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Ir-Rh thin films as high-temperature electrodes for surface acoustic wave sensor applications

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ABSTRACT

The achievement of surface acoustic wave (SAW) devices stable in high-temperature oxidizing atmospheres requires the development of conductive thin film electrodes that can withstand such harsh conditions. Recent studies have demonstrated the suitability of Pt-based alloys, multilayers or nanocomposite films for temperatures up to 800 °C. Electrodes based on new materials and structures still have to be developed for applications taking place at higher temperatures. In this perspective, thin films based on iridium could be good candidates regarding the high melting point, and thus the low diffusion coefficients of this noble metal. In particular, Ir-Rh bulk alloys have shown superior performance as spark plug electrodes, which have to resist concurrently to physical and chemical wear such as high-temperature SAW electrodes. Consequently, this paper deals with the high-temperature behavior of Ir-Rh thin films. Ir-Rh alloys and multilayers films, with an Ir atomic ratio between 10 and 50%, are deposited by one-gun electron beam evaporation method. The impact on the films of a 4 days annealing treatment at 800 °C in air is studied by X-ray diffraction, scanning and transmission electron microscopy, electron energy loss spectroscopy and four-points probe resistivity measurements. It turns out that all the films oxidized during the annealing period. The post-annealing electrical properties are highly dependent of the initial composition of the film: the higher is the Ir rate in the film, the lower is the electrical resistivity after annealing. Moreover, an Rh_2O_3 overlayer, with a thickness of some tens of nanometers, forms at the surface of the film, confirming previous observations made on Ir-Rh bulk alloys. First SAW measurements made on devices based on $Ir_{30}Rh_{70}$ alloy electrodes are very promising as a SAW signal is still clearly visible after the 4 days annealing process, while no agglomeration phenomenon can be observed.

1. Introduction

There is currently a strong industrial request to develop wireless measurement solutions for a variety of industrial applications taking place between 200 °C and 1000 °C that cannot be addressed by wired sensors. For instance, in metallurgy domain, such sensor could allow an accurate monitoring of the temperature at different positions on a steel band undergoing a thermal annealing treatment cycle, and thus a better adjustment of the annealing line parameters. Surface acoustic wave (SAW) technology could meet this need as SAW devices are passive components, thus requiring neither embedded electronics nor power source to be wirelessly interrogated. These devices are composed of a piezoelectric substrate on which are built interdigited conductive electrodes called interdigital transducers (IDTs). The typical thickness of the thin film from which the IDTs are lithographed is one hundred nanometers. One of the main challenges to face in order to achieve high-temperature wireless SAW sensors is the development of the constitutive materials able to withstand elevated temperatures up to 1000 °C and corrosive atmospheres, starting with hot air. Concerning the piezoelectric substrate, langasite ($La_3Ga_5SiO_{14}$; LGS) is currently the most appropriate candidate. It has been intensively studied, demonstrating a good stability at high temperature [1,2]. However, it has to be noted that, under high-temperature vacuum

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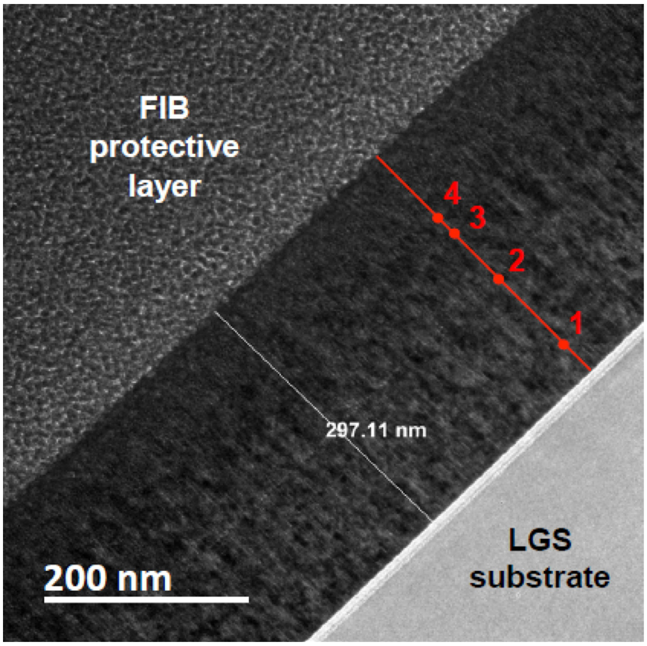

Fig. 1. Cross-section bright-field TEM image of an as-deposited 300 nm-thick $Ir_{10}Rh_{90}$ thin film. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article).
Red points indicate the EELS measurements locations.

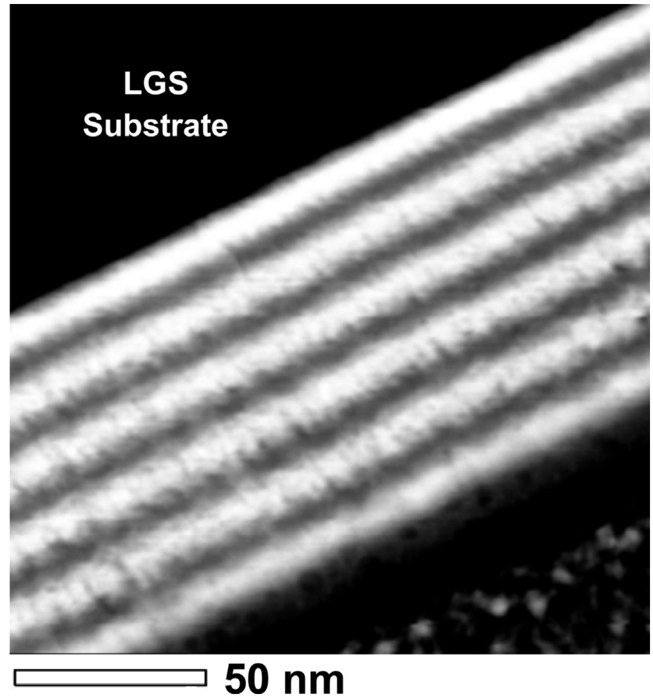

Fig. 2. Cross-section bright-field TEM image of an as-deposited 12 × 10 nm $Ir_{50}Rh_{50}$ multilayers thin film.

conditions, LGS suffers from surface degradation related to gallium and oxygen losses [3–5]. Regarding the IDTs, the use of a material having good corrosion resistance seems required. First studies naturally turned to platinum regarding its exceptional noble character and a melting temperature as high as 1773 °C [6,7]. However, it was observed that 100 nm-thick Pt thin films agglomerate at temperatures higher than 700 °C [8–10]. Agglomeration is a phenomenon specific to thin films, related to their large surface tension, which transforms a continuous thin film in a more stable configuration from the thermodynamics point of view, namely in a collection of separate beads. Agglomeration kinetics depend both on the film thickness and the nature of the film material. The thicker is the film, the slower are agglomeration phenomena. On the other side, as agglomeration is driven by atomic diffusion phenomena, it is slower for metals with higher melting temperature and thus smaller diffusion coefficients. Consequently, agglomeration phenomena can be hindered by adding impurities in pure metal films in order to decrease diffusion coefficients. This strategy was successfully employed by Pereira da Cunha et al. who significantly increased the performance of Pt-based IDTs by the use of Pt-Rh alloys, Pt-Rh/ZrO_2 multilayers and Pt-Rh/ZrO_2 nanocomposites. This group successfully operated an LGS-based SAW device with Pt-10%Rh/ZrO2 nanocomposites electrodes for more than 5 months at 800 °C in air atmosphere [11]. Since then, new electrode materials and structures have been investigated by this group towards establishing stable operation of LGS-based SAW high-temperature sensors at 900 °C and beyond [12].

In order to achieve electrodes that are able to operate in corrosive atmospheres at temperatures of 800 °C and more, it could be relevant to replace platinum by a noble metal with lower intrinsic diffusion coefficients. In this context, iridium is an appropriate candidate as its melting temperature (2447 °C) is significantly higher than that of platinum. First investigations on pure Ir electrodes were very promising, confirming the high resistance of iridium thin films to agglomeration phenomena [10–13]. However, these results were obtained under vacuum conditions as Ir transforms into volatile IrO_3 above 800 °C in oxidizing environments [14]. This drawback could be overcome by alloying iridium with another element. Indeed, Osamura et al. studied the high-temperature behavior of spark plug electrodes made of various Ir-based alloys, including Ir-W, Ir-Re, Ir-Mo, Ir-Ru, Ir-Hf, Ir-Pt, Ir-Pd, Ir-Ni and Ir-Rh [15]. Material challenges to face for this application are very similar to those encountered in the development of high-temperature SAW IDTs. Indeed, it is necessary to employ a conductive material with a high melting point to improve the sparking wear resistance. Moreover, as spark plugs are exposed to a hot environment (temperature can exceed 2000 °C) containing large amounts of oxygen, it is important that the material from which spark plug electrodes are made be highly resistant to oxidation.

Osamura et al. found that among all the investigated Ir-based alloys, Ir-Rh alloys show the best performance in such conditions. In particular, Ir-Rh alloys show a far better resistance to oxidation than pure iridium above 800 °C. This phenomenon was explained by the formation of a thin rhodium oxide passivation layer at the surface of the samples during annealing. In fact, Osamura et al. assumed that Ir atoms close to the surface transform into IrO_3 and fly off, so that only Rh atoms remain close to the surface. The latter also oxidize into Rh_2O_3 which is a stable oxide that hinders O diffusion deeper inside the sample. Unfortunately, the thickness of the Rh_2O_3 passivation layer is not given in this study. Note that the Rh protection effect is maximized as soon as the Rh content reaches 10 wt%. Very few other studies were dedicated to Ir-Rh alloys and none of them to Ir-Rh thin films with a thickness in the range of some hundreds nanometers.

Based on Osamura's study, it appears that Ir-Rh thin films could be good candidates for SAW applications taking place in high-temperature oxidizing atmospheres. Consequently, the goal of this study is to make a first investigation on the high-temperature behavior of such Ir-Rh thin films, to determine their relevance for the targeted application.

2. Experimental

Ir-Rh thin films were deposited on Y and Y + 50°-cut LGS substrates (Witcore Co. Ltd., Jinan, China) by one-gun electron-beam evaporation method (Edwards Auto 306). The base pressure was