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Experimental studies on the effect of ultrasonic waves on single drop liquid–liquid extraction

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ABSTRACT

The influence of ultrasonic waves on hydrodynamics and mass transfer of circulating drops in liquid–liquid extraction process was studied. The recommended chemical systems of toluene–acetic acid–water with mass transfer resistance mainly in the organic phase, and cumene–isobutyric acid–water in the aqueous phase were used. An extraction column, equipped with an ultrasonic emitter of 35.40 kHz real frequency and 0.37 mW/cm^2 intensity, was employed. The ultrasound properties were measured using the hydrophone standard method. Drops terminal velocity was comparable with the Grace model. In mass transfer study, significant enhancement was revealed in overall mass transfer coefficient for different drop sizes and for the both mass transfer directions by using ultrasonic waves. The average and maximum enhancements were, respectively, 20.8 and 31.7% for toluene–acetic acid–water, and 40.3 and 55.1% for cumene–isobutyric acid–water. Small drops exhibited a higher enhancement percentage. Regarding the mass transfer direction, the system of cumene–isobutyric acid–water with continuous to dispersed phase direction, was benefited more as the consequence of creating effective agitation in continuous phase than in dispersed phase.

1. Introduction

Liquid–liquid extraction, a prevalent separation method, has found many applications in different industries. The industrial extractors involve two major types of mixer-settlers and columns. Columns are most conventional equipments for higher inputs and for adjusting contact of phases by drops, which are produced by distributors and provide a wide contact area.

Up to now, the influence of several parameters and operating conditions have been studied on the hydrodynamics and mass transfer of drops in different extraction columns. Recent works include investigations on the influence of contaminants [1], aqueous phase pH [2], presence of salts [3], temperature [4], presence of nanoparticles both with single drop investigations [5–7] and swarm of drops [8,9] and nanoparticles in the presence of magnetic field [10]. However, among these investigations, the lack of a fascinating phenomenon in process intensifying and also a flexible alternative parameter for energy efficient processes by "ultrasonic waves", is still distinctive while it is attainable. Applying ultrasound waves, have been found efficient in diverse chemical, medical, biological and food industries [11,12].

Many phenomena may result from propagation of ultrasonic waves into a fluid and particularly into a liquid medium. The most important favorite impacts of ultrasound in mass and heat transfer are due to acoustic streaming and acoustic cavitation. Acoustic streaming arises further gradients in momentum by ultrasonic waves, and thereby the fluid currents will be more established. Acoustic cavitation, on the other hand, is a phenomenon that may arise from the dissipation of ultrasonic waves into a liquid. It is the formation, growth, oscillation and powerful collapse of gas bubbles into a liquid. Some experimental results and photographic studies show the impact of a collapsing cavitation bubbles could last 10^{-7} s and reaching a local pressure of up to 193 MPa. This cavitation explains many phenomena involved in chemistry, biology, engineering and so forth [13,14].

Ultrasound waves are often classified according to their frequency or power. The classification can be roughly expressed with respect to the application as following [14–16]:

- Low frequency ultrasound, between 20 and 100 kHz waves are defined as "power ultrasound" devoted to the study of high-intensity applications. Indeed, it is usually transferred at a high power level (a few tens of Watts). Power ultrasound finds uses in various processes like cleaning, plastic welding, sonochemistry, homogenization, extraction and so forth. It is also generally used for heat and mass transfer process intensification.
- Intermediate frequency range of 100 kHz–1 MHz, is less used than power ultrasound to promote transport phenomena.

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Nomenclature

c	viscometer constant
C	concentration
d	drop diameter (mm)
E	extraction fraction
Eö	Eötvös dimensionless number, $Eö = g\Delta\rho d^2/\gamma$
g	standard gravity (m/s^2)
H	dimensionless group defined by Grace et al. $H = (4/3)Eö\, M^{-0.149}(\mu_c/\mu_w)^{-0.14}$
k	dispersed and continuous phase local mass transfer coefficient (μm/s); viscometer constant
K_{od}	overall dispersed phase mass transfer coefficient (μm/s)
M	Morton dimensionless number, $M = g\mu_c^4\Delta\rho/\rho_c^2\gamma^3$
N_{PG}	inverse of Morton dimensionless number
Re	drop Reynolds number ($Re = \rho_c u_t d/\mu_c$)
T	temperature (°C)
t	drops contact time and efflux time in viscometer (s)
u_t	drops terminal velocity (m/s)
We	drop Weber number ($We = du_t^2\rho_c/\gamma$)

Greek Symbols

γ	interfacial tension (mN/m)
Δ	difference
μ	viscosity (mPa·s)
ρ	density (kg/m^3)

Subscripts

c	continuous phase
d	dispersed phase
f	final value
i	initial value
od	overall dispersed phase
w	water

Superscripts

$*$	equilibrium

- High frequency ultrasound of above 1 MHz, is considered as "low power ultrasound" (usually less than 10 W), does not affect the medium of propagation. Consequently, it is especially used for medical diagnosis or nondestructive material control.

In this regard, different kinds of reactors and contactors are equipped with ultrasound devices. Among the several types of sonicator systems, mostly bath and probe–types are used [15]. With respect to the desired uniform and diffusive vibration in fluids, a variety of investigations have been conducted on the subject of ultrasonic assisted mass transfer in gas–liquid, solid–liquid and liquid–liquid transfer cases.

In the work of Riera et al. [17], concerning on CO_2 supercritical extraction of oil from different substrates, it was reported that power ultrasound would enhance mass transfer as a result of producing small scale agitation. In their work a transducer working with frequency of about 20 kHz and power of 50 W, located inside a high-pressure extractor, caused an increase of 30 and 20% in the kinetic and the oil extraction yield, respectively. In addition, they introduced this technique as an inimitable applied manner for producing agitation, because exerting mechanical stirrers was not available in their set-up. In a similar work, the effect of operational parameters for extracting oil from adlay seed, in an ultrasound assisted supercritical fluid extraction was investigated by Hu et al. [18]. They reported that ultrasonic waves create vibration and an enhancement of about 14% was achieved in the extraction.

In gas–liquid operations, Asgharzadehahmadi et al. [19] employed a contactor equipped with an ultrasonic horn. The effect of ultrasonic intensity on volumetric gas–liquid mass transfer coefficient as a function of gas flow rate and temperature was investigated. It was reported that during augmentation of ultrasonic power, mass transfer coefficient was increased and reached a maximum value with 320–360 W power; however, further increase was led to a decrease in mass transfer as a result of uncontrolled temperature rise (caused by high ultrasonic dissipation) and gas hold up in the system.

In the field of solid–liquid operation, Jiao et al. [20] presented a new computational quantitative model for enhancing mass transfer coefficient in the presence of ultrasound in a sonochemical reactor working with the frequencies within 40–100 kHz and power range of 10–50 W. They studied the impact of more effective parameters of ultrasonic power and frequency as well as other parameters including temperature, transducer diameter and distance between reactor and transducer (filled with fresh water). A piece of solid silicon was placed in KOH solution where the reaction between them was performed. The results indicate that mass transfer coefficient on silicon surface in the reactor with ultrasound was higher and mass transfer coefficient increased with increasing the ultrasound power. However, mass transfer coefficient did not increase steadily with the frequency. In the condition of ultrasound power of 50 W, the maximum enhancement was at 60 kHz and the minimum at 80 kHz.

Concerning liquid–liquid contact, recent applications are mainly concerned on dispersion of a liquid phase small droplets in another phase under ultrasound assistance until the initial heterogeneous liquid–liquid system is made uniform. This process is known as "homogenization" or "emulsification" and is a well-documented process in both the analytical and industrial fields. However, mass transfer between two immiscible phases is arguable if one considers the ability of this form of energy to form stable emulsion. Probably for this reason, analytical chemists have been reluctant to test ultrasound as a means for improving liquid–liquid extraction (LLE). In fact, ultrasound application may cause stable emulsions that results in long times phase separation, generally non-favorable in practice [21].Despite these attempts, no investigation has been made on applying ultrasound in liquid–liquid extraction columns. The device can provide a desired vibration and agitation in a column for mass transfer intensification. The aim of this study was therefore to investigate the hydrodynamic and mass transfer of different single drops in a liquid–liquid extraction column equipped with low intensity ultrasonic irradiation. Two chemical systems of toluene–acetic acid–water and cumene–isobutyric acid–water were used for this aim. Both systems are well-known and recommended for liquid–liquid extraction investigations. The most prominent difference of these chemical systems is the slope of equilibrium curves. In the former system with a rather high interfacial tension, mass transfer resistance exists mainly in the organic dispersed phase ($K_{od} \approx k_d$), whereas in the latter system with medium interfacial tension, there is mass transfer resistance mainly in the aqueous continuous phase ($K_{od} \approx k_c$).

2. Experimental

2.1. Chemicals

To perform experiments, the used chemicals were: toluene (mass purity > 99.9%), acetic acid (> 99.9%), isobutyric acid (> 99.5%), cumene (> 99.5); all of them from Merck Company. Fresh deionized water (with conductivity of 0.08 μS/cm) were obtained from a