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Thin Film Electrodes for High Temperature Surface Acoustic Wave Devices

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Abstract

Wireless surface acoustic wave devices based on langasite allow sensor operation in high-temperature environments. While langasite shows piezoelectric behavior up to temperatures close to its melting point of 1470 °C, the stability of thin film electrodes for excitation of acoustic waves limits the application temperature. Different metal and ceramic based thin films are tested regarding their applicability as electrode materials for wireless surface acoustic wave devices at temperatures above 600 °C. The devices should withstand temperatures of 800 °C for at least several hours. The high-temperature stability of platinum, platinum-rhodium, iridium as well as lanthanum strontium manganite (LSM) based electrodes is characterized using electrical measurements, x-ray diffraction (XRD) measurements and scanning electron microscopy (SEM) imaging. SAW devices with Ti/Pt thin film electrodes are operated up to temperatures of 800 °C. LSM/Pt films exhibit a good stability up to temperatures of 800 °C. Above 600 °C its resistance is comparable to platinum films .

Keywords: high-temperature, langasite, electrode materials, surface acoustic wave

1. Introduction

Piezoelectric single crystals like langasite enable the development of high-temperature stable surface acoustic wave devices for sensor applications in harsh environments. Especially the option of wireless sensor interrogation permits applications in systems, which are currently not accessible with conventional wired sensors. Although e.g. langasite shows piezoelectric behavior up to its melting point at 1470 °C [1], the high-temperature operation of piezoelectric devices is limited by the stability of electrode materials and packaging.

Typical reasons for failures of thin film electrodes are oxidation, evaporation and agglomeration. Although platinum group metals are in general chemically stable, they often form a thin oxide layer on the surface [2]. While an evaporation of the pure metal in this material group is very unlikely due to the low vapor pressures of the platinum group metals [3], the formed oxides might be volatile, which results in a constant mass loss of the electrodes [4].

Agglomeration is the dominant failure mechanism of most thin films of the platinum group metals [5]. During the deposition process, the thin metal layers shifts its lattice constant towards the lattice constant of

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the substrate or adhesion layer, which induces stress in the layers. At elevated temperatures, the film relaxes due to a lower stiffness and shifts its lattice constant towards the bulk value. This results in the formation of insulated platinum areas and therefore in the destruction of the electrode.

Potentially suited materials and material systems are investigated with respect to their applicability at temperatures above 600 °C. At 800 °C and above, the devices should be operational for at least several hours. With respect to the specific application background the thickness of heavy platinum electrodes should not exceed 150 nm due to its influence on the surface acoustic waves. The electrodes should remain stable under harsh conditions while exhibiting a good adhesion and high conductivity. The tested materials include metal based electrodes like platinum, platinum/rhodium, palladium and iridium as well as conductive ceramics like lanthanum strontium manganite (LSM), which are already used as conductive materials in high-temperature fuel cells [6].

2. Experimental

The preparation of thin films for degradation tests is performed using pulsed laser deposition (PLD) or magnetron sputtering. In contrast to other deposition methods, layers prepared using PLD show commonly negligible shifts in cation stoichiometry from target to substrate, making it suitable even for complex ceramic materials like LSM. For electrode structuring of the final SAW devices nano imprint lithography (NIL) is used, which enables low cost nano-structuring on planar substrates. NIL is based on the principle of defining a pattern on a planar substrate by pressing a pre-patterned template into a thin polymer film (resist). Here, the metallization is applied using electron beam deposition.

The high-temperature stability of the tested thin films is characterized using electrical resistance measurements with a Keithley 1000 multimeter in a four terminal setup to eliminate the wire resistance, x-ray diffraction (XRD) measurements and scanning electron microscopy (SEM) imaging.

3. Results

3.1. Platinum based electrodes

Different platinum and platinum-rhodium (10 %) thin films with 10 nm titanium adhesion layers are applied on polished langasite substrates. The resistance of the films at temperatures up to 1100 °C in steps of 100 K and a dwell time at every temperature of 6 hours is measured. Figure1 shows the time dependent resistance of the tested films.

The Ti/Pt electrode with a total thickness of 110 nm shows a stable resistance with no resistance increase up to temperatures of 800 °C. At 900 °C, a strong degradation of the electrode is observed. The latter is caused by the formation of agglomerates (figure 2, top). By applying a thin alumina protection layer of 15 to 20 nm, the stability of platinum based electrodes can be increased significantly. The alumina layer prevents

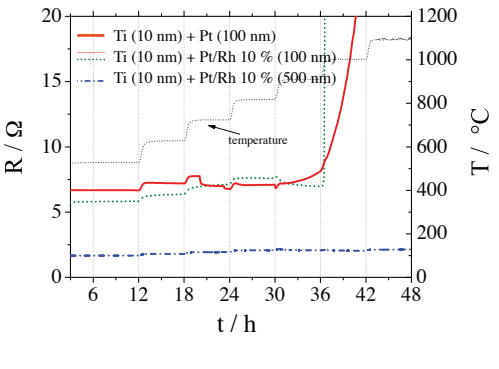

Figure 1: Temperature and time dependent resistance of different platinum based thin films.

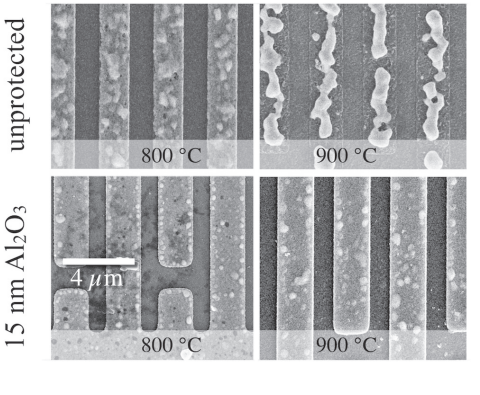

Figure 2: Ti/Pt thin film after annealing at 800 °C and 900 °C without protection layer (top) and with an 15 nm alumina layer applied on the whole device (bottom).

or at least suppresses the agglomeration process and shifts the stability range to at least 900 °C (figure 2 bottom).

Another possibility to increase the high-temperature stability of platinum-based electrodes is the application of a platinum-rhodium alloy instead of pure platinum, since platinum-rhodium exhibits a higher melting point and leads to an increased oxidation resistance [7]. The titanium/platinum-rhodium (10 %) film with a total thickness of 100 nm shows a significantly higher stability at temperatures of 900 °C. Above that, the platinum-rhodium also agglomerates and therefore looses its electrode functionality.

3.2. Iridium electrodes

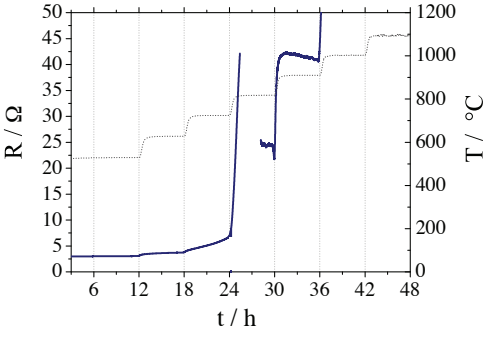

Figure 3: Temperature and time dependent resistance of an iridium thin film (100 nm).

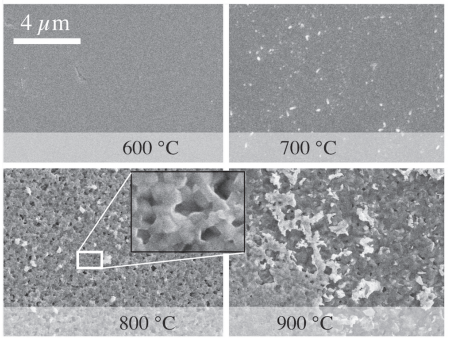

Figure 4: SEM pictures of iridium thin films (thickness 200 nm) after annealing for 6 hours at different temperatures.

Iridium films with a thickness between 100 nm and 200 nm are prepared. Compared to the tested platinum based films, iridium shows a good adhesion on the substrate even without an additional adhesion layer. A possible explanation is the yield strength of iridium, which is much higher than that of other noble metals.

Iridium exhibits a very good high temperature stability up to temperatures of 650 °C. After 150 hours at this temperatures, no significant degradation of the thin films is observed. At 700 °C, the resistance measurements indicate a significant degradation followed by a total destruction of the layer at 800 °C (figure 3). The SEM pictures (figure 4) show, that the degradation process is not caused by agglomeration. The crystallite structures marked in figure 4 are identified as IrO_2 by XRD, which is formed at temperatures of 700 °C and above. At 800 °C, a temporary loss of the resistance signal is observed, which might be related to the transition of Ir to Ir_2O_3. After the recovery of the signal, the film resistance is about one order of magnitude larger than before, which is comparable to the expected resistance value for Ir_2O_3 [8]. At temperatures around 1000 °C, volatile IrO_3 is formed and leads to an evaporation of the thin film [9].

3.3. Ceramic electrodes

In order to test ceramic electrodes, LSM ($La_{0.65}Sr_{0.35}MnO_3$) is chosen. At temperatures of 600 °C and above, LSM electrodes with 200 nm thickness shows a resistance, which is only by a factor of 5 to 10 larger than that of the metal films. Up to temperatures of 900 °C, a stable resistance is found (see figure 5).

Beside pure LSM layers, a co-deposited layer of LSM and platinum is prepared using a segmented PLD target (volume ratio LSM:platinum 1:1). The thickness of the LSM/Pt film is 100 nm. Compared to pure LSM electrodes, a relatively low resistance is observed at temperatures below 600 °C. At temperatures of 700 °C, the resistance is comparable to values obtained for pure metal electrodes. First degradation effects are visible at temperatures of 900 °C. A further optimization of this material combination is expected to deliver higher stability and lower resistances.

3.4. High-temperature surface acoustic wave measurements

First measurements with Ti/Pt coated SAW resonators are performed at temperatures up to 800 °C. Figure 6 shows the spectra of an resonator type device up to temperatures of 800 °C. An operation at this temperature is possible for at least three hours, while the device remains fully functional. A shift to higher operation temperatures is expected by application of above mentioned protection layers.

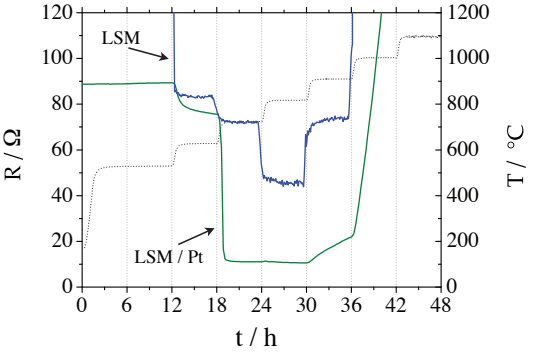

Figure 5: Temperature and time dependent resistance of LSM and LSM/Pt thin films.

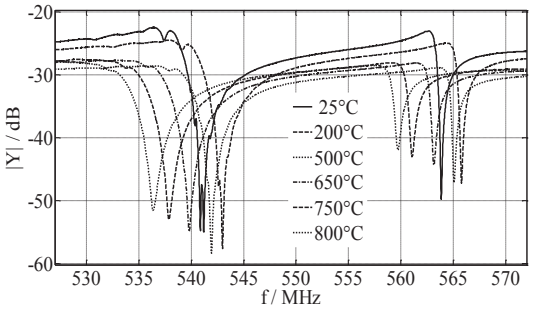

Figure 6: Admittance spectra of a titanium/platinum (100 nm) coated langasite one-port synchronous SAW resonator cut with Euler angles (0°,22°,31°) at temperatures up to 800 °C.

4. Conclusions

The high-temperature stability of metal and ceramic thin films for langasite based surface acoustic wave devices is tested. Among the metal based films, Pt/Rh (10 %) exhibits the best high-temperature stability for a given film thickness. A stability of the film is demonstrated up to temperatures of 900 °C. Platinum films are long-term stable up to temperatures of 700 °C. A stability in the range of hours is given up to 800 °C and is therefore suitable for short-term operation. The degradation process of the platinum based films is predominantly caused by agglomeration. This process can be significantly suppressed by deposition of an additional alumina protection layer. The investigation of iridium thin films show a degradation at temperatures of 700 °C, which is dominated by the oxidation of the material. Promising results are obtained for thin LSM and LSM/Pt films. The latter are stable up to temperatures of 800 °C and show a good conductivity at temperatures above 600 °C.

Ti/Pt coated langasite SAW resonators are successfully operated at temperatures up to 800 °C for several hours.

5. Acknowledgements

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