

**Research note**

## **Experimental Study of Phase Inversion in n-Butylacetate-Water, Kerosene-Water, and n-Heptane-Water Systems in a Rotating Disc Contactor Column**

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### **Abstract**

*An experimental study of the phase inversion phenomenon was carried out in a rotating disc contactor (9.3 cm diameter). The effect of energy input via agitation and physical properties of the liquids upon inversion holdup and also delay time (time of inversion) was investigated at the constant input flow rate of the dispersed phase under no mass transfer conditions. Water was chosen as the continuous phase, and it was kept stagnant inside the column. The critical dispersed phase holdup was found to be decreased by increasing the energy input via agitation. It was observed that the lower both the interfacial tension and the density difference of two phases, the greater the resistance of the system to inversion. Moreover, a reduction in delay time was observed by increasing the energy input via agitation.*

**Keywords:** *Phase Inversion, Rotating Disc Contactor (RDC), Holdup, Delay Time*

### **1. Introduction**

Liquid-Liquid extractions are used in many different applications from chemical production to environmental clean-up. Dispersion of immiscible liquids is indispensable to liquid-liquid extractions.

A dispersed phase with the maximum holdup is required to maximize the productivity in liquid-liquid extractions, although the holdup can be increased up to a critical value, beyond which phase inversion occurs. Phase inversion in liquid-liquid dispersions is the phenomenon in which the dispersed phase becomes continuous phase and vice versa

under conditions determined by system properties, phase ratio and energy input via agitation [1-5].

The change of the continuous phase will lead to a system with different properties (e.g. rheology, drop size) which can be desirable in some cases like production of margarine, but in other cases is unwanted, for example in exothermic aromatic nitrations, phase inversion can cause a sudden increase in the reaction rate and produced heat [2]. Whether desired or undesired, control over the phase inversion process is essential for successful and profitable manufacturing.

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Phase inversion may occur whenever the equilibrium between drop coalescence and redispersion (breakage) in an agitated two phase system shifts towards coalescence [6,7]. Even though coalescence is not well understood, it is clear that with increasing the dispersed phase holdup, generally there is greater coalescence and eventually drops grow at a faster rate than they break [8,9]. This condition is considered to cause phase inversion, for example, with increasing amounts of oil, an oil-in-water (O/W) dispersion becomes unstable and spontaneously changes into a water-in-oil (W/O) one.

If for any reason the velocity of the continuous phase is increased, this will increase the drag force on the droplets and will cause the smaller droplets to rise (or fall) more slowly. As the continuous phase velocity is increased further, a point is reached where a significant number of droplets stop rising (or falling), forming a dense region which, eventually, as a result of drop coalescence, forms a second interface in the column; which is an inoperable condition [10].

The same phenomenon can be caused at constant continuous phase velocity by inducing the formation of smaller droplets through increasing input energy via agitation or change of the dispersed phase distributor. These smaller droplets can no longer overcome the drag force and holdup, thus inducing flooding which corresponds to phase inversion [11]. Although phase inversion is a spontaneous phenomenon, it was found that when a small amount of the dispersed phase (sufficient to cause inversion eventually) is added to a system already

close to inversion, phase inversion occurs a significant time after the addition. The delay time (time of inversion) varies from 5 to 1500s depending on a wide range of parameters [12].

Due to the fact that a large amount of liquid-liquid dispersions is produced within stirred vessels, the majority of phase inversion studies have been largely focussed on agitated systems [3,4]. As yet, phase inversion studies in liquid-liquid contactors have been very limited.

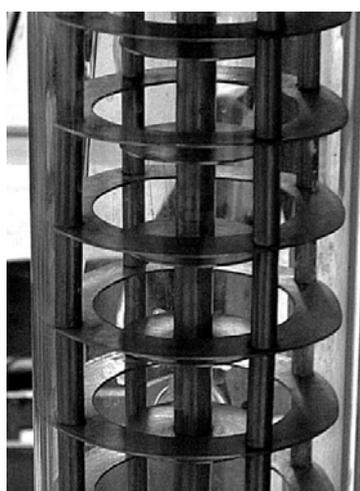
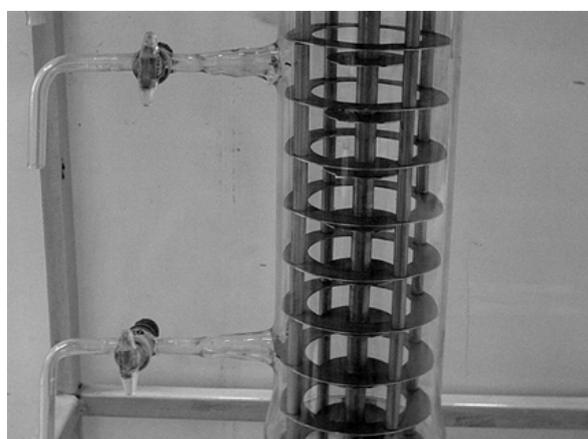
The aim of this study is to investigate the effect of energy input via agitation and also the effect of physical properties on phase inversion phenomenon in a rotating disc contactor.

## **2. Experimental**

### **2-1. Apparatus**

A pilot plant RDC, 9.3 cm diameter, liquid-liquid extraction column was used in this study. The main body of the rotating disc contactor, all inlet and outlet valves and also the connective lines were made of Pyrex glass to resist any corrosive chemicals. The column interior which was manufactured from stainless steel consisted of 30 compartments. The dispersed phase reservoir provided with adjustable rotary valve, was located on a plate at the upper part of the supporting metallic frame of the column. The continuous phase was stagnant inside the column for all runs. The organic dispersed phase entered from the lower part of the column through a single glass nozzle of 0.4 millimeter internal diameter. The nozzle was gradually declined from its vertical position, 10 millimeter lower than the first stator ring to provide a better distribution of inlet drops

to the column. The tip of the nozzle, which was the place of drop formation, was made completely smooth and flat. The discharge valves were located at equal distances of column height. Exactly every five rotor discs or every five stator rings were situated between two successive discharge valves. The view of the column is shown in Fig. 1. Two aqueous phase discharge valves were located at the lower part of the column. The main dimensions of the column interiors are given in Table. 1.



**Figure 1.** View of the experimental RDC column used for this work.

**Table 1.** The main dimensions of the RDC column

Item	Symbol	Dimensions (cm)
Column Diameter	$D_C$	9.3
Stator Ring Diameter	$D_S$	6.27
Disc Diameter	$D$	4.6
Compartment Height	$h_C$	3
Column Working Height	$Z$	100

## 2-2. Liquid-Liquid systems

The chemicals n-butylacetate, n-heptane and kerosene were obtained from Merck with purities of more than 99% and were used as received without further purification. Distilled water was prepared in our laboratory and used throughout all experiments as continuous phase.

Densities were measured by a picnometer of 25 milliliters volume and repeated three times for each phase and then averaged. Viscosities were determined by the Cannon-Fenske Viscometer, Petrotest Instrument, with the use of a stopwatch of 0.01 seconds precision. Measurements for each phase were made three times, and then averaged. Interfacial tensions were measured by a digital tensiometer, Krüss K10 T, Wilhelmy plate. Because of the great sensitivity regarding this method, measurements were made 10 times, and then the appropriate quantity that had been observed was taken as correct value [13].

A listing of used liquid-liquid systems and the accompanying physical properties is shown in Table 2.

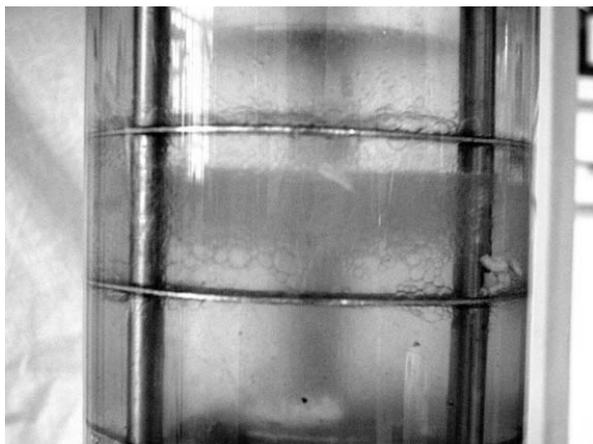
**Table 2.** Physical properties of the used liquid-liquid systems at P= 1 bar and 298.2 K

System	Aqueous phase		Organic phase		$\sigma(mN m^{-1})$
	$\mu(mPa s)$	$\rho(Kg m^{-3})$	$\mu(mPa s)$	$\rho(Kg m^{-3})$	
n-butylacetate/water	0.980	998.67	0.732	881.72	14.1
kerosene/water	1.010	998.14	1.684	810.31	32.2
n-heptane/water	0.958	996.80	0.393	683.19	50.1

### 2-3. Experimental Procedure

In order to fulfill the overall experimental objective, many runs were carried out to study the effect of energy input (agitation level or rotor speed) and physical properties of the liquids on phase inversion. Additionally, delay time (time of inversion) was investigated. The experiments were carried out in the absence of surfactants and, under no mass transfer conditions. All experiments were for the organic phase dispersed to avoid large-scale coalescence on the column interiors. Before each run, all the internal parts of the column and the dispersed phase reservoir were washed with acetone and freshly made warm 10% (w/v) chromic acid. After that, they were rinsed completely with warm distilled water. During this process, any probability of leakage was detected and removed quickly. No lubricants were used for valves and joints. The agitator shaft of the column was driven by a motor, controlled by a variable voltage transformer. The effective speed range was in the range of 500 to 1000 rpm. A digital tachometer photo / contact type was used to record the speed of the rotor. In order to prevent miscibility effects from interfering with experimental measurements two phases were mutually saturated by repeated circulation through the column for approximately 10 minutes before each run. The column was slowly filled with

the continuous aqueous phase until it reached a previously set level (1-2 mm above the top stage). The continuous phase was stagnant and the dispersed phase was entered (as single drops) from the bottom of the column by using a nozzle with definite internal diameter. The flow rate of dispersed phase was fixed in a specific value (3 l/min) by use of a rotameter. The experimental procedure involved operation of the column at the fixed rotor speeds, with the dispersed phase holdup was increased incrementally until inversion occurred. When the extraction column reached near flooding conditions as indicated by creation of a dense layer of droplets (dispersion bed) between coalesced dispersed phase and adjacent continuous phase, and the layer height reached one centimeter, inversion was observed to occur [14,15]. At inversion point, the dispersed phase inlet valve was quickly shut. The inversion holdup values which correspond to the holdup of the dispersed phase once phase inversion occurs were measured by sampling via the discharge valve nearest to the dispersion bed [13]. Fig. 2 shows the typical phase inversion behavior of n-butylacetate/water system for an O/W dispersion. As observed from Fig. 2, phase inversion is characterized by creation of a dense layer (dispersion bed) of the coalesced dispersed phase droplets.



**Figure 2.** Illustration of the phase inversion occurrence (dispersion bed) in *n*-butylacetate/water system

Moreover, delay time (the time of inversion), which is the time taken by the dispersed phase droplets to coalesce and form a homogenous phase, was recorded for each run.

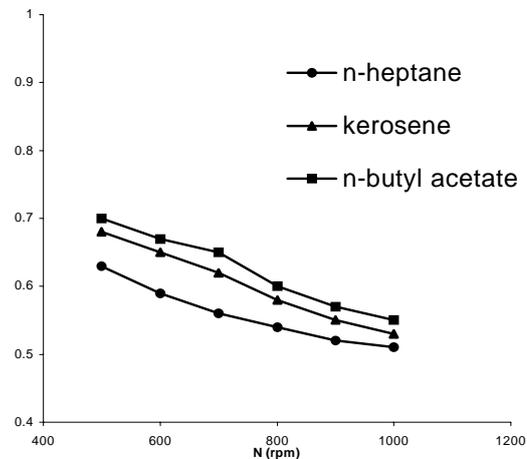
### 3. Results and discussion

The effects of energy input on inversion holdup were examined experimentally using three liquid-liquid systems with different physical properties. In addition, the effect of energy input on delay time was investigated using the above-mentioned liquid-liquid systems. Results are shown in Figs. 3 and 4.

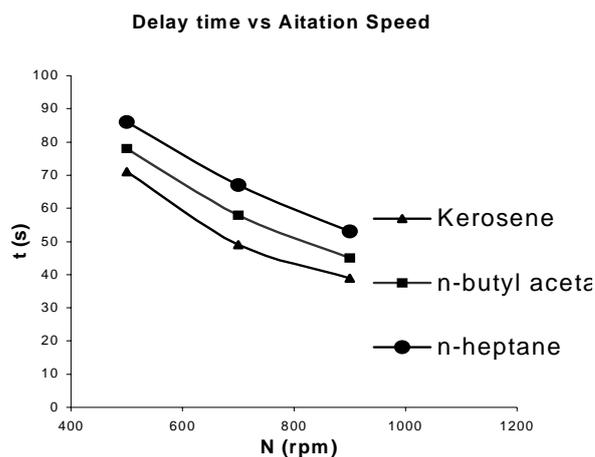
#### 3-1. Effect of energy input

The effects of energy input via agitation on inversion holdup and also delay times for all three liquid-liquid systems are illustrated in Figs. 3 and 4, respectively. With all runs represented in Fig.3, inversion holdups in the RDC column were significantly decreased by increasing the energy input. These results are in agreement with the findings of researchers [16-18]. The breakage of a drop in the

turbulent flow occurs if the dynamic pressure



**Figure 3.** Dispersed phase holdup versus energy input via agitation



**Figure 4.** Delay time versus energy input via agitation speed

due to turbulence exceeds the pressure due to surface tension. High energy input and, thereby, intensive flow results in a high breakage rate that prevents formation of large droplets. Since phase inversion is a catastrophic phenomenon, it is conceivable that the swarm of droplets created at high impeller speeds will coalesce at inversion point. Fig. 4 shows that at high energy input levels the small drops produced require a

short time to coalescence. Inversion delay time gets shorter with increasing input energy. This observation is in agreement with the findings of Gilchrist et al. [12] and Nienow et al. [19]. It can be discerned that the strengthened centrifugal force associated with the intensive flow vortices cause the drop coalescence to promote.

### **3-2. Effect of system properties**

Three Liquid-liquid systems were used to check the effect of the system properties on the phase inversion holdup and delay time. As shown in Fig. 3, the inversion holdup for n-butylacetate/water is higher than that for kerosene/water, and also the inversion holdup for kerosene/water is higher than that for n-heptane/water. It can be concluded that at a constant energy input, inversion holdup increases with decrease in interfacial tension. A low interfacial tension yields high drop breakage which causes low coalescence rates [13], thereby commensurately increasing the inversion holdup.

In constant rotor speed, inversion holdup increases directly versus density difference between the phases. High density difference between the phases will inevitably result in the complete separation of two phases and the low drainage of the intervening film of continuous phase trapped between drops. Furthermore, an increase in the density difference between the dispersed and continuous phases leads to higher relative velocities between the phases. Thus, the increased shear causes the droplets to break and, consequently, phase inversion occurs at lower dispersed phase holdup. Similar trends were also found by other investigators [10,20].

No systematic effect of viscosity on inversion holdup was found in these experiments. It was seen that when the viscosity of the organic phase decreased, the delay time of inversion increased. This observation is in agreement with the findings of Falco et al. [21] and Guilinger et al. [22] who reported that there is a maximum in the dispersed phase viscosity at or near the region of phase inversion, thus the dispersed phase viscosity is a property which indicates when phase inversion occurs but is not a factor which controls the phenomenon.

### **4. Conclusions**

The effect of energy input via agitation and physical properties of the liquids upon inversion holdup and also delay time was studied experimentally in an RDC column with a stagnant continuous phase at the constant input flow rate of the dispersed phase under no mass transfer conditions.

The main conclusions are summarized as follows:

- Phase inversion was characterized as the formation of a dense layer of the dispersed phase droplets between the coalesced dispersed phase and the adjacent continuous phase. Phase inversion took place near RDC extraction column flooding points. Creation of a high dispersed phase holdup (in the region of 0.50 to 0.70) is the prerequisite of Phase inversion, because the collision probability generally increases with an augmentation of holdup. This is concomitant with an increase in coalescence rate. Thus, phase inversion occurs.
- The inversion holdup and delay time are

considerably affected by the rate of input energy via agitation, and also, the physical properties of both liquids. The increase in the energy input via agitation results in the decrease of inversion holdup and inversion delay time. Moreover, in a system with larger viscosity, the inversion process will proceed more slowly than in a low viscosity system.

- The low interfacial tension means small drop sizes, thereby causing the efficiency of the RDC extractor to improve. RDC columns are suitable for liquid-liquid systems with low interfacial tension, because it is perfectly feasible to operate at a high dispersed phase holdup far from the flooding conditions.
- The lower both the interfacial tension and the density difference of the two phases, the greater the resistance of the system to inversion. When both the interfacial tension and density difference of phases decreases, the drainage time of the continuous phase film between two colliding droplets increases. The result is a lower coalescence rate. Thus, for the phase inversion to occur the input energy via agitation must be increased.

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