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Hydrodynamic Sonochemistry in Food Processing

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Abstract: - The paper presents a detailed analysis of the physico-chemical effects of acoustic cavitation used in food processing. The mechanism of interaction between acoustic cavitation and food media is discussed. An overview of recent studies carried out on dairy processing using acoustic cavitation is provided along with a full analysis of these studies. The acoustic cavitation reactors available in the market are not suitable for large-scale food processing despite positive results obtained with laboratory and pilot scale experiments. Considering a new approach to the theory of cavitation in rotary machines, it has been suggested that hydrodynamic cavitation can be an alternative to acoustic cavitation in food processing applications involving large volumes. A model that has been developed is suitable for the construction of new generation cavitational rotary disintegrators.

Keywords: - *Food sonochemistry, hydrodynamic cavitation, cavitational rotary disintegrator.*

I. INTRODUCTION

Ultrasonic sonochemistry, despite its young age in science, has firmly taken the place of a separate section in high energy chemistry [1]. Now it has separate research areas such as ultrasonic food processing that are actively developing [2-5]. It promises to solve many problems faced by the food industry, such as the efficient replenishment of moisture lost during the storage and primary processing of edible raw materials. The mankind has been forced to keep the ever-increasing supplies of raw food materials in the dried and frozen form. Therefore, the effective binding of water with food biopolymers – hydration process – is one of the major problems in the modern food industry. The advantages derived from the addition of water during food processing were established by Henry IV Bolingbroke.

A global scientific community for the first time focused its attention to the importance of water in food in 1974 at the International Symposium «Water relations of food» in Glasgow. Then the Proceedings of the Science Forum, edited by Professor RB Duckworth of the University of Strathclyde [6], was released which is now more popular among professionals. Biochemists at the symposium reported that chemically pure protein can theoretically bind up to 40% water by weight as a result of hydration. Hydration shells of protein increase their affinity for water during the precipitation of colloidal systems that can further enhance the hydration of ground biomass.

The hydration process and any reversible chemical reactions move to energetically favorable conditions in accordance with the principle of Le-Chatelier-Brown. As hydration is an exothermic process, it is better when the hydration shell of the protein is built from individual water molecules at the initial stages of the reaction, which can be achieved by pre ultrasonic treatment. Its action is based on the distribution of water in the periodic pressure pulses that are under the influence of elastic ultrasonic waves emitted by microscopic gas inclusions (cavitation bubbles). Ultrasonic treatment can lead to hydration without heating and does not affect the properties of water, such as its solvation power, structure, etc. Similar mechanisms of action of cavitational restructuring hydration shells of ions in real solution may be responsible for the denaturation of biopolymers in their colloidal solutions and even dispersed phase of sols, and emulsions, that is, in any process in which the object is formed by the impact ion-dipole and dipole-dipole interactions structural connections. Many useful reactions are induced by ultrasound in liquid media: food processing is based on similar such reactions [3, 4]. It is established that the formation of dense and strong hydration shells raise the dissolved thermoresistance valuable nutrients and vitamins, preventing them from thermal denaturation at the subsequent heat treatment [5].

II. RESULTS OF PRELIMINARY STUDIE

During the ultrasonic processing of food ingredients, routine sonochemical reactions are not desirable. Products generated by reactions in the gas phase inside the cavitation bubbles and subsequently in the liquid phase (induced by primary radicals) should not be present [2, 4] during food processing. In connection with this concept, some studies have identified [4, 7] the safe limits on the usable frequency and intensity of ultrasound. Recommended frequency is 20 kHz and the average amplitude of the reactor sound pressure should be under 2 bar.

For food processing applications, where processing is subject to small amounts of food to several tons a day, ultrasonic equipment is available in various scales. For example, Hielscher Systems GmbH [9] has built ultrasonic equipment for small to large scale processing. One of the Hielscher products, a flow type reactor, meets the engineering prediction of one of the founders of the cavitation. Professor Knapp from University of California, who once wrote that the industrial use of cavitation will be possible only when reactors are built that can handle continuous flow of fluid [10]. At University of Melbourne has used a 4 kW flow through ultrasonic unit for the processing of dairy fluids (Fig 1) [11].

It was found that, in spite of the desired frequency ultrasound (20 kHz), with reduced production of free radicals and desired reduction in the viscosity and increased heat stability of protein concentrates, such units have limitations in terms of processing volume (6 L/min). So, where volumes are handled tens of tons per day or more, the use of ultrasonics becomes a problem. For processes with small production quantities, such as moistening the grain before grinding or «wet» salted minced meat [12, 13] in studies conducted in Moscow [4] the ultrasonic cavitation reactor РКУ-0,63 was used. It is intended for brine meat and food products from minced meat, where process volume of 80 tons per day of 1-2 tons per day could be achieved.

Fig 1. Pilot-scale experimental setup used in dairy processing using *Hielscher Systems GmbH* UIP-400 (Figure Adapted from [11])

In this regard, it has to be recognised that hydrodynamic cavitation was introduced more than half a century ago in the processes of homogenization, dispersion and emulsification [2, 14]. Cavitation is generated due to constrained fluid flow. Technology in this field has made a great progress in recent years. One of the first explorers of the hydrodynamic cavitation, Dr. J. Hint in Estonia, continued to engage in traditional rotary hydrodynamic disintegrantors. J. Hint, was the first, who noticed in the process of mechanical disintegration that not only dispersion, but some other important physico-chemical properties of processed materials are changed: this has introduced the concept of «*disintegrational activation*». Later he began to also write about the activation of liquid media in mechanical disintegrators [15]. The phenomenon of activation of the rotary-pulse devices will soon become uniquely associated with the action of cavitation [16]. But only in this century, the disintegration of fluids as a result of erosive effects of hydro [17] or acoustic cavitation has been highlighted and an independent term «*cavitational disintegration*» [18] has been established.

In studies involving the phenomenon of cavitational disintegration of oil-water emulsions produced on board of a *Pacific Ocean Shipping Company Ltd*. Ferry, the performance characteristics and flow of emulsion with high water content was noticed. The effect can be attributed solely to the hydration reaction. Bunker oil is a substance with polar molecules such as naphthenic acids and resin-asphaltene compounds. These are different from petroleum hydrocarbons of higher molecular weight and the presence of oxygen, sulfur and nitrogen hetero-atoms [19] provides them with the polar groups. In the process of emulsification in the rotary-pulsed apparatus, they react with the hydrated water, which seems, is still subject to cavitation disintegration, and deprived of its own structure. This hydration increases the hydrophilicity of these substances and contributes to the formation of these structural and mechanical layer at the interface of emulsion, increase its dispersion and improve the conditions of combustion. These hydration reactions are also important in food processing applications of acoustic cavitation. After all, the components of the protein molecules in the hydration reaction of amino acids involved active polar centers represented the carboxyl -COOH, hydroxyl -OH and amino $-NH_2$ groups. The binding of water to protein molecules results in hydration structures (Fig 2).

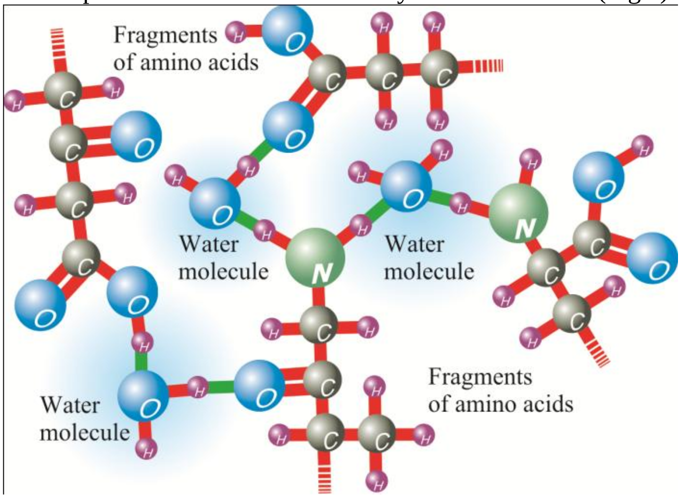
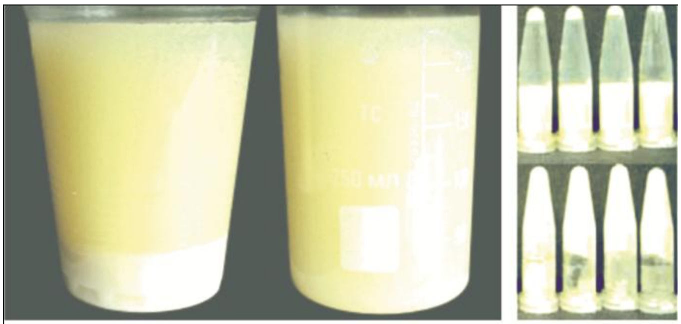


Fig 2. Hydration and structuring of proteins

Studies have shown that ultrasonically treated water results in better hydration of proteins, stabilized against separation and precipitation, resulting in complete dissolution of proteins (Fig. 3a). However, the structuring of the protein leads to an increase in the viscosity of protein solutions and changing the conditions of occurrence of syneresis in them, which is not always a positive feature. Some investigations on the effects of ultrasound on whey proteins were aimed at expanding the parameters used in subsequent stages of heat treatment technology and to facilitate the further process of ultrafiltration. This involves the need to reduce the viscosity and prevent the formation of protein aggregates [3, 20].

The desired effects cannot always be achieved by a separate water treatment, as previously recommended. The ultrasonic treatment of aqueous solutions of dry whey protein WPC 80 (Figure 3b) resulted in an increase in viscosity under specific experimental conditions. It can be clearly seen that the sonication effect in terms of increasing the solubility of proteins differ from the effect of heat stability.



a). b).

Fig 3. a) Reconstituted whey, left without sonication, right with the sonication at 22 kHz, 90 W/L, 1 min. b) solution of whey protein from WPC 80 in the top row without treatment, the bottom with treatment at 20 kHz, 31 W, 60 min. Post heating to +80 ° C for 10, 12, 14 and 16 minutes (left to right)

III. THEORY AND COMPUTER EXPERIMENT

Due to the shortcomings of ultrasonics processing, a new look at the hydrodynamic method has emerged for producing cavitation and for the possibility of its use in food processing. Here, we describe hydrodynamic cavitation disintegrators, which are based on the hypothesis of Hint, linking the basic principles of building construction disintegrants. It can be expressed by the following quote from his essay: «... *the more the number of strokes imparted by particulate matter, the greater the impact velocity and the smaller the interval between successive blows, the more there is activity*». From this assessment, similar representations of disintegration are set forth in the development of rotary-pulse devices, where the efficiency of hydrodynamic cavitation increases by increasing the rate of fluid flow and interruptions in its flow rate. The main factor is the kinetic energy of the fluid flow, which is proportional to the square its velocity [15, 16]. It is easy to realize in such rotor-switching device that processing 10 t/h of fluid, at a speed of 300 s^{-1} using a rotor diameter of 150 mm can be achieved using a 10×20 mm channel: the flow of the processing liquid occurs as long as these channels have openings with little more than a tenth of a millimeter. When gas bubbles move from a high to a low pressure zone, they do not survive [10]. The sudden drop in pressure leads to the growth and the eventual collapse of these gas bubbles, referred to as hydrodynamic cavitation. In roto disintegrators, the effects are not just due to the liquid flow alone – additional forces are generated due to hydrodynamic cavitation. Here, we should use a slightly different approach to assessing efficiency than commonly used. The pulsations of cavitation bubbles and the shock waves that emanate from them can be major forces causing cavitation erosion [21,22], which in the theory of vibrations and waves can be estimated by finding the value of the deformation of rarefaction and compression, that is, determining the magnitude of the potential component of the scattered energy in a fluid [10]. Cavitation power depends proportional to the square of periodic changes in pressure in the fluid. It depends not so much on the changes in pressure caused by the discontinuity of fluid flow through channels in the radial direction, but it is generated by movement relative to the profile of transversal section of the rotating rotor, which was due to the presence of periodically repeating holes.

The latter can be confirmed by looking at the homogenizers manufactured by *Aquametro AG*, in which the fluid does not flow in the radial direction but axially in the gap between deaf equipped with grooves facing each other on the walls of the rotor while the stator cross-sectional area of which the diameter remains constant (Fig 4a).

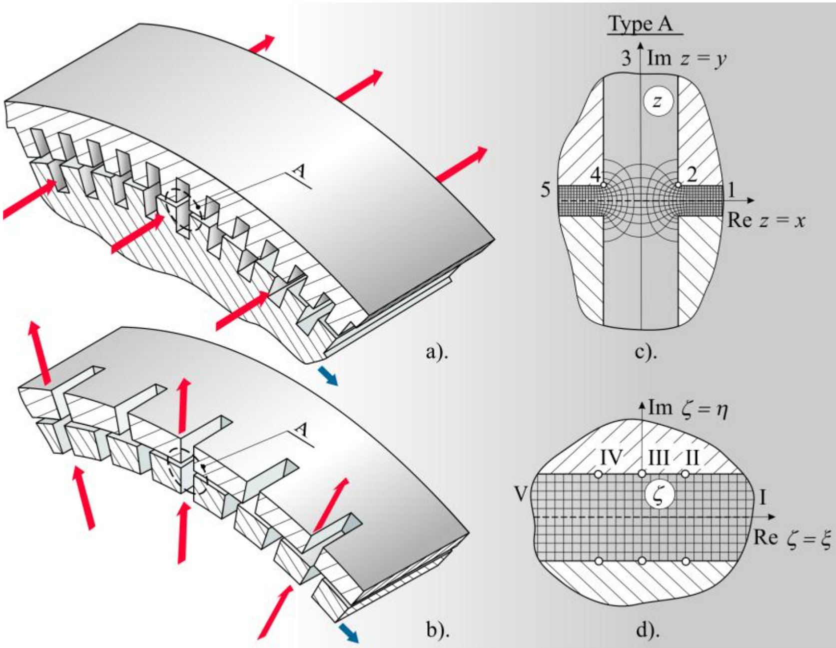


Fig 4. a). Schematic design of rotor and stator homogenizer (Swiss company Aquametro AG). b). The device with the rotor and stator in the form of cylindrical shells with the rectangular holes. Red arrows indicate the direction of fluid motion, blue - the rotation of the rotor. b). Is shown the cross section - the lines show stress-strain fluid having a rheological equation of state of the limit nature, as in an absolute elastic body, and the current without friction, obtained by conformal mapping d). Is shown of the invariant for section c). Arabic and Roman numerals indicate the corresponding angles.

But among the rotary disintegrators widely used, for example, where the rotor and stator are used with one or more cylindrical shells of finite length at the bottom, fluids flow through the hole of rectangular shape. The rotor and stator create a working volume between the top and the bottom of one another, fluid flow through the axis to the periphery of the construction in the radial directions by opening and closing of the rotating liquid flow channels (Fig 4b). Mathematical model of cavitation in a disintegrator may be based on differential equations of Hickling-Plesset or Rayleigh-Plesset [21.23] for vapor cavity wall motion under the influence of strain and stress, as described in some problems of solid mechanics theory of functions of complex variables. Description of this occurs when a mechanical stress in the fluid or the force of the surface area, that is, the pressure at any point in the volume of fluid can be made on the basis of conformal mapping. In order to apply them to a flat region, z profile of the treated liquid in a rotary disintegrator, with shaping the profile elements to display its conformal invariance, which has a uniform distribution of stresses, strains, such as an infinite strip of constant width (Fig 4c, Fig 4d) the Schwarz-Christoffel integrals can be used.

Such a model should consider the liquid friction on the structural elements of the cage, and thus the rheological equation of state has a limit, i.e., it is completely flexible. In calculating the absolute values of the characteristics of these conditions are not quite correct, but in the comparison of similar operations on the same liquid, they are quite acceptable, especially if the cavitation bubble on the wall behaves as a Newtonian fluid according to Hickling-Plesset equation. Mechanical stress at any point can be expressed in terms of the pressure in the working volume, p_0, derivative of the mapping function to map this point onto the invariant □:

$$\sigma = \frac{p_0}{\dot{z}^2}, \text{ where } \quad \dot{z} = \frac{dz}{d\zeta} \tag{1}$$

It is clear that the exponent at $\dot{z}$ is 2 only if all the profiles of fluid flow plane-parallel to each other. The nature of the elastic strain and stress, as well as their behavior in a batch of microscopic inclusions of steam in the liquid can be determined by numerical simulations. They allow for numerical comparisons. The behavior of the cavitation bubble was performed by numerical integration of Hickling-Plesset Runge-Kutt method. The periodic variation of pressure in the liquid was substituted appropriately (1) with a derivative of the function display strip to the diametrical cross section of one of the holes in the rotor, one of the holes in the stator and the gap between them:

$$z = \frac{\delta}{\pi}\,\mathrm{Arth}\,\frac{\mathrm{sh}\,\frac{\pi}{2}\zeta}{\sqrt{\mathrm{ch}^2\frac{\pi}{2}\zeta + \frac{a^2}{\delta^2}}} + \frac{a}{\pi}\,\mathrm{arctg}\,\frac{a\,\mathrm{sh}\,\frac{\pi}{2}\zeta}{\sqrt{\delta^2\mathrm{ch}^2\frac{\pi}{2}\zeta + a^2}}, \tag{2}$$

where: a – width of the opening (the size of the diameter); δ □ the gap between the stator and rotor; ζ □ coordinate on the invariant expressed by the complex number $\xi+\varphi\eta$. Its derivative is equal to ζ:

$$\dot{z} = \sqrt{1 + \frac{a^2}{\delta^2(2 + e^{\pi\zeta} + e^{-\pi\zeta})}} \tag{3}$$

Reciprocal of the square of $\dot{z}$ is proportional to the tensile deformation of the fluid at any point of the section, causing a pressure change:

$$p \sim \frac{\delta^2(2 + e^{\pi\zeta} + e^{-\pi\zeta})}{\delta^2(2 + e^{\pi\zeta} + e^{-\pi\zeta}) + a^2} \tag{4}$$

To calculate the change in pressure points on the real axis for a total period of the invariant, the coordinate must vary in the range □ξ. The time during which a corresponding change in pressure occurs will be $T = \frac{\xi}{\omega\pi R}$, where R – outer radius of the rotor, and is the minimum allowable period of pressure on the elastic deformation of the liquid. ξ is a root of the transcendental equation:

$$\varepsilon = 1 - \frac{\delta^2(2 + e^{\pi\xi} + e^{-\pi\xi})}{\delta^2(2 + e^{\pi\xi} + e^{-\pi\xi}) + a^2}, \tag{5}$$

where $\varepsilon = 0{,}05$ – margin of error in relative units.

The quantification of the effectiveness of cavitation can be estimated from the increment of the erosion of power that characterizes its action [22]. A comparative assessment of its value can be estimated using conditional parameters, Δ□ – erosion of unit capacity, that is, the extra power released in the pressures on the wall during the collapse, $p_{max} > p_0$, the maximum for the pulse of its volume V_{max} conventionally located in each channel in the middle of the stator and rotor clearance a cavitation:

$$\Delta P = \frac{\beta}{2}\omega V_{max}\,Nn\,(p_{max} - p_0)^2, \tag{6}$$

where: β □ adiabatic compressibility of the fluid; ω □ rotor speed; N – number of channels in the rotor, n – in the stator. The model was implemented in a computer program. Water was chosen as the liquid with the values of the equation of its state.

A computer experimental analysis was made on the behavior of bubbles with a diameter of 10 □ m using the model in accordance with (1) - (6) and on the basis of information contained in [17]. It was also verified by the measurement of power of the sound near the stator of the experimental cavitational disintegrator (Fig 5), on the frequency in which the cavitation noise was expected, calculated as:

$$f = 2\pi R\frac{\omega}{\delta z}, \text{ at } \zeta = \xi \tag{7}$$

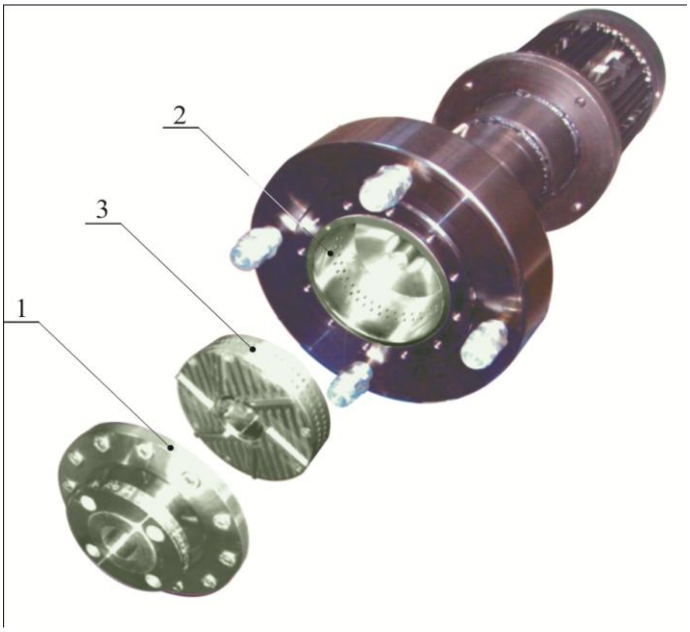
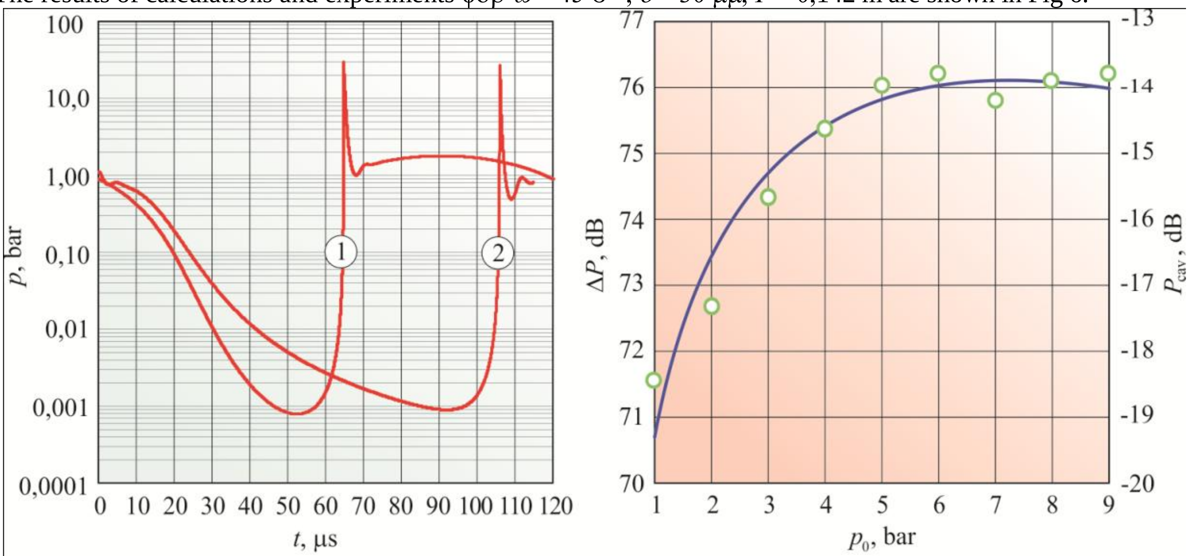


Fig 5. Photos of the prototype rotary cavitation disintegrator with a cover sealing the lid of the working volume 1 and withdrawn from stator 2 the rotor 3 and left.

The results of calculations and experiments φορ $\omega = 45\ \sigma^{-1}$, $\delta = 30$ μμ, $P = 0{,}142$ m are shown in Fig 6.



a). b).

Fig 6. a). Pressure on the surface of a cavitation bubble as a function of time 1 at the antinode of acoustic waves of frequency 20 kHz with an amplitude of sound pressure 1,2 bar; 2 in the gap between the stator and rotor of the disintegrator, b). calculated ΔP (blue), measured with low gain P_{cav} (green).

IV. CONCLUSIONS

The research outcome that is obtained to a scientific problem in a scientific laboratory using laboratory-scale equipment may not always be useful in large-scale processing. Such problems can be overcome by appropriate alternatives that are suitable for large scale processing. Experimental and theoretical studies have shown that cavitation rotary disintegrators, designed and manufactured by *Oil Tech Production OY*, may be suitable for large-scale processing in food industry, where hydrodynamic cavitation is found to be an alternative to acoustic cavitation for processing large volumes of liquid media. The concept of the hydrodynamic disintegrator fully meets the established requirements of the frequency and intensity of ultrasound in food processing.

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