

Liquid-liquid Phase Separation in Batch Settling with Inclined Plate

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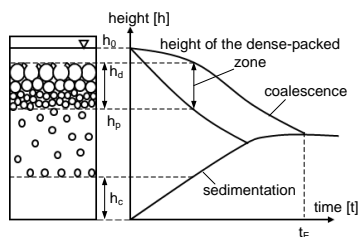
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Graphical abstract



Abstract

Liquid-liquid dispersions which are occurring in the petrochemical and chemical industry are commonly separated in gravity settlers. To improve the settler design, phase separation of the dispersions was studied. The parameters having the greatest effect on the phase separation under gravity are drop size, drop-size distribution, as well as driving-force parameters for sedimentation and coalescence, namely density difference, viscosity and interfacial tension. The effect of the driving-force parameters on coalescence and sedimentation was characterized experimentally using a standardized batch-settling cell. The viscosity of the aqueous phase was increased by adding polyethylene glycol. This not only changed the viscosity but also the interfacial tension, having a significant impact on drop size. In high-viscosity systems the settling speed of swarm droplets will significantly slow down and droplet size will be smaller. Furthermore, internals in gravity settlers also have significant impact on the phase separation. The influence of internals was studied for inclined plates. It can be seen from the experiments that internals speed up coalescence for different viscosities and volume ratios between the phases up to a factor of two.

Keywords: Liquid-liquid separation; sedimentation; coalescence; internal; viscosity

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1.0 INTRODUCTION

Liquid-liquid extraction is a separation method applied in chemical industries particularly for temperature-sensitive components. It can be used to purify a desired product without heat consumption or substance transformation. Liquid-liquid extraction makes use of two liquid phases usually, an organic and an aqueous phase which are immiscible or partially immiscible. One option for phase separation is the use of mixer-settlers.

Henschke [1, 6] studied the behavior of sedimentation and coalescence in batch settling experiments in order to describe two-phase separation and finally improved the design of settlers. The sedimentation and coalescence over time can be described by the sedimentation and the coalescence curves as illustrated schematically in Figure 1.

After mixing, a disperse and a continuous phase exist in the sedimentation zone. The disperse phase starts to sediment and subsequently to coalesce. Coalescence can be subdivided into drop-drop coalescence and drop-interface coalescence. The latter occurs when droplets join with their own phase. If disperse droplets sediment faster than they coalesce, a close-packed layer will occur. The separation process finishes at the so-called settling time t_E .

There are various research activities on the effective properties influencing the settling behavior. One important parameter significantly affecting settling is the system viscosity. It

can be seen from the result of Anursan, *et al.* [3] that in highly-viscous systems the separation efficiency is reduced due to the increasing settling time. It is also known that packings inside the settler affect the settling behavior and hence the settling time [5, 7]. In this study the influence of viscosity in batch-settling cells with internals was investigated. Inclined plates were chosen as the internals. The settling times of the viscous system with and without inclined plates are compared and discussed in the following.

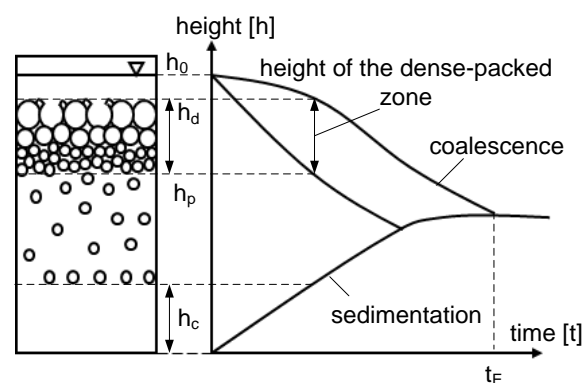


Figure 1 Schematic representation of a settling experiment according to Henschke [1]

2.0 EXPERIMENTAL

Analytical grade cyclohexanone was obtained from Carl Roth GmbH, Germany. Ultrapure water was used as aqueous phase using the same batch in all experiments. The viscosity of the aqueous phase was increased by adding Polyethylene glycol (PEG 4000, batch-production number:12H220008) from VWR BDH Prolabo, Germany.

The experiments were carried out in a standardized settling cell [1, 5-6], which is shown in Figure 2.

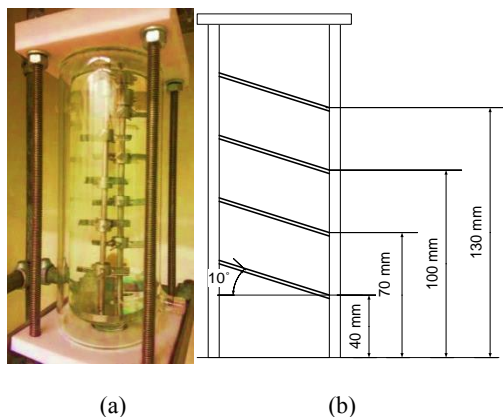


Figure 2 Settling apparatus: (a) stirred vessel, (b) design of inclined plate in settling cell

The equipment consists of two parts. The upper part is a stirred vessel used for mixing a saturated cyclohexanone phase and an aqueous phase at 800 rpm for 30 seconds. The lower part is a settling cell. It is connected to the stirred vessel by a drain ball valve. The inclined plates are made of stainless steel with a thickness of 2 mm and a diameter of 78 mm. The 10° inclined plates were placed in the settling cell in order to study the influence of internals on settling time (see Figure 2b). The viscosity of the aqueous phase was increased by adding PEG4000 in different concentrations: 4%wt, 13%wt and 19%wt. The viscosities of the aqueous phase at these concentrations were 2.07 mPa.s, 4.87 mPa.s and 9.78 mPa.s respectively. A video-recording camera was installed to observe the changing system behavior.

3.0 RESULTS AND DISCUSSION

The systems of cyclohexanone + water were investigated first without any additional PEG at different phase ratios. In Figure 3, the results are compared with data from Pattarawut [2] and Hülswitt [5] at the same temperature. By changing the phase ratio, the settling time was slightly changed but not the general settling behavior of the system. For phase ratios of organic to aqueous (o/a) = 1/2 to 1/5, cyclohexanone is the disperse phase. For the ratio o/a = 2/1 a phase inversion occurs. Henschke [1] found the same behavior for the cyclohexanone systems.

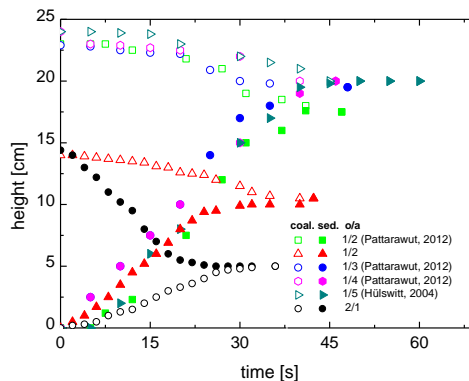


Figure 3 Comparison of experimental data of cyclohexanone + water at different phase ratios at 20°C

Then the viscosity was varied by adding PEG 4000. The sedimentation and coalescence curves of cyclohexanone + water with PEG 4000 for different viscosities of the aqueous phase are plotted in Figure 4. The settling time was found to be dependent on the viscosity of the aqueous phase. Similar observations were made even at the phase ratio o/a = 2/1 when the organic was the disperse phase.

The modeling of sedimentation for drop swarms developed by Henschke and Henschke, *et al.* [1, 6] has been applied successfully for the settling cell in the past, and thus it was tested here. The swarm sedimentation velocity (v_s) of the droplets can be determined from the slope of the sedimentation curve which is a linear. From this, the sauter mean diameter (d_{32}) can then be calculated.

$$d_{32,0} = \frac{Re_s \eta_c (1 - \varepsilon_0)}{\rho_c v_s} \quad (1)$$

The Reynolds number for sedimentation can be calculated from,

$$Re_s = \frac{3q\varepsilon_0}{c_w \zeta (1 - \varepsilon_0)} \left[\left(1 + Ar \frac{c_w \zeta (1 - \varepsilon_0)}{54q^2 \varepsilon_0^2} \right)^{0.5} - 1 \right] \quad (2)$$

with Archimedes number

$$Ar = \frac{\rho_c \Delta \rho g d_{32,0}^3}{\eta_c^2} \quad (3)$$

the parameter ζ in Equation 2,

$$\zeta = 5K_{HR}^{-3/2} \left(\frac{\varepsilon_0}{1 - \varepsilon_0} \right)^{0.45} \quad (4)$$

and the friction coefficient

$$c_w = \frac{Ar}{6Re_\infty^2} - \frac{3}{K_{HR} Re_\infty} \quad (5)$$

The Hadamard–Rybczynski factor is

$$K_{HR} = \frac{3(\eta_c + \eta_d)}{2\eta_c + 3\eta_d} \quad (6)$$

The parameter q in Equation 2, which depends on the disperse phase volume fraction, can be calculated from,

$$q = \frac{1 - \varepsilon_0}{2\varepsilon_0 K_{HR}} \exp\left(\frac{2.5\varepsilon_0}{1 - 0.61\varepsilon_0}\right) \quad (7)$$

and finally the Reynolds number in an infinitely extended fluid is

$$Re_x = 9.72 \left[(1 + 0.01Ar)^{4/7} - 1 \right] \quad (8)$$

This is modeled with appropriate models accounting especially for local hold-up [1, 6]. Then the model of Henschke and Henschke, *et al.* [1, 6] was used to evaluate the data which required the physical system properties to be measured.

The results showed that as expected when the system viscosity was increased, the drop size (d) decreased. Moreover, the increased viscosity leads to an increased lag time at the beginning of the experiment. At PEG 4000 concentration of more than 20 %wt. the system is opaque. This makes it difficult to use any optical evaluation technique to identify the sedimentation zone.

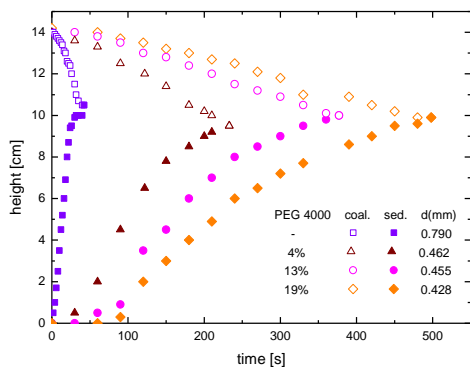


Figure 4 Comparison of coalescence and sedimentation curves of cyclohexanone+water with $o/a = 1/2$ at different concentrations of PEG4000

For improving the separation efficiency and studying the influence of internals, the inclined plates were placed in the settling cell. The system viscosity was increased by adding 4%wt. PEG4000. The experimental results are shown in Figure 5 in which each individual settling and coalescence contour between the plates is indicated. It can be observed that sedimentation and coalescence occur between the plates as well as at the major interface. The inclined plates force the droplets to coalesce before moving on in the direction of their mother liquid. This behavior also leads to holdup changes between every plate. At phase ratio 2/1 experiments show similar results when the organic was the disperse phase.

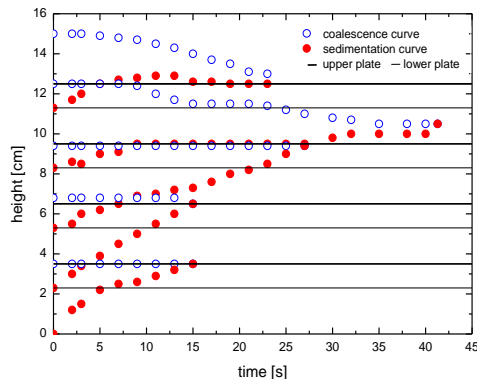


Figure 5 Experimental variations in the heights of sedimenting and coalescing interfaces with 4 inclined plates: the system of cyclohexanone+water +4%wt PEG 4000, $o/a = 1/2$

In order to investigate the influence of the distance between the plates, it was varied to 25, 30 and 40 mm as shown in Figure 6. To compare the efficiency of the varied plate distances a normalized settling time $\tau = t_{E,im}/t_E$ is utilized, which is defined as the ratio of the settling times with and without plates, minimizing the system-specific influence on coalescence e.g. induced by trace impurities. The results show that the settling time is increasing with plate distance, which means that the separation efficiency is reduced. The inclined plates speed up coalescence for different viscosities and volume ratios between the phases up to a factor of two.

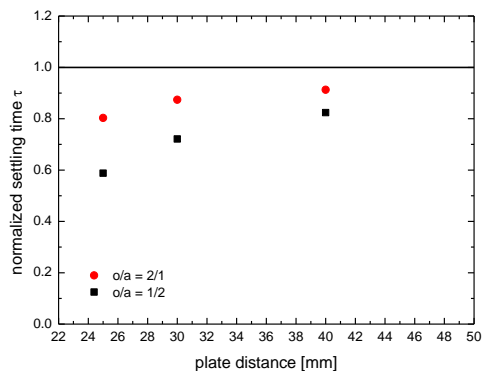


Figure 6 Variation of the normalized settling time of cyclohexanone + water + 4%wt PEG 4000 with plate distance

4.0 CONCLUSION

The phase ratio of the organic and aqueous phase is one parameter which affects the direction of phase dispersion. For high ratios the aqueous phase will be disperse, while for low ratios the organic phase will be disperse. Higher-viscosity systems have longer settling and dead times than lower-viscosity systems. Higher viscosity systems lead to smaller droplet sizes. When comparing viscous system separation with and without internal plates, it was found that internal plates enhance the separation significantly. They speed up the coalescence of the viscous system by reducing the settling time up to a factor of two. The evaluation shows that the model of Henschke is valid to describe the settling and coalescence

behavior also for viscous systems as a basis for knowledge-based settler design.

Greek letters

ε	volume fraction of dispersed phase
η	viscosity, Pa·s
ξ	Dimensionless group defined by Henschke [1] (Equations 2 and 4)
ρ	density, kg/m ³
Δ	difference

Nomenclature

Ar	Archimedes number
c_w	friction coefficient
d_{32}	Sautermean diameter, m
h	height, m
H	Hamaker coefficient, N·m
K_{HR}	Hadamard–Rybczynski factor
q	Dimensionless group defined by Henschke [1] (Equations 2 and 7)
Re	Reynolds number
t	time, s
v	velocity, m/s

Subscripts

0	initial value (after mixing is stopped)
∞	in infinite extended fluid

a	aqueous phase
c	continuous phase
d	dispersed phase
E	end
i	at the interface
p	dense-packed zone
s	sedimentation

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