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Mass transfer and hydrodynamic studies in a 50 mm diameter centrifugal extractor

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A B S T R A C T

Centrifugal extractor is a preferred device for process intensification in solvent extraction. The complexities of high radiation damage to solvent and resultant degradation of solvent performance also necessitate the need for fast contactors like centrifugal extractors in the nuclear fuel recycle operations. In this study, mass transfer performance as well as hydrodynamic characterization was reported for a 50 mm diameter annular centrifugal extractor. Mass transfer studies included extraction and stripping experiments with solvent-aqueous pair of 30% Tri-*n*-butyl phosphate/*n*-dodecane and aqueous nitric acid solutions for different operating conditions like rotor speed, combined throughput and phase flow ratios. Flooding zone was characterized. Dispersed-phase holdups in annular as well as rotor regions and interfacial area for mass transfer were measured at different operating conditions. Non-ideal effects were studied by conducting residence time distribution analysis using aqueous phase as well as organic phase pulse tracer experiments. Various models like dispersion and tanks-in-series models were applied to experimental data. The contactor in study, was approximated as *N* number of tanks in series with a finite dead volume.

1. Introduction

Solvent extraction is one of the non-energy intensive mass transfer operations, widely used in chemical and nuclear industries [1]. Mixer-settlers, rotary columns and pulse-agitated columns are among the conventional solvent extraction equipment in vogue. The solvent extraction process is based on principle of difference in distribution coefficients of constituent species in a biphasic liquid-liquid immiscible system containing aqueous and organic solvent. Recently work on aqueous biphasic systems also has been reported by many researchers for separation of specialty chemicals. In the settlers of columns and mixer-settlers, separation of two immiscible phases happens due to density difference of individual phases. Larger is the difference, better will the gravity driven separation. Therefore, for aqueous-solvent systems having little density difference, gravity based settling does not provide a clean separation. This problem can be easily overcome by using a centrifugal settler which operates at higher speeds producing acceleration practically few hundred times the gravity and yields a high degree of clean separation.

Centrifugal extractors are a rotary version of mixer-settler. A practical combination of a paddle mixer and rotary centrifugal extractor was demonstrated in the SRL design [2]. The separated aqueous and organic liquids exited the settler through a complex air-pressure controlled weir design. Later at ANL, researchers modified the SRL design by incorporating an annular mixing chamber at the periphery of rotary settler and air-controlled weir was replaced by a robust fixed weir design. However, fixed weir dimensions could work only in a narrow range of density differences and adoptability of SRL design for practically any aqueous-solvent pair was lost.

1.1. Description of the equipment

In the annular centrifugal extractor, as shown in Fig. 1, two immiscible liquid feeds enter at respective inlets into the annular mixing zone between the stationary outer cylinder and rotating inner cylinder. Due to shear caused by a velocity gradient in the annular liquid zone, power is dissipated (in the range of 1–500 kW/m^3) to liquid and results into a very fine dispersion of one of the phases into another phase. The phase converted into fine drops is

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Abbreviations: ANL, Argonne National Laboratory; DD, *n*-dodecane; NPH, normal Paraffinic hydrocarbon; rpm, revolutions per minute; RTD, residence time distribution; SRL, Savannah River Laboratory; TBP, tri-*n* butyl phosphate; TIS, tanks in series.

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Nomenclature

C	Concentration
E	Exit age fraction
E_θ	Normalized exit age fraction
N	Number of tanks
V_d	Dead volume (mL)
θ	Dimension less time
υ	Volumetric flow rate (mL/min)
τ	Residence time (min)

called dispersed phase, while the other phase is termed as continuous phase. The mixture of two phases (i.e. dispersion) flows first downwards in the annular region, and then flows radially inwards in the region below the rotating cylinder and finally enters the central opening of the rotating settler cylinder. A deflector baffle prevents direct upwards entry into settler. The deflected flow now fills the bottom of settler. The cross-baffles are provided in the settler to prevent the whirling during rotation. The rotating cylinder imparts the liquid a practically rigid body rotation the inner surface of which is almost vertical shape because of high 'g' except a small parabolic portion at the bottom. The dispersion entering at the bottom gets separated as it moves upwards in the centrifugal flow field. The rate of separation depends upon the drop size distribution, their settling velocities under the centrifugal action ($r\omega^2$), density difference between heavy and light phases as well as viscosities and coalescing behaviour of the two phases. After separation light phase exits extractor through its weir into lighter phase collecting chamber. Heavier phase exits the extractor after passing through underflow and flowing over heavy phase weir.

For an exhaustive review of the development of centrifugal extractors and status of concurrent research, reader is referred to Vedantam and Joshi [3] and Leonard [4]. Duan et al. [5] reported recent advances in centrifugal extractor and its application in nuclear industry. The versatility of the centrifugal extractor lies in the tuneable energy dissipation during mixing of aqueous–solvent pair in the annular mixing zone. The required energy input for mixing is usually a function of interfacial tension between the aqueous and solvent pair. If excessive energy is delivered during mixing, the mixed-phase may turn into an emulsion and may not settle in the setting zone.

Unlike conventional extractors, the operation of centrifugal extractors has strictly limited operational zone. The other-phase carryover or entrainment is fixed at strict 1% limit and extractor is said to be flooded if this limit is exceeded. The combined throughput at flooding is taken as nominal maximum capacity and operating capacity is usually fixed at 60% of this flooding capacity.

Arafat et al. [6] reported observation of small entrainment in the product streams during proof-of-concept tests at ANL. Kumar et al. [7,8] reported submicron size other-phase carry over (entrainment) in the aqueous and organic phases respectively with 30%TBP diluted with *n*-dodecane/nitric acid systems. Mass transfer performance of extractor unit has to be measured prior to deployment into operation in industry. Mandal et al. [9] and Meikrantz and Garn [10] reported mass transfer efficiencies approaching unity for both extraction and stripping in 125 mm centrifugal extractor. Recently useful modelling of centrifugal extractors was reported by Drewery et al. [11].

Due to positive attributes of low floor-space requirement, low head space, high capacity and high mass transfer efficiency, centrifugal extractors are used in several liquid-liquid extraction based commercial chemical industries like recovery and purification of metals in mining industries.

Dead volume, by-pass and recirculation are common problems in the contactors resulting in poor performance. These problems are generally characterized by degree of aggregation, earliness of mixing and residence time distribution. Holdup and drop size distribution are required to calculate interfacial area of mass transfer at different operating combinations. Schuur et al. [12] reported liquid holdups, residence time distribution, phase behaviour and drop size distributions of centrifugal contactors for various liquid-liquid systems. Tamhane et al. [13] reported computational modelling of RTD in centrifugal contactors of various dimensions. Recently centrifugal extractors have also been used as multifunctional reactor-separator in the pilot synthesis of biodiesel fuels from vegetable oils [14].

Successful operation of centrifugal extractor without phase contamination, depends on operating conditions. It requires identification of operating envelop at different operating conditions once geometrical parameters are fixed. Extensive mass transfer, hydrodynamic experiments including holdup, drop size distribution are performed in this work, residence time distribution experiments are conducted to identify ills in the contactor and quantified them using low cost pulse tracer experiments.

2. Experimental procedure

Experimental setup consisted of two ISMATEC pumps coupled to FMI-Q3 heads (max. flow rate 2300 mL/min) and associated feed/product tanks connected to a single-stage 50 mm diameter centrifugal extractor. Details of centrifugal extractor used in this study are reported in Table 1.

2.1. Chemicals used

Tri-*n*-butyl phosphate (HWP, Talcher, India, 99%+) was diluted with *n*-dodecane (Sigma Aldrich, 99%+) to make 30% TBP solution which was used as a solvent. Nitric acid (69–72% AR, Chemspure) was used to make the aqueous solution of 0.01 N and 4 N nitric acid concentration. AR grade *n*-hexadecane (Sigma Aldrich) was used as organic tracer in the organic phase RTD experiments. AR grade Sodium dodecyl sulphate (Merck) was used for stabilization of dispersed phase during drop size measurement experiments. Normal paraffinic hydrocarbon mixture (~98% *n*-dodecane, Orion Chemicals, Mumbai) was used as organic phase in RTD experiments.

2.1. Mass transfer studies

Before starting the experiment, the centrifugal extractor was flushed with water. Continuous phase was pumped first and

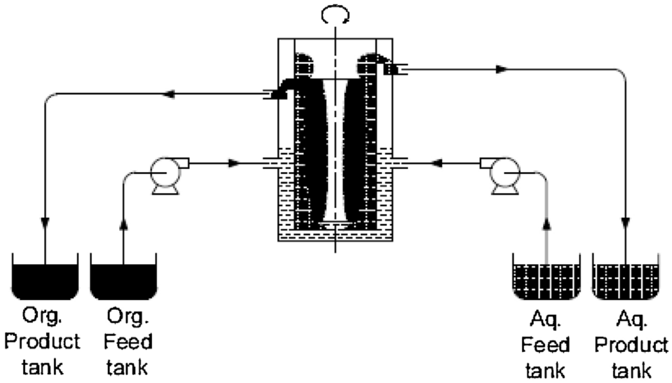

Fig. 1. Schematic of experimental setup.