



Predictive model for the design of reactive micro-separations

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ABSTRACT

Solvent extraction at micro-scale has known increased interest in recent years. Consequently, numerous theoretical and experimental studies focused on diagnosis, detection and recovery of either biological substrates or base metals, isotopes and rare earths have been reported so far in the last few years.

Within this context, the present work reports a thorough analysis of the coupling between the hydrodynamics, mass transfer and chemical reaction kinetics in multiphase micro-systems; the predictive mathematical model describes under non-steady state, fluid-wall interaction and interaction between the fluids in contact to finally predict the solute mass transport rate between the flowing liquid phases when a fully developed profile and a flat interface are not initially assumed unlike previously reported works. This approach allows tracking the interface along the complete micro device geometry and makes the model adaptive to the specific needs of each micro fluidic application. The analysis has been developed for a model system based on the mass transfer of Cr (VI) from an aqueous feed phase in a stratified 2-layer flow system in a Y-Y shape micro device. Two different scenarios have been considered: (a) two homogeneous phases, where water is the receptor phase (b) an heterogeneous system where the solute moves from the feed solution to a receptor phase composed of Shellsol D-70 and Alamine 336 as the selective extractant. Model simulations accounting for the system reaction parameters already reported have been assessed against a set of experimental runs; the two systems under consideration provided data that satisfactorily fitted simulations with an error less than 10%, thus, validating model calculations. Thus, this rigorous and flexible model seeks to provide a useful tool for the design of micro separation processes by predicting the technical performance for numerous applications at micro scale.

1. Introduction

Microfluidics has aroused great expectations about its countless applications and strengths during the last few years. Several remarkable benefits of this mighty technology have sparked a tremendous interest in harnessing its potential [1,2]. Scaling down allows for handling smaller volume of fluids, which entails small quantities of reagents and samples reducing waste products and hazardous substances and therefore, decreasing costs [3]. Besides, high surface-to-volume ratio enhances mass transfer and thermal dissipation, which makes liquid-liquid extraction a suitable technique to be combined with microfluidic devices [4]. In fact, micro-solvent extraction possesses a large number of applications in diverse fields that range from food safety control in applications such as extraction and pre-concentration of pesticides from juice samples [5] to forensics, detecting different antidepressant compounds in human urine and plasma samples [6]. Besides, scaling down promising advantages have also been applied to nuclear applications as the work developed by Mayur et al. [7] where uranium is extracted in microbore tubes. As many materials employed in microfluidics as PDMS

or SU-8 report biocompatibility [8], there are several studies focused on the development of microfluidic analytical techniques of biological substrates, isolation of leukocyte and erythrocyte cells from blood cells [9], detection of cocaine and its derivatives in hair samples [10] or even tumour progression tracking [11].

The crux of solvent extraction (SX) techniques often lies in molecular diffusion. Since down scale predominantly implies laminar flow ($Re < 1$) with no turbulences and a streamlined flow, net transport of molecules occurs due to their random motion [12]. Based on this principle, several studies regarding the separation of solutes have been applied in two differentiated systems: homogenous, where two aqueous phases are contacted, and solute transport occurs by simple diffusion, and heterogeneous systems, where an aqueous phase is brought into contact with an organic receptor phase and the separation is carried out by facilitated transport.

Regarding homogeneous systems, since the pioneering work of Brody and Yager. [14] reporting the diffusive transport of carboxy-fluorescein in an aqueous phase, different authors have contributed with interesting studies on molecular diffusion combining experimental

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runs and computational techniques as recently reported by Gómez-Pastora et al. [15] who studied the solute mass transport performance in Y-Y shaped microchannels as function of flow patterns and mass transport kinetics. Moreover, microfluidic techniques have been also applied to protein extraction in aqueous two-phase systems, Novak et al. [16]. In this context, mathematical models to characterize microchannels mixing and flow quality and models aimed to determine the necessary operation conditions to verify laminar flow and predicting diffusivity and concentration profiles of the solute have been already reported [17–20]. Different solutes such as glucose, benzoic acid, sucrose and glycine among others were contacted with water and the experimental results were satisfactorily validated with the predicted ones. Afterwards, Ciceri and co-workers [21] focused on the diffusion of Co (II) from an aqueous feed solution to an aqueous buffer and validated the experimental diffusion study with model simulated results based on the previous work of McCulloch et al. [22] who, using “Instaspec” III software, developed an analytical solution starting from Fick's second diffusion law in order to predict the target species concentration in the receptor phase.

Concerning heterogeneous systems, Sato et al. [23] firstly experimentally demonstrated the molecular transport of Ni (II) into a chloroform reservoir and Bowden et al. [24] reported the rapid hydrocarbon extraction to a hexane phase. In addition, different reports [13,21] developed a treatment to control the degree of hydrophilicity/hydrophobicity of the microdevices walls. They further reviewed stabilizing methods of stratified micro flows and studied the molecular diffusion to describe the solute transfer across the water/oil interface. Recently, experimental micro extraction has been applied by Kolar et al. [25] to validate the viability of rare earth elements extraction into Cyannex 572. Furthermore, experimental work coupled with computational techniques has been reported as in the work of Kuban et al. [26] to determine the influence of physical parameters such as interfacial area, density, viscosity and flow velocity on the performance of micro-solvent extraction processes. Despite most solvent extraction processes are developed and simulated in 2 layer microdevices, Surmeian et al. [27] and Tetala et al. [28] carried out the simultaneous forward and backward extraction steps in a water/oil/water system. While Surmeian et al. assured the stability between the 3 phases and achieved a rapid transport of methyl red into cyclohexane, Tetala et al. studied the extraction effectiveness of alkaloids from plants extracts. Phase stabilization in a 2 phase system was also reported by Žnidaršič-Plazl and Plazl [29], who estimated the diffusion coefficients by correlations and mathematically described a non-linear equation system where the esterification of isoamyl acetate took place at the interface between n-hexane and an aqueous phase. Moreover, Mason et al. [30] focused on heterogeneous transport and developed a simplified model under steady state conditions; they assumed a stable interface and estimated the mass transfer coefficients by four different approaches to determine the best correlation to describe the solute transfer to a receptor phase. This analysis gave rise to a subsequent work carried out by Ciceri et al. [31] to determine the extraction kinetics of Co (II) into DEHPA. A numerical model was constructed and solved using CFD techniques assuming a flat interface between the fluids in contact as well as a streamline flow along the device. In addition, a no-slip velocity condition was considered as boundary condition on the walls of the micro device and a pressure driven gradient set the flow rate of each phase.

On the basis of previous studies, this work pursues the advance on micro-extractors design solving the coupling between fluid dynamics and mass transfer kinetics and allowing the interface tracking along the complete geometry of the microdevice. The analysis has been developed for the transport of hexavalent chromium as target solute from an aqueous phase flowing through a Y-Y shaped microchannel and considering two different scenarios: (i) an homogeneous system, where the solute is separated by simple diffusion across two aqueous phases and, (ii) an heterogeneous system where facilitated transport promoted the

solute transport across the aqueous-organic interface. ANSYS FLUENT software was used to develop a flexible model that solves under dynamic conditions both Navier-Stokes and species balance equations; the model also implements the surface tension between the liquid phases that had been experimentally determined, and the fluid-wall interaction through the measurement of the contact angle. Furthermore, a set of experimental runs was carried out for the two cases of study and the results fitted satisfactorily to simulated data using CFD modelling together with physical-chemical parameters already reported [32]. Consequently, the rigorous and flexible model here developed constitutes a useful tool for the design of micro-separators and is widely applicable to different separation systems in numerous application fields at micro-scale.

2. Methodology

2.1. Materials

KCrO₄ (99%, Panreac Quimica S.A.) was used to prepare the feed solution and Shellsol D-70* (Kremer) and Alamine 336* (BASF) were employed for the organic phase of the heterogeneous system. Hydrochloric acid (37%, Panreac) was also added to the initial solution to adjust the pH. Furthermore, in order to verify the interface stabilization of the homogeneous system, sodium fluorescein, C₂₀H₁₀Na₂O₅, (Scharlau) was employed as fluorescent tracer to better visualize the aqueous phases in contact. All aqueous solutions were prepared with milli-Q water.

2.2. Microfluidic device

A Y-Y shaped microdevice made of SU8 as substrate was used to perform the experimental procedure. The contact zone after the Y-inlet was 2 mm long, 300 μm wide and 300 μm deep and the angle between both inlets was 60° (same angle as between the outlet branches) as shown in Fig. 1. The cross-sectional shape was rectangular and the interfacial area between phases was approximately 0.6 mm².

2.3. Experimental set-up and measurements procedure

The micro device was placed in a polymethyl methacrylate (PMMA) holder (16 × 8.5 mm) with a magnetic closure and interchangeable inlets and outlets connections of 1/4" UNF (MicruX Technologies). At the same time, two 50 mL stainless steel syringes (Harvard Apparatus) were loaded with their respective solutions, placed in two infusion pumps (KD Scientific Legato series 200) and connected to the micro device inlets through Tygon® tubes of 0.8 mm of internal diameter (Saint-Gobain). For each scenario, the fluid phases were brought in contact along the microdevice and for both cases of study, fluids co-flow and the interface track were controlled on a stereo microscope (Nikon SMZ18) equipped with a green fluorescence filter (light wavelengths of around 550 nm) and a Jenoptik ProgRes C5 camera. Images were taken using the ProgRes® CapturePro software (CapturePro V2.10.0.0).

Samples were collected in Eppendorf tubes (1 mL) and the chromium concentration of the aqueous phases was measured by atomic absorption spectroscopy (Perkin Elmer 3110). Absorption standards were prepared with a chromium standard solution of 1000 mg/L (PanReac). Experiments were executed in triplicate to verify their

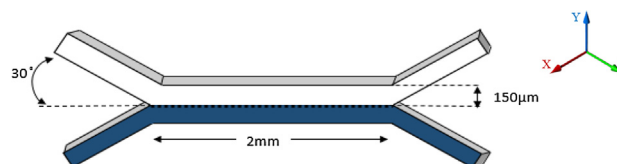


Fig. 1. Micro device geometry and dimensions.

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