DIELECTRIC RESONATORS WITH "WHISPERING-GALLERY" WAVES IN INVESTIGATIONS OF SMALL-VOLUME BINARY SOLUTIONS

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A new method of investigation of the parameters of aqueous binary solutions is proposed. It is applicable for small amounts $((4\div8)\times10^{-3}$ ml) of a solution. The method uses quasioptical dielectric resonators, in which "whispering-gallery" modes are excited. The frequency shift and Q-factor of the HE mode of a resonator depend on the complex dielectric permittivity of the sample under study.

1. Introduction

On the study of various substances, including complicated biological structures, microwave methods are widely used [1, 2]. Mostly used as the objects under investigation are biological tissues, biopolymers, and others. The aim of such studies is to get the information about intermolecular interactions and to determine the parameters and characteristics related to them [3].

The majority of biomaterials is liquids (blood, physiological solutions, etc.) characterized by high values of the dielectric permittivity and great losses in the microwave range, including the millimeter one. Moreover, their properties are very sensitive to changes in temperature [3]. Therefore, it is necessary to develop the reliable methods of measurement that would allow one to identify the properties of the studied objects. They include the methods aimed at the measurement of the complex dielectric permittivity $\varepsilon^* = \varepsilon' - i\varepsilon'' = \varepsilon'(1 - i\tan\delta)$ of liquids with great losses ($\tan\delta \approx 1$), where ε' and ε'' are the real and imaginary parts [2]. In this case, the question of the study of liquids in small volumes, especially biological liquids, gains importance.

The existence of a great number of the methods of measurement of ε and tanδ in the microwave range is defined by the availability of various transmission lines, the possibility to choose different parameters convenient for the measurement, and the possibility to use the samples of different forms and to choose their position in the system [2, 4—10]. We can divide these methods into three groups:

— methods, in which guided waves are used;

— methods, in which waves in the free space are used;

— resonance methods.

Most widespread are the resonance methods which differ from one another by the resonance system type and the type of excited modes in it. The popularity of the resonance methods is explained by their higher sensibility which is ensured by the "many-run" character of the interaction of a microwave field with the substance under study during resonance measurements. The possibility to measure samples of sufficiently small volumes is also of importance [6—8, 10]. However, the application of such structures leads to some difficulties [3, 7] during the measurement of dielectric materials made of substances with great losses, which is related, first of all, to a reduction of the Q-factor of resonators.

Recently, the great interest has been paid to open dielectric resonators [11] and, in particular, to quasioptical dielectric resonators (QDRs) with waves of the whispering-gallery type [12—14]. The results of studies of such resonators, which were obtained up to now, indicate the possibility of their application in the

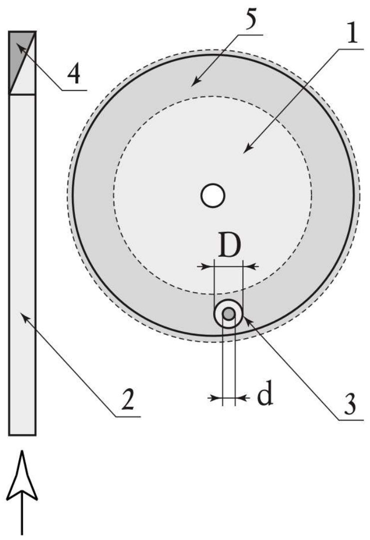


Fig. 1. Structural scheme of the measuring cell: *1* — disk-shaped dielectric resonator; *2* — dielectric waveguides with matched load; *3* — capillary; *4* — matched load; *5* — resonator field caustic (its boundaries are marked by a dashed line)

investigation of liquids with any values of losses. The authors of works [14, 15] showed that, while using a disk-shaped QDR with a capillary positioned in the field caustic region, pure liquids with great losses could be examined.

Many solutions under study are two-component (binary) water compounds, whose radiophysical parameters (dielectric permittivity and absorption coefficient) depend on the concentration of a solution and can be used as a concentration index [2, 3]. The strong dependence of the dielectric permittivity ε on the concentration of such a solution is caused by a high value of ε of water, as compared with the permittivity of the substances dissolved in it, and by the dependence of ε of a solution in the millimeter range of wavelengths on the structure of a solution and intermolecular bonds [1].

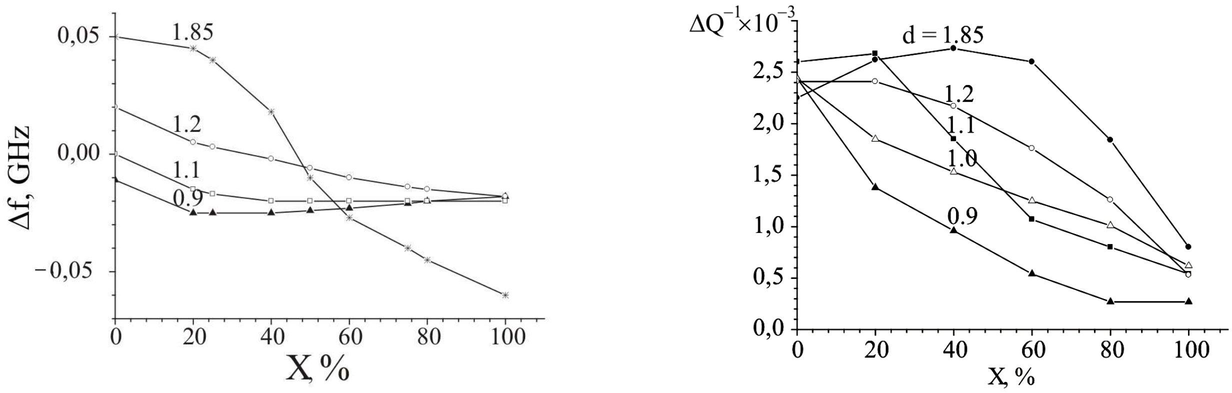
Taking into account all the above-said and the results obtained in the investigations of pure liquids, including liquids with great losses, with the use of disk-shaped QDRs [14, 15], we investigated a resonator with a two-component solution on the basis of water as a strongly absorbing liquid with the purpose to clarify the possibility to use a resonator of this type to determine the dielectric parameters of solutions (ε and $\tan\delta$) in a wide range of the quantities under study. Ethyl alcohol was chosen as the second component of a solution.

2. Experimental Technique

The investigation of the concentration dependence was conducted in a frequency range of 37—40 GHz with the use of an oscillator with the subsequent computer analysis of the results obtained. The structural scheme of a measuring cell is presented in Fig. 1. A disk-shaped QDR of the radius R=39.0 mm and the height h=7.2 mm is made of teflon (ε=2.04). Through the region of distributed coupling in resonator (1), the dielectric waveguides (2) made of the same material as the resonator excite a traveling wave of the HE_{mns}-type at a frequency of 38.34 GHz, where m, n, and s are the azimuth, radial, and axial indices, respectively ($m = 35\div38$, n=1, and $s \sim 0$), in which the E component of the field was mainly directed along the capillary axis (the z axis). Small levels of the coupling between a waveguide and a resonator ($\beta < 0.1$) were obtained at distances between them, being close to λ. The difference of the frequencies between neighbouring oscillations, whose Q-factor was about 1600—1800, in the measurement range was equal to 0.89 GHz. One of the ends of a dielectric waveguide was matched with a metallic waveguide, and the matched load *4* was positioned at the second end (the matching coefficient k_m=1.17). In the measurement, we used the transmission scheme. The solution under study filled capillary *3* that was located in a hole with diameter $D< \lambda$ (λ is a wavelength in the resonator) parallel to the resonator axis in the maximum of the traveling wave field limited by the resonator field caustic *5*. During the experiment, the capillary diameter ($d< D$) was changed within the limits 0—1.85 mm. Moreover, we registered a shift of the resonance frequencies $\Delta f = f_{\rm liq} - f_{\rm air}$ and the quantity $\Delta Q^{-1} = Q_{\rm liq}^{-1} - Q_{\rm air}^{-1}$ of the resonator ($Q = f_0/\Delta F$, where f_0 and ΔF are, respectively, the resonance frequency and the width of the resonance line of oscillations in the resonator) on the filling of the capillary with air and a solution, respectively. The solution concentration was expressed in volume percents X of the alcohol content in water. The measurement errors of the frequency and, respectively, the Q-factor depended on the metrological characteristics of the setup and did not exceed 10%. The measurement was realized at a temperature of the environment and the water-alcohol solution of 20 ($\pm$0.1) °C.

3. Results of Experimental Studies and Their Analysis

The dielectric properties of liquids are sensitive to their molecular structure and thus can be used to verify the



a **b**

Fig. 2. Frequency shift $\Delta f(a)$ and inverse quality-factor ΔQ^{-1} (b) in the resonator with a capillary filled by a water-alcohol solution versus the concentration X (%) of ethyl alcohol at various values of the capillary diameter d (in mm)

models of the structure and to get information about the processes of molecular reorientation. The dielectric properties of water were and are rather widely examined up to now [16, 17], as well as the properties of alcohol. However, alcohol and water are the liquids with hydrogen bonds, and their structure is not studied sufficiently well. The measurement of the dielectric permittivity of this solution can be a good indicator of the alcohol concentration. For example, an increase of the alcohol concentration in water leads to a reduction of the dielectric permittivity of the solution, which is caused, first of all, by the reduction of the number of water molecules in the unit volume and, secondly, by the union of some part of water molecules with dissolved molecules of the solute. Therefore, a rather simple and convenient method of measurement of the properties of the substances characterized by great losses and their solutions could consist in the measurement of a frequency mainly determined by ε' and of losses at the resonance frequency which is mainly defined by ε''. Such an approach is not strict, because the resonance frequency and the intrinsic quality of a resonator depend in general case on both parts of a complex permittivity, i.e. on ε' and ε''. Therefore, it is important to determine the conditions, under which the indicated influence of the solution permittivity on the resonator properties remains analogous to that of a solution with small losses.

The results of measurements of the frequency shift Δf and the quantity ΔQ^{-1} as functions of the volume concentration X of a water-alcohol solution are presented in Fig. 2(a, b). As seen, an increase in the alcohol concentration leads to both a shift of the resonance frequency and a change of the quantity ΔQ^{-1} which are characterized by different steepnesses of their concentration dependence on different sections. Moreover, we observe the specific features at the points $X \sim 20\%$ and $X \sim 40 \div 50\%$ characteristic of the given solution. In the first case, tanδ reaches the maximal value. In the second case, we have $\varepsilon'=\varepsilon''$. What is more, the behavior of the concentration dependence varies depending on the capillary diameter (the volume of the solution under study), which can be defined by the depth of penetration of a field into the liquid, as was shown in work [15]. For example, the concentration dependence is close to a linear one at a certain value d (d=1.2 mm for Δf, and d=1.0 mm for ΔQ^{-1}), though the steepness of curves is somewhat greater in the region of concentrations up to 20%. That is why, we may suppose that there exist such values of the capillary diameter d, at which the dependences of Δf and ΔQ^{-1} on X have linear character. In the authors' opinion, this circumstance is important for the determination of the parameters of a resonator meant for the monitoring of the solution concentration by its dielectric properties. The change of the sign of the frequency shift Δf correlates with the dependence given in work [14] for pure liquids.

Using the formulas describing the dependence of ε', ε'', and tanδ on the solution concentration and their frequency dependence in the measurement range given in works [18,19], we carried out the calculations of their concentration dependence for the frequency, on which a measuring resonator was excited (see the insert in Fig. 3,a), and performed the comparative analysis of the concentration dependence for a water-alcohol solution with the obtained experimental results for Δf (Fig. 3,a) and ΔQ^{-1} (Fig. 3,b).

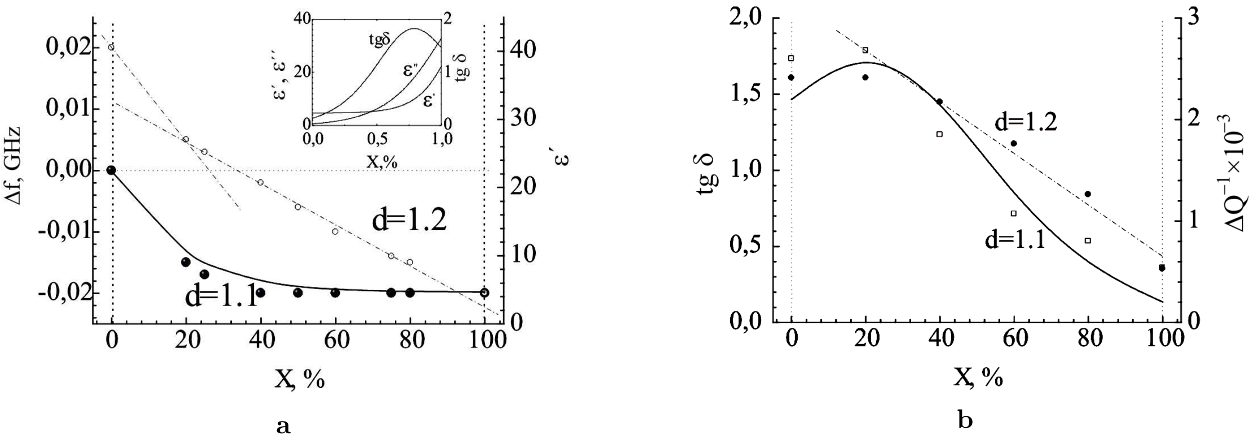


Fig. 3. Frequency shift Δf (a) and inverse quality-factor ΔQ^{-1} (b) in the resonator with a capillary filled with a water-alcohol solution versus the concentration X (%) of ethyl alcohol (points are experimental data; a continuous line is calculated for ε', according to the data in [19]. The insert gives the concentration dependences of the dielectric parameters of a water-alcohol solution calculated on the basis of the data in [18, 19]

As was mentioned above, the dependence of tanδ on X has a characteristic point in the region of concentrations close to 20%. The behaviors of the curves in the regions $X<20\%$ and $X>20\%$ are different, which is related to the domination of different mechanisms of the relaxation [18—20].

The comparison and comparative analysis showed that the concentration dependences of Δf and ΔQ^{-1} for a capillary with the diameter d=1.1 mm are the closest to the calculated ones. This fact can be used in the determination of the dielectric parameters of a given solution with regard for the calculated data. The degree of the agreement of the experimental and calculated dependences will be influenced by the measurement accuracy of the frequency (Q-factor) of the resonance, the solution concentration, and the temperature nonstability of the resonator cell filled with a liquid, which affects both the resonator parameters and the properties of the solution under study. In order to determine the characteristics of solutions on the use of capillaries with different values of d, it is convenient to use the method of comparison, by calibrating the measuring system by standard solutions. In this case, we can use such parameters as the resonance frequency and losses defined by values of ε' and ε'', respectively.

Thus, on the basis of the results which are obtained during the investigation of a water-alcohol solution concerning the concentration dependence of the frequency shift and the reciprocal Q-factor of a resonator, we can draw the following conclusion. By using the capillary diameter d≈1.1 mm for the given QDR and the calculated dependence, we can determine the dielectric parameters, whose accuracy will depend mainly on the frequency measurement accuracy. Moreover, we can also use the calibrating curves of water solutions with the known dielectric characteristics.

4. Conclusions

The results presented in this work testify that the use of disk-shaped QDRs can be successfully applied to the investigation of both the dielectric properties of pure liquids with an arbitrary value of dielectric losses (tanδ) [14] and the concentration dependence of the properties of two-component solutions on the basis of such strongly absorbing liquid as water. It is worth noting that the obtained results well correlate with the results of investigations of the molecular structure of water-alcohol solutions [18—20] and with the specific features of dielectric characteristics observed in the present work.

The determination of the concentration dependence of the dielectric parameters can be performed with help of the method of comparison of experimental data with calculated ones, those obtained on the basis of the known formulas or with the use of calibrating curves of the known solutions. The construction of the given resonator is such that it allows one to measure the parameters of a solution both in a stationary mode and the pumping mode.

The measurement of water-based strongly absorbing substances by the resonance method with help of a disk-shaped QDR allows one to enhance the sensitivity and efficiency of the investigation of objects in the microwave range, which is important for biology, medicine, and other branches of science. The possibilities of measuring the properties of solutions in very small volumes ($(4\div8) \times 10^{-3}$ ml) with the use of a QDR and the continuous monitoring of their dielectric parameters as functions of the concentration are of practical interest for solutions characterized by the strong absorption of a microwave energy.

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ДІЕЛЕКТРИЧНІ РЕЗОНАТОРИ З ХВИЛЯМИ "ШЕПОЧУЧОЇ ГАЛЕРЕЇ" В ДОСЛІДЖЕННЯХ БІНАРНИХ РОЗЧИНІВ МАЛИХ ОБ'ЄМІВ

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Р е з ю м е

Запропоновано новий підхід для дослідження параметрів бінарних розчинів на основі води в малих об'ємах $((4\div8) \times 10^{-3}$мл). В основі підходу лежить застосування дискового квазіоптичного діелектричного резонатора, в якому збуджуються хвилі "шепочучої галереї". Зсув частоти та добротність *HE*-моди резонатора залежать від комплексної діелектричної проникності досліджуваної речовини.