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Mass transfer studies in a horizontal pulsed sieve-plate column for uranium extraction by tri-n-octylamine using axial dispersion model

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ABSTRACT

In this work, uranium extraction from an aqueous sulfate solution with 0.25M concentration acid, by using 5% v/v tri-n-octylamine (TOA) solvent, 90% v/v kerosene diluent and 5% v/v decanol modifier is studied. The effect of operation parameters (i.e., phase flow rates and pulsation intensity) on the volumetric overall mass transfer coefficients and axial dispersion in a horizontal pulsed sieve-plate column is evaluated using axial dispersion model (ADM). According to the results, increasing pulsation intensity and phase flow rates leads to the enhancement of the mass transfer performance. The continuous phase axial dispersion is found to be significantly higher than that in the dispersed phase. Axial dispersion of a phase is found to be mostly influenced by pulsation intensity and the flow rate of the phase itself and is minor influenced by the second phase flow rate. The overall height of mass transfer unit (HTU_{oc}) is obtained to be less than 0.2 m, indicating high performance of a horizontal pulsed sieve-plate column for solvent extraction of uranium. Furthermore, new correlations are proposed for prediction of $K_{oc}a$, E_c and E_d , which are in satisfactory agreement with the experimental data with AARE values of 1.03%, 3.35% and 1.89% respectively.

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1. Introduction

It is of importance to extract uranium from its containing ores due to its radioactivity and toxicity risks on surface and ground water. Uranium has become an important actinides element for the industrial applications and nuclear energy plans especially from environmental perspective (Karve and Rajgor, 2008; Lothongkum et al., 2009). Solvent extraction is a classical separation process that is conducted in most of industries including petroleum refining, biochemistry, nuclear fuel processing, pharmaceuticals, metal extraction, food industry, and waste management (Akhgar et al., 2017; Amani et al., 2017). It is because of the fact that solvent extraction offers various advantages including high selectivity, high product purity, high recycling capability, low cost, simplicity of process and high production capacity (Benedict et al., 1981).

http://dx.doi.org/10.1016/j.pnucene.2017.02.010 0149-1970/© 2017 Elsevier Ltd. All rights reserved. Generally, pulsed columns compared to mixer-settlers are desirable from both safety and economic perspective, in particular their higher throughput, less space consumption, no internal moving parts, minimum leakage, and simplicity of design. The advantages are more beneficial especially while processing corrosive or radioactive solutions since the pulsing unit can be remote from the column. Pulsed columns can be divided into the vertical and horizontal types. The vertical columns meet the needs for industrial applications, but when height limitation, especially in indoor applications (i.e., nuclear industry), is a concern it is required to use the horizontal columns.

The design and optimization of an extraction column require determination of independent parameters including the crosssectional area and the column length in order to accommodate required flows without flooding as well as meeting the desired extraction performance. In addition, the axial dispersion in the dispersed and continuous phases are among the important parameters which leads to the reduction of mass transfer performance and consequently the enhancement of the required length for meeting desired extraction (Pratt and Baird, 1983). Many

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models have been proposed for predicting the mass transfer in solvent extractors, such as the axial dispersion, plug flow, the mixing stages, the forward mixing, the backflow and drop population balance models (Panahinia et al., 2017b). Among these models, axial dispersion model, which considers all non-ideal parameters in a single parameter based on Fick's law and the concentration profile, has been widely employed for mass transfer characteristics in extraction columns (Asadollahzadeh et al., 2017).

Taking into account the great applicability of horizontal extraction column in industrial applications that we previously investigated the hydrodynamic and mass transfer of such columns (Khajenoori et al., 2015a, 2015b, Panahinia et al., 2017a, 2017b), this article concerns the applicability of a horizontal pulsed sieve-plate extraction column in solvent extraction of uranium. In addition, the effect of operating parameters including pulsation intensity and flow rates of phases on mass transfer performance and axial mixing parameters has been evaluated using axial dispersion model.

1.1. Previous work on uranium extraction

Investigations conducted on uranium (VI) extraction from nitric acid solutions to ionic liquids can be classified into three distinctive mechanisms of the extraction (i.e., cation exchange, anion exchange (Chelating) and solvation) which depend on the nature of extractant, counterion concentration, structure of the ionic liquid as well as the aqueous phase composition (Dietz and Dzielawa, 2001; Wei et al., 2003). It has been revealed that liquid-liquid extraction technique is the most effective separation method to extract and purify the solutions containing uranium (VI) in the nuclear industry (Billard et al., 2011). Various types of extractants have been used so far for purification of uranium which are given in Table 1. Most common extractants are di (2-ethyl hexyl) phosphoric acid (D2EHPA), tri n-butyl phosphate (TBP) and tri-n-octylamine (TOA) (Datta et al., 2016; Yan-Zhao et al., 2003; Zhijun et al., 2003). However, TOA is an environmentally-friendly extractant which has been used in many applications due to its high selectivity and efficiency especially for uranium recovery from various sources (Goldenberg and Abbruzzese, 1983).

Mixer-settlers and columns are the only extraction equipment which have been used so far in the industrial applications for solvent extraction of uranium (Movsowitz et al., 1997). The only published type of columns applied in uranium solvent extraction is the Bateman pulsed column and only two reports are available in the literature which compare the performance of extraction between a pilot plant and industrial columns (Miller and Kleinberger, 2000; Movsowitz et al., 2000). Both reports deal with the Bateman pulsed columns: one involves in extraction of uranium in Olympic Dam, Australia which presented by Movsowitz et al. (2000) and the other one is focused on production of phosphoric acid by extraction in Haifa Chemicals, Israel, which proposed by Miller and Kleinberger (2000). According to Movsowitz et al. (2000), the performance of the columns was excellent and the extraction yield was similar to the 4 mixer-settlers in series - about 98%. It is

Table 1

Types of extractants for uranium extraction.

achieved that the operating costs in solvent extraction columns are lower because of smaller losses of solvent. Therefore, the installation of the Bateman pulsed columns in order to purify the uranium solutions has shown that these columns provide a beneficial advantage over the mixer-settlers. The approximate capital expenditure of the pulsed column was about 20% lower than that of similar size plants employing mixer-settlers (Jahya et al., 2009; Movsowitz et al., 1997). The successful use of pulsed columns in the uranium industry is leading to more interest in pulsed columns in order to extract other metals such as cobalt, zinc, nickel and copper. For instance, applying pulsed columns in the GORO Nickel project in New Caledonia has exhibited the entrance of pulsed columns into the base metals industry, resulting in application of pulsed columns for other metals (Vancas, 2003).

2. Experimental

2.1. Extraction column description

The experiments were conducted in a semi-industrial horizontal pulsed sieve plate column. The active part of column was a pipe housing an internal plate cartridge including 25 pair of sieve plates made from 304 stainless steel. Pulsations were provided by applying a fluctuated air pressure (controlled by two solenoid valves) to the air space above a liquid leg connected to the bottom settler of the extraction column. A 9 cm-diameter settler was installed on both ends of the column in order to separate two phases. The interface between the light and heavy phases in the upper settler was controlled by an optical sensor, which was connected to a solenoid valve in outlet streamline of downer settler. A schematic diagram of the setup is given in Fig. 1 and geometrical characteristics of the column are listed in Table 2.

2.2. Chemicals and reagents

The aqueous solution was a leach liquor supplied from the Bandar Abbas Uranium Production Plant (BUP) with the concentration of 196 ppm. Inductively Coupled Plasma (ICP) used for analysis of the leach liquor that results is presented in Table 3.5% v/ v tri-n-octylamine (TOA) solvent, 90% v/v kerosene diluent and 5% v/v decanol modifier were the dispersed phase. The commercial extractor tri-n-octylamine was prepared from Sigma Aldrich Company. Decanol and Kerosene (non-aromatic) were used as diluents without further purification. The heavy and light interfacial tension measurements were obtained via a Krüss tensiometer. Density and viscosity of organic and aqueous phases were measured by the picometer method and with DVI-Prime viscometer respectively. Physical properties of the phases are listed in Table 4. It should be noted that, under mass transfer conditions, a degree of uncertainty surrounds the estimation of physical properties (particularly interfacial tension), since these vary not only with the inlet solute concentrations, but also along the column. In the present research, the values of physical properties have been

Cationic extractant	Anionic extractant	Neutral extractant
High density polyethylene	Alamine 336	Tri-n-butyl phosphate
β -diketones	Tri-laurylamocum chbride	Tri-n-octylphosphine oxide
Cyanex 272	Di —trimethyl hexyl ammonium chloride	Tri-butylphosphine oxide
Cyanex 301	Aliquat 336	Cyclic polyether di-benzo-18 -crown-6 (DBC)
Cyanex 302	Tri-dentate diglycolamide	Cyanex 921
Hydroxyl oximes	2-DGA	Cyanex 923
Thi-ocarbazones	Tri-n-octylamine	Cyanex 925

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