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Acoustic wave liquid sensors enhanced with glancing angle-deposited thin films

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ABSTRACT

The application of nanostructured thin films grown by glancing angle deposition (GLAD) to enhance the sensitivity of Love wave liquid sensors was investigated. The effect of high-angle GLAD films, with and without nanostructure modification by ion milling and clustering, on device response was studied. Ion milling was used to prevent clustering of individual nanostructured posts, and was compared to films that were intentionally clustered. Both sets of modified films were shown to eliminate signal losses due to damping from the non-rigid, as-deposited posts. Sensitivity enhancement was tested by viscous loading with varying mixtures of glycerol and de-ionized water. Frequency shifts were found to be non-linear with the square root of the density–viscosity product, $(\rho\eta)^{1/2}$, and were modeled with exponentials. The sensitivity was shown to increase with film thickness, but decreased with increasing values of $(\rho\eta)^{1/2}$. There was also an increase in signal loss with film thickness under a liquid environment. The balance between sensitivity enhancement and signal loss must be carefully optimized with consideration for the intended application.

1. Introduction

In the past two decades, surface acoustic wave (SAW) devices have been thoroughly studied as a sensing platform [1–3]. They have found application in gas sensing [4–7], as well as chemical and biological sensing in liquid environments [8–10]. While the most common SAW devices use Rayleigh waves, these waves are unsuitable for operation in liquids due to the significant radiation losses into the liquid [11,12]. As a result, shear-horizontal (SH) polarized waves have been employed because they do not elastically couple with the liquid. In particular, Love wave devices have been extensively studied because of their high sensitivity [13,14]. These devices consist of a piezoelectric substrate and a thin guiding layer, which confines the energy of the wave to the surface. A variety of materials have been used for the waveguide layer, including silica [15,16], polymethyl methacrylate (PMMA) [17,18], and photoresist [19,20].

Nanostructures provide a high surface area-to-volume ratio, which makes them ideal for coupling with SAW sensors to increase sensitivity. They can be fabricated from a variety of techniques, such as physical vapor deposition (PVD), chemical vapor deposition (CVD) and the vapor–liquid–solid (VLS) method. Various groups have employed nanostructures on SAW devices to increase effective surface area, and in turn, the sensitivity [21–23]. However, almost all reports have been on enhancing SAW gas sensors, with few studies on the enhancement of liquid sensors [24–26]. One reason for this is that ordered nanostructures tend to cluster or clump from the liquid capillary forces, which has caused them to be deemed unsuitable for sensors in a liquid environment [27,28]. We have shown, in a previous study, that ion-milling is an effective way of eliminating clustering for vertically aligned nanostructures grown by glancing angle deposition (GLAD) [29]. These modified structures are thus promising candidates for integration with sensors operating in a liquid environment.

In this study, we investigate the use of high surface area, nanostructured thin films fabricated by GLAD to enhance the performance of Love wave SAW liquid sensors. Nanostructured thin films are first deposited on top of the SAW devices. One set of devices is then irreversibly clustered while another set is modified by ion-milling for comparison. We report on the how these film modifications affect the device response, in comparison to bare Love wave devices, and the effect of film thickness on response. Sensitivity enhancement from the increased surface area is investigated by viscous loading of varying mixtures of glycerol and water, and compared to bare Love wave devices.

2. Experimental

2.1. Love wave devices

Love wave SAW devices were fabricated as per our previous study [30]. Briefly, ST-cut quartz (Sawyer Technical Materials, LLC) was used to fabricate delay-line SAW devices. The interdigitated transducers (IDTs) were made of a Cr/Au layer and consisted of 50

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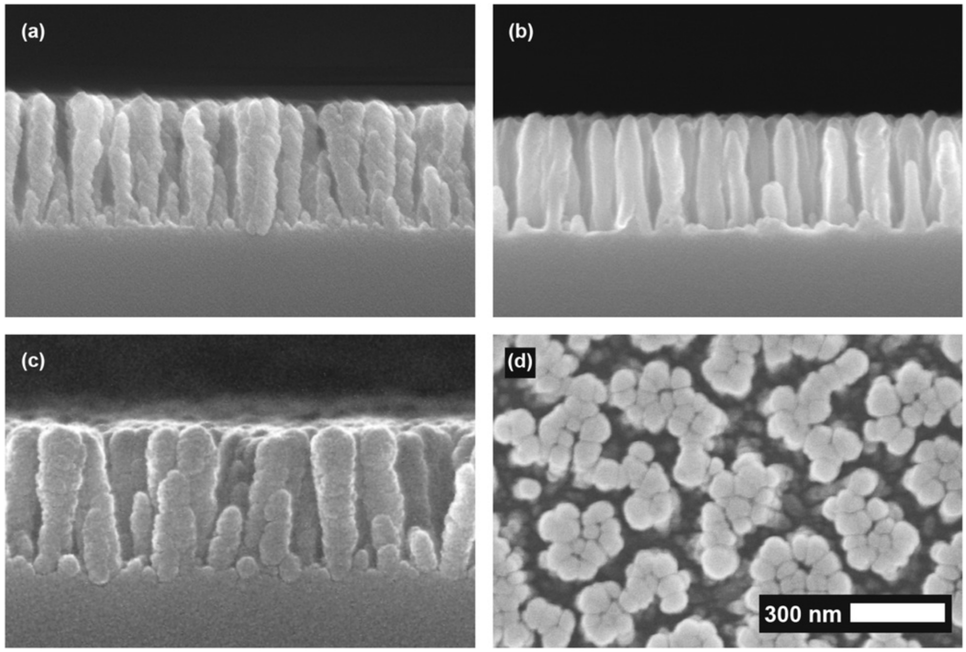

Fig. 1. SEM images of (a) an as deposited GLAD film, followed by (b) ion-milling, or (c) clustering. (d) Top-down SEM image of (c). Scale bar is applicable to all images.

pairs of split electrodes with a period of 40 μm, a metallization ratio of 0.5, and an aperture of 3 mm. The center-to-center spacing of the IDTs was 5 mm. The propagation direction was aligned perpendicular to the crystallographic *x*-axis. These parameters gave rise to a measured fundamental frequency of ∼125 MHz.

Polymethyl methacrylate (PMMA) was used as the wave guide material. Prior to PMMA deposition, the devices were cleaned by sonicating in acetone for 10 min, followed by rinsing with acetone, isopropyl alcohol (IPA), and de-ionized (DI) water. 495PMMA A11 (MicroChem) was spun onto the SAW devices at 4500 rpm for 50 s, followed by a bake at 185 °C for 3 min. The PMMA on the electrode pads was carefully removed with a razor blade. The thickness of the guiding layers was about 1.1 μm, as measured with a KLA-Tencor Alphastep IQ profilometer. The fundamental frequency of the guided devices shifted down to ∼123 MHz.

2.2. Glancing angle deposition

Vertical post nanostructured thin films were fabricated using GLAD. Details of this PVD technique can be found elsewhere [31,32]. An electron-beam evaporation system (Kurt J. Lesker AXXIS system) using SiO_2 source material (1–3 mm pieces, 99.99% purity from Cerac, Inc.) was used to deposit onto the Love wave devices (active area and IDTs). A 30 nm layer of solid film was first grown at an angle of 30°, followed by films deposited at an angle of 86° relative to the substrate normal with a pitch of 5 nm. An angle of 86° was used to produce highly porous films with well separated structures. Films were deposited at a deposition rate of 1 nm/s as measured by a quartz crystal microbalance (QCM). Half the devices were then ion-milled while the other half were clustered. An example of an as-deposited film can be seen in Fig. 1(a).

Ion-milling was carried out using an Ionfab 300 Plus (Oxford Instruments) at the University of Alberta NanoFab. Samples were milled at normal incidence relative to the substrate surface, with a beam voltage of 950 V at a current of 120 mA over an area with a diameter of 14 cm. The substrate holder was rotated at 20 rpm during milling to maximize ion-beam exposure uniformity. Milling times varied from 25 s to 40 s (increasing time with thickness), and were chosen with consideration of the minimum milling time required to prevent clustering, as per our earlier work [29]. Ion-milling has the effect of shortening the height and smoothening the sides of the nanostructures by removing material from the tops of the post and redepositing the material on the sides and base of the nanostructures. Some material is completely removed from the substrate as well. The diameter at the base of the nanostructures is also increased by the redeposition of material, and combined with the smoothening effect, these result in more robust nanostructures that will not cluster under capillary forces. Fig. 1(b) shows an example of an ion-milled film used in this study.

Clustering of the nanostructured films was achieved by placing a small droplet of DI water on top of the film, and then allowing it to dry off at room temperature. The devices were then rinsed with isopropyl alcohol to wash off residue that may have been present on the film. The effects of clustering can be seen in Fig. 1(c) and (d).

2.3. Measurement setup

The transmission characteristics of all devices were measured with a Hewlett-Packard 4396B network analyzer. The insertion loss and phase were examined immediately before and after film deposition and then ion-milling. A scanning electron microscope (Hitachi S4800) was used to image and characterize film structures and thicknesses.

The Love wave devices were incorporated into an oscillating circuit and the signal was coupled out to a frequency counter (Agilent 53132A) that was interfaced with a computer. The oscillating circuit included two amplifiers (Minicircuits ZFL-500), a low-pass filter (Minicircuits SLP-150), and a directional coupler (Minicircuits ZFDC-10-1). Sensitivity to viscous loading was investigated by flowing varying mixtures of DI water/glycerol (Fisher Scientific) over the active area of the devices with a custom flow cell with a Viton o-ring seal. The film on top of the IDTs and on the contact area between the o-ring and the device was carefully removed using a cotton-tipped applicator. A NE-4000 double syringe pump (New Era Pump Systems, Inc.) was used to exchange mixtures over the active area. The fluid under test was held in place for 10 min during each measurement, which allowed signals to settle. Temperature was kept at 25 °C using an Octagon 20 Advance incubator (Brinsea).