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Synthesis of hydrometallurgical processes for valorization of secondary raw materials using ant colony optimization and key performance indicators

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ABSTRACT

An algorithm-based method for synthesis of hydrometallurgical processes using limited amounts of experimental data is presented. The method enables simultaneous selection and sequencing of unit operations and optimization of operating parameters. An ant colony optimization (ACO) based algorithm is used to identify the most economic process alternative in an iterative manner. Key performance indicators are used for comparison of candidate processes: a purification performance index measures purity improvement and a separation cost indicator is used as an objective function in process optimization. Computational times were reduced significantly with the suggested method compared to an algorithm which evaluates all the possible process options. The practical applicability of the method to hydrometallurgy is demonstrated by investigating zinc recovery from argon oxygen decarburization dust with two alternative leaching methods and recovery of lanthanides from nickel metal hydride (NiMH) batteries. In the first zinc recovery process, 150 min normal batch leaching with 0.5 M H_2SO_4 is used, and in the other one 270 min batch leaching with H_2SO_4 is done by controlling the pH (>3.0). In both cases the leachate is extracted with D2EHPA at pH 4.27, and stripped with circulating solution from zinc electrolysis. For lanthanides recovery the algorithm suggested a process in which the raw material is leached with 1.3 M HCl, the leachate is extracted with D2EHPA at pH 2.2, organic phase is stripped with 2.0 M HCl and 99% pure Ln-oxalates are precipitated with oxalic acid at pH 0.6. Compared to previously suggested process for the same raw material, the algorithm suggests operating the leaching step such that higher selectivity is achieved by sacrificing some yield.

Abbreviations

ACO	ant colony optimization
AOD	argon oxygen decarburization
CPU	central processing unit
EDR	energy dissipation rate
IRR	internal rate of return
KPI	key performance indicator
PPI	purification performance index
SCI	separation cost indicator

Notation

A	flow rate, m^3/s
c_i	concentration of contaminants, kg/m^3
E	concentration of extractant, m^3/m^3
K	cost, item of expenses, €/kg
k_L	specific cost of a leaching step, €/kg
$k_{pur,l}$	specific cost of a purification step, €/kg
L	probability
M	number of components in a chemical system
N	number of ants in a colony
n	number of process steps
P	number of discrete values of operating parameters
SL	solvent loss in solvent extraction
T	concentration of a target metal in the system, kg/m^3
t_b	batch time in leaching, s
U	number of unit operations
V_L	volume of leaching vessel, m^3
x	purity
Y	yield

Greek symbols

α	the degree of importance of the pheromones
ξ	parameter used to control the scale of the global updating of the pheromone
$\tau_{l,u,p}$	amount of pheromone in a cell
ρ	pheromone decay factor

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Subscripts

0	initial
b	batch
chem	chemicals
el	electricity
extr	extractant
f	final
i	contaminant
L	leaching
l	process step
org	organic phase
p	value of operating parameter
raf	raffinate
sol	solvent
str	stripping
tot	total process SCI
u	unit operation

Superscripts

a	aqueous phase
k	ant
raf	raffinate

1. Introduction

Hydrometallurgical process development usually starts with analysis of the raw materials to be treated, i.e., chemical composition, mineralogy, state, particle size, volume etc. (Forsén and Aromaa, 2013). The process itself typically consists of three consecutive main steps: leaching, concentration and purification, and final product recovery. Several alternative unit operations are available for each process step. For instance solvent extraction, ion exchange, and/or selective precipitation can be employed for purification and concentration, and crystallization, chemical precipitation or electrowinning for product metal recovery. Moreover each unit operation can be run under a wide range of operating conditions (pH, pressure, phase ratio, solvent type, etc.). A key to successful and efficient hydrometallurgical purification is identification of the most suitable sequence of unit operations and the most effective combination of operating parameters to obtain the desired purification and yield with minimum (economical) effort.

Cisternas (1999) identified lack of works devoted to design of complete process due to complexity of the problem and great number of variables and restrictions to consider in his extensive review on synthesis of processes in extractive metallurgy and inorganic chemistry. To decrease the size of the problem process steps are usually designed individually (Cisternas, 1999), so that there are many methods and techniques available for design of each process step (Gálvez et al., 2004; Alonso et al., 2001; Trujillo et al., 2014). However, synthesis of complete processes is potentially more efficient since process step interactions are taken into account (Angira and Babu, 2006; Cisternas, 1999).

When a new hydrometallurgical process is being developed, comparison between process alternatives and process optimization is usually done based on the experience of scientists and engineers, as well as on extensive experimentation (Rintala et al., 2011). Over-expenditure on reagents, experimental biases, complicated data processing and the complexity of considering several process parameters simultaneously prolong the course of hydrometallurgical process development at its early stages and contribute to inefficiency.

Hydrometallurgical purification process development is usually based on scale-up of processes established on a laboratory or pilot scale. Conceptual design or process synthesis in the early stage is thus viewed as the most important stage of process development (Cziner et al., 2005). Major decisions affecting the lifecycle of the process are made during development of the first process flowsheet. Experience-based process synthesis can often result in overall suboptimal processes with inefficient utilization of energy and auxiliary materials (Nfor et al., 2009). Therefore, systematic process development based on identification of justified optima is essential for efficient utilization of time and resources.

Nfor et al. (2009) identified four types of process synthesis strategies applicable to chemical industries: heuristics or knowledge-based strategies (Cziner et al., 2005), optimization-based strategies (Steimel et al., 2013; Grossmann and Daichendt, 1996), high-throughput experimentation strategies (Bhambure et al., 2011; Schuldt and Schembecker, 2013) and a combination of the aforementioned strategies (Ahamed et al., 2006). Each approach has strengths and weaknesses as discussed elsewhere (Nfor et al., 2009). Mathematical optimization based method can offer significant advantages to hydrometallurgical process development: clarification of interactions between unit operations, utilization of validated models for process optimization, user-independence after formulation of the search space, and the ability to identify the optimal process meeting the set criteria (Nfor et al., 2009). Application of mathematical optimization requires a superstructure of process alternatives and the availability of useful objective functions.

Numerical measures for assessment of process performance are required for efficient application of optimization based method. These measures have to reflect the main features of the alternative unit operations and form a reliable base for comparison. The main criteria for decisions on process synthesis in extractive metallurgy are technical feasibility and economic potential, along with environmental, safety and other aspects (Linninger, 2002; Chakraborty et al., 2004). It is desirable to base process synthesis decisions upon costs over the complete process. However, at the very beginning of process development, before process concepts are available, such information is not available and the profitability of a process or its internal rate of return (IRR) cannot be precisely estimated. The use of the key performance indicators (KPIs) introduced by Winkelnkemper and Schembecker (2010) offers a potentially effective approach to address this problem. The KPIs were developed for rating purification and cost-efficiency on the basis of single step purity improvement, yield and specific costs. The indicators do not require complete mass and energy balances and can be applied from the beginning of experimental investigation. Although the KPIs were first introduced for evaluation of pharmaceutical bio-separation processes, they are equally valid for hydrometallurgy.

Solution of an optimization problem requires a suitable and efficient algorithm that is capable of identifying the minimum value of the target function and the corresponding sequence of unit operations and their operating parameters. Mathematical programming algorithms and methods available for synthesis of chemical processes have been the subject of a number of reviews (Grossmann and Daichendt, 1996; Grossmann et al., 1999; Acevedo and Pistikopoulos, 1998). Algorithms based on extensive searches for the optimal solution are computationally not preferred due to the high computational efforts required (Raeesi et al., 2008). However, the problem can be addressed in an efficient manner by using meta-heuristics to find approximate solutions (Raeesi et al., 2008; Biswas et al., 2009). The stochastic meta-heuristic ant colony optimization (ACO) algorithm has been found to be promising for efficient synthesis and optimization of processes (Raeesi et al., 2008; Chunfeng and Xin, 2002).

The objective of this research was development of an algorithm-based method for synthesis of hydrometallurgical processes using experimental data. Key theoretical aspects of in silico hydrometallurgical process development using ant colony optimization (ACO) and key performance indicators (KPIs) are discussed and the developed method and algorithm are presented. The efficiency of the algorithm is demonstrated, and utilization of the method is examined based on two case studies, namely recovery of Zn from argon oxygen decarburization (AOD) dust, and extraction of lanthanides from spent nickel metal hydride (NiMH) batteries.