Microfluidic Acoustic Metamaterial SAW Based Sensor

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Abstract

Introduction. Microacoustic sensors based on surface acoustic wave (SAW) devices allow the sensor integration into a wafer based microfluidic analytical platforms such as lab-on-a-chip. Currently exist various approaches of application of SAW devices for liquid properties analysis. But this sensors probe only a thin interfacial liquid layer. The motivation to develop the new SAW-based sensor is to overcome this limitation. The new sensor introduced here uses acoustic measurements, including surface acoustic waves (SAW) and acoustic methamaterial sensor approaches. The new sensor can become the starting point of a new class of microsensor. It measures volumetric properties of liquid analytes in a cavity, not interfacial properties to some artificial sensor surface as the majority of classical chemical and biochemical sensors.

Objective. The purpose of the work is to find solutions to overcome SAW-based liquid sensors limitations and the developing of a new sensor that uses acoustic measurements and includes a SAW device and acoustic metamaterial.

Materials and methods. A theoretical analysis of sensor structure was carried out on the basis of numerical simulation using COMSOL Multiphysics software. Lithium niobate (LiNbO3) 127.86° Y-cut with wave propagation in the X direction was chosen as a substrate material. Microfluidic structure was designed as a set of rectangular shape channels. A method for measuring volumetric properties of liquids, based on SAW based fluid sensor concept, comprising the steps of: (a) providing sensor structure with the key elements: a SAW resonator, a high-Q set of liquid-filled cavities and intermediate layer with artificial elastic properties between them; (b) measuring of resonance frequency shift, associated with the resonance in liquid-filled cavity, in the response of weakly coupled resonators of SAW resonator loaded by periodic microfluidic structure; (c) determination of volumetric properties of the fluid on the basis of a certain relationship between the speed of sound in liquid, the resonant frequency of the set of liquid-filled cavities, and the geometry design of the cavity.

Results. The new sensor approach is introduced. The eigenmodes of the sensor structure with a liquid analyte are carried out. The characteristic of sensor structure is determined. The key elements of introduced microfluidic sensor are a SAW structure, an acoustic metamaterial with a periodic set of microfluidic channels. The SAW device acts as electromechanical transducer. It excites surface waves propagating in the X direction lengthwise the periodic structure and detects the acoustic load generated by the microfluidic structure resonator. The origin of the sensor signal is a small frequency change caused by small variations of acoustic properties of the analyte within the set of microfluidic channels.

Conclusion. The principle of the new microacoustic sensor, which can become the basis for creating a new class of microfluidic sensors, is shown.

Key words: acoustic metamaterials, surface acoustic wave, microfluidic sensor

Известия вузов России. Радиоэлектроника. 2019. Т. 22, № 4
Journal of the Russian Universities. Radioelectronics. 2019, vol. 22, no. 4

ОРИГИНАЛЬНАЯ СТАТЬЯ

Приборы и системы измерения на основе акустических, оптических и радиоволн
Measuring Systems and Instruments Based on Acoustic, Optical and Radio Waves

ORIGINAL ARTICLE

Introduction. Microacoustic sensors based on surface acoustic wave (SAW) devices allow the sensor integration into a wafer based microfluidic analytical platforms such as lab-on-a-chip. Currently exist various approaches of application of SAW devices for liquid properties analysis [1–5]. The basic concept of a SAW sensor utilizes an interrogation of external measuring substance with propagating along the waveguide acoustic wave. Initially developed Rayleigh wave SAW sensors were applied for the organic gas detection utilizing polymer sensitive layers [6]. However, an application of the same approach for liquid sensors was associated with a considerable wave attenuation caused by irradiation of normal component of Rayleigh wave into the liquid volume. Further development of SAW devices discovered a various modes of surface acoustic waves such as horizontally polarized shear waves that were applied for non-polar liquids measurements. Unfortunately, the wave attenuation was still too high because of a significant mismatch of dielectric properties of substrate (commonly used quartz substrates) and liquid. As a result, the wave confinement at the waveguide surface was deteriorated. The current problem was later solved by utilization of substrate materials with dielectric constant that is considerably higher than quartz [7, 8]. The application of lithium niobate and lithium tantalite substrates of different cuts allowed the sensor to operate with high permittivity liquids even in cases where it is directly applied to the waveguide surface. Thus, the SAW sensor based on $36°$ YX cut of $LiTaO_3$ wafer was utilized as a biosensor to detect amounts of an enzyme immobilized on a surface during the catalytic reaction [9]. Later, another type of shear horizontal SAW liquid sensor was developed with the use of Love surface wave. The Love wave is a surface acoustic wave that is localized within the overlayer deposited atop of the SAW waveguide. This type of wave appears in a layered structure and localizes in the overlayer because of its low acoustic wave velocity in comparison to the waveguide. Such acoustic wave localization allows decreasing the surface wave attenuation caused by scattering into the bulk of the substrate. The adjustment of the waveguiding layer thickness may create an acoustic film resonance that significantly improves the sensor sensitivity to mass loading [10].

The SAW-based sensor concepts were broadly utilized for biosensor applications. In most cases, the sensor structure is formed on a solid substrate basis with a tailored wave propagation path (surface modification with recognition layers), which allow for the realization of liquid sensors that are specifically sensitive to certain targeting substances. The detecting variation of mass load in this case is extended by altering the functional layer properties caused by the adsorption in a sensitive layer or binding to the recognition layer. In contrast to near-surface detection mechanisms, the velocimetry-based sensor approaches detect the variation of the speed of sound of the liquid analyte. Excitation of liquid pressure resonances and control of the resonant response of liquid-containing volumes is one of the most convenient ways to measure the analyte velocity of sound. The application of cavity-based approaches on a basis of SAW sensor platform is rather challenging because of the unavoidable scattering of acoustic waves into the substrate volume. Therefore, the velocimetry-based analysis of liquids on a SAW sensor platform is rather challenging approach.

Another approach is sensors based on phononic crystals. Composite periodic structures, also called phononic crystals, allow to develop the composite arrangements with artificial acoustic properties that are defined not only by the material properties of the structure constituents, but by the design (geometry, symmetry, periodicity) [11, 12]. A propagation through such structures of elastic waves is featured by the wavelength regions, within which sound cannot propagate through the structure (bandgap); therefore, almost complete reflection or scattering of incident acoustic waves occurs. For the frequencies corresponding to a bandgap region, the periodic structure can be described in terms of high acoustic impedance for an incident acoustic wave. Thus, one of the most advantageous features of phononic structures is an ability to be applied in those cases, where rather high acoustic impedance boundaries are required and application of standard materials (such as tungsten) is limited.

Since among other parameters the acoustic properties of the composite arrangement depend on material properties of structure constituents, their variation causes a change in a structure transmission behavior. That feature allows to apply solid-liquid periodic composite arrangements for liquid sensor purposes that were already demonstrated in several previous works [13–15]. A control of the frequency

ОРИГИНАЛЬНАЯ СТАТЬЯ

Известия вузов России. Радиоэлектроника. 2019. Т. 22, № 4
Journal of the Russian Universities. Radioelectronics. 2019, vol. 22, no. 4

ORIGINAL ARTICLE

Приборы и системы измерения на основе акустических, оптических и радиоволн
Measuring Systems and Instruments Based on Acoustic, Optical and Radio Waves

position of isolated narrow transmission bands is more beneficial rather than deviation of bandgap edges, as it was demonstrated earlier. For that reason, the idea of phononic crystal based liquid sensors was focused on the obtainment of the structure isolated transmission peaks (dips) that correspond to material properties of the liquid constituent. In contrast to well developed microacoustic liquid sensors, the phononic crystal liquid sensor approach enables the detection of the velocity of sound of liquid analyte. Similarly to ultrasonic velocimetry sensor approach, the phononic crystal based sensors allow to evaluate thermodynamic quantities of the liquid analyte analyzing the speed of sound at a certain range of pressures. The reaction on molecule interactions is reflected as a change in a liquid compressibility that can be detected by probing the analyte with ultrasonic velocimetry methods. Proposed approach allows to keep the advantages of velocimetry based methods and at the same time to apply the measurement principle based on a control of structure resonances similar to microacoustic sensor devices [16].

Several of previous works have already demonstrated phononic crystal based sensor designs and confirmed the concept showing a direct correlation of the periodic structure response to volumetric material properties of the analysed liquid (more precisely, speed of sound) [17, 18]. Depending on the application field, certain advantages of the phononic crystal based sensor approach can be underlined. In [19, 20] it was shown that application of periodic arrangements for the detection of properties of hydrocarbon blends is advantageous in several aspects. It was shown that the speed of sound properties of analytes vary in a distinct manner depending on the composition, and the deviation between different blends is much sufficient for the detection.

Materials and Methods. A new SAW based sensor which idea is introduced here detects a change in resonance frequencies of the system of piezoelectric transducer loaded by periodic structure of microfluidic channels filled by liquid. The resonance frequencies of the SAW with microfluidic system strongly depend on acoustic properties of liquid. Detecting of its changes allow to obtain qualitative and quantitative information of the composition of liquid and thermodynamic properties including molecular interactions within the (free) liquid.

The key elements of the new microfluidic SAW based sensor are a SAW resonator, an acoustic metamaterial and a high-Q set of liquid-filled channels. The SAW resonator acts as electromechanical transducer. It excites acoustic waves propagating towards a periodic microfluidic structure and detects the acoustic load generated by liquid resonator.

Application of resonator based SAW platform enables the efficient readout of microfluidic structure sensing modes at the resonance conditions of the SAW resonator that leads to significant improve of the sensor response. The application of that approach is associated with challenges related to the appearance of couple resonances and as a result broadening of spectral properties and loss of sensitivity. In order to decouple the resonances of the SAW structure and the microfluidic structure modes, we propose to introduce the intermediate layer with artificial elastic properties that allows to tune the coupling between the resonating part of the arrangement in a predefined manner. The result of the structure coupling is the achievement of structure readout in a form of narrow-band resonance that is sensitive to the variation of material properties of the periodic structure liquid constitute.

Lithium niobate (LiNbO$_3$) 127.86° Y-cut with wave propagation in the X direction was chosen as a substrate material. It has an efficient coupling to Rayleigh wave and is a typical material for broadband SAW devices. Electrodes with an equal aperture of 100 of SAW wavelengths (interdigitated transducers IDTs), an equal electrodes width, and a period along the wave propagation were used in the current sensor design. Microfluidic structure with a set of rectangular shape channels are made of silicon.

Results and Discussion. The modelling of designed structure was based on solving a system of equations that includes an equation for the propagation of acoustic waves in anisotropic media

$$\rho \frac{\partial^2 u_i}{\partial t^2} = \sum_j \sum_k \left[e_{kij} \frac{\partial^2 \varphi}{\partial x_j \partial x_k} + \sum_l c_{ijkl}^E \frac{\partial^2 u_k}{\partial x_j \partial x_l} \right],$$

where ρ is the density; u_i are the components of the displacement field; e_{kij} is the piezoelectric tensor; φ is electric potential; c_{ijkl}^E is the elastic modulus tensor determined with a constant electric field; i, j, k

Известия вузов России. Радиоэлектроника. 2019. Т. 22, № 4
Journal of the Russian Universities. Radioelectronics. 2019, vol. 22, no. 4

ОРИГИНАЛЬНАЯ СТАТЬЯ

Приборы и системы измерения на основе акустических, оптических и радиоволн
Measuring Systems and Instruments Based on Acoustic, Optical and Radio Waves

ORIGINAL ARTICLE

and l are indices running from 1 to 3; and the Helmholtz equation for a pressure wave in a liquid

$$\nabla\left(-\frac{1}{\rho}\nabla p\right) - \frac{\omega^2 p}{\rho v^2} = 0,$$

where ω is the circular frequency; v is the speed of sound in a liquid; p is pressure.

Conditions at the boundaries of the "solid-liquid" section are as follows:

$$\mathbf{F} = -\mathbf{n}_s p; \quad \left(\mathbf{n}_f \cdot \mathbf{u}\right)\omega^2 = -\mathbf{n}_f\left(-\frac{1}{\rho}\nabla p + \mathbf{q}\right),$$

where $\mathbf{F}$ is the force per unit area representing the load on the cylinder walls; $\mathbf{n}_s$ is the normal vector directed from a solid; $\mathbf{n}_f$ is the normal vector directed from the fluid volume; $\mathbf{u}$ is the mechanical displacement vector in a solid; $\mathbf{q}$ is the acceleration vector reported by the fluid.

The simulations were completed with an acoustic module of COMSOL$^{\mathrm{TM}}$ Multiphysics software (Burlington, MA, USA).

The structure geometry was divided into separate domains of three different types, each of which is described with a separate system of equations. The "piezoelectric material" domains are described as anisotropic piezoelectric materials. The 127.86° Y cut is defined with a rotational coordinate system that recalculates the respective material properties in accordance with Euler angles prescribed in the rotation coordinate system of the model. The computational domains of the microfluidic structures and IDTs are described as isotropic materials that are mechanically coupled to the piezoelectric waveguide. These domains are described as "linear elastic material" domains. The liquid that fills the microfluidic channels is described by the pressure acoustic model. It specifies the propagation of pressure waves in the liquid domains and contains the liquid material properties that are targeted for sensing (such as speed of sound and density). Model boundaries in the Z planes are prescribed with periodic boundary conditions that make the whole arrangement infinitely long in the Z direction. This boundary condition allows for a significant reduction of the computational model to complete the simulation tasks within a meaningful time duration. The surface acoustic wave excitation is completed with prescribed periodic potential and ground boundaries along the X direc-
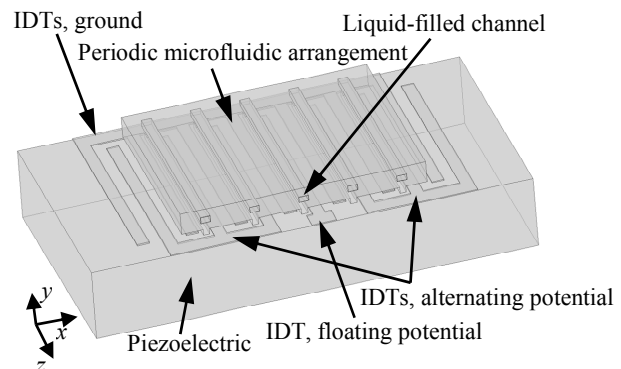

Fig. 1. Microfluidic SAW based sensor structure

tion of the waveguide surface. Receiving IDTs-waveguide boundaries are prescribed with "ground" and "float potential" conditions. The receiving electrodes are in the middle of the structure.

Fig. 1 depicts a general scheme of microfluidic SAW based sensor. These are the principle structure of sensor, connected to the measuring circuit. The key elements of microfluidic SAW based sensor are a SAW resonator (piezoelectric plate with electrodes), intermediate layer with artificial elastic properties and a set of liquid-filled cavities. The most important in this sensor design is the system itself, consisting of the said three structural components. The design is determined by wide possibilities for optimizing characteristics, in particular for the fluids under study. The sensor device is supplemented with measuring means for measuring at least two electrical responses selected from the group consisting of resonant frequencies, damping, admittance (or impedance).

The SAW device acts as an electromechanical transducer. It excites acoustic waves propagating towards the periodic microfluidic structure and detects the acoustic load generated by the intermediate layer and the set of liquid cavities.

The important element of fluidic SAW based sensor is an intermediate layer with artificial elastic properties that separates the piezoelectric resonator and liquid-filled cavities. The main purpose of inputting of this layer is creation of conditions for weak mechanical coupling of two resonators. By changing the reflection coefficient of the intermediate layer, it is possible to control the degree of mechanical coupling between the resonators and adjust the sensitivity and resolution of the sensor. Thus, the three requirements hold for the intermediate layer. First, it must effectively control the propagation of acoustic waves between the piezoelectric transducer and mi-
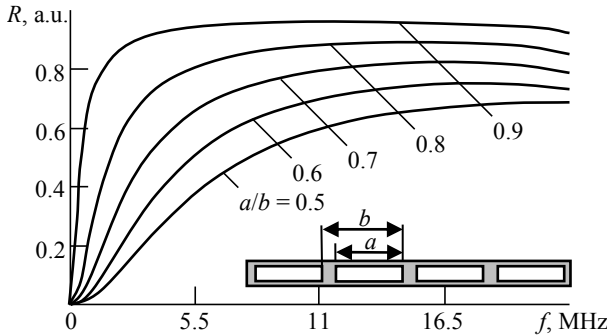
ОРИГИНАЛЬНАЯ СТАТЬЯ

ORIGINAL ARTICLE

Известия вузов России. Радиоэлектроника. 2019. Т. 22, № 4
Journal of the Russian Universities. Radioelectronics. 2019, vol. 22, no. 4

Приборы и системы измерения на основе акустических, оптических и радиоволн
Measuring Systems and Instruments Based on Acoustic, Optical and Radio Waves

Fig. 2. Frequency dependence of the reflection coefficient (*R*) of an acoustic metamaterial in the form of a periodical system of rectangular cavities in a solid-state plate

Fig. 4. Admittance spectrum of the sensor device for two different sound velocities of the liquid

crofluidic resonator. Second, design and materials must define a passive control. Third, it must not introduce additional acoustic energy losses. Acoustic artificial materials are suitable for these tasks. In fact, this sensor device does need neither a full bandgap nor a complete suppression of the respective waves. We need a reflection coefficient (*R*) in the range of 80–95 % for the shear waves in the operating frequency range of the sensor. Our studies prove that one layer of periodically spaced rectangular cavities is sufficient. Fig. 2 shows the frequency dependence of the reflection coefficient of a periodic arrangement of those cavities in silicon for shear waves. Width and position on the frequency scale can be set by the length of the cavity, *a*, and the lattice period of the structure, *b*. The larger the filling factor a/b, the higher the reflection coefficient. As can be seen from Fig. 2, the values of the filling factor from 0.7 to 0.9 are suitable for our task in the frequency range from 14 to 21 MHz, which covers the sensor's operating range.

Fig. 3 shows possible variations of the intermediate layer structure: an arrangement of rods or voids of rectangular, cylindrical or triangular shape; a composite or a porous material; a multilayer struc-
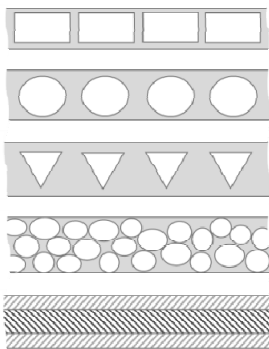
ture. Despite the various possible designs and materials, the most significant and common for the invention is its functional purpose of weak mechanical coupling.

Fig. 4 shows a frequency dependence of admittance (its module, |Y|) of the microfluidic acoustic metamaterial SAW based sensor, shown in Fig. 1. The two curves show the response of microfluidic SAW based sensor for different values of speed of sound in liquid. The figure demonstrates the joint work of the two weakly coupled resonators. The peak A frequency depends on speed of sound in liquid. The peak B corresponds to the resonance of the piezoelectric transducer. Its frequency does not depend on the properties of the liquid (Fig. 5).

Fig. 5 shows a dependence of characteristic frequencies of the system of weakly coupled resonators of SAW resonator loaded by solid-fluid structure on the speed of sound in liquid. Curves A and B in Fig. 5 correspond to peaks A and B in Fig. 4, respectively. Fig. 6 shows the vibrational modes. Colors represent displacement (in solid) and pressure (in liquid) fields with white node lines. Fig. 6, *a* shows the situation at the first resonance. The complete overlayer vibrates
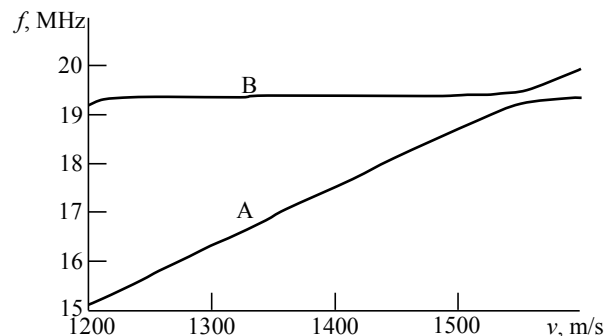

Fig. 3. Possible variations of the intermediate layer structure

Fig. 5. Dependence of Eigenfrequencies on the speed of sound for microfluidic structure (A) and SAW (B)

Известия вузов России. Радиоэлектроника. 2019. Т. 22, № 4
Journal of the Russian Universities. Radioelectronics. 2019, vol. 22, no. 4

ОРИГИНАЛЬНАЯ СТАТЬЯ

Приборы и системы измерения на основе акустических, оптических и радиоволн
Measuring Systems and Instruments Based on Acoustic, Optical and Radio Waves
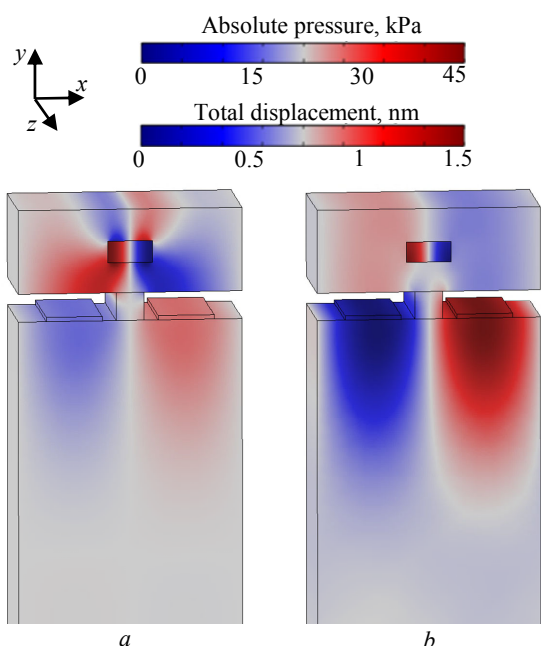
ORIGINAL ARTICLE

Fig. 6. Displacement and pressure fields distribution for microfluidic structure resonance (*a*) and SAW resonance (*b*)

now in a way that resembles film resonance. One however should note that the displacement profile differs significantly from that in the homogeneous counterpart. Fig. 6, *b* illustrates the second resonance. Obviously, the displacement diminishes already half the way through the intermediate layer. The phase shift at the SAW-structure interface is small. This finding has an important engineering impact since the geometrically extraordinary thick overlayers become acoustically similar to layers of common thickness in some SAW applications.

In accordance with curve A on Fig. 5, we see a linear dependence of the frequency on the speed of sound. The corresponding equation for the shift of the resonance frequency $\left(\Delta f\right)$ from the change in the speed of sound in liquid $\left(\Delta v\right)$ is:

$$\Delta f = k_1 k_2 \Delta v,$$

where k_1 is a constant depending on the material properties and the geometry of cavity; k_2 is a constant depending on the elastic properties of intermediate layer.

Applying acoustic metamaterial concepts in the construction of an intermediate layer and a liquid-filled cavity, we can increase the values of k_1 and k_2, increasing and the sensitivity and resolution of the SAW based sensor.

Conclusion. The principle of the new microacoustic sensor, which can become the basis for creating a new class of microfluidic sensors, is shown.

The principle of measuring volumetric properties of liquids, based on SAW based fluid sensor concept, comprising the steps of:

(a) providing sensor structure with the key elements: a SAW resonator, a high-Q set of liquid-filled cavities and intermediate layer with artificial elastic properties between them;

(b) measuring of resonance frequency shift, associated with the resonance in liquid-filled cavity, in the response of weakly coupled resonators of SAW resonator loaded by periodic microfluidic structure;

(c) determination of volumetric properties of the fluid on the basis of a certain relationship between the speed of sound in liquid, the resonant frequency of the set of liquid-filled cavities, and the geometry design of the cavity.

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ОРИГИНАЛЬНАЯ СТАТЬЯ

ORIGINAL ARTICLE

Известия вузов России. Радиоэлектроника. 2019. Т. 22, № 4
Journal of the Russian Universities. Radioelectronics. 2019, vol. 22, no. 4

Приборы и системы измерения на основе акустических, оптических и радиоволн
Measuring Systems and Instruments Based on Acoustic, Optical and Radio Waves

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