



# Experimental investigation of the effects of the hydrophilic silica nanoparticles on mass transfer and hydrodynamics of single drop extraction



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## ABSTRACT

In this study, the effect of hydrophilic silica nanoparticles on mass transfer in liquid-liquid extraction process was experimentally investigated using the ternary chemical system toluene-acetic acid-water. In order to examine the dispersed phase drop sizes, single drop experiments with a range of nozzle diameters within 0.1–0.51 cm were carried out. Hydrophilic SiO<sub>2</sub> nanoparticles with different volume concentrations of 0.005–0.1 vol% were added to the continuous phase. The results demonstrated that mass transfer coefficient reduced in presence of hydrophilic SiO<sub>2</sub> nanoparticles and maximum deterioration of 22% was observed using 0.1 vol% silica nanofluid for drops generated by 0.2 and 0.4 cm nozzles. Viscosity increase, steric hindrance of nanoparticles at the liquid-liquid interface, reduction in both free volume of mass transfer path and liquid-liquid contact area because of solid nanoparticles presence are introduced as the main reasons of observed behavior. Moreover, it can be concluded from the experimental results that silica nanoparticles do not significantly influence hydrodynamic parameters of the extraction system.

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## 1. Introduction

Liquid-liquid extraction (LLE) is widely used in petroleum, pharmaceutical and chemical industries [1]. Enhancing mass transfer in LLE operations has attracted interest and different techniques such as chemical and mechanical methods and use of nanoparticles have been examined. The influences of aqueous phase pH, electrolyte salts and binary surfactants on the mass transfer for extraction of acetone in toluene-acetone-water system were investigated using single drop experiments [2–4]. Extraction rate reduction was observed by increasing aqueous phase pH that was attributed to adsorption of hydroxyl ions onto the drop interface and consequently retardation of the solute mass transfer [2]. Addition of different electrolyte salts in the aqueous phase enhanced mass transfer rate remarkably. Hydration of ions was reported to facilitate solute transfer to the aqueous phase [3]. The effects of operating temperature on single drop extraction in toluene-acetic acid-water and 4-methylpentan-2-one - acetic acid - water chemical systems were examined in single drops [5,6]. Results demonstrated remarkable effect of temperature on the overall mass transfer coefficients. Molecular diffusivity enhancement in

drops with temperature was introduced as the cause for the LLE enhancement.

Application of nanotechnology in mass transfer enhancement in different operations and geometries, including diffusive and convective mass transfer has attracted interest and limited studies on the subject were recently performed [7–11]. Among these researches, few are devoted to applying nanoparticles to liquid-liquid extraction processes [12–20].

Bahmanyar et al. [12,13] investigated the effect of hydrophobic SiO<sub>2</sub> nanoparticles on mass transfer performance and hydrodynamic characteristics of a pulsed liquid-liquid extraction column (PLLEC). Their experimental results indicated that mass transfer augmented 4–60% by using 0.01–0.1 vol% of nanoparticles. Brownian motion of nanoparticles was introduced as the responsible mechanism for the mass transfer enhancement.

Under the conditions matching previously mentioned works by Bahmanyar et al. [12,13], Khoobi et al. [14] examined size, shape and distribution of the organic drops in a PLLEC. They found that nanoparticle presence had a remarkable influence on the drop geometrical shapes and make drops undergo shape change from ellipsoidal to spherical.

Moreover, Ghafoori Roozbahani et al. [15] examined the influence of hydrophobic SiO<sub>2</sub> nanoparticles on the static and dynamic hold-up using the same chemical system in the extraction column

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## Nomenclature

### Latin letters

$C_D$	drag coefficient
$d$	drop diameter (cm)
$D$	diffusivity ( $\text{cm}^2 \text{s}^{-1}$ )
$E$	extraction fraction
$E\ddot{o}$	Eötvös dimensionless number ( $g\Delta\rho d^2/\gamma$ )
$H$	dimensionless group defined by Grace et al.
$K_d$	dispersed phase mass transfer coefficient ( $\mu\text{m s}^{-1}$ )
$K_{od}$	overall dispersed phase mass transfer coefficient ( $\mu\text{m s}^{-1}$ )
$K^*$	partition coefficient
$M$	mass (g)
$Mo$	Morton dimensionless number ( $g\mu_c^4\Delta\rho/\rho_c^2\gamma^3$ )
$n_i$	number of the drops with diameter $d_i$
$Re$	drop Reynolds number ( $\rho_c u_i d/\mu_c$ )
$Sc_d$	dispersed phase Schmidt number ( $\mu_d/\rho_d D_d$ )
$Sh_d$	dispersed phase Sherwood number ( $k_d d/D_d$ )
$t$	time (s)
$u$	velocity ( $\text{m s}^{-1}$ )

### Greek symbols

$\kappa$	ratio of dynamic viscosities = $\mu_d/\mu_c$
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$\mu$	viscosity ( $\text{mPa s}^{-1}$ )
$\rho$	density ( $\text{kg m}^{-3}$ )
$\gamma$	interfacial tension ( $\text{mN m}^{-1}$ )
$\Delta$	difference
$\phi$	particle volume concentration

### Subscripts

$bf$	base fluid
$d$	dispersed phase
$A,d$	solute in dispersed phase
$A,c$	solute in continuous phase
$f$	final value
$i$	initial value
$nf$	nanofluid
$p$	particle
$t$	terminal
$w$	water

### Superscript

*	equilibrium
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used in [12,13]. Their results indicated that dynamic hold-up increased up to 70%, as a result of the nanoparticles presence.

Mirzazadeh Ghanadi et al. [16] explored the effect of nanoparticles on mass transfer in a simple liquid-liquid extraction column while employing different dropping and jetting flow modes. Their results indicated that concentration and type of the nanoparticles and also the flow mode influence mass transfer augmentation. They reported that mass transfer increased with nanoparticles concentration and an effective absorption value of 2, as the maximum mass transfer enhancement, was observed for ZnO nanoparticles.

The influence of hydrophobic  $\text{SiO}_2$  nanoparticles on mass transfer in an irregularly-packed liquid-liquid extraction column was investigated by Nematbakhsh and Rahbar-Kelishami [17]. Maximum mass transfer coefficient enhancement (peak enhancement) of about 42% was observed at 0.05 vol% for the smallest nanoparticles. They attributed such observation to Brownian motion and believed that aggregation and viscosity effects are liable for the decrement observed after the peak enhancement.

One of the most recently published study in this field is Ashrafmansouri and Nasr Esfahany's article [18] on the influence of hydrophobic silica nanoparticles on hydrodynamics and mass transfer of a spray liquid-liquid extraction column. The results demonstrated that hydrodynamic parameters were not found to be influenced significantly by silica nanoparticles, whereas maximum augmentation of 47% in overall mass transfer coefficient was observed making use of 0.001 vol% nanoparticles for larger drops. Induced microconvection and Brownian motion of nanoparticles was introduced as the dominant mechanism for mass transfer enhancement. Moreover, nanoparticle aggregation and reduction in free volume because of solid nanoparticles presence were considered as the main presumable reasons liable for decreasing trend in higher nanoparticle concentrations.

Saien and Bamdadi [19] studied liquid-liquid extraction in the presence of nanoparticles in single drops. They observed a maximum augmentation of 157% and 121% for magnetite and alumina nanoparticles (average LLE enhancement of about 72% and 75%) at nanoparticle weight fraction of 0.002%, respectively. They observed that smaller drops experience higher mass transfer

enhancement while the size of the drops was not found to be remarkably affected by the nanoparticles presence.

In another recently published survey, Saïen et al. [20] examined the effects of oscillating magnetic field on extraction from organic drops loaded with magnetic nanoparticles. Maximum enhancement of 121% was observed in their experiments.

Single drop experiments appeared to be a fundamental step toward the design of liquid-liquid extraction processes. However, it is still difficult to predict the rate of mass transfer as a function of major physical parameters, even for single drops. The limited investigations of nanoparticles effects on LLE process focused on the dispersion of nanoparticles in the dispersed phase. In the previous investigations hydrophobic nanoparticles have been used. In the present study, the influences of hydrophilic  $\text{SiO}_2$  nanoparticles dispersed in aqueous continuous phase, on the mass transfer and hydrodynamics in single drop extraction were investigated. Experiments were performed utilizing the chemical system toluene - acetic acid - water and various dispersing nozzles and finally presumable reasons for the observations have been discussed based on the experimental results.

## 2. Experimentation

### 2.1. Materials and methods

#### 2.1.1. Materials

Chemical system of toluene-acetic acid-water was used in the present study. Toluene, acetic acid, NaOH (as titrant) and KHP (potassium hydrogen phthalate for standardization of NaOH solutions) were purchased from Merck (Germany). De-ionized water with conductivity of  $18 \mu\text{S cm}^{-1}$  was provided by a deionizer (FINETECH SWPS-3001M, Korea). Hydrophilic  $\text{SiO}_2$  nanoparticles 7–13 nm and density of  $2100 \text{ kg m}^{-3}$  were purchased from PlasmaChem (Germany).

The physical properties of the dispersed and continuous phases are given in Table 1 (W, AA and T stand for de-ionized water, acetic acid and toluene, respectively).

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