Formation of the structure of gas-liquid, food dispersion non-equilibrium liquid systems in conditions of hydrodynamic and acoustic cavitation

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Abstract

The present article illustrates the principles of the transition from macro static equilibrium to the equilibrium of micro static volume of liquid dispersion systems under conditions of the combination of two types of cavitation actions. In the process of food dispersion systems production, a non-equilibrium state is created. Properties of disperse systems are explained on the example of invert syrup. In static conditions were determined the principles of formation of aggregates: from sucrose molecules at the stage of sugar dissolution to formation of voids and gas-phase bubbles. Conditions of creation of the dynamic non-equilibrium state of syrup by combining hydrodynamic and acoustic cavitation are taken into consideration. The character of bubble state transformation in conditions of geometry flow in gas-liquid systems and sound oscillations excitation in them is shown. After the collapse of bubbles in the gas phase the nature of the conversion of highly concentrated gas-liquid systems is also represented. That was the determining factor in the formation of materials of new types. The practical output of the present work was the production of products in the conditions of combined cavitation effects.

Keywords

Macromolecules, cavitation, kinetic reactions, invert syrup

1. Introduction

From the formal point of view, all existing methods for food products processing may be classified by their properties into several classes like mechanical, thermal, biotechnological, biochemical and chemical (Aksenova, Kochetov, Lisitsyn et al., 2015; Oganesyants, Khurshudyan, Galstyan et al., 2018). That classification of technological processes is based not only on the type of energy supply and energy redistribution, however, also on energy dissipation in the technological systems. This means that the incoming energy fluxes are being consumed by useless losses and changes in a food system. These two factors determine the final product quality despite the previous formal separation.

Such features are evident in existing traditional technologies of production of food conglomerates production, in particular confectionery (Aksenova, L.M., Kochetov, V.K., Lisitsyn,
A.B., et al., 2015; Kondratyev, N.B., Savenkova, T.V., Radetsky, 2007; Krylova, E.N., Savenkova, T.V., Mavrina, 2012; Nurumukhanbetova D.E., 2018; T. V. Savenkova, 2003). It means that similar technological processes proceed relatively slow. In this case, at each moment some macroscopic state equilibrium is established in the system. Thus, we can describe this state with macroscopic parameters as density or molar concentrations for multiphase flow and temperature. The impact on the medium begins on the molecular level and finishes on the macroscopic spatial level in all technological volumes. Furthermore, the kinetic reactions last until they reach the values of the appropriate local equilibrium state.

Under perfect circumstances, energy losses would be desirable for us. It should be noted that the biological properties of the medium due to may deteriorate to excessive heat treatment. Unlike to traditional approach, we would like to make local changes in the structure of the treated medium. In the end, it should change drastically its physical and chemical properties.

In our opinion, the issue can be solved only in the framework of a complex approach (Caruso et al., 2009; Suslick & Price, 1999). Hereinafter, we are going to discuss one of the possible solutions to the problem due to the features of structure formation in liquid high concentrated food dispersed systems.

It should be noted that in structured systems at any speed, two opposite processes occur - destruction and reconstruction of the structure. The resulting characteristic describing the equilibrium state between these processes in the steady state flow is the effective viscosity.

The nature of the technological processes is reflected by the dependence of the effective viscosity on the degree of destruction of the structure and is described by a complete rheological curve, from an uncured to a completely destroyed disperse system (fig. 1).

![Complete rheological curve of disruption of structure of disperse systems](image)

**Fig. 1. Complete rheological curve of disruption of structure of disperse systems**

### 2. Structural properties of liquid food dispersed systems

The structural properties of liquid dispersion systems (LDS) are widely used in the food industry. Distinctive features of such systems are 1) increased content of dry substances, at least 70%;
production of solid particles with high dispersion; 3) complete absence of translational movement. The subject of the study is invert syrup which is widely used in the confectionery industry. The syrup contains 80% of reducing sugars with 100% decomposition of sucrose into glucose and fructose. This syrup belongs to one of the simplest macromolecular media where the processes of structures forming can be demonstrated easily.

During the initial stages of invert syrup production, while the sugar syrup is heated up to 90°C to be dissolved, a large number of particles of the dispersed phase with the formation of aqueous shells from the dispersion medium around them are obtained. Due to the uncompensated molecular forces in the interphase surface layer, the retention strength of hydration shells with particles of the dispersed phase is determined by molecular adhesion forces. The process of heating which constantly leads to the increasing amount of fructose and glucose create the most favorable conditions for maximum convergence of particles of the dispersed phase. Meanwhile, the hydration shells break, they adhere to form aggregates of various spatial shape and size.

As a result, specific surface area decreases as far as its’ density with the simultaneous formation of voids of various values. All this indicates that the syrup may have a compressibility property. (Karimov, Taleysnik, Savenkova, & Aksenova, 2019; Mark, 2007; Uriev, Taleysnik, 1985). Such voids in their physical essence can be cavitation embryos, whose physical characteristics can change over time. Different emulsions in which the density during mechanical action decreases over time have similar features (Uriev, Taleysnik, 1985). It could be said that a fundamental feature of liquid media with a high solids content serves the presence of a significant number of cavitation nuclei, which under certain conditions can become macroscopic gas bubbles. After the maximum possible dissolution of sugar and the gas-liquid system formation with a large number of aggregates from sucrose molecules, further inversion is carried out under conditions of creating a dynamic and non-equilibrium state of the syrup.

During the transition from the macroscopic to the microscopic level of LDS, it becomes possible to control various technological flows. It is possible by combining fast and slow
hydrodynamic processes (Aksenova, Kochetov, Lisitsyn et al., 2015; Uriev, Taleysnik, 1985). For example, these processes (fig.2) can occur in a laboratory cavitation installation where two types of cavitation are used (hydrodynamic and acoustic). According to the hydrodynamics laws under hydrodynamic cavitation conditions (HDC), flow rate rockets and a dramatic pressure drop occurs. With surface energy accumulating gas bubbles grow, aggregates are destroyed.

Acoustic cavitation, resulting from generated oscillations from a ultrasonic transducer with 18-24 kHz frequency, provides turbulent movement of solid particles of disperse system in direction of flow movement. Due to a sharp increase in the speed of movement and a decrease in pressure in the cycle of oscillations of the cavitation bubble at the rise of the wave, its volume increases significantly. A short-term rarefaction and further accumulation of surface energy are created (fig.3).

\[ V_1 < V_2; P_1 < P_2 \]

V1, P1 speed and pressure at the rise of the wave
V2, P2 speed and pressure at the descent of the wave

Fig. 3. Scheme of oscillation cycle in gas phase bubble under conditions of acoustic cavitation

At the descent of the wave the pressure increases sharply. Under the conditions of deformation processes bubbles are compressed. Moreover, energy is released with the formation of a shock wave during the collapse, which causes maximum dispersion of aggregates from solid-phase particles and their uniform distribution in the dispersion medium. At the outlet of the reactor, when the pressure increases, the bubbles collapse with the release of stored energy, which leads to the destruction of the aggregates. Such selective exposure can initiate various kinetic reactions and, from now on, change the structure of the medium.

At this rage bubble collapse process may be sufficient to agitate, ionize, dissociate water molecules (Aksenova, Kochetov, Lisitsyn et al., 2015; Uriev, Taleysnik, 1985). We suppose that the particles of the dispersion medium, colliding with fragments of broken macromolecules, will be included in their composition with the possibility of forming new types of substances.

3. Bubble dynamics growth in the non-equilibrium environment during the cavitation

In simple liquids like water, the development of gas bubbles may be accompanied by coagulation of those bubbles, their emergence from the medium under the action of buoyant force (Babaeva et al., 2016). One should take into account the fundamental difference in the dynamics of simple liquids and LDS caused by the structural features of highly concentrated systems. The molecules along the interface between gas-liquid phases being exposed to constant frequency sound waves conduct themselves as dipoles. Each of the ends concentrates a charge of different signs (Babaeva et al., 2016).
It can be assumed that the interacting molecules are stretched in the same direction. The signs of their charges on each side are also the same. As a result, a membrane effect (which prevents the transit of certain molecules and ions) occurs at the gas-liquid interface. That border forms a so-called amphiphilic structure with basic proprieties similar to cell membranes. The amphiphilic structure prevents gas bubbles in their convergence from amalgamation. Where adjacent bubbles are separated by the film, a large number of bubbles start to form a foam. This mechanism determines the structure of many food liquid dispersed systems.

![Flow diagram in a limited volume](image)

**Fig. 4.** Flow diagram in a limited volume (Karimov et al., 2019)

Coagulation in non-equilibrium liquid dispersed systems is impossible because of the amphiphilic effect. On account of the revealed acoustic effect when the total dynamic pressure changes in particular the hydrodynamic pressure, the value of the surface energy increases as well as and the size of the gas medium bubbles. A change in the configuration volume limiting the flow affects via increasing velocity and a hydrodynamic pressure drop (Fig. 4). However, the acceleration of the flow can be replaced by its inhibition. The kinetic energy of the liquid turns into a thermal form. As a result, the pressure increase will occur, leading to a collapse of the bubble with the release of stored energy in the so-called singular point (Karimov et al., 2019; Suslick & Price, 1999).

**4. Practical results**

At present, technically, the combination of hydrodynamic and acoustic effects, under which conditions the passage of physicochemical processes is initiated, is implemented on a laboratory cavitation facility, ensuring maximum energy storage and its release with an intense technological effect (see Fig. 7) (Aksenova, Kochetov, Lisitsyn et al., 2015; Karimov et al., 2018). Here, the flow is realized the velocity distribution $V_3 > V_2 > V_1$ and the pressure distribution $P_3 < P_2 < P_1$ (see Fig. 5).
The effectiveness of combining two types of cavitation effects was reflected in the production of invert syrup with 100% inversion of sucrose into fructose and glucose. Application of cavitation effect for the first time ensured production of invert syrup with high dispersion, 100-400 nm. (see fig. 6). A distinctive feature of invert syrup is degassing, after collapse of bubbles at reactor outlet, with energy release (Aksenova, Kochetov, Lisitsyn et al., 2015).

Distinctive features also include the lack of conditions for sedimentation of aggregates, due to the high density of the syrup.

In the process of structuring the syrup, the presence of a large number of microaggregates (formed as a result of sucrose inversion and dispersed under cavitation conditions) is a determining factor in their interaction with the formation of the structure by forming coagulation and point contacts with a number of fundamentally new modified properties.

The positive effect of combining two types of cavitation action is confirmed in the production of one of the leading representatives of flour confectionery products - sugar cookies (Dorn, Savenkova, Sidorova et al., 2015; Savenkova, Osipov, Kazantsev et al., 2016). Under cavitation conditions at the emulsion production stage, the size of solid particles of the dispersed phase decreases to 6 μm with a
significant increase in the number of dispersed particles of sugar sand surrounded by a coating from the dispersion medium.

Finished cookies are prepared with modified properties and improved quality indicators: absorptivity up to 230% (according to the classical technology 180-200%), density (20% decrease), increased porosity and crumbling (see Fig. 7).

Fig. 7. Appearance of cookies obtained in different ways

Invert syrup was the basis for creating new types of marmalade using fresh fruits and vegetables. A distinctive feature of the technology for obtaining these types of products is the relatively low temperature regime of about 70 °C (instead of boiling out using existing technologies), this allowed to significantly preserve native vitamins and minerals in the products. For example, when using carrots and invert syrup obtained under cavitation conditions, the amount of β-carotene in the products became 14.2 mg/100 g, according to the existing marmalade technology, the amount of β-carotene didn’t exceed 1.5-2%. Thus, the prospect of creating food products with given properties and new products of a specialized purpose opens up.

References
Aksenova, L.M., Kochetov, V.K., Lisitsyn, A.B., Nikolsky, K.N., Panfilov, V.A., Podhomutov, N.V., Semenova, A.A., Taleysnik, M. A. (2015). Food technology and nano transformations of biopolymers. Krasnodar: Diapazon-V.

Babaeva, N. Y., Berry, R. S., Naidis, G. V., Smirnov, B. M., Son, E. E., & Tereshonok, D. V. (2016). Kinetic and electrical phenomena in gas–liquid systems. High Temperature. https://doi.org/10.1134/s0018151x16050059

Dorn, G., Savenkova, T., Sidorova, O., & Golub, O. (2015). Confectionery goods for healthy diet. Foods and Raw MaterialsFoods and Raw Materials, 3(1), 70–76. https://doi.org/10.12737/11240

Karimov, A. R., & Schamel, H. (2001). Influence of initial velocity field on the formation of coherent structures in simple hydrodynamic flows. Physics of Plasmas, 8(4), 1180–1192. https://doi.org/10.1063/1.1352059

Karimov, A R, Taleisnik, M. A., Savenkova, T. V, & Aksenova, L. M. (2019). The influence of velocity field on simple chemical reactions in viscous flow. Physica Scripta, 94(4), 045002. https://doi.org/10.1088/1402-4896/aafc15
Kirsanov, E.A., Matvienko, V. N. (2016). Non-Newtonian Behavior of Structured Systems. Moscow: Tekhnosfera.

Margulis, M. A. (2000). Sonoluminescence. Physics-Uspekhi, 43(3), 259–282. https://doi.org/10.1070/PU2000v043n03ABEH000455

Savenkova, T. V. (2003). Scientific basis of creation of confectionary products of dietary purpose. Confectionery Manufacture, 2, 12–13.

Savenkova, T. V., Osipov, M. V., Kazantsev, E. V., Kochetkova, A. A., Vorobieva, V. M., Vorobieva, I. S., & Kiseleva, T. L. (2016). The production technology of diabetic confection with modified carbohydrate profile. Research Journal of Pharmaceutical, Biological and Chemical Sciences, 7(6), 3123–3130.

Sucrose Molecule. (2016). Retrieved from https://www.worldofmolecules.com/foods/sucrose.htm

Uriev, N.B., Taleysnik, M. A. (1985). Food disperse systems. Moscow: Agropromizdat.