Preparation of Emulsions Using Impinging Streams

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Two-phase dispersions consisting of a liquid dispersed within a continuum of another liquid are frequently encountered in industrial applications. The industries where dispersions play an important role include foods, medical and pharmaceuticals, cosmetics, agriculture, varnish, paints, and polymers. In many applications of emulsions the stability of droplets in a continuous phase is of paramount importance. The stability of an emulsion is an extremely intricate phenomenon but as a rule the smaller the size of droplets and the narrower the size distribution, the more stable is the emulsion.

Narrow size distribution is of specially great interest in the case of emulsions that are cosmetics, pharmaceuticals, or food products. In this case it is important to obtain the highest possible interfacial area from a given quantity of dispersed material (that is, the highest specific area of the disperse phase), which ensures effective swallowing of the active substance by a living organism. From that point of view it is especially important to break the biggest droplets, which may contain a considerable part of the total volume of the disperse phase. The size and size distribution of droplets in an emulsion also influence its rheological properties, which play an important role in many process operations like mixing or pumping.

The scope of the work covered investigations of the emulsions formed by impingement of two streams of droplet spray flowing one toward the other, containing atomized liquid components. In this method of producing emulsions, which was first proposed by Tamir and Sobhi (1985), particular conditions in the zone of impingement of two streams were applied that led to a breakup of droplets created by pneumatic atomizers.

The method has not been yet investigated in detail, although a few articles have been published by now (Kembłowski et al., 2000, 2001; Tamir, 1994; Tamir and Sobhi, 1985). The influence of the distance between the atomizers on the monodispersivity of the emulsion was not investigated. Tamir and Sobhi (1985) did not notice any considerable changes in the mean surface-area droplet diameter d_{20} with the distance that varied in their work in the range 20–170 mm. The technique of diameter measurement by Tamir and Sobhi did not allow them to measure smaller particles and the accuracy of measurements was rather low; the droplet of the diameter of 3 μm on the photograph was represented by a circle of diameter 1 mm. Their results showed, however, that the droplets were smaller than those obtained by one stream directed toward a wall, which they explained by the droplet coalescence on the wall.

Probably the reason for the low level of interest of investigators in this problem is one serious disadvantage of the method: it is more energy-consuming than the conventional methods of producing emulsions because of the application of pressurized air. In some cases, however, its advantages may be predominant over the costs, such as when the price of the product is high because of expensive stocks and the cost of energy does not substantially influence the final costs (some medicines and pharmaceuticals).

Experimental

The experimental part of the work was carried out using the setup described by Kembłowski et al. (2000, 2001). It was a modified version of the apparatus used by Tamir and Sobhi (1985). The main part was a chamber with two typical pneumatic atomizers facing each other. Both fluids were conveyed to the two atomizers and thus both of them produced the same or very similar aerosols. The distance between outlets of the atomizers was adjusted. During some preliminary tests performed in the same experimental setup using the same media (Kembłowski et al., 2001) it was found that at the pressure equal to 0.1 MPa the mean droplet diameter was only 30% higher than that at 0.3 MPa and the pressure increase over 0.3–0.35 MPa did not change the structure of the aerosols. Thus, the pressure of 0.4 MPa was applied in all experiments, which resulted in a volumetric flow rate of 0.9 to 1.0 × 10^{-3} m^3/s for each atomizer.

The distance h between the air exit in the nozzle and the

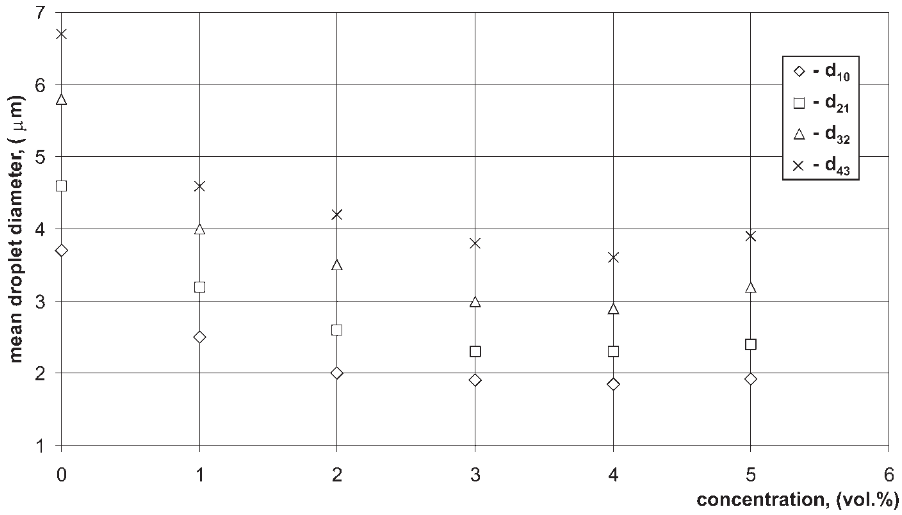

Figure 1. Dependency of the mean droplet diameter on the emulsifier concentration in water.

spray outlet was also investigated and presented in the same article (Kembłowski et al., 2001). It was found that the influence of the distance h on the aerosol droplet diameter was only slightly noticeable: the mean droplet diameter varied in the range from 1.9 to 2.2 μm with the minimum in the range between 1 and 1.6 mm; however, for $h = 1.6$ mm the monodispersivity was the best. Thus, the distance 1.6 mm was maintained in all tests.

Water containing the emulsifier Rokafenol N22/30 (produced by Rokita, Poland) and kerosene were used as experimental media. Preliminary tests were made for the influence of the emulsifier concentration in water on the mean droplet diameter at the distance between the atomizers equal to 25 mm. Based on the results shown in Figure 1 the concentration of 3% was used in all tests. The density and viscosity of the aqueous phase were practically equal to those of pure water; its surface tension was 0.044 N/m. The density of the kerosene was 860 kg/m^3, viscosity 0.00265 Pa·s, and surface tension 0.032 N/m. Only emulsions of the type O/W were obtained. Emulsions of the 7 and 35% kerosene contents were produced. The distance between the atomizer outlets varied in the range from 4 to 150 mm. The distance of 4 mm was the least one at which the normal work of the atomizers was possible; at smaller distances it was not possible to stabilize the outflow, probably because both outflowing streams disturbed each other. Two samples were taken from each run, from which six micrographs were taken using a microscope equipped with lens of 100-fold magnification and a digital camera. The digital photographs were analyzed using Microscan v.1.4 program. About 1,000 droplets were analyzed from each run. The mean droplet diameters were next calculated using a Shortver v. 0.9 program (Bobiński, 2002) and the droplet diameter distribution was evaluated. Four mean diameters: d_{10}, d_{21}, d_{32}, and d_{43}, defined by Eq. 1, were calculated

$$d_{jk} = \frac{\sum_i d_i^j}{\sum_i d_i^k} \qquad \text{for } j = k + 1,\ k = 0, 1, 2, 3 \tag{1}$$

It is clear that the manner in which the mean diameter is defined influences its value. The arithmetic mean value d_{10} is not sensitive to the presence of big droplets that, although not numerous, may constitute most of the volume of the disperse phase. Because the bigger droplets issued from the atomizer were rather scarce, the influence of their further dispersion by the impinging streams on the d_{10} was poorly noticeable. The mean values based on higher power of the diameter are influenced by bigger droplets to a greater extent. Thus, comparison of the mean values defined in different ways allows us to compare the degree of polydispersivity of various emulsions (Shimizu et al., 2001).

To compare the emulsions obtained by impinging streams with the emulsions produced by mechanical stirring, an emulsion containing 35 vol % of kerosene was produced using a homogenizer of the type Ultra Turrax® at the speed of 20,500 rpm for 10 min.

Results

Figure 2 presents the dependency of four mean diameters defined by Eq. 1 on the distance between the sprayer outlets for the emulsion of 7% kerosene contents. The results for the 35% emulsion were similar, so for the sake of clarity of the plot they are not included. It may be observed that the mean droplet diameter decreases when the distance diminishes, reaching a minimum at the distance of about 8 mm. That confirms the influence of the impingement of aerosol streams on the droplet size. The effect may be explained mainly as a result of the droplet disintegration through a rapid deceleration during the impingement with the countercurrent stream of gas, but it may also be attributed to the droplet collision. When the distance between the atomizers increases, the velocity in the impingement zone as well as the volumetric content of droplets in the aerosol decreases. For that reason the ability of droplets to break up is lower. The closer the sprayers, the higher the velocity in the zone of impingement and the higher are the inertial forces that deform the droplets, and also the ability of droplet collision is greater. On the other hand, at the distance between the atomizers comparable with the outlet diameter (3.6

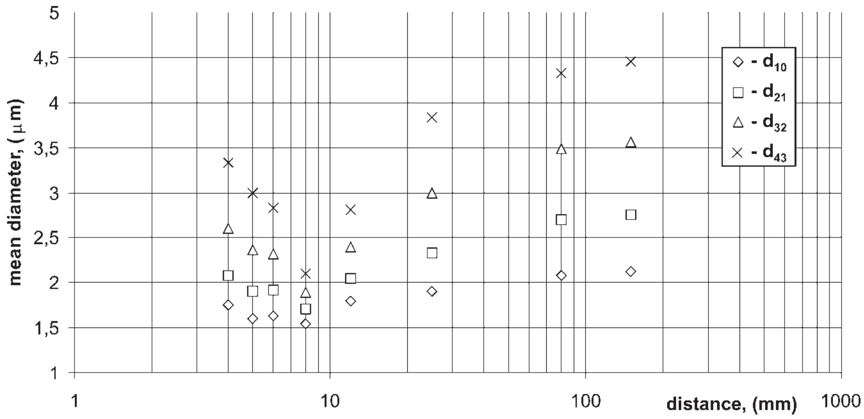

Figure 2. Dependency of the mean droplet diameter on the distance between the atomizers for the emulsion containing 7% of kerosene.

mm), instabilities in the gas outflow occur that may worsen the effect of impingement and the efficiency of droplet disintegration. One may also expect that at the smallest distances the intensity of microturbulence is the highest. That may lead to a higher frequency of collision of droplets moving in the same direction, which leads to coalescence of droplets and to the increase of their mean size.

It is obvious that the effect of droplet impingement with a stream of gas flowing toward it is more noticeable in the case of bigger droplets because of higher inertial forces during the impingement. The comparison of the arithmetic mean droplet diameter d_{10} with the other mean diameters defined using a higher power of the diameter (Figure 2) confirms that conclusion. The arithmetic mean d_{10} changes with the distance between the atomizers not more than 35% from the minimum to the maximum value, although other kinds of the mean value, which are more greatly influenced by the biggest droplets, change with the distance to a higher degree. This confirms that rather few droplets break up, although those are the larger ones.

The same conclusion follows from the diameter distribution for the optimum (8 mm) and maximum (150 mm) distance between atomizers (Figure 3). The plot presents the relative number of droplets in different diameter ranges Y defined according to Eq. 2 vs. diameter:

$$Y = \frac{n_i}{n\Delta d_i} \tag{2}$$

where n_i is the number of droplets in the diameter range Δd_i, n is the total number of droplets, and Δd_i is the width of diameter range.

It is observed in the plot that the emulsion produced at the optimum distance contains a smaller number of larger droplets and higher number of smaller droplets (diameter < 1.5 μm). It is characteristic and also noticeable in the case of 35% emulsions that the diameter distribution is bimodal: at about 2–2.3 μm there is a small second peak.

Figure 4 presents the results for the emulsion containing 35% of kerosene produced using a high-speed rotational homogenizer at 20,500 rpm. It can be seen that the emulsion from the impinging streams contains a higher number of smaller droplets < 1.7 μm and a smaller number of larger droplets $>$ 3 μm. The droplet size distribution in this emulsion is narrower. This result shows that the method of impinging streams enabled better atomization and led to more monodisperse emulsions than very intensive and long-lasting mechanical stirring.

All emulsions produced using impinging streams were very stable: the results of the microscopic measurements of droplet

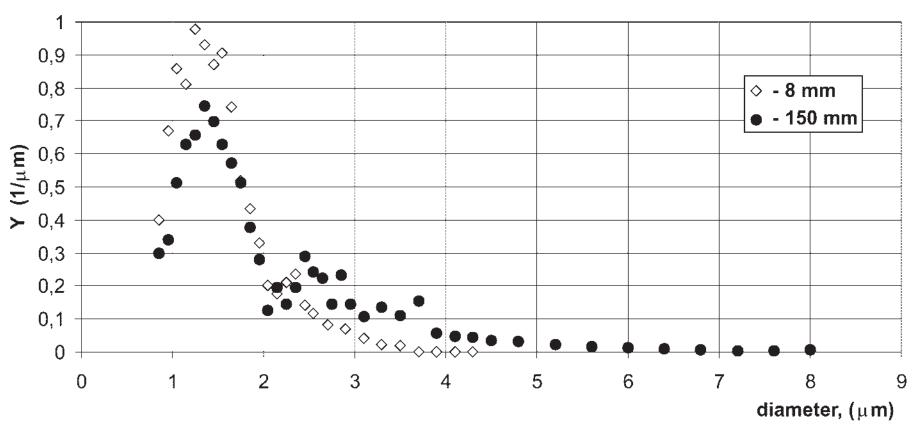

Figure 3. Droplet diameter distribution of the emulsion containing 7% of kerosene produced at distances between the atomizers equal to 8 and 150 mm.

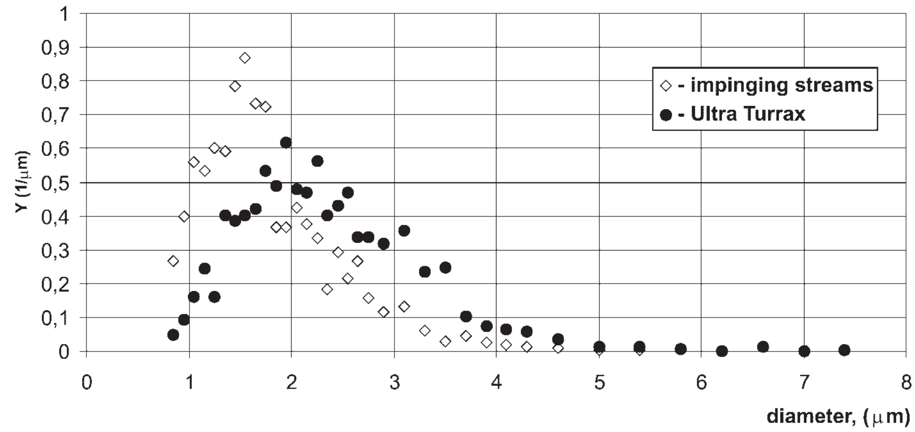

Figure 4. Droplet diameter distribution of the emulsions produced by impinging streams and a homogenizer of the type Ultra Turrax.

diameter as well as the rheological properties measured in a steady and oscillating flow did not change considerably over a period of 6 months. Both 7 and 35% emulsions were Newtonian liquids whose viscosity was only slightly higher than that of water. Thus, their viscosities would not be noticeably changed by the structure instability. Because flocculation and coalescence occur in the cream layer, which contains many more droplets than the bulk emulsion, the emulsion was concentrated to 70% by centrifuging and its flow curve during steady flow testing as well as the storage modulus G' during oscillatory testing were measured. Thus the concentrated emulsion was a shear-thinning liquid obeying the power law in the range of shear rate from 0.2 to 200 s^{-1}.

$$\tau = k\dot{\gamma}^n$$

where $n = 0.61$ and $k = 2.1$ Pa·s$^{0.61}$

The measure of elastoviscous properties, that is, the storage modulus, was practically constant in the range of frequency between 0.1 and 30 Hz and equal to about 100 Pa.

Conclusions

The results show that the impingement of aerosol streams may lead to the decrease of the mean droplet size and also render the emulsion more monodisperse, which is an advantage of the method. There is an optimum distance between the outlets of atomizers at which the best disintegration of droplets can be achieved. The method enabled even better atomization of droplets than a long-lasting, very intensive stirring using a rotational homogenizer.

Acknowledgments

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