proceedings

Proceedings

Diamond Coated LW-SAW Sensors-Study of Diamond Thickness Effect †

L. Drbohlavová [1,2,*], J. C. Gerbedoen [3], A. Taylor [1], A. Talbi [3], L. Fekete [1], P. Ashcheulov [1], A. Soltani [3,4], V. Bovtun [1], M. Kempa [1], J. Bartoň [5,6], P. Cígler [5] and V. Mortet [1,2]

[1] Institute of Physics, Academy of Sciences Czech Republic v.v.i, Prague 8, Czech Republic; taylor@fzu.cz (A.T.); fekete@fzu.cz (L.F.); ashcheulov@fzu.cz (P.A.); bovtun@fzu.cz (V.B.); kempa@fzu.cz (M.K.); mortetv@fzu.cz (V.M.)
[2] Czech Technical University, Faculty of Biomedical Engineering, Kladno, Czech Republic
[3] University Lille, Centrale Lille, UVHC, ISEN, LIA LICS/LEMAC-IEMN UMR CNRS 8520, F-59000 Paris, France; jean-claude.gerbedoen@iemn.univ-lille1.fr (J.C.G.); Abdelkrim.Talbi@iemn.univ-lille1.fr (A.T.); ali.soltani@iemn.univ-lille1.fr (A.S.)
[4] LN2, Université de Sherbrooke, 3IT, 3000 Bld de l'Université, Sherbrooke, QC, Canada
[5] Institute of Organic Chemistry and Biochemistry, v.v.i., Academy of Sciences of the Czech Republic, Flemingovo nam. 2, 166 10 Prague 6, Czech Republic; Barton@uochb.cas.cz (J.B.); cigler@uochb.cas.cz (P.C.)
[6] Faculty of Science, Charles University, Hlavova 2030, 128 40 Prague 2, Czech Republic
* Correspondence: drbohlavova@fzu.cz; Tel.: +420-266-052-634

† Presented at the Eurosensors 2017 Conference, Paris, France, 3–6 September 2017.

Published: 9 August 2017

Abstract: This study focuses on the fabrication and characterization of Love wave surface acoustic wave (LW-SAW) sensors with a thin nano-crystalline diamond (NCD) coating with an integrated microfluidics system. The effect of diamond layer thickness on the acoustic wave phase velocity and the sensor's sensitivity have been investigated experimentally and compared with theoretical simulations. The fabricated sensors have been tested with a several liquids using a home-made microfluidics system.

Keywords: Love waves; acoustic sensors; CVD diamond; sensing in liquids

1. Introduction

Biosensors are a very promising tool for bacteria detection, which can potentially replace conventional time consuming and labour intensive microbiological techniques [1]. Amongst acoustic sensors, Love wave surface acoustic wave (LW-SAW) sensors are the most sensitive one which can operate in liquids. Elastic waves generated by the piezoelectric substrate are coupled to a guiding layer with a lower phase velocity than the substrate's one. In LW-SAW, the displacement of particles is parallel to the surface and perpendicular to the direction of propagation, which allows waves to travel in contact with liquids without acoustic radiative losses in liquids [2,3]. When the LW-SAW sensor operates in liquid, the wave propagation is affected mainly by mass loading, and could be also affected by viscous loading and acoustoelectric coupling between electric field and ions and dipoles in liquid leading to the changes in propagation velocity, attenuation of signal and variation of resonant frequency and insertion loss [2]. Real-time monitoring sensors require a long term stability of attached bio-receptors. Several chemical functionalization schemes were developed in last decades that enabled biomolecule attachment onto diamond surface [4,5]. Recent study demonstrated prolonged stability of attached biomolecules in comparison to silicon or gold surfaces [6]. For these reasons, in this work, we investigate the properties of diamond coated LW-SAW sensors for their potential use as bio-sensors.

2. Materials and Methods

LW-SAW sensors were fabricated on ST-cut quartz crystals with 200 nm thick aluminum interdigital transducers (IDTs) with two different spatial periods—λ = 16 and 32 µm using photolithography and lift-off techniques. An amorphous SiO_2 layer (h_{SiO2} = 1.45 µm) acting as a guiding layer was deposited using plasma enhanced chemical vapour deposition (PECVD). Thin nano-crystalline diamond (NCD) layers were deposited using a microwave linear antenna plasma enhanced chemical vapour deposition (MW-LA-PECVD) system. To prevent diamond growth on the IDTs contact pads, a selective diamond seeding process using clean lab tape was used. All diamond depositions were carried out at low temperature (<500 °C) to preserve the piezoelectric properties of the quartz crystal. The effect of diamond layer thickness was investigated by deposition of consecutive diamond layers. Surface roughness and surface morphology of the diamond coating were investigated using a Dimension Icon ambient atomic force microscope in Peak Tapping mode. Diamond layers were also characterized by Raman spectroscopy using Renishaw InVia Raman microscope with 488 nm laser. Sensor sensitivity has been determined by measurement of the frequency response of the diamond coated (100 nm thick) LW-SAW sensor as a function of the thickness of a thin polymer deposited on the sensor surface. The polymer consists of a multi-layer of diluted lift-off resist (LOR) deposited by spin coating at 5000 rpm for 30 s and subsequently baked for 110 °C for 150 s on a hot plate. IDTs contact pads were also protected for LOR deposition by clean lab tape. Frequency response of the LW-SAW sensors were carried out using a vector network analyzer Agilent E8364B and Summit 9000 Analytical Probe Station with Infinity Probes at room temperature. LW-SAW sensors were tested with four different liquids—water, methanol (MeOH), isopropylalcohol (IPA) and phosphate buffer saline (PBS composition: 8 g of NaCl, 0.2 g of KH_2PO_4, 1.27 g of Na_2HPO_4 and 100 mL (0.02% (*w*/*v*)) EDTA in 1 l of deionized water) using an home-made microfluidics set-up.

3. Results and Discussion

3.1. Effect of NCD Layer Thickness

Figure 1a shows AFM picture of a 30 nm thick NCD layer deposited on a LW-SAW device. The NCD layer is well coalesced and nearly closed. Figure 1b) shows the Raman spectra of NCD layers with different thicknesses. The diamond peak at 1332 cm^{-1} can be clearly observed. No significant fraction of sp^2 carbon phase is observed. The peak at 1490 cm^{-1} is attributed to transpolyacetylene [7]. On Raman spectra for 30 nm thick NCD layer, one can observe SiO_2 substrate's peaks at 800 and 1100 cm^{-1}. Figure 2a shows the centre frequency shift and phase velocity of a LW-SAW sensors with a silica normalized thickness h_{SiO2}/λ = 0.1 as a function of the diamond layer thickness. It is clearly observed that both the phase velocity and the centre frequency increase with the increasing surface's stiffness of thick NCD layers. This intuitive result is consistent with theoretical modelling [8].

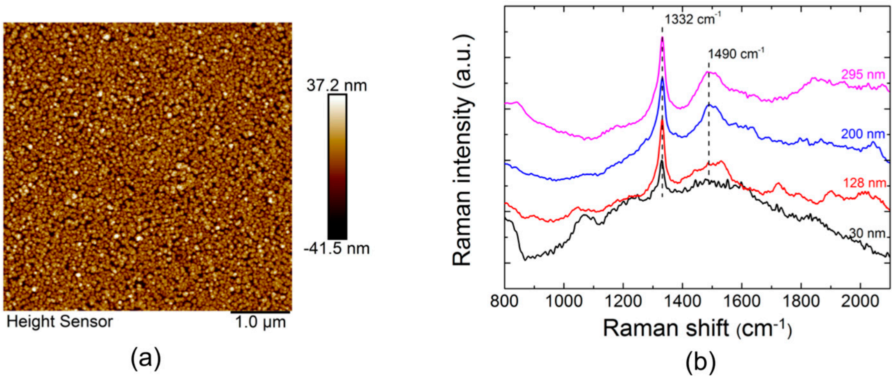

Figure 1. (**a**) AFM picture of 30 nm thick NCD layer deposited on a LW-SAW device; (**b**) Raman spectra of NCD layers with different thicknesses.

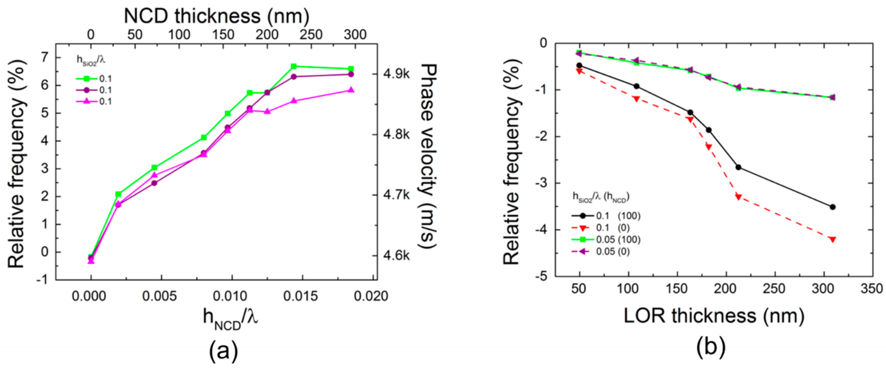

Figure 2. (**a**) Relative frequency shift and phase velocity of LW-SAW sensors with various thicknesses of NCD coating for a silica's normalized thickness h_{SiO2}/λ = 0.1; (**b**) Relative center frequency shift as a function of the LOR thickness

3.2. Sensitivity

The measurement of sensor's sensitivity was carried out on a 100 nm thick NCD coated LW-SAW sensor with two different silica normalized thicknesses h_{SiO2}/λ = 0.05 and 0.1 and compared to uncoated (i.e., no diamond layer) sensors. Figure 2b) clearly shows the sensor centre frequency decrease with the increasing the LOR thickness and its mass loading. The frequency shift of uncoated and NCD coated sensors are comparable for both normalized thicknesses. The LW-SAW sensors with h_{SiO2}/λ = 0.1 exhibit the highest sensitivity of 1170 cm^2/g compared to 340 cm^2/g of the sensor with h_{SiO2}/λ = 0.05.

3.3. Sensor Operation in Liquids

The frequency response of LW-SAW sensors with two normalized thicknesses h_{SiO2}/λ = 0.05 and 0.1 with 100 nm thick NCD layer was also tested with four liquids using an home-made microfluidics set-up. All liquids have a larger dielectric constant than the quartz substrate ($\varepsilon_r \approx 4.5$) and different viscosity (see Table 1). The insertion loss increases with liquids of high dielectric constant. The thin amorphous SiO_2 guiding layer does not reduce capacitive loading so the impedance mismatch of the devices increase the insertion loss and alter the frequency response [9]. The highest insertion loss was observed for PBS buffer, which was caused by presence of salts ions in comparison to distilled water. Elevation of baseline level was also observed with higher dielectric constant. Out of band rejection was slightly lower for diamond coated LW-SAW sensors in comparison to uncoated ones. Contribution to acoustic losses have also mechanical properties of liquid such as viscosity or density. It was also observed, that there is no frequency shift between frequency response of sensor integrated in microfluidic set-up without liquid and with liquid loading for uncoated as well as NCD coated LW-SAW sensor.

Table 1. Liquid properties at 20 °C.

Liquid	Dielectric Constant (-)	Viscosity (mPa·s)
Water	≈ 80.1	1.002
PBS buffer	≈ 79	-
MeOH	≈ 33.1	0.58
IPA	≈ 17.9	2.2

4. Conclusions

In this work, we investigated properties of diamond coated LW-SAW sensors. The center frequency as well as phase velocity of Love waves increase with thickness of deposited NCD layer as expected from theoretical simulations. Remarkably, NCD coated LW-SAW sensors shows sensitivity comparable to uncoated sensors. A sensitivity as high as 1170 cm^2/g has been determined for a diamond coated (100 nm thick) sensors and normalized amorphous silica thickness $h_{SiO2}/\lambda = 0.1$. These counter intuitive results are highly promising for potential bio-sensing applications. The sensors were also tested in various liquids. High insertion loses have observed and attributed to dielectric mismatch between the sensor and the liquid. As a result, the ST-cut quartz piezoelectric substrate is not an ideal substrate for LW-SAW sensors operating in water due to its low dielectric constant. Nonetheless, no frequency shift was observed due to liquid loading. Piezoelectric substrate with higher dielectric constant such as $LiTaO_3$ or $LiNbO_3$, or increasing of thickness of the silica guiding layer are possible solutions to resolve this issue.

Conflicts of Interest: The authors declare no conflict of interest.

References

1. Singh, A.; Glass, N.; Tolba, M.; Brovko, L.; Griffiths, M.; Evoy, S. Immobilization of bacteriophages on gold surfaces for the specific capture of pathogens. *Biosens. Bioelectron*. **2009**, *24*, 3645–3651, doi:10.1016/j.bios.2009.05.028.
2. Du, J.; Harding, G.; Collings, A.; Dencher, P. An experimental study of Love-wave acoustic sensors operating in liquids. *Sens. Actuators A Phys*. **1997**, *60*, 54–61, doi:10.1016/S0924-4247(96)01424-0.
3. Herrmann, F.; Weihnacht, M.; Büttgenbach, S. Properties of shear-horizontal surface acoustic waves in different layered quartz–SiO2 structures. *Ultrasonics* **1999**, *37*, 335–341, doi:10.1016/S0041-624X(99)00012-8.
4. Krueger, A.; Lang, D. Functionality is Key: Recent Progress in the Surface Modification of Nanodiamond. *Adv Funct Mater*. **2012**, *22*, 890–906, doi:10.1002/adfm.201102670.
5. Mochaling V.N.; Shenderova, O.; Ho, D.; Gogotsi, Y. The properties and applications of nanodiamonds. *Nat. Nanotechnol*. **2012**, *7*, 11–23. doi:10.1038/nnano.2011.209.
6. Vaijayanthimala, V.; Cheng, P.-Y.; Yeh, S.-H.; Liu, K.-K.; Hsiao, C.-H.; Chao, J.-I.; Chang, H.-C. The long-term stability and biocompatibility of fluorescent nanodiamond as an in vivo contrast agent. *Biomaterials* **2012**, *33*, 7794–7802, doi:10.1016/j.biomaterials.2012.06.084.
7. Azevedo, A.F.; Baldan, M.R.; Ferreira, N.G. Nanodiamond Films for Applications in Electrochemical Systems. *Int. J. Electrochem*. **2012**, *2012*, 1–16. doi:10.1155/2012/508453.
8. Talbi, A.; Soltani, A.; Rumeau, A.; Taylor, A.; Drbohlavová, L.; Klimša, L.; Kopeček, J.; Fekete, L.; Krečmarová, M.; Mortet, V. Simulations, fabrication, and characterization of diamond-coated Love wave-type surface acoustic wave sensors. *Phys. Status Solidi A.* **2015**, *212*, 2606–2610. doi:10.1002/pssa.201532188.
9. Turton, A.C.; Bhattacharyya, D.; Wood, D. High sensitivity Love-mode liquid density sensors. *Sens. Actuators A Phys*. **2005**, *123–124*, 267–273, doi:10.1016/j.sna.2005.02.010.