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Structural Evolution of a Multicomponent Liquid: Modelling and Analyse

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1. Statement of the Problem

At the present stage of creating new generation materials and devices, the task is not only to control their properties but also to use them effectively in various fields, including medicine. The purpose of the article is to develop and use in practice, for example, forensic examination, an effective non-destructive method that allows analyzing the structure of liquids, in particular, blood serum. This goal was realized by solving interrelated tasks, namely: to improve the experimental technique based on the quartz weighing method described in [1-3]; to identify the structure of a blood drop, to find out its topology; to analyze the physicochemical properties and morphology of the substance.

2. Evolution of a Multicomponent Liquid

The analysis of the structural evolution of heteroatomic liquids (in particular, blood) from the standpoint of theoretical and experimental approaches allows us to open new horizons in the practical plane, to determine the phenomenology of the process based on modern research methods [1-3]. The structural kinetics of a liquid, such as blood serum or blood itself, is a complex multistage process. The latter can be divided into two stages: events occurring during the evaporation of free water, and the actual structure formation. The latter is the stage of structure formation associated with water evaporation. Experiments show that a liquid that dries on a solid wettable substrate (under room conditions) takes on a specific appearance. For an example, see (Figure 1).

Similar results were obtained in other laboratories [4,5] and by us. The reason for this is a complex of complex physicochemical and mechanical processes, which is determined by the self-organization of dehydration [5]. The kinetic characteristics of a liquid that dries on a substrate reflect the morphological state of the object to which it belongs. For example, if we analyze the biological fluid of the human body, the interpretation of the results allows us to use the phenomenon as an additional criterion not only in functional electronics [3], but also in biomedical diagnostics [6] and, most importantly, in medicine. Based on the results of the study of the stages of nonlinear processes occurring in such systems using materials Based on the methods of natural science research [3,7], it is important to identify and consider the mechanisms that determine and have signs of nonlinear processes at each stage of the formation of the spatial and temporal structure in a multicomponent liquid, in particular, blood serum. The results obtained allow us to note the following. The multistage process of drying a drop of blood serum on a rigid substrate occurs in two stages: 1) evaporation of free water, which lasts 20-35 minutes (for drops of 3-5 ml); 2) further evaporation of water, which lasts 2-3 days; 3) evaporation of water is accompanied by an increase in deformation and the formation of additional cracks that form the final morphological appearance of the drying film, suitable for microscopic studies.

Among the mechanisms responsible for the formation of morphological features of droplets at both the first (liquid) and second (solid) stages of drying are the following: a) interaction of the droplet with the substrate; b) distribution of components with different surface activities in the droplet; c) phase transitions in the droplet. At the beginning of the drying of a liquid droplet on a solid substrate, the colloidal phase is carried to the periphery, and the ratio of components in the liquid (inner) part of the droplet changes. As a result of water evaporation, the ionic strength of the solution increases, and the volume fraction of the colloidal phase decreases due to its removal to the periphery. The forces of attraction between particles increase due to a decrease in the Debye radius and an increase in the density of surface charges. The radius of interaction between the particles decreases. The colloidal phase gradually loses its hydrate shells, the charge of the molecules approaches the isoelectric point, and the solution goes into a metastable state with subsequent coagulation, as shown in. Colloidal particles can form different structures- from colloidal glasses, with a very high-volume fraction of colloid and a weak force of interaction between particles, to colloidal gels, with a very small volume fraction of colloid and a large force of attraction between particles. Before the formation of a gel, colloidal particles form fractal clusters, which subsequently combine into a spatial lattice - a gel. Any deviation from the fractal growth of nanoclusters (a detailed analysis of such systems was carried out by one of the authors, see monograph [3]) leads to a violation of gel formation. Given the existing relationship between salt concentration and pH value in protein solutions, it can be expected that a shift in the isoelectric point of albumin when it is loaded will cause a change in the coagulation kinetics.

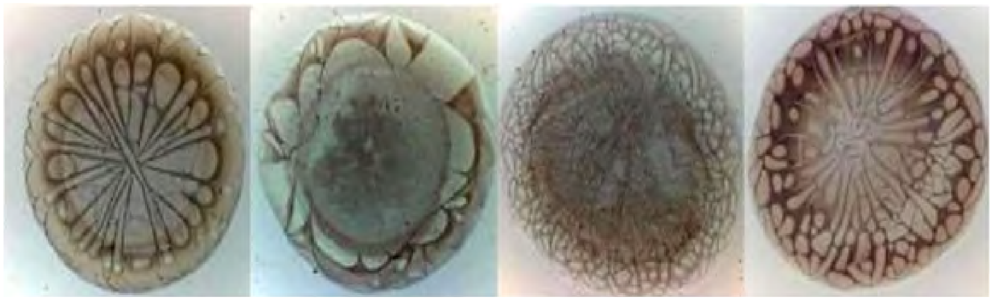

Figure 1: Dried serum drops [4]; 28 X magnification: a) practically healthy person; b), c), d) people with different types of diseases

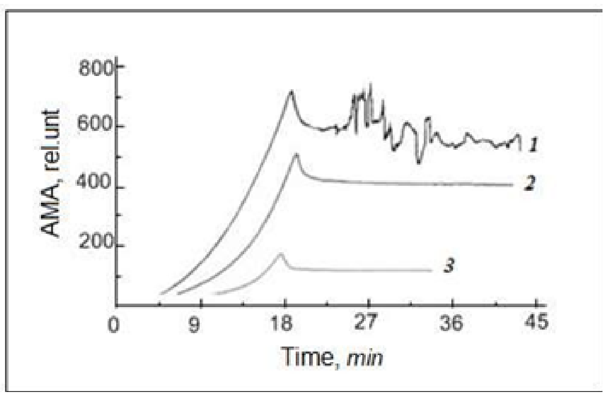

Figure 2: Dynamics of AMI of drying droplets in the blood serum of a person who died from a drug overdose (1); a practically healthy donor (2); a person who died from alcohol intoxication (3).

3. Methods for Analyzing the Kinetic Self-Organization of a Liquid

The process of self-organization of the drying liquid makes it technically possible to record the dynamics of this process. These processes can be combined. On this basis, we propose a technology that opens up new prospects for the study of liquid media, as well as for the development of a number of practical applications in medicine based on it. Let us consider a method for analyzing multicomponent liquids based on a sensor device. The main feature of this method is obtaining electronic signatures of liquids. Here, blood serum is a good example, as it is suitable for identification, attestation, and certification. The information basis of the method is the dynamics of complex processes of self-organization of drying droplets, which is critical for the composition and structure of the liquid. Recording these dynamics in the form of acoustic- mechanical impedance (AMI) allows to obtain quantitative differences between the compared liquids, which can be used to control their quality by comparing them with a reference. To analyze the self-organization of liquid droplets, a resonator in the form of a quartz plate with an xys/1030′ cut of 48.0-4.5-1.2 (mm) is proposed. The resonator performs longitudinal oscillations of the compression-expansion type. In the process of measuring the operating vibration frequency of the quartz plate, it is forced to maintain constant and equal to the resonant frequency of the unloaded resonator - 60 kHz. The vibration amplitude of the unloaded resonator is sinusoid ally distributed along the length of the plate. A drop of the liquid under study is placed near the end of the plate. Estimates have shown that the distribution of the vibration amplitude along the length of the plate practically does not change when the resonator surface interacts with the object (drop). In other words, the drying droplet does not introduce errors into the distribution of the oscillatory velocity along the length of the resonator. The measured value is the complex electrical conductivity of the resonator loaded with the drop, while the bridge circuit of the device subtracts the resonator's own capacitance and the capacitance of the connecting cable. The AMI value of a drop of liquid (blood serum) is calculated from the measured electrical conductivity and displayed on the screen in real time. At the stage of droplet evolution, the AMI modulus value is measured, displayed and recorded. AMI is the value of the acoustic or mechanical impedance of an object that loads quartz in the irrigation oscillation mode. In our case, we study such a specific object as a drop of liquid (blood serum) drying on the surface of quartz (substrate). Therefore, it can be assumed that AMI(A) integrally includes such physical characteristics of the object as viscosity, elasticity, friction, and mass with varying degrees of adhesion to the substrate. The analytical results of the measurements proved that the initial and final values of the AMI can be mathematically described by the expressions presented and substantiated in detail by the authors of [3]. However, such simple situations as "liquid sample at the beginning" and "solid residue at the end" do not always occur.

4. Structural Analysis of a Liquid on a Sensor

The kinetics of drying a liquid on a solid wettable substrate is a natural model of a self-organizing system with an infinite variety of process variants depending on the composition and structure of the liquid. The drying process is determined by the initial parameters of the solution: surface tension, wetting, viscosity, internal structure, dispersion, thermal conductivity, ionic strength, and pH. These factors affect such processes as aggregation, precipitation, sedimentation, gelation, and crystallization that accompany the drying process of a multicomponent liquid, as described by the authors of [4]. As a result, the physical properties of the liquid change, the dynamics of which is displayed as a curve in the AMI-time coordinates. Our research has shown that the shape of the AMI curve is a passport characteristic of the liquid under investigation, such an important component in forensics as blood serum. An important, clearly controlled parameter of the liquid in our technology is its volume. It is this parameter that must be the same for the liquids being compared. The liquid is applied by means of a microscopic dispenser and its guide arms to a specific location on the quartz plate from a low height, which prevents splashing. The splash area is not limited, as it reflects the degree of wetting of the substrate and is one of the important characteristics of the liquid. Therefore, a uniform sensor surface quality is critical for this technology. Before and after the measurement, the sensor surface is successively treated with water and isopropyl alcohol and then thoroughly dried. This procedure for treating the quartz surface allows for high reproducibility of results when measuring the same liquid repeatedly. Thus, the parameter that ensures the formatting of the proposed technology is the wetting of the sensor surface. With the same volume of samples of the liquids being compared and different degrees of wetting, the droplets of these liquids will have different shapes: one will be flatter, with a larger base area, and the other will be more convex, i.e. with a smaller base area. This will certainly provide a difference both in the initial signal value and in the dynamics of liquid structuring during the drying process.

Another important parameter affecting the dynamics of AMI is the physical properties of the adsorption layers. It has been experimentally proven that in a multicomponent liquid, the components of the liquid phase are redistributed according to their surface activity. The concentration and qualitative composition of surfactants forming the adsorption layer at the interface determine the mechanical properties of the liquid, such as elasticity and density, limiting to varying degrees the evaporation of water and, consequently, its structuring. The surfactant also reduces the surface tension at the liquid-substrate interface and reduces the rate of gelation in the colloidal phase by forming a structural and mechanical barrier. This is what ensures the occurrence of pressure during the formation of blood nanoclusters. These processes contribute to the dynamics of the AMI signal, since the size of nanoclusters significantly affects the evolution of structure formation in the drying liquid. The self-organization processes in the drying droplets of multicomponent liquids reflected by the AMI signal are completely reversible and well reproducible [5,6]. It should be noted that the model description of the processes occurring in a liquid during drying is still an extremely difficult task from a technological point of view. The quartz resonator technique described in [2], which allows for quantitative analysis of blood serum (and other fluids: sweat, semen, etc.) of patients, can be used to form a database. The analysis of the process of self-organization (by drying) of a drop of a multicomponent liquid occurs in several stages. This is evidenced by the AMI values for the donor's blood serum. The first stage is the formation of a protein roller along the periphery and flattening of the drop dome. Then, a gel is formed and the process of salt crystallization in the gel matrix takes place. The process ends with the evaporation of the remaining free water. At the same time, the maximum (i.e., saturation) of the AMI signal is observed. The whole process takes 40 minutes. It is the ratio between the mass of the droplet and the freely bound water that characterizes the adhesion of the precipitate to the surface of the quartz plate.

(Figure 2), as an example, shows the dynamics of the AMI of drying droplets in the serum of patients with various diseases. For comparison, (Figure 2) shows the stages of drying blood serum from a practically healthy donor (curve 2) and people with pathologies (drug addiction - curve 1, alcohol intoxication - curve 3). Curve 2 reaches saturation and maintains it due to the constant weight and sufficient elasticity of the formed gel with loosely bound water. It is this curve that should be considered the reference for quantitative analysis of the liquid structure. This approach to the study of multicomponent fluids allowed us to create a patient database. Typical AMI curves of multicomponent fluids of patients who died from an overdose of drugs, alcohol, sugar, etc. can be compared with those of a healthy donor. In particular, high levels of drug metabolites, such as those found in the blood of a sick person, prevent gelation in the drying serum droplets, making them extremely fragile. Even small loads on the quartz (compression and tension) lead to intense cracking of the remaining droplets and detachment of their fragments from the sensor surface. This chaotically changing mechanical load is displayed on the monitor screen in the form of unbalanced zigzag lines (Curve 1). The AMI curves of people who died of alcohol intoxication (curve 3) differ from those of a healthy donor by low AMI values. Thus, the study of the kinetics of liquid drying on the surface of an oscillating quartz resonator, due to controlled physical and chemical parameters that integrally affect the shape of the AMI curve, provides information about the liquid sufficient for its identification. Thus, this method of registering the impact of a number of physical factors (magnetic field of UV radiation, odors, X-rays) on a liquid opens up the possibility of quality control of wines, juices, milk and other liquids. Among other things, the identification technology we describe can be a convenient tool for detecting counterfeit medicines using an express method, as well as for one-step determination of the Ratio, the ratio of soluble solids to acidity, the main indicator of juice quality used in expert laboratories.

5. Conclusion

The results of the research described in this article are systematized as follows: the quartz weighing method was analyzed; the structure of blood serum was identified; the topology of the multicomponent liquid was determined; and the physical and chemical properties of the liquid were analyzed. The tasks we solved made it possible to determine the morphology of the liquid material, and hence the multicomponent substance. The method of analyzing the morphology of multicomponent liquids is based on the use of a sensor device, a characteristic feature of which is the acquisition of electronic signatures of liquids suitable for their identification and certification. The information basis of our proposed method is the dynamics of complex processes of self-organization of drying droplets, which is critical for the composition and structure of the liquid. Capturing these dynamics and expressing it in the form of AMI curves allows us to obtain quantitative differences between the compared liquids, which can be used to control their quality by comparing them to a standard. It should be noted that the structural evolution of drying droplets of biological liquids is a complex multistage process in which two stages can be conditionally distinguished: events occurring during the evaporation of free water and structure formation associated with water evaporation. The stages of structure formation at the first stage are as follows: interaction of the liquid with the substrate, subject to wetting, formation of an attachment line to the substrate and development of centrifugal capillary flow; formation of a vitreous layer on the periphery of the drop; distribution of dissolved components according to their surface properties and formation of adsorption layers along the interface; a cascade of phase transitions; crystallization of salt in the gel matrix.

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