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Dynamic modelling of an industrial copper solvent extraction process

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Abstract

The dynamic behaviour of an industrial copper solvent extraction mixer–settler cascade is modelled to develop an advanced process control system. First, the process is introduced and the dynamical models are formulated. The testing environment is described and the successful results presented. Only industrially measured variables are required and plant-specific McCabe-Thiele diagrams are utilized to predict copper concentrations. The results with constant and adapted parameters are compared and the importance of parameter adaptation is discussed. Testing the simulator with adapted parameters over a period of 1 month of industrial operating data gave data that followed the real process measurements closely. In the future, the mechanistic models will be used for control system development and testing. The model can be used on all copper solvent extraction plants by modifying the flow configuration and adapting parameters.

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Keywords:
Copper solvent extraction; Dynamic simulation; Modelling; Mechanistic models; Mixer–settler; Process control

1. Introduction

The copper leaching, solvent extraction and electro-winning (LX/SX/EW) process is one of the most important options in the production of copper from low-grade oxidized ore. Research on the copper LX/SX/EW process has so far mainly focused on the process equipment and control of the plants relies on basic control loops. Advanced control of a solvent extraction plant would make it possible to keep the process variables close to their optimal values, thus increasing the amount of copper produced and reducing the amount of chemicals and energy consumed (Kordosky, 2002; Jämsä-Jounela, 2001; Bergh et al., 2001; Hughes and Saloheimo, 2000).

The motivation of this study is to model the dynamic behaviour of an industrial copper solvent extraction mixer–settler cascade in order to develop an advanced control system for the process. During the last decade, the modelling and control research of copper liquid–liquid extraction has focused on extraction columns (see review by Mjalli et al., 2005).

According to Wilkinson and Ingham (1983) and Ingham et al. (1994), the most common dynamic models for mixer–settler cascades consist of two steps, mixing and settling, the mixer being modeled as an ideal mixer and the settler as separated plug flows for aqueous and organic phases. The mixer model is based on the two-film theory of interfacial mass transfer, and requires the individual film coefficients for both phases and the equilibrium curve. Similar models have also been developed for extraction columns (Steiner and Hartland, 1983).
A dynamical, pulsed flow model for a rare-earth solvent extraction cascade has been developed by Wichterlová and Rod (1999). The model for one mixer–settler pair is separated into one mixing and several settling steps, in which each of the steps has equal time intervals. In the mixing step, the metal is transferred between the aqueous and organic phases according to the linear equilibrium relationship. The settling steps are modelled as ideal mixers, separately for each phase.

A mathematical steady state simulation model for a copper SX/EW pilot plant has been developed by Aminian et al. (2000). In the solvent extraction part, only mixing is modeled. The mixer model is based on mass conservation and it includes both transfer to the interfacial surface of the phases and the reaction rate over the surface.

The challenge and novelty of this study is to develop mechanistic models of liquid–liquid extraction that require only industrially measured variables and utilize plant-specific McCabe-Thiele diagrams to predict the copper concentrations of an industrial copper solvent extraction plant. In this paper, the process is first briefly introduced, the modeling is then described in detail and, finally, the simulation results are presented and discussed.

2. Process description

The aim of the copper solvent extraction process is to concentrate aqueous copper solution from a few g/l to about 40–50 g/l and to purify the solution from ferrous, manganese, chloride and other impurities harmful for the electrowinning process. This continuous process consists of extraction and stripping processes, both of which may contain several unit operations. In the extraction steps, copper is transferred from the lightly acid aqueous solution (PLS) to the organic solution and in the stripping is transferred to strongly acid aqueous electrolyte solution. Copper transfer between the aqueous and organic phases takes place in mixers, and the phases are separated in settlers.

Generally, the process has two input flows, the pregnant leach solution (PLS) and the lean electrolyte (LE), and one recycling flow, the organic solution. In the extraction stage, the copper is extracted from the PLS to the barren organic (BO) solution. In the stripping stage, copper is stripped from the loaded organic solution (LO) to lean electrolyte (LE) in the stripping unit. The resulting rich electrolyte solution (RE) is led to the electrowinning process. The barren organic solution (BO) is recycled back to the extraction unit.

In industry, mixer–settler types of equipment are used (Robinson, 2003). The mixer–settler design varies considerably; for example, in the Outokumpu VSF technology, the organic and aqueous flows are combined in a dispersion pumping unit followed by two mixers and a large settler (Nyman et al., 2003).

Typical process instrumentation includes flow rate, temperature and level measurements. In addition, conductivity in the mixers, pH and copper and impurity online concentration measurements may be included. The copper and impurity assays, phase ratio, phase separation time and other diagnostic measurements are performed by the process operators and by the laboratory.

3. Dynamic modelling

The solvent extraction process studied consists of four unit operations, three of which are for extracting copper from the aqueous phase to the organic phase and one that strips copper from the organic to the electrolyte. The inputs of the process are pregnant leach solution (PLS) flow rate and concentration, lean electrolyte flow rate and concentration, and the flow rate of the organic solution. The organic solution is recycled in the process, but the flow rate can be manipulated through the organic storage tank (Fig. 2).

The solvent extraction process is modelled considering only the mass transfer of copper between organic and aqueous phases. Each unit process is modelled as combination of an ideal mixer and plug flow with online estimated parameters. The model requires online measurements of flow rates, copper concentrations and organic tank level. The parameters related to the McCabe-Thiele diagrams require offline measurements of the copper concentrations and chemical organic to aqueous ratios from all the mixers, pH of the PLS, acidity of the electrolyte and maximum loading of the organic.
The solvent extraction process. The inputs are the concentration of PLS (cPLS) and lean electrolyte (cLE), and flow rates of PLS (fPLSS and fPLSP), organic (fLO) and lean electrolyte (fLE). The output variables are the concentration of the raffinates (cRaffP and cRaffS), rich electrolyte (cRE), and the concentration of the recycled organic stream, barren organic (cBO) and loaded organic (cLO).

The basic assumptions, described by Wilkinson and Ingham (1983), are applied in this study: perfect mixing in the mixer, immiscibility of the two phases, no mass transfer and plug flow in settler. The modifications to these assumptions are that the equilibrium curve in the mixer is a plant-specific non-linear McCabe-Thiele isotherm, and the output value is calculated on the basis of the equilibrium isotherm. The mass transfer coefficients \( K_0 \), equilibrium isotherm parameters \( A \) and \( B \) for extraction, and \( C \) and \( D \) for stripping and efficiency coefficients \( \alpha \) are estimated from the offline plant measurements.

The copper transfer is calculated from the ideal mixing equations, where the equilibrium value \( c^* \) is determined on the basis of the incoming flow rates, concentrations and plant-specific McCabe-Thiele diagram with estimated parameters. The variables are marked as follows: flow rates \( F \), the mixing volumes \( V_{mix} \), organic concentrations \( c_{org} \), aqueous concentrations \( c_{aq} \), mass transfer coefficients \( K \) and efficiency parameters \( \alpha \). The settler, always following the mixer, is described by a pure time delay \( t_c \).

In extraction, copper is transferred from the aqueous to the organic phase. Each of the three extraction unit operations are modelled by differential equations of the concentrations for both organic (\( dc_{org}^{aq}(t)/dt \)) and aqueous phases (\( dc_{aq}^{aq}(t)/dt \)):

\[
\frac{dc_{org}^{aq}(t)}{dt} = \frac{F_{org}^{aq}(t)}{V_{mix,1}(t)} \cdot [c_{org}^{aq}(t-t_0) - c_{org}^{aq}(t)] \\
+ K_1 \left[ c_{org}^{aq}(t) - c_{org}^{aq*}(t) \right]
\]

(1)

\[
\frac{dc_{aq}^{aq}(t)}{dt} = \frac{F_{aq}^{aq}(t)}{V_{mix,1}(t)} \cdot [c_{org}^{aq}(t-t_0) - c_{org}^{aq}(t)] - K_1 \left[ c_{org}^{aq}(t) - c_{org}^{aq*}(t) \right]
\]

(2)

\[
c_{org}^{aq*}(t) = g(c_{org}^{aq}(t-t_0), c_{org}^{aq}(t), F_{org}^{aq}(t), F_{aq}^{aq}(t), \alpha_1, A, B)
\]

(3)

\[
\frac{dc_{org}^{el}(t)}{dt} = \frac{F_{org}^{el}(t)}{V_{mix,4}(t)} \cdot [c_{org}^{el}(t-t_0) - c_{org}^{el}(t)] \\
+ K_3 \left[ c_{org}^{el}(t) - c_{org}^{el*}(t) \right]
\]

(4)

\[
\frac{dc_{aq}^{el}(t)}{dt} = \frac{F_{aq}^{el}(t)}{V_{mix,4}(t)} \cdot [c_{org}^{el}(t-t_0) - c_{org}^{el}(t)] - K_3 \left[ c_{org}^{el}(t) - c_{org}^{el*}(t) \right]
\]

(5)

\[
c_{org}^{el*}(t) = g(c_{org}^{el}(t-t_0), c_{org}^{el}(t), F_{org}^{el}(t), F_{aq}^{el}(t), \alpha_2, A, B)
\]

(6)

The first raffinate (cRaffP) concentration is the time delayed value of the aqueous concentration from the first unit process; second raffinate (cRaffS) is the time delayed value of the aqueous concentration from the second unit process; and loaded organic (cLO) is the time delayed value of the organic concentration from the third unit process.

\[
c_{RaffP}^{org} = c_{org}^{aq}(t-t_1)
\]

(10)

\[
c_{RaffS}^{org} = c_{org}^{aq}(t-t_2)
\]

(11)

\[
c_{LO}^{org} = c_{org}^{aq}(t-t_3)
\]

(12)

In the stripping, copper is transferred from the organic to the electrolyte solution. The stripping unit operation is modelled by differential equations of the concentrations for both electrolyte (\( dc_{ele}^{aq}(t)/dt \)) and organic (\( dc_{ele}^{org}(t)/dt \)) phases:

\[
\frac{dc_{ele}^{el}(t)}{dt} = \frac{F_{ele}^{el}(t)}{V_{mix,4}(t)} \cdot [c_{ele}^{el}(t-t_0) - c_{ele}^{el}(t)] \\
+ K_4 \left[ c_{ele}^{el}(t) - c_{ele}^{el*}(t) \right]
\]

(13)

\[
\frac{dc_{ele}^{org}(t)}{dt} = \frac{F_{ele}^{org}(t)}{V_{mix,4}(t)} \cdot [c_{ele}^{org}(t-t_0) - c_{ele}^{org}(t)] \\
- K_4 \left[ c_{ele}^{el}(t) - c_{ele}^{el*}(t) \right]
\]

(14)

\[
c_{ele}^{el*}(t) = h(c_{ele}^{el}(t-t_1), c_{ele}^{el}(t), F_{ele}^{el}(t), F_{ele}^{org}(t), \alpha_4, C, D)
\]

(15)
Rich electrolyte concentration (cRE) is the time delayed value of electrolyte concentration and barren organic (cBO) is the time delayed value of organic concentration in the stripping unit.

\[
c_{RE}(t) = c_{el}^1(t-t_4)
\]

\[
c_{BO}(t) = c_{org}^4(t-t_4)
\]

3.1. Equilibrium values

The theoretical equilibrium values for extraction and stripping are determined from the McCabe-Thiele diagram, presented in Fig. 3. The isotherm curves for extraction and stripping are assumed to be constant, or changing remarkably slower than the other process dynamics, whereas the operating lines are changing every sampling time according to the organic to aqueous volume ratio in the mixers. Each step in the diagram presents one mixer–settler pair.

The equilibrium value in one mixer–settler is the coincidence point of the equilibrium isotherm and the inversed operating line, weighted by the efficiency coefficient \( \alpha \). In extraction, the equilibrium isotherm is non-linear

\[
c_{org} = \frac{Ac_{aq}}{B + c_{aq}}
\]

and in stripping linear

\[
c_{org} = C \cdot c_{aq} + D
\]

In extraction, the theoretical equilibrium point (100% efficiency) is determined by:

\[
y = Ax/(x + B) = ax + b \Leftrightarrow ax^2 + (Ba-A+b)x + Bb = 0 \Rightarrow x^* = 1/2a \left( -(Ba-A+b) - \sqrt{(Ba-A+b)^2 - 4ab} \right).
\]

The equilibrium value for the aqueous concentration is the efficiency weighed theoretical value:

\[
c_{aq}^* = \alpha x^* + (1-\alpha) c_{aq}^0
\]

The equilibrium value for the organic concentration is the efficiency weighed theoretical value:

\[
c_{org}^* = \alpha y^* + (1-\alpha) c_{org}^0
\]

Above \( A \) and \( B \) are the equilibrium isotherm parameters, the slope of the operating line is:

\[
a = -F_{aq}/F_{org},
\]

and \( b \) is the linear term combining the input concentrations of the organic and aqueous phases

\[
b = c_{org}^0 - a \cdot c_{aq}^0
\]

In stripping, the isotherm parameters are \( C \) and \( D \), and \( a \) and \( b \) as before; the equilibrium point is solved by:

\[
y = Cx + D = ax + b,
\]

resulting in the theoretical equilibrium concentration of the aqueous phase \( x^* \):

\[
x^* = \frac{b-D}{C-a}.
\]

The equilibrium values for the aqueous and organic concentrations are calculated from Eqs. (21) and (22).

3.2. Efficiency

The efficiency of extraction and stripping is affected by the reagent concentration in the organic solution and the acidity of the aqueous solution. The retention time in the mixers and settlers also have an effect, especially if they are too short (Ritcey and Ashbrook, 1998).
The efficiency for each unit process is estimated on the basis of the theoretical equilibrium value $x^*$ (aqueous) and $y^*$ (organic), the input concentration $c_{in}$ and the actual equilibrium value $c_{out}^*$. The efficiency parameter has usually values close to 1.

$$\eta_E = \frac{c_{org}^* - c_{org}^{aq*}}{c_{org}^*} = \frac{c_{org}^* - c_{in}^*}{c_{org}^{aq*} - c_{in}^*}$$ (27)

Calculation of efficiency for stripping unit is presented in Fig. 4. The concentrations of rich electrolyte (RE) and barren organic (BO) are determined by drawing an inversed operating line from the input point (lean electrolyte, loaded organic) towards the stripping isotherm. The point where this line and the isotherm overlap is the theoretical equilibrium value for rich electrolyte and barren organic. These values are then weighed with the experimental efficiency parameter to get the true output values RE* and BO*.

3.3. Mass transfer coefficient

The mass transfer coefficient $K$ describes the speed of concentration change in the mixers. It is inversely related to the process time constant: the higher $K$, the shorter time constant. The mass transfer coefficient $K$ is approximated from the process model by setting it to the difference term $(c_{out}^* - c_{out})$. 

$$V \frac{dc_{out}}{dt} = F(c_{in} - c_{out}) - K(c_{out}^* - c_{out})V = 0 \Leftrightarrow K = \frac{F(c_{in} - c_{out})}{V(c_{out}^* - c_{out})} = \frac{F(c_{in} - c_{out})}{V \cdot \varepsilon}.$$ (28)

Using this procedure, the $K$ values are between 50 and 400 l/s for the studied process. Wilkinson and Ingham (1983) suggest giving arbitrary high value for the $K \cdot V$ term. The modelling approach in Ingham et al. (1994) is slightly different, using constant mass transfer coefficient $K = 25$ l/s.

3.4. Equilibrium isotherms

The extraction and stripping equilibrium isotherms are nonlinear functions of various process variables, such as temperature, pH and reagent strength. In this study, the equilibrium isotherm data available included only reagent volume % in the organic, pH of the PLS solution and acidity of the lean electrolyte. The equilibrium isotherm parameters $A$, $B$, $C$ and $D$ are estimated daily from the corresponding laboratory measurements: The following model structures were found to best describe the plant equilibrium isotherms:

The extraction equilibrium isotherm was estimated:

$$y^* = \frac{A \cdot x^*}{B + x^*} = \frac{0.9877 \cdot ML \cdot x^*}{cH^{1.1338} \cdot (0.2131 \cdot cPLS - 0.0298 \cdot cBO) + x^*}$$ (29)

where ML is the maximum load, $cH$ is $10^{-pH}$, $x^*$ is the aqueous equilibrium concentration and $y^*$ the organic equilibrium concentration.

Stripping equilibrium isotherm was estimated as:

$$y^* = Cx^* + D = 0.0972 \cdot x^* + 52.2116 \cdot \frac{\text{vol}^0.3}{\text{acid}^0.5}$$ \quad -10.9432 (30)

where vol% is the maximum load (ML) times the upload factor, acid is the acidity of the lean electrolyte, $x^*$ is the electrolyte equilibrium concentration and $y^*$ the organic equilibrium concentration.

4. Simulation model

A dynamic simulator of the process was constructed on the basis of the mechanistic models presented above. The aim of the simulator is to study the dynamic

| Table 1 |
| Inputs, parameters and outputs |

<table>
<thead>
<tr>
<th>Variable name</th>
<th>Abbreviation</th>
</tr>
</thead>
<tbody>
<tr>
<td>Inputs</td>
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</tr>
<tr>
<td>PLS concentration</td>
<td>cPLS</td>
</tr>
<tr>
<td>Lean electrolyte Concentration</td>
<td>cLE</td>
</tr>
<tr>
<td>PLSP flow rate</td>
<td>fPLSP</td>
</tr>
<tr>
<td>PLSS flow rate</td>
<td>fPLSS</td>
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<tr>
<td>Organic flow rate</td>
<td>fO</td>
</tr>
<tr>
<td>Lean electrolyte flow rate</td>
<td>fLE</td>
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<tr>
<td>Parameters</td>
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</tr>
<tr>
<td>Mass transfer coefficients</td>
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</tr>
<tr>
<td>Efficiency coefficients</td>
<td>$\alpha_1, \alpha_2, \alpha_3, \alpha_4$</td>
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<tr>
<td>O/A correction coefficients</td>
<td>$c_{f1}, c_{f2}, c_{f3}, c_{f4}$</td>
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<tr>
<td>Extraction isotherm parameters</td>
<td>$A, B$</td>
</tr>
<tr>
<td>Stripping isotherm parameters</td>
<td>$C, D$</td>
</tr>
<tr>
<td>Outputs</td>
<td></td>
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<td>Loaded organic concentration</td>
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<tr>
<td>Barren organic concentration</td>
<td>cBO</td>
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<tr>
<td>Rich electrolyte concentration</td>
<td>cRE</td>
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<td>Raffinate P concentration</td>
<td>cRaffP</td>
</tr>
<tr>
<td>Raffinate S concentration</td>
<td>cRaffS</td>
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</table>
behavior of the solvent extraction process and, in the future, to provide a test bench for the control system. The unit operation models were implemented in Matlab Simulink according to the plant configuration.

The inputs to the model are the flow rates of PLS, organic and electrolyte, and copper concentrations for PLS and lean electrolyte. The outputs are the copper concentrations of raffinates and rich electrolyte. Barren

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**Fig. 5.** Input concentrations, PLS and lean electrolyte, scaled values.

---

**Fig. 6.** Input flow rates, PLS series, PLS parallel, organic and electrolyte, scaled values.
and loaded organic concentrations are states. The inputs, parameters and outputs are listed in Table 1.

The flow rates, copper concentrations and organic tank level were measured every 10 min. The parameters were estimated daily from the offline measurements of the copper concentrations and chemical organic to aqueous ratios from all the mixers, pH of the PLS, acidity of the electrolyte and maximum loading of the organic. The organic to aqueous ratio is corrected on the basis of the chemical O/A ratio measurement from the mixers.

Since the industrial data was extremely noisy, especially for the organic concentration and flow rate measurements, it was necessary to apply filtering. The first step was to remove clear outliers, i.e. those over the maximum or under the minimum limit. The aim of filtering was to remove part of the noise, but to keep the dynamics of the variables. Zero-phase digital filtering was assumed to be appropriate for the simulation purposes and therefore Matlab's filtfilt method with moving averaging was chosen. Several averaging periods were tested and compared to each other by visual and statistical examination of the residuals between real data and filtered data. The best results were obtained with a 4-h averaging period for the copper concentration and flow rate measurements.

5. Testing results

The simulator was tested with one month of operating data. The first aim was to validate the ability of the mechanistic models to describe the process, i.e. to follow the process trends. The second aim was to compare the performance of the simulator with adapted and constant parameters. The results of the different test runs were compared by absolute mean of the residual and the standard deviation of the residuals, calculated as percentage of the mean value of the corresponding variable.

The inputs are show in Figs. 5–7, first the concentrations of PLS and lean electrolyte, then the flow rates and then the adapted parameter values. The PLS concentration level rises and the lean electrolyte concentration varies a lot during the experiment. The flow rates also vary remarkably with clear step changes in the PLS flow rates. Due to process changes, the isotherm parameters are rising during the experiment, while efficiency and

![Fig. 7. Adapted values of the efficiency, mass transfer and isotherm parameters, scaled values.](image-url)
mass transfer parameters seem to be varying more independently.

In the nominal test (EKI), all the parameters were adapted and the simulated outputs were compared against real process measurements. The statistical results, shown in Tables 2 and 3, were good; the mean residual well below 2% for organics and rich electrolyte and around 6% for raffinates, which is very good considering the poor measurement accuracy of these streams. Also visual examination of the results concludes that the modelling was successful; the outputs followed smoothly the dynamic trends, as shown in Figs. 8–12.

To study further the effect of each of the parameters, in the following tests, some of the parameters were kept constant while the others were changing. The

<table>
<thead>
<tr>
<th>Test</th>
<th>LO</th>
<th>BO</th>
<th>RE</th>
<th>RaffS</th>
<th>RaffP</th>
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<td>1.25</td>
<td>6.09</td>
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<tr>
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<td>1.25</td>
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<td>5.98</td>
</tr>
<tr>
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<td>2.34</td>
<td>1.26</td>
<td>8.18</td>
<td>6.03</td>
</tr>
<tr>
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<tr>
<td>– (%)</td>
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<td>4.32</td>
<td>1.04</td>
<td>18.56</td>
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</table>

<table>
<thead>
<tr>
<th>Test</th>
<th>LO</th>
<th>BO</th>
<th>RE</th>
<th>RaffS</th>
<th>RaffP</th>
</tr>
</thead>
<tbody>
<tr>
<td>EKI (%)</td>
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<td>2.46</td>
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<tr>
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<tr>
<td>E (%)</td>
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<tr>
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<tr>
<td>– (%)</td>
<td>2.88</td>
<td>5.33</td>
<td>1.33</td>
<td>19.18</td>
<td>10.65</td>
</tr>
</tbody>
</table>

**Fig. 8.** Loaded organic concentration, measured and simulated (dashed), scaled values.

**Fig. 9.** Barren organic concentration, measured and simulated (dashed), scaled values.
Fig. 10. Rich electrolyte concentration, measured and simulated (dashed), scaled values.

Fig. 11. Raffinate series concentration, measured and simulated (dashed), scaled values.

Fig. 12. Raffinate parallel concentration, measured and simulated (dashed), scaled values.
constant parameters are mean values of the adapted parameters. The input concentrations and flow rates were the same in all the experiments.

In the second test (EI), the mass transfer parameters were kept constant and the results are almost as good as with nominal test, which implies that adapting this parameter might not be particularly important. In the third test (EK), the isotherm adaptation is dropped out, but both efficiency and mass transfer parameters are adapted. Now the results are clearly worse than the base case—which concludes that the isotherm parameter adaptation is important.

The fourth test (E) was run by only adapting efficiency parameters. The results are very similar to the previous case with efficiency and mass transfer parameter adaptation, but still worse than the nominal case. When efficiency parameters are kept constant (fifth and sixth test runs), the results are drastically decreased for the organic and raffinate concentrations. In the fifth run (I), only the isotherms are adapted and in the last run (–) all the parameters are constant. This concludes that the parameter adaptation, especially for the efficiency parameters, is utmost important for the modelling accuracy.

6. Conclusions

A dynamic process simulator was constructed on the basis of the mechanistic models. The simulator with adapted parameters was tested with 1 month of industrial operating data. The simulated data smoothly followed the real process measurements during the test period and the residuals were sufficiently small. The effect of the parameter adaptation was studied by keeping some of the parameters constant and by comparing the results to the nominal case. The results clearly indicated the necessity for efficiency parameter adaptation.

The model accuracy can be further be increased by using nonlinear parameter estimation methods such as extended Kalman filtering. In the future, the mechanistic models will be used for control strategy and algorithm development. The simulator will be utilized as test bench for the control system. The model can be applied to all copper solvent extraction plants using mixer–settlers by modifying the flow configuration and adapting the parameters.

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