Numerical and Experimental Study on Oil-water Dispersion in New Countercurrent Centrifugal Extractor

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Abstract

A new countercurrent centrifugal extractor, in which multiple theoretical stages are formed by Taylor-vortexes has been developing. A continuous extraction of Zn(II) with D2EHPA was carried out with different rotating speeds of inner rotor and different property of rotors. A numerical simulation was also carried out by changing physical properties such as surface tension and lipophilicity. As a result, the formation of multiple theoretical stages was confirmed and the extraction efficiency was optimized by increasing the rotating speed of inner rotor. The results of numerical simulation suggests the dispersion was promoted by increase in the lipophilicity of the rotor surface.

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Keywords: Centrifuge extractor, Taylor-Cuette, D2EHPA, Numerical Simulation, Volume of Fraction Model;

1. Introduction

Recycling precious metals and rare-earth metals from the industry is more and more important both for economical and strategic points of view. M. Ozawa et al. suggests special nuclear fuel cycle for production of rare-earth metals from nuclear spent fuels [1][2]. In this case, large extractors are not applicable due to strong radioactivity of solution and criticality safety. Many extractants and extractors have been developed for nuclide separation and extraction. One typical extractor is a mixer-settler extractor, which has a strong agitation power and is easy to operate. However, for an advanced extraction with multiple theoretical stages, contentiously connected mixer-settler extractors are needed. Furthermore, secondary radioactive wastes are correspondingly increased. Hence, pulsed-column extractors can be used for multi-stage extraction in e.g. the reprocessing plants.
for spent nuclear fuels. However, the size of pulsed-column extractor becomes too huge to achieve higher extraction performance, and the residence time becomes larger. Small sized extractor is advantageous from the view point of criticality safety, and shorter residence time alleviates radiolytic decomposition of extractants. Therefore, small and high-performance extractor is needed. Takeshita et al. have been developing liquid-liquid counter-current centrifugal extractor with many theoretical stages [3]. For the multi-stage extraction with a single extractor, understanding of oil-water interfacial behavior is important because concentration gradients of each chemical compound and stable countercurrent flow should be maintained during the extraction operation. Otherwise percent extraction never exceeds that obtained with the single mixer-settler extractor. However, the effect of inner rotor or interfacial tension is not known. Therefore, the relation between the interfacial behavior and the percent extraction is discussed by numerical and experimental approaches.

2. Experimental Setups

The extractor mainly consists of three parts; an inner rotor, a mixing part and a settling part at the top as shown in Fig.1. When the rotating speed exceeds critical Taylor’s number which is the dimensionless number indicating the effect of centrifugal force in rotating fluids, a series of ‘Taylor vortexes’ is observed. By the formation of the Taylor vortexes as shown in Fig.1-(b), a formation of concentration gradient, an effective dispersion of the organic phase and an increase of organic phase in the extractor are expected. Organic phase is 10mM of bis(2-ethylhexyl) phosphoric acid (D2EHPA) diluted by n-dodecane and aqueous phase is nitric acid solution dissolving Zn(NO₃)₂. The pH and ionic strength of the aqueous phase are 2.5 and 0.02, respectively. The ionic strength is conditioned by sodium nitrate. Organic phase is injected from the bottom and aqueous phase from the top of the extractor, as shown in Fig.1(b). Feed speeds of inlet and outlet of both phases are 10 cc/min and rotating speed is 400, 800 and 1200rpm. Two types of inner rotors are tested to survey the effects of the property of the rotor surface. One is made of Teflon and the other is made of stainless steel whose surface is polished. Though the surface roughness of the stainless steel is not known, the lipophilic property of stainless steel is worse than that of Teflon. The effect of the roughness of the stainless steel surface is not considered in this study.

Fig. 1. (a) picture of extractor; (b) geometry of extractor and Taylor vortexes; (c) mesh created with ICEM-CFD
3. Calculation Condition and Models

To simulate the behavior of oil-water interfaces, a commercial CFD code ‘ANSYS FLUENT 13.0’ is used because of its excellent convergence ability. Before the series of simulations, preliminary tests were done with changing mesh fineness, physical models such as turbulent models, wall functions and other options to resolve Taylor vortexes in the extractor. In this study, low-cost turbulent model is needed to simulate with Volume of Fraction (VOF) model to reduce calculation time because VOF model needs fine mesh and consequently calculation time becomes larger. It is reported that the simulation with Reynolds Stress Model (RSM) can predict flow behavior[4][5], but lighter burden is appropriate. Finally, calculation condition was selected as listed in Table.1. Considering them, Renormalization Group (RNG) k-ε model in which the transport equations of turbulent kinetic energy ‘k’ and energy dispersion rate ‘ε’ are calculated with the alleviation of overestimated turbulent viscosity is selected. In VOF model calculation, transport equations (Navier-Stokes equations) of volume factor which is 0 to 1 are calculated in each cell. From the distribution of volume factor, oil-water interfaces are reconstructed by visualization software ‘AVS Express/Dev 7.3’ which algorithm is based on marching cubes method [6]. Material property for calculation is shown in Table.2. The extractants, metal ions and other chemical compounds are ignored and physical values of water and n-dodecane are used.

Table 1. Calculation Condition and Settings

<table>
<thead>
<tr>
<th>Dimension</th>
<th>Mesh</th>
<th>Time Stepping Method</th>
<th>Turbulent Model</th>
<th>Wall Function</th>
<th>Rotating Speed</th>
<th>Inlet Velocity of Liquid</th>
<th>Outlet Velocity of Liquid</th>
</tr>
</thead>
<tbody>
<tr>
<td>3D</td>
<td>3 million</td>
<td>Variable(*)</td>
<td>RNG k-epsilon</td>
<td>Standard</td>
<td>400,800,1200(rpm)</td>
<td>10 (cc/min)</td>
<td>10 (cc/min)</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Solver Method</th>
<th>Unsteady Formulation</th>
<th>Gradient Option</th>
<th>Velocity Formulation</th>
<th>Momentum</th>
<th>Surface Tracking</th>
</tr>
</thead>
<tbody>
<tr>
<td>PISO</td>
<td>1st Order Implicit</td>
<td>Least Square Cell Based</td>
<td>Absolute</td>
<td>2nd Order Upwind</td>
<td>Volume of Fraction Method</td>
</tr>
</tbody>
</table>

(*)Global Courant Number is fixed to be 2 as constant value and time step is automatically adjusted.

Table 2. Material Properties for calculation

<table>
<thead>
<tr>
<th>Aqueous phase</th>
<th>Density</th>
<th>Viscosity</th>
<th>Organic Phase</th>
<th>Density</th>
<th>Viscosity</th>
<th>Contact Angle</th>
<th>Surface Tension</th>
</tr>
</thead>
<tbody>
<tr>
<td>Water-liquid</td>
<td>998.2 kg/m³</td>
<td>0.001003 k g/ms</td>
<td>n-dodecane</td>
<td>748.7 kg/m³</td>
<td>0.001383 k g/ms</td>
<td>30,90,120degree</td>
<td>0 – 25 mN/m</td>
</tr>
</tbody>
</table>

4. Results and Discussion

4.1. Behavior of oil-water dispersion

Fig.2 shows the simulated results of oil-water dispersion from 0 to 1 second after starting rotation of the inner rotor with VOF model under the conditions of 1200rpm of rotating speed, 5mN/m of surface tension and 30degree of contact angle. In the results, organic phase is colored by the tangential velocity and therefore, blue part indicates 0 m/s of tangential velocity of organic phase. Due to the huge calculation time, initial state is set as shown in (a) and sensitivity survey was done. 0-1 seconds seems to be short, but to see the effect of each parameter such as rotating speed, property of inner rotor and interfacial tension. With the inducement of Taylor
vortexes, organic phase is dispersed following 3 stages. Firstly, separation of oil columns by tangential interface vibration (Reyleigh instability) is occurred. Secondly, interface vibration in the direction of long axis is observed along with the formation of the Taylor vortexes. Finally, dispersion of organic phase by the strong shearing stress by the Taylor vortexes and the interfacial area increases effectively. And the dispersed organic phase flows consequently through the series of Taylor vortexes. In the extraction tests, similar behaviour was observed.

4.2. Effect of the surface tension and the property of inner rotor surface

To see the effect of the surface property of inner rotor on dispersion behavior, contact angles between fluids and the outer wall are changed as shown in Fig.3(a), (c) and (d), and surface tension between organic and aqueous phases in Fig.3(a) and (b). The surface tension of n-dodecane and water is known as 25 mN/m at ambient temperature. However, calculation under 25 mN/m shows insufficient dispersion of organic phase. This is because that metal complex is formed in the actual extractor and it shows the interfacial activation effect [7][8]. Therefore, it is confirmed that the formation of metal complex and its amount has a great importance on extraction performance. In (c) and (d) of Fig.3, the effect of contact angle of liquid is shown. From these results, it is clarified that the inner rotor has a great impact on the oil-water dispersion by numerical sensitivity analysis. Thus, to study the actual effect of the surface property of inner rotor, Teflon and stainless steel are tested as the material of inner rotor and contentious extraction tests were done. In (a) to (d) of Fig.4 show the relation between the actual dispersion behavior and property of inner rotor surface. In case of 400rpm with Teflon surface, organic phase adheres to the inner rotor and dim spiral flow was observed, but in case of stainless steel rotor, clear upwards and spiral flow was observed. This means that stainless steel has better lipophilic character and has lower contact angle. Under the condition of 800rpm, stainless steel rotor shows the better dispersion. From these results, it is clarified that the property of the inner rotor surface is important by the experimental approach.
Fig. 3. Sensitivity study of surface tension and contact angle of inner rotor on oil-water dispersion, (a) 25 mN/m and 90-degree; (b) 0 mN/m and 90-degree; (c) 25 mN/m, 30-degree; (d) 25 mN/m, 120-degree.

Fig. 4. Actual behavior of oil-water dispersion; (a) 800 rpm with Teflon rotor; (b) 800 rpm with stainless steel rotor; (c) 400 rpm with Teflon rotor; (d) 400 rpm with stainless steel rotor; (e) n-dodecane on the Teflon rotor; (f) n-dodecane on stainless steel rotor (cannot stay onto the rotor).
4.3. Extraction efficiency

The results of continuous extraction tests using Teflon and stainless steel rotors were shown in Fig.5(a) and (b). Dotted lines in these figures stand for the percent extraction under the condition that the number of equilibrium stage is equivalent to one. The concentrations of each chemical compound in organic and aqueous phases were adjusted as the percent extraction became approximately 50 % with the batch extraction tests. The percent extractions for the stainless steel rotor were higher than those for the Teflon rotor. These results indicate that the improvement of lipophilicity of inner rotor surface is suitable for higher percent extraction (theoretical stages) and also indicates that the surface property such as roughness of the rotor surface may also affect on the behavior of oil-water dispersion and thus the percent extraction is affected. When the rotating speed is increased from 800 to 1200 rpm, the percent extraction is decreased for the extractor with the stainless steel rotor. This is because the entrainment of organic phase into aqueous phase occurred greatly. If the oil entrainment is restrained by the use of an inner rotor, on which oil can be adsorbed easily, the extraction performance will be further improved.

![Fig. 5. Effect of inner rotor property on percent extraction(a) Teflon; (b) stainless steel](image)

5. Conclusions

- Oil-water dispersion behavior in the liquid-liquid centrifugal extractor is simulated using commercial CFD code and results were visualized. The similar interfacial behavior is observed in extraction tests.
- By the sensitivity study using numerical simulation, it is clarified that the decrease in surface tension by the formation of metal-extractant complex has a great impact on oil-water dispersion behavior.
- By numerical and experimental approaches, it is clarified that the property of inner rotor surface affects greatly on the oil-water dispersion behavior and results in the difference of percent extraction.
- The inner rotor with lipophilic surface is suitable for fine dispersion of the organic phase and better extraction performance due to the increased interfacial area were confirmed.
- When the rotating speed is too high, the occurrence of entrainment made percent extraction worse.

By the further study to clarify the effect of inner rotor surface such as roughness, contact angle or functional groups on the inner rotor surface, better extraction performance by the single extractor can be achieved. Research cost will also be reduced greatly by applying sensitivity analysis with numerical simulation. The combination of numerical approach and experimental approach is the most effective way to develop a small and high performance extractor.
Reference


