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Two-stage ultrasonic irradiation for dehydration and desalting of crude oil: A novel method

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ABSTRACT

In this study, a new methodology is proposed based on using two periods of ultrasonication for the dehydration and desalting of heavy crude oil. For this purpose, ultrasonic irradiations are used under specified conditions such as irradiation input power, irradiation time and gap time after irradiation within a batch standing-wave resonator reactor. A serious problem with the commonly used single stage operation is that the enlarged water drops containing soluble salts in oil need to be settled for at least 60 min. To reduce this time, we proposed "two-stage ultrasonic irradiation at the equal irradiation times". This methodology proved to be an efficient way with the least settling time required. The results showed that the ultrasonic irradiations with irradiation input powers of 75 W for primary irradiation and 50 W for secondary irradiation (75–50 W) at the equal irradiation times of 45 s considerably decreased the settling time to 5(×2) min. Moreover, dehydration rate was more than 96% and also the final salt content was up to 2.56 PTB, which can meet the need of refineries. Micrographs obtained also confirmed efficiency of the methodology which caused the water drop size to increase by 40% after each step of the process.

1. Introduction

Up to recently, ultrasonic irradiation was regarded as an efficient method in dehydration and desalting of heavy crude oil. This method was first used in refinery oil dehydration by Yu et al. [1] to remove water in crude oil emulsion, which is mostly made by deliberate injection of water into the crude oil in order to dissolve soluble salts especially NaCl. Their experimental study on the influence of sound field parameters on the water in crude oil emulsion behavior indicated that the efficient response of the dispersed water phase to ultrasonic irradiation to drive, coalesce, and segregate was similar to the behavior of suspended particle [2,3], droplet [4], and bubbles [5,6] in sound field. If highly powered ultrasound is applied to an emulsion at low frequencies (<30 kHz), it can cause to split an emulsion into its component aqueous and oil phases [7,8]. A one-dimensional sound field that is imposed on water in oil emulsion can push the water drops into the parallel bands of high pressure sound waves and can eventually organize them on pressure nodes because the density of water is bigger than that of the crude oil [9–11]. Hence, the acoustic field will produce water drop-free and water drop-rich streams in the emulsion. In this situation, demulsification will happen when the water drops in oil are collapsed and coalesced by ultrasound waves [11].

Although other studies [12] investigated the effect of ultrasonic waves on the crude oil emulsions separation acceleration, they could not solve the problem of stability of crude oil emulsions due to presence of emulsifying agents (e.g. asphaltenes, resinous substances, oilsoluble organic acids) [13]. In fact, these agents, which are found in heavy crude oil more than light one, usually occur as a film on the surface of the dispersed droplets of water dissolving salts in crude oil [14]; therefore, they can reduce probability of coalescing droplets to form larger ones. The influence of ultrasonic irradiation to demulsify heavy crude oil emulsion was considerably improved by Cai et al. [15] when they applied an ultrasonic standing wave field to dehydrate crude oil. In this known field, the acoustic path length must be equal to odd times of the ultrasonic half wavelength and accordingly all of the reflection waves have the same frequency and amplitude as the radiating. As a result, a resonant acoustic field was created in which demulsification really occurred efficiently. However, a long time was needed as settling time to settle the enlarged droplets. Ye et al. reported the demulsification process under irradiation of resonant ultrasonic standing wave as a pretreatment process [10,16,17]. Although Ye et al. [10,17] achieved some successes in their experiments, the operating conditions applied were not acceptable to refineries for two reasons: firstly, the temperature of current pretreatment of crude oil are usually 130–140 °C in refinery [17], but the different temperatures

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Nomenclature

English letters

x	distance from a pressure antinode of a standing wave (m)
t	time (s)
f	frequency (s^{-1})
$F_{1,ac}$	primary acoustic force (N)
$F_{2,ac}$	secondary acoustic force (N)
$F_{net(g-b)}$	net effect of gravity (N)
$F_{v,s}$	viscous and surface tension forces (N)
a	drop radius (m)
E_{ac}	average energy density of the acoustic field (J/m^3)
F	acoustic contrast factor (dimensionless)
g	gravitational constant (m/s^2)
$\boldsymbol{U}t$	Stokes velocity (m/s)
V	volume of drop (m^3)
r	center-to-center distance between the drops (m)
E_{ir}	irradiated input energy density (J/m^3)

Greek letters

γ_f	compressibility of continuous phase (m^2/N)
γ_d	compressibility of drop phase (m^2/N)
k	wave number of the driving acoustic pressure (m^{-1})
$\Delta\rho$	density difference between the continuous and dispersed phases (kg/m^3)
μ	viscosity of the continuous phase (kg/m s)

tested by them were below the boiling point of crude oil (below 80 °C [17]). When the used temperature in ultrasonically dehydration and desalting process was less than the optimum range, crude oil emulsion viscosity would be highly increased. In that situation, an efficient acoustic field could not be established. In fact, as the viscosity increases, the attenuation of the propagating sound pressure increases; as a result, the intensity of the sound which reaches the interface is reduced. Hence, a large fraction of energy gets dissipated as viscous dissipation [18]. Secondly, as a physical method, gravity settling, which would be applied after ultrasonic irradiation, requires large residence times (settling time of 90 min [10,16,17]) and large physical spaces. In order to be sure that the commonly used ultrasonic irradiation methods were really unable to meet the needed settling time of refineries, Check and Mowla [11] tried to keep the main parameters including ultrasonic irradiation and also operating parameters in optimal values. Hence they designed a batch standing-wave resonator reactor as desalter/dehydrator, in which crude oil was subjected to ultrasonic irradiation at temperature of 100 °C (above 80 °C). But finally they managed decrease settling time only to 60 min.

In the present study, two-stage ultrasonic irradiation technique was proposed to investigate the ability of the second period of ultrasonication to decrease settling time and also further coalescence in the ultrasonically dehydration and desalting of heavy crude oil. For this purpose, the new methodology of "two-stage ultrasonic irradiation at the equal irradiation times" was experimentally developed. Also this methodology was optimized to find its optimal values including the irradiation values (input power and time) and settling time.

2. Theoretical background

In a standing wave field, when ultrasonic irradiation is applied to water droplets contained with soluble salts in crude oil, at one time, the drops migrate toward the pressure nodes of the standing wave field under the influence of the primary acoustic force [3,9]. It is given by:

$$F_{1,ac} = 4\pi a^3 k E_{ac} F \sin(2kx) \tag{1}$$

where, a is drop radius (m), κ is the wave number (m^{-1}), E_{ac} is energy density of the acoustic field (J/m^3), x is the position relative to a pressure node (m), and F, the acoustic contrast factor (dimensionless), quantifies the density and compressibility difference between the two phases. It has a positive value for water droplets dispersed in oil.

Since the density of the droplet phase is more than that of the continuous phase (crude oil), the net effect of gravity–buoyant forces ($F_{net(g-b)}$) will cause droplets to move downward, in direction of gravity. However the lift forces that are due to droplet rotation and shear forces can be neglected when a droplet is falling in a stagnant fluid [19].

The magnitude of the above mentioned body forces ($F_{1,ac}$ and $F_{net(g-b)}$), are much more than the surface tension forces ($F_{v,s}$) which resist the drops to move freely. So, the comparison of them derives an inequality which demonstrates the relatively fast movement of drops to the collection plane (see Eq. (2)).

$$\frac{F_{1,ac} + F_{net(g-b)}}{F_{v,s}} \gg 1 \tag{2}$$

As a reasonable assumption, droplets take only a few seconds to migrate to the collection plane for typical values of acoustic energy density, frequency and droplet sizes [20]. Then coalescence occurs when droplets located within the collection plane interact due to the secondary acoustic force. This force acts along the line of centers of the two drops and, since it increases in magnitude as the drops approach, the secondary acoustic force is expected to contribute to coalescence [9,21]. It is given by:

$$F_{2,ac} = \frac{k^2 E_{ac}}{2\pi}\left(1 - \frac{\gamma_d}{\gamma_f}\right)^2 \frac{V_2 V_1}{r^2} \tag{3}$$

where V_i is the volume of drop i ($i = 1, 2$), r is the center-to-center distance between the drops and γ_d and γ_f are compressibility of droplet phase and continuous phase respectively.

Gravitational separation of the enlarged drops then follows as governed by the Stokes' equation:

$$U_t = \frac{2r^2 \Delta\rho g}{9\mu} \tag{4}$$

where $\boldsymbol{U}_t$ is terminal velocity (settling rate) of drops, r is radius of drop, $\Delta\rho$ is density difference between the continuous (crude oil) and dispersed (water contained with soluble salts) phases, g is gravitational constant and μ is viscosity of the external phase (crude oil). Therefore, from Eq. (4), it is clear that the intensification of settling results from increasing the drop size, increasing the density difference between the phases and decreasing the continuous phase viscosity. The density difference and viscosity of the phases can be controlled by diluents and temperature respectively [22], whereas the drop size can be controlled by applied ultrasonic standing wave field. It is, therefore, the applied ultrasonic standing wave field that greatly controls the salt/water removal efficiency. Additionally, it also could be realized from Eqs. (1) and (3), the dependency of the primary and secondary acoustic forces with E_{ac} for coalescing. The energy density is the product of irradiation input power and irradiation time per unit volume of the acoustic field [11]. Although increasing the energy density is expected to enhance the rate of coalescence, in practice the application of a high energy density to an emulsion is also expected to enhance the probability of occurrence of cavitation [21] and acoustic streaming within the chamber containing the emulsion. Cavitation is collapse of coalesced water drops in crude oil due to rapid significant change in pressure, while