MEASUREMENT OF LIQUID HOLD-UP OF THE DISPERSED PHASE BY AN ULTRASONIC METHOD

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Received July 14th, 1982

A new method has been proposed of the continuous detection of the volume hold-up of the dispersed phase in a liquid–liquid system. The method utilizes different velocities of sound in the organic and the water phase and has been tested on four liquid systems typical for liquid extraction. These experiments have shown the dependence of the velocity of ultrasound on the hold-up to be given by Eq. (7) as well as its independence on the size of the droplets forming the dispersion. Further the experiments have revealed no interaction between the effect of the hold-up and a third component dissolved in the liquids on the velocity of ultrasound in the dispersion. The net effect may thus be computed by the additivity rule. Thanks to its simplicity and reliability the method is suitable not only for laboratory measurements but also for monitoring hold-up in large scale industrial equipment.

Volume hold-up of the dispersed phase in liquid extraction is one of the most important quantities controlling the rate of the process. At present there are several methods at hand for continuous monitoring of the hold-up. The pressure method[1] detects the hydrostatic pressure head of the dispersion, which depends on the difference of densities of the continuous and the dispersed phase and the ratio of their volumes. The relative sensitivity of the method is given by

$$R = (\varrho_1 - \varrho_2)/\varrho_1 \,, \qquad (1)$$

and hence the accuracy of the method decreases with the decreasing difference of densities.

An instrument with a point electrode [2] has been devised to measure local hold-up in a system with the dispersed organic phase. A moving droplet upon contacting the electrode reduces the current in an electric circuit; the hold-up is then evaluated from the frequency and duration of changes of the current.

Optical methods are based on the absorption, reflection and refraction of light in the dispersion. The measured quantity is the extinction of a light beam passing through the dispersion. The extinction due to the absorption in the droplets is proportional to the hold-up. A color dye is usually added[3] in order to enhance absorption compared to the remaining optical phenomena. In contrast, extinction of the light beam due to the reflection and refraction on the droplets is proportional to the interfacial area[4]. Such an arrangement has been achieved by a perfectly parallel narrow beam of light impinging on a light sensitive detector. The refraction and reflection method is not well suited for the measurement of liquid hold-up.

An analog of the optical absorption method is the measurement of the intensity of γ-rays passing through the dispersion[5]. Yet another method utilizing radioactive rays is the measurement of the intensity of γ-rays emanated from the dispersion containing a dissolved radioactive isotope of iodine[6].

From the standpoint of measurement of liquid hold-up in industrial equipment, where the liquids contain various compounds and contaminants, the electric and optical methods are of little use due to the fouling of the probes associated with a hard to control distortion of the signal. The choice of the methods is also limited by the construction of the apparatus containing the dispersion. To give an example, in a vibrating-plate extractor, where the dispersion is mixed by vibrating perforated plates evenly spaced along the column shaft, the measurement may be disturbed by the motion of the plates. In this case the pressure method is not suitable either as the periodic changes of pressure brought about by the motion of the plates considerably exceed the monitored hydrostatic pressure. Also the measurement of electric quantities, dependent on the hold-up, may be disturbed by the motion of the metallic plates. An alternative method of measuring the hold-up is thus highly desirable.

The aim of this study has been to test the possibility of detecting liquid hold-up through the measurement of the velocity of ultrasound in the dispersion.

THEORETICAL

The arrangement of molecules is not compact. The sound spreads through the space between molecules considerably more slowly than through the molecule itself; the molecule is passed essentially instantaneously[7]. Accordingly, the velocity of ultrasound in a liquid may be expressed as a function of the ratio of the total volume of liquid and the free volume by

$$\mathscr{V} = (V/V_\mathrm{f})^{1/3}\,\mathscr{V}_\mathrm{g}\,, \tag{2}$$

where $\mathscr{V}_\mathrm{g}$ is the velocity of the sound in the gas. It is apparent that practical utilization of Eq. (2) encounters the problem of determinig the free volume V_f, similarly as the theory of Jacobson[8], who derived the following expression in terms of the intermolecular distance, L_f

$$\mathscr{V}_\mathrm{l} L_\mathrm{f} \varrho^{1/2} = K\,. \tag{3}$$

Eq. (3) is used in the study of the velocity of sound in pure liquids or solutions.

For the velocity of sound one can write generally the following relation

$$\mathscr{V} = 1/\sqrt{(\varrho\beta)}\,. \tag{4}$$

In heterogeneous dispersed liquid–liquid systems the time necessary for a sound wave to travel the distance L is determined by the velocities in individual liquid phases and the volume fractions of the water and the organic phase. One can write for the total time that

$$t = t_1 + t_2 \, . \tag{5}$$

The partial times may be expressed by means of the fractions of the corresponding distances and velocities

$$L/\mathscr{V} = LX/\mathscr{V}_1 + L(1 - X)/\mathscr{V}_2 \, . \tag{6}$$

A simple arrangement yields the following relationship for the calculation of the net velocity

$$\mathscr{V} = \mathscr{V}_1\mathscr{V}_2/[(1 - X)\,\mathscr{V}_1 + X\mathscr{V}_2] \, . \tag{7}$$

As follows from Eq. (*3*) the velocity of sound in liquids depends on temperature. The method of compensation of the thermal dependence has been described *e.g.* in ref.[9]. There are essentially two alternatives. The first employes the principle of differential measurement while the second compensates the temperature electronically on the basis of simultaneous measurement of the velocity of sound and temperature[10].

EXPERIMENTAL

The velocity of sound in the dispersion was measured by an ultrasonic analyser of solutions UAR 4, developed by the Research Institute of Mechanisation and Automation[10]. The analyser measures velocities of ultrasound between a transmitter and a receiver 53 mm apart with an accuracy of ± 1 m/s. The transmitter and the receiver, equally as an electronic thermometer, are located in a probe immersed in the liquid. The instrument permits linear temperature compensation of the mesaured velocity.

Materials. The liquids selected for measurements are common in liquid–liquid extraction. Apart from the one-component liquids we used also a solution of toluene in 1,2 dichlorethane with the density close to that of water (solution R 1) and aqueous solutions of ammonium sulphate at concentrations up to 470 g/l (solution R 2). The temperature dependence of the velocity of sound measured in single-phase liquids is shown in Table I. In the investigated range this dependence was found linear:

$$\mathscr{V} = \mathscr{V}_0 + C(T - T_0) \, . \tag{8}$$

The coefficient of temperature dependence, C, assumes positive values for water and negative values for organic liquids. In the dispersion, accordingly, the coefficient decreases from positive to negative values with increasing fraction of the organic phase and remains zero in a certain region of hold-ups.

Procedure. Table II shows two-phase systems in which the velocity of sound was measured. 1·2 litres of dispersion was placed in a cylindrical glass vessel 13 cm in diameter. Eccentrically suspended in the vessel were a four-blade impeller and the ultrasonic probe. The ultrasound signal passed vertically roughly through one half of the height of the dispersion avoiding thus regions near the bottom and the top of the mixing vessel. The speed of revolution of the impeller was adjusted by gradual increasing to a value where the hold-up appeared uniform along the height of the mixer. Further increase of the speed of revolution of the impeller had no effect on the velocity of ultrasound. This suggests that the signal is independent of the size of the droplets as the latter decreases with increasing speed of revolution. Near the bottom of the mixer the intensity of mixing was less intensive.

During dispergation of the heavier of the two liquids droplets clinging to the bottom were observed. The heavier phase had the tendency to accumulate here and its fraction in the liquid sensed by the probe was therefore less than the mean fraction in the whole mixer.

RESULTS

The velocities measured in systems 1–3 (Table II) in dependence on the fraction of the organic phase in the mixer are plotted in Fig. 1 together with the theoretical dependence (7). The first two systems exhibit deviation from Eq. (7) always toward the value of the velocity of the lighter phase. This is probably caused by the above described difference between the mean hold-up in the mixer and the hold-up in the region sensed by the probe. This hypothesis was confirmed by measurement in a system with almost equal densities of phases (Fig. 1*c*) where the hold-up was homogeneous in the whole mixer and no systematic deviation from the theory appeared.

TABLE I

Temperature dependence of the velocity of ultrasound $T_0 = 23$°C

Liquid	$\mathscr{V}_0$, m/s	C, m/s °C	Temperature range, °C
Water	1 480·2	4·23	20–28
Toluene	1 344·0	–5·56	20–28
1,2-dichlorethane	1 243·3	–5·40	20–30
R 1 Solution	1 273·3	–5·14	20–25

TABLE II

Dispersed systems

Reference number	Organic phase	Water phase	ϱ_1 kg/m^3	ϱ_2 kg/m^3	ϑ_1 m/s	ϑ_2 m/s	T °C
1	toluene	water	866	998	1 344·0	1 480·2	23·0
2	1,2-dichlorethane	water	1 246	998	1 243·3	1 480·2	23·0
3	R 1 solution	water	1 034	997	1 271·0	1 482·4	23·5
4	1,2-dichlorethane	R 2 solution 100 g/l	1 246	1 051	1 242·5	1 562·7	23·2

The validity of Eq. (7) was verified also by measuring the velocity of ultrasound in static liquids. In these static experiments a layer of the lighter phase was placed on top of the heavier layer and the hold-up was adjusted by positioning the horizontal interface between the transmitter and the receiver. The results of the static experiments are shown in Figs 1*a* and 1*b*.

System 4 was used (Table II) to test the effect of a dissolved salt on the velocity of ultrasound in the dispersion in the range of hold-ups corresponding to the conditions of extraction. For this system too the dependence on the hold-up is given by Eq. (7) as shown in Fig. 2. The effect of concentration of the salt was first investigated on

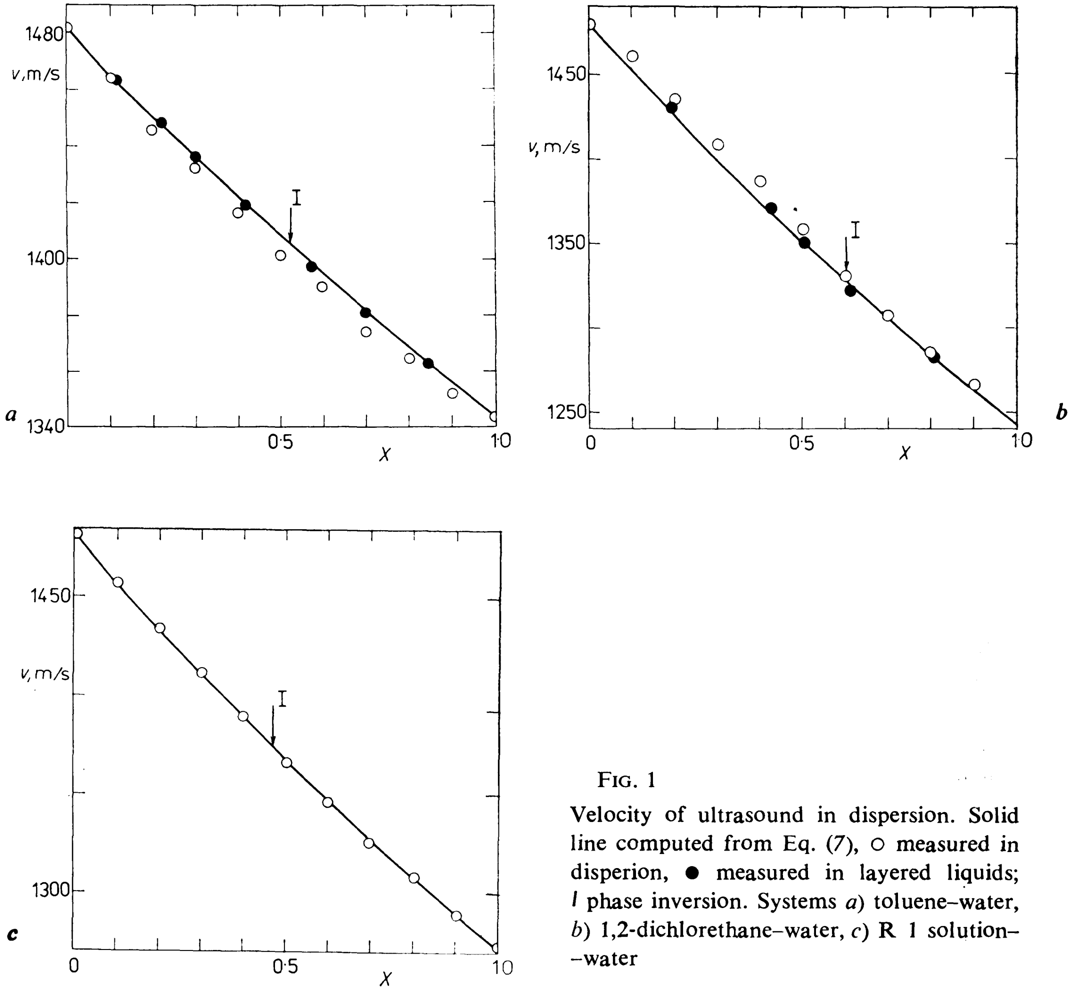

FIG. 1

Velocity of ultrasound in dispersion. Solid line computed from Eq. (7), ○ measured in disperion, ● measured in layered liquids; *I* phase inversion. Systems *a*) toluene–water, *b*) 1,2-dichlorethane–water, *c*) R 1 solution–water a single-phase system. Twenty experimental points were fitted by a calibration curve shown in Fig. 3:

$$\mathcal{V}_2(w) = 1\,485{\cdot}1 + 0{\cdot}777\,w\,, \quad 0 \leqq w < 167$$
$$\mathcal{V}_2(w) = 1\,496{\cdot}6 + 0{\cdot}708\,w\,, \quad 167 \leqq w \leqq 467\,. \qquad (9)$$

The deviations of the calibration measurements (not shown in Fig. 3) from the calibration curve are within experimental error. For the hold-up of the organic phase $X = 0{\cdot}3$ were than measured the velocities of ultrasound in the dispersion at five different concentractions of $(NH_4)_2SO_4$ as shown in Fig. 2. Then, using Eq. (7), corresponding velocities, $\mathcal{V}_2$, in the single-phase solutions were computed and shown in Fig. 3. The deviations of these velocities from the calibration curve are within experimental error.

These results indicate a simple additivity of the effect of changes of the hold-up and the changes of the concentration of the salt on the velocity of ultrasound. This permits compensation of the effect of concentration of the salt by means of an in advance found calibration relationship in single-phase liquid.

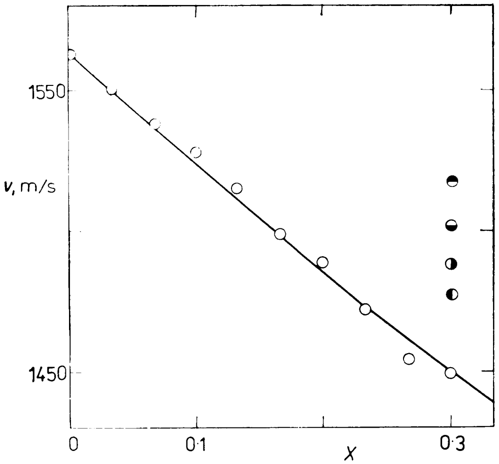

FIG. 2

Velocity of ultrasound in 1,2 dichl~~o~~ethane -aqueous solution of ammonium sulphate dispersion. Solid line computed from Eq. (7) for the concentration of 100 g/l. Concentration of the salt: ○ 100 g/l, ◐ 142·3 g/l, ◑ 183 g/l, ⊖ 222·7 g/l, ⊖ 261·7 g/l

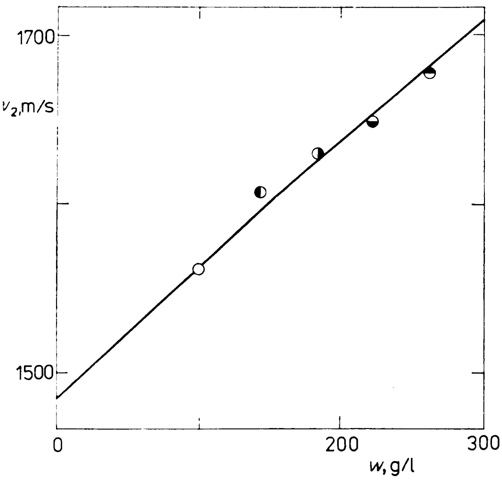

FIG. 3

Velocity of ultrasound in aqueous solution of ammonium sulphate at 23·2°C. Solid line is the calibration curve, Eq. (9). Same caption as for Fig. 2

/or O

CONCLUSION

The performed experiments indicate that the velocity of ultrasound in a dispersion depends on the velocities in individual phases forming the dispersion and their volume fractional presence, as expressed in Eq. (7). The velocity of ultrasound is thus independent of the droplet size and appears therefore suitable for the determination of hold-up of the dispersed phase in a two-phase liquid–liquid system.

It may be assumed that the ultrasound detection of the hold-up will be used primarily in industrial apparatuses as it appears less sensitive to disturbances (contaminants in liquids, deposits on the probe, fluctuation of intensity of the signal source, *etc.*) than the currently used methods.

The authors wish to thank Dr Z. Figura for the information about the measurement of the velocity of ultrasound in liquids and to the Institute of Mechanisation and Automation in Nové Mesto nad Váhom for providing the measuring technique.

LIST OF SYMBOLS

C	coefficient of temperature dependence, m/s°C
K	temperature dependent constant, $(\mathrm{kg\ m})^{1/2}/\mathrm{s}$
L_f	intermolecular free distance, m
R	relative sensitivity
t	time, s
T	temperature, °C
$\mathcal{V}$	velocity of ultrasound, m/s
V	volume of liquid, m^3
V_f	free intermolecular volume of liquid, m^3
w	concentration of salt, g/l of solution
X	volume fraction of organic phase in dispersion
β	adiabatic compressibility factor, $\mathrm{ms}^2/\mathrm{kg}$
ϱ	density, $\mathrm{kg/m}^3$

Subscripts

1	organic phase
2	water phase
o	reference value
g	gas
l	liquid

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Translated by V. Staněk.