface energy than that of oxides. This indicates that Pt can adhere well on crystalline oxide surfaces and do not require adhesion layer like on amorphous SiO_2, which presents very low surface energy resulting in Pt delamination.

Therefore, the direct deposition of Pt on LGS surface was tested. In order to check Pt adhesion, some typical tests have been driven: the deposition didn't delaminate/damage at all after scotch-tape, sonication, typical annealing, vacuum ambient pressure cycles, deposition on top, lithography, etching, stripping. Therefore, Pt adhesion is considered strong enough on LGS surface to survive to an entire device fabrication process. After annealing at 800 °C for 48 h, only very initial process of agglomeration present at the grain boundaries could be observed (Fig. 3). Note that, after annealing, Pt agglomeration is consirebaly slower directly on LGS than on buffer layers. No interdifussion between Pt and LGS was present (Fig. 4). Thus, the use of LGS/Pt stack is confirmed to be reliable for the purpose of SAW devices operating at high-temperature. However, a few defects were observed at the interface directly related to the quality of the LGS surface (Fig. 4). In order to maximize the robustness of the stack, further experiments have been performed to ameliorate LGS surface quality.

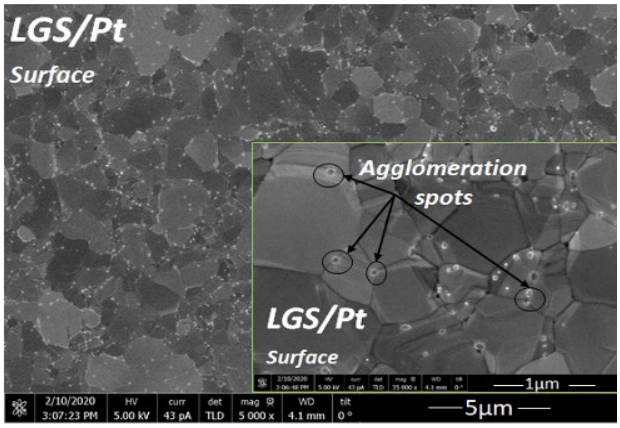

Fig. 3 SEM picture of LGS/Pt stack surface after 48h at 800°C. Inset zooms on the small agglomeration spots

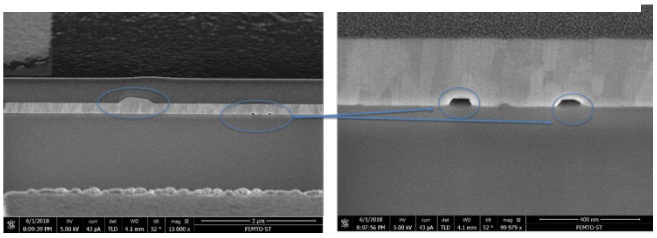

Fig. 4 SEM picture of LGS/Pt stack cross-section after 48h at 800°C. Right image zooms on the small interface defects.

B. LGS surface quality

Different LGS orientation (standard, Y, YX rotated) as well as langatate (LGT) with Z orientation were tested in order to study the surface quality and the surface energy effect of different cuts on the Pt agglomeration process. The latter showed a smoother surface than LGS (no matters the orientation considered). Following Pt growth and annealing, considerably lower amount of Pt agglomerated on Z-LGT was revealed, which highlights the fact that the surface of the substrate influences the Pt stability. This difference could be related to the higher surface energy of LGT than that of LGS, as smother surface facilitates the mobility and thus, the agglomeration should be increased. Note that LGT has not

been used further because of its higher instability related to Ga_2O_3 in comparison to LGS.

As-received LGS has a low roughness but it presents many surface defects. Microscopic picture shows some spot which are assumed to be due to the well-known Ga oxide migration towards LGS surface, which resulted in the interface defects illustrated in Fig. 4. This subsurface migration offers Pt a dirty growth. Considering that Ga_2O_3 is a volatile compound at elevated temperature, a first set of experiments consisting in LGS annealing have been realized. For the purpose of improving the quality of LGS (here Y-LGS is considered), annealing at different temperatures and for different durations have been performed. Fig. 5 presents AFM scan and microscopic pictures of Y-LGS as-received , annealed for 150h at 700°C, annealed for 48h at 900°C, annealed 90h at 900°C, and finally annealed 48h at 1100°C.

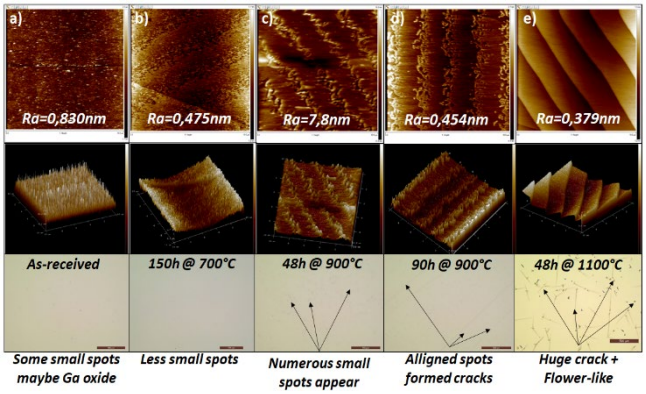

Fig. 5 Y-LGS surface characterization by AFM and optical microscope after different annealing treatments: as-received (a), after 150h at 700°C (b), after 48h at 900°C (c), after 90h at 900°C (d), after 48h at 1100°C (e). AFM scan size is 10x10µm² (top) and optical microscope scales bar represents 20µm in length (bottom).

After a long annealing at temperature below Tamman temperature, the surface is quite similar to that of the as-received substrate: low roughness (Ra=0.475nm) but with many surface defects (Fig. 5.b). One can see that the temperature is not high enough for surface mobility and for creation of terrace. The role of the annealing time can be seen in Fig. 5 c and d: after 48h, only rough teraces are observed and surface restructuration is not finished (Ra=7.8nm) whereas after 90h annealing, some cracks appears on the surface but the terraces are better structured and the surface is really smooth (Ra=0.454nm). Furthermore, a perfectly smooth and designed surface is obtained after 48h at 1100°C showing the cleanest terraces and a roughness Ra=0.379nm. However, the cracks are big and some new features appear. The smoothness of the surface is assumed to come from crystal rearrangement. A contrario, the cracks/flowers/spots are related to gallium oxide diffusion, confirmed by several studies dealing with LGS annealing and composition of the observed features [13]–[15]. The Ga diffusion coefficient (D) has been determined in air [16]: at 750°C, D is four orders of magnitude lower than at 1000°C which tends to validate that Ga_2O_3 is the cause of LGS surface damaging. Obviously, Ga/Ga oxide migrations damage the perfect stoichiometry of the compound so the acoustic properties and stability of the device. To summarize, the annealing close to Tamman temperatures allows to make terraces appearing and limit the Ga migration from the bulk so the clean surface without macroscopic defects, similar to as-received LGS can be obtained. After an appropriate cleaning step and annealing step, the cleaner surface with the diminution of spots (assumed to be Ga oxide) than as-received samples can be achieved. At T>Tamman temperature, volume diffusion is activated and Ga from the bulks migrates towards the surface and forms different features, damaging LGS composition and surface.

The pre-treatment of LGS surface by plasma have been tested, as well.Fig. 6 shows SEM images of Pt surface, deposited on LGS treated by different plasma processes followed by the typical annealing process. The LGS surface was treated by Reactive Ion Etching using CHF_3 gas (1 nm and 40 nm, Fig. 6 c and d) and by using Nanoplas (Fig. 6 b).

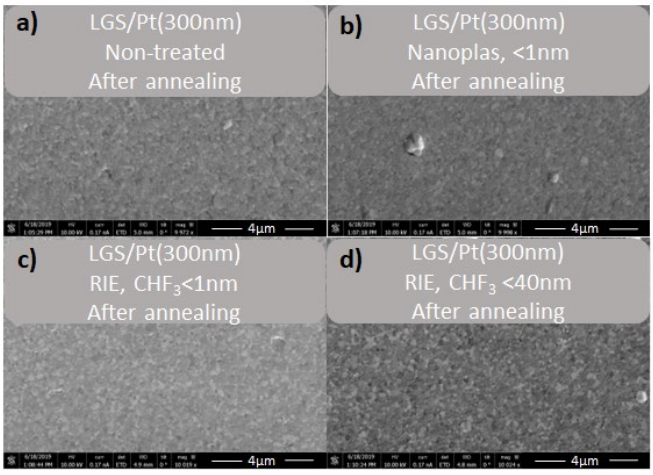

Fig. 6 SEM images of Pt surface on plasma treated LGS, annealed for 48h at 800°C: non-treated (a), Nanoplas <1nm (b), Reactive Ion Etching <1nm (c), Reactive ion Etching <40nm (d).

No significant difference was observed between Pt quality on plasma. Thus, these treatments were not further used.

C. Cladding layer and SAW device stability

To further ameliorate the Pt stability, the surface mobility and the agglomeration can be limited by using the cladding layer. The use of a cladding layer has already proven its efficiency in MEMS history. Ref [1], [4]–[7], [10] have investigated high temperature SAW devices using cladding layers. The stability of the stack got improved, so the same approach has been used in this work. The thermodynamical considerations of the reactivity of possible cladding layers (ZnO, TiO_2, ZrO_2, Al_2O_3, Ta_2O_5, SiO_2) with oxide components contained in LGS substrates already allow to understand which compounds are compatible (do not react) together. SiO_2 seems to offer the best properties as it is an LGS solution compound. In the same way Ta_2O_5 cladding layer would be preffered for LGT crystals.

Primarily, an LGS/Pt/SiO_2 stack consisting of continuous films have been deposited and annealed. SEM imaging allowed to prove the high-stability surface (Fig. 7 top) and interface (Fig. 7 bottom) of this heterostructure (Fig. 7 top).

Finally, SAW device fabrication and measurements have been performed using the optimized SiO_2/Pt/LGS stack. Optical image of the latter after typical annealing at 800 °C is shown in Fig. 8 a, where no damage is observable. Single-port resonator conductance has been measured after several curing times and are presented in Fig. 8 b. Note that a conditioning at operational temperature takes place during the first annealing process and the sensor frequency presents a shift. After a total of 384h annealing (spaced in days) at 600°C, the SAW

response remains stable in the limit of measurement errors, (between 248.85MHz and 548.89MHz).

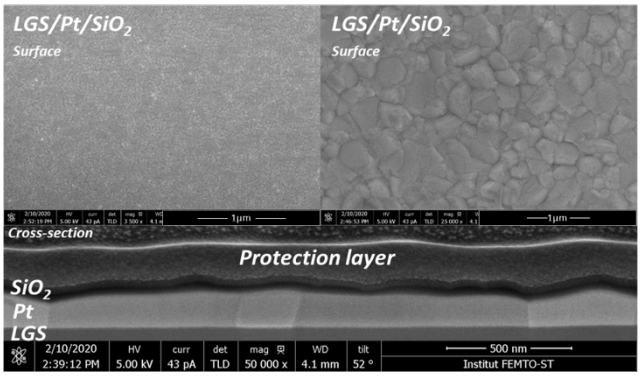

Fig. 7 SEM images of LGS/Pt/SiO_2 stack before (left) and after typical 800°C for 48h annealing (right, bottom): surface images on the top and cross-section on the bottom

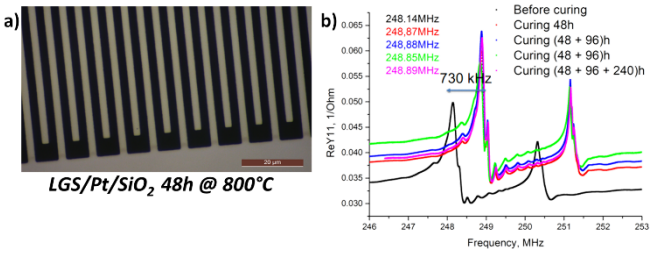

Fig. 8 Optical image of SAW device consisting of LGS/Pt/SiO_2 stack a), measurement of the device after several curing duration spaced in days for a total of 384h at 600 °C.

IV. Conclusion

Moreover, the stack has been limited to 3 materials (LGS, Pt, SiO_2), and the quality of LGS/Pt interface has been studied to optimize Pt adhesion. In that way, interdiffusion have been cancelled and the material issues are limited to agglomeration of IDTs. The use of a cladding layer allowed to seal Pt structure and incredibly decrease agglomeration. The measurement of a complete SAW sensor gives a promising proof of reliability concerning this new approach, although it needs deeper investigation to make the most of each layer.

A new approach consisting on the deposition of Pt directly on LGS has been studied for high temperature SAW devices. The study of materials reactivity, surface, crystallinity, allowed to attain an acceptable LGS/Pt interface quality for device processing. Good LGS surface quality for Pt growth could be obtained by an appropriate cleaning process and an annealing below Tamman temperature. The use of an adhesion layer has been given up as direct Pt growth on LGS ameliorated hetrostructure stability at high-temperature. SiO_2 cladding layer completely sealed Pt structure by limiting surface mobility and preventing agglomeration perfectly. Moreover, SiO_2 being a part of LGS did not present any interdiffusion issues. Eventually, SAW device measurement after annealing at 600°C for a total of 384h showed very good SAW frequency stability.

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References

[1] S. Moulzolf, R. Behanan, T. Pollard, R. J. Lad, and M. Cunha, *Capacitively coupled IDT for high temperature SAW devices*. 2013.

[2] M. Cunha, T. Moonlight, R. J. Lad, G. Bernhardt, and D. J. Franke, *P4L-1 Enabling Very High Temperature Acoustic Wave Devices for Sensor & Frequency Control Applications*. 2007.

[3] T. Aubert *et al.*, "Behavior of Platinum/Tantalum as Interdigital Transducers for SAW Devices in High-Temperature Environments," *IEEE transactions on ultrasonics, ferroelectrics, and frequency control*, vol. 58, pp. 603–10, Mar. 2011, doi: 10.1109/TUFFC.2011.1843.

[4] M. Cunha, T. Moonlight, R. Lad, D. Frankel, and G. Bernhard, *High temperature sensing technology for applications up To 1000°C*. 2008.

[5] D. J. Frankel, G. P. Bernhardt, B. T. Sturtevant, T. Moonlight, M. Pereira da Cunha, and R. J. Lad, "Stable electrodes and ultrathin passivation coatings for high temperature sensors in harsh environments," in *SENSORS, 2008 IEEE*, Oct. 2008, pp. 82–85, doi: 10.1109/ICSENS.2008.4716388.

[6] S. Moulzolf, D. Frankel, M. Cunha, and R. Lad, "High temperature stability of electrically conductive Pt–Rh/ZrO2 and Pt–Rh/HfO2 nanocomposite thin film electrodes," *Microsystem Technologies*, vol. 20, Apr. 2014, doi: 10.1007/s00542-013-1974-x.

[7] R. Behanan *et al.*, "Thin films and techniques for SAW sensor operation above 1000°C," in *2013 IEEE International Ultrasonics Symposium (IUS)*, Jul. 2013, pp. 1013–1016, doi: 10.1109/ULTSYM.2013.0260.

[8] G. Bernhardt, C. Silvestre, N. LeCursi, S. C. Moulzolf, D. J. Frankel, and R. J. Lad, "Performance of Zr and Ti adhesion layers for bonding of platinum metallization to sapphire substrates," *Sensors and Actuators B: Chemical*, vol. 77, no. 1, pp. 368–374, Jun. 2001, doi: 10.1016/S0925-4005(01)00756-0.

[9] A. Ababneh *et al.*, "Electrical and morphological characterization of platinum thin-films with various adhesion layers for high temperature applications," *Microsystem Technologies*, Nov. 2015, doi: 10.1007/s00542-015-2715-0.

[10] D. Frankel, S. Moulzolf, M. Cunha, and R. Lad, "Influence of composition and multilayer architecture on electrical conductivity of high temperature Pt-alloy films," *Surface and Coatings Technology*, vol. 284, Sep. 2015, doi: 10.1016/j.surfcoat.2015.08.074.

[11] X. Xu *et al.*, "Single crystalline highly epitaxial Pt thin films on (001) SrTiO3," *Applied Physics Letters*, vol. 92, pp. 102102–102102, Mar. 2008, doi: 10.1063/1.2890844.

[12] A. Kahsay, M. C. Polo, C. Ferrater, J. Ventura, J. M. Rebled, and M. Varela, "Growth of epitaxial Pt thin films on (001) SrTiO3 by rf magnetron sputtering," *Applied Surface Science*, vol. 306, pp. 23–26, Jul. 2014, doi: 10.1016/j.apsusc.2014.01.123.

[13] T. Aubert and O. Elmazria, *Stability of langasite regarding SAW applications above 800°C in air atmosphere*. 2012, p. 2101.

[14] M. Seifert, G. Rane, B. Kirbus, S. B. Menzel, and T. Gemming, "Surface Effects and Challenges for Application of Piezoelectric Langasite Substrates in Surface Acoustic Wave Devices Caused by High Temperature Annealing under High Vacuum," *Materials*, vol. 8, pp. 8868–8876, Dec. 2015, doi: 10.3390/ma8125497.

[15] M. D. Malinkovich, Yu. N. Parkhomenko, E. A. Skryleva, and N. Yu. Shulga, "XPS study of gallium loss from langasite crystal surface under vacuum annealing," *Sensors and Actuators A: Physical*, vol. 180, pp. 63–66, Jun. 2012, doi: 10.1016/j.sna.2012.04.017.

[16] M. Schulz, H. Fritze, H. L. Tuller, and H. Seh, "Diffusion-related implications for langasite resonator operation," *IEEE Transactions on Ultrasonics, Ferroelectrics, and Frequency Control*, vol. 51, no. 11, pp. 1381–1387, Nov. 2004, doi: 10.1109/TUFFC.2004.1367476.