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Research Paper

Surface Acoustic Wave Interaction with a Mixture of Oxygen and Nitrogen

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Parameters of surface acoustic waves (SAW) are very sensible to change of physical conditions of a propagation medium. In the classical theory formulation, the waves are guided along the boundary of semi-infinity solid state and free space. A real situation is more complex and a medium commonly consists of two physical components: a solid substrate and a gaseous or liquid environment. In the case of stress-free substrate, the strongest impact on SAW properties have surface electrical and mechanical conditions determined by solids or liquids adhering to the boundary. This impact is utilised for constructing sensors for different gases and vapours e.g. (Jakubik *et al.*, 2007; Hejczyk *et al.*, 2011; Jasek *et al.*, 2012). The influence of gaseous environment on the SAW properties is usually very weak and ignored. However, in certain condition it can be significant enough to be applied to sensor construction. In general, it concerns Rayleigh wave devices where energy leakage phenomenon is perceptible, especially when the gas being detected considerably changes the density of environment. The paper presents the results of experiments with oxygen-nitrogen mixture. Their primary aim was focused on finding the dependence of resonant frequency and attenuation in SAW resonator on parameters and concentrations of the gas in the environment.

Keywords: surface acoustic wave sensors; oxygen detection; leaky Rayleigh waves.

1. Introduction

Rayleigh wave being a superposition of bulk transversal and longitudinal waves can propagate only along subsurface layers of solids. At the distance of few wavelengths from the surface both bulk waves can propagate separately. The surface amplitude of the Rayleigh wave that propagates from a point source of excitation vanishes theoretically with distance r from this point proportionally to $1/\sqrt{r}$ – much weaker than the both mentioned bulk waves. The energy vanishes as a result of radiation to the volume of the solid substrate. The waves carrying energy from surface into the depth of the substrate are called Rayleigh leaky waves. When the substrate abuts on a gas environment then small part of energy is also radiated into the gasous volume. The analysis of waves that propagates from a point source located at the solid-gas border shows that in an ideal case the source emits nine types of waves: two bulk in the solid, two head waves in solid and in gas, one longitudinal (compressional) in gas, Rayleigh wave in a solid subsurface, Scholte wave in gas and two waves leaking to the solid and to the gas (Zhu *et al.*, 2004). The waves are illustrated in Fig. 1.

The mechanism of energy leakage from a solid SAW substrate to a gaseous (liquid) environment was analysed in literature using three different approaches. In the first approach mechanic continuum equations were used. In this approach any change in boundary conditions leads to modification of a Rayleigh characteristic equation by introducing to its structure an additive complex term responsible for additional attenuation (Zhu *et al.*, 2004; Cheeke, 2002). The second approach uses quantum-mechanical methods that treat SAW as a flux of phonons splitting between a solid substrate and a gas (Borman *et al.*, 1987). The last approach uses probabilistic methods and utilises some models of gas atoms or particles distributions over solid

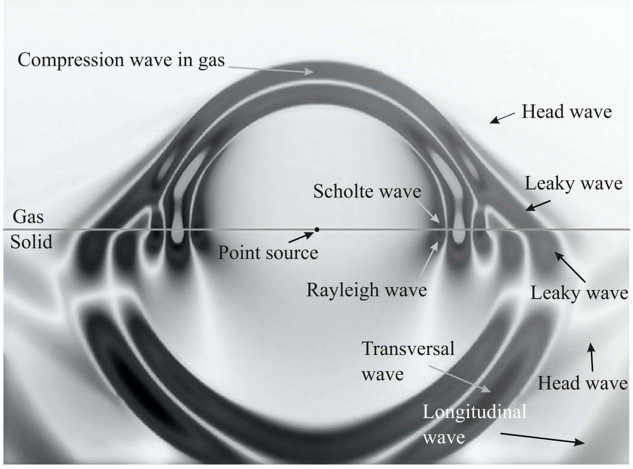

Fig. 1. Waves generated by point source located at solid – gas border.

surface and statistical mechanisms of kinetic energy transfer form a surface wave to a gas (Aleksandrov *et al.*, 1995). The gas properties in all mentioned approaches are described using Maxwell-Boltzmann ideal gas model.

It is interesting that all the theories mentioned above predict additional attenuation which is proportional to the square of SAW amplitude but postulate different frequency dependences. According to the classic continuum model the attenuation is proportional to f^2 (however some earlier approximations lead simply to f (Arzt *et al.*, 1967)), from the phonon approach to $f^{3/2}$, and the probabilistic methods predict just f. The experimental results described in (Slobodnik Jr., 1972) show that for SAW frequency values in the order of 1 GHz the dependence is linear. However, the experimental characteristic lies between the linear functions obtained from continuous theories (Terry, Strandberg, 1981; Aleksandrov, Seleznev, 1991).

The discrepancies should be explained but, regardless of the kind of the frequency and attenuation dependences between SAW and gas parameters, they are strong enough to be used to discriminate the components of a gas mixture.

2. Gas loaded SAW substrate

Classical approach to the Rayleigh wave analysis assumes free semi-space over the surface of a wave substrate and consequently vanishing normal and tangential stresses at the border.

Substitution of the free semi-space by a gas introduces to the boundary conditions some kind of continuity. In this case velocity boundary conditions are used. The velocities are considered in Eulerian mean field category. If the viscosity of the gas is neglected the boundary condition concerns only normal velocity component. Compression waves in the gas are generated when the phase-matched condition is satisfied:

$$\cos\theta = \frac{k_R}{k_G}, \qquad (1)$$

where k_R and k_G are wave vectors of Rayleigh wave and gas longitudinal wave respectively, θ is the angle between the vectors. The schematic view of the interface is shown in Fig. 2.

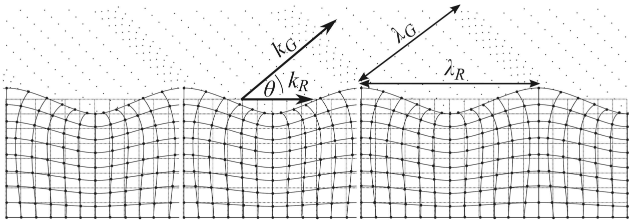

Fig. 2. Schematic view of the solid – gas interface. Indexes R and G indicate Rayleigh wave and gas compressive wave parameters.

The pressure waves in the gas are radiated at an acute angle to the Rayleigh wave. The angle depends on the wave numbers ratio. The frequencies of SAW and the radiated wave are equal.

The complete Rayleigh dispersion relation in the case of a fluid load was described in (Vannest, Büchler, 2011). It has the following form:

$$4\sqrt{1-\frac{v_R^2}{v_l^2}}\sqrt{1-\frac{v_R^2}{v_t^2}}-\left(2-\frac{v_R^2}{v_t^2}\right)^2 + i\frac{\rho_G v_R^4}{\rho_S v_t^4}\sqrt{\frac{1-\frac{v_R^2}{v_l^2}}{\frac{v_R^2}{v_G^2}-1}} = 0, \qquad (2)$$

where v_R, v_l, v_t are Rayleigh, longitudinal and transversal wave velocities in a solid, respectively, v_G is sound velocity in a gas, ρ_G, ρ_S are the gas and solid densities, respectively, $i = \sqrt{-1}$. It is worth to note that under loading the Rayleigh wave velocity is insignificant; lower than nominal. This equation has one real root and one complex root. The real root corresponds to a Scholte wave and the complex root corresponds to a modified Rayleigh wave. For ideal gas, one can express the density and velocity of sound in terms of pressure and temperature. Then the attenuation coefficient for Rayleigh waves can be written in the form

$$\alpha \approx 0.0869\frac{fp}{\rho_S v_R}\sqrt{\frac{\gamma M}{RT}} \quad [\mathrm{dB/cm}], \qquad (3)$$

where M is a molecular weight the gas, p is gas pressure, R is universal gas constant, T is temperature and γ is ratio of the specific heat at constant pressure to the specific heat at constant volume. The coefficient α

defines the energy loss resulting from the transmission of a bulk wave into the fluid over a distance x as being $\exp(-\alpha x)$.

The quantitative analysis of Eq. (2) shows that for liquids the attenuation coefficient may achieve values of the order of 10^3 dB/cm but for gases it is about three or even four orders of magnitude lower. However, the parameter is measurable and significantly different for different gases, so it allows their differentiation, especially in binary mixtures. In the case of oxygen and nitrogen, the attenuation calculated from Eq. (3) are equal 0.307 and 0.288 dB/cm, respectively.

3. Results of measurements

The measurement set consists of two mass-flow controllers (MFC) for precise gas mixing, molecular sieve for drying of a gas and a dedicated electronic system for a precise frequency characteristic analysis (SFCW). All parts of a gas delivery path as well as SAW gas measurement cell were thermally stabilised. Two-port SAW resonators were used as sensors. The SAW devices have 4 mm gap between interdigital transducers and are dedicated to sensor application (Soluch, 2008). Schematic view of the system is shown in Fig. 3.

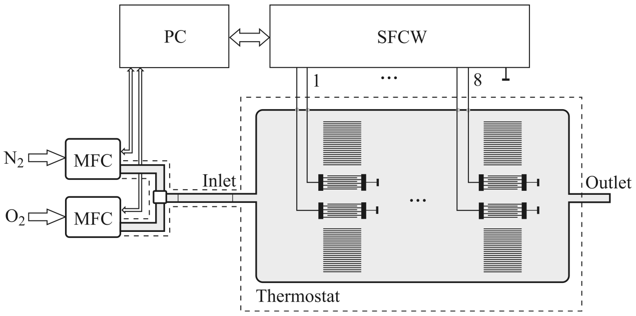

Fig. 3. Measurement set.

The measurement set allows for 8 simultaneous sensors measurement. It was described in more detail in (Jasek *et al.*, 2013). The SFCW system is a dedicated 8-chanell network analyser that measures SAW devices transmission characteristics in the frequency band encompassing the main resonance peaks of the resonators. The examples of the transmission characteristic for two devices having electrically open and shorted surface between transducers are shown in Fig. 4.

The short-circuited surface was obtained by deposition of 50 nm layer of Ni. The resonators have tree resonance peaks related to two ends and centre of planar reflectors reflection characteristics. The influence of oxygen concentration on the devices characteristics was observed around the peaks having maximal amplitudes and frequency shift values (dotted square in the Fig. 4). During the measurements the oxygen concentration was gradually increased and decreased in the range of 0 to 100%.

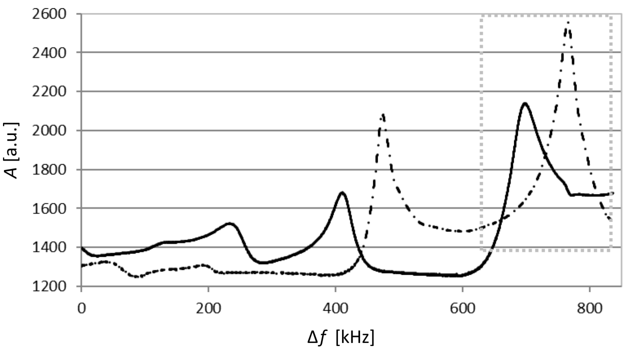

Fig. 4. Exemplary characteristics of sensors with open (dashed line) and shorted (solid line) surface between transducers. The frequency shift is related to 194 MHz. Amplitudes are given in bits of 12-bit ADC. Dotted rectangle indicates the region in which oxygen concentration influence is observed.

It was observed that resonant frequencies stayed nearly unaffected for all measured devices, both with short-circuited and open surfaces. Regarding amplitudes significant changes were noticed. The changes for two exemplary resonators are presented in Fig. 5.

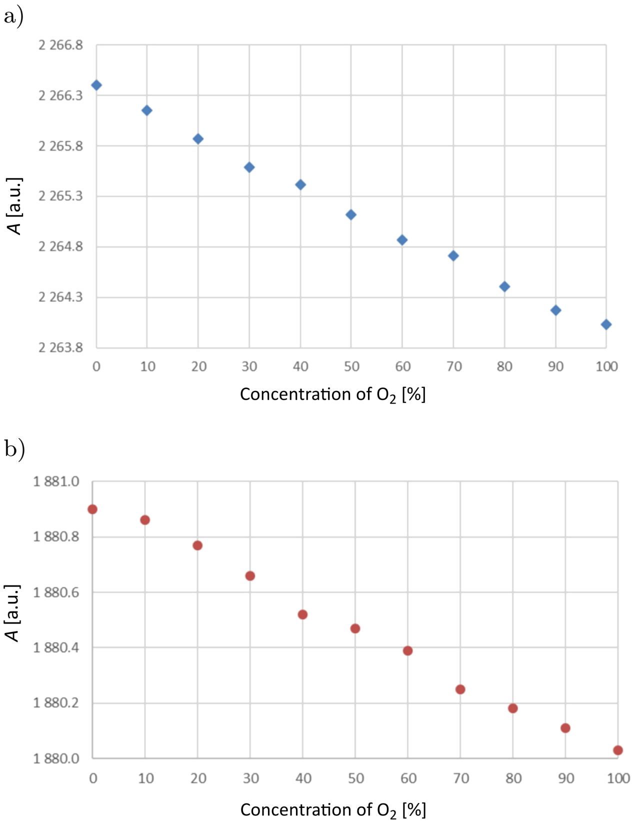

Fig. 5. Amplitude changes *vs* oxygen concentration for exemplary resonators with electrically open surface (a) and short-circuited surface (b).

The changes of amplitude are proportional to the oxygen concentration and are higher for the electrically open surface but are observable for all devices. The changes are repeatable and have no hysteresis.

4. Conclusions

The experimental measurements confirm that a change of oxygen concentration in nitrogen affects the SAW resonance amplitude in a linear way in the case of both open and short-circuited substrate. The changes were demonstrated using a binary mixture of gases having similar atomic weights, nitrogen and oxygen. In general, the higher atomic weight difference of mixture components the stronger the changes of the amplitude *vs* concentration. This phenomenon can be used for an observation of concentration changes in simple gas mixtures.

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