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Optimization of leaching parameters for the extraction of rare earth metal using decision making method

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ABSTRACT

The optimization of leaching operation of Rare earth bearing ores is a complex process since many attributes simultaneously affect the operation, with some of them being conflicting in nature. Therefore a proper selection of the leaching process with pertinent attributes is crucial for the user in order to maximize the percentage recovery with minimal operating costs. In this paper a methodology is proposed for evaluation, comparison and ranking of various leaching process alternatives which we define as leaching candidates, in order to select the best candidate from the available options. Coding scheme for 28 attributes is proposed in order to evaluate the available candidates. A three stage procedure with elimination search, technique for order preference by similarity to ideal solution (TOPSIS) followed by line graphs and spider diagrams, is used for the optimal selection of candidate leaching system. The proposed methodology is illustrated with an example by choosing a few pertinent attributes from the attributes. The suitability index for the best leaching candidate was calculated to be 0.5225 and the coefficient of similarity (COS) values for the best candidate based on line graph and spider diagram obtained were 0.6183 and 0.2711 respectively.

1. Introduction

Since rare earth elements (REEs) have become an indispensable part of modern life, their extraction procedures now hold huge importance. Recovering REEs is a complex process which involves mining the ores which contain REE bearing minerals like Monazite and Bastnaesite, followed by mineral dressing, chemical upgrading and refining procedures (London, 2010). It is important that the ultimate refining procedures actually bring about maximum possible recovery of the REEs present in the ore, while optimizing the costs involved in all the operations.

Physical beneficiation techniques alone such as froth flotation, magnetic separation, gravity separation etc. are not optimal methods for obtaining a market grade concentrate (Girgin and Gunduz, 1996). These techniques need to be followed up by processes such as leaching and solvent extraction, which have subsequently shown a higher percentage recovery of REEs from the pretreated ore (Green and Harbuck, 1996). Leaching is a widely used extractive metallurgical technique for the extraction of the metal of interest from its ore or concentrate in the presence of a solvent known as the leaching agent. Generally, strong acids were employed for the greatest recovery of REEs (Vancouver, 2009), which dissolves the desired REEs present in the ore into the solution. These hydrometallurgical operations have got several advantages; they may start on a small scale and can expand on to a larger one, and these operations also allow greater control in processing ores for the recovery of valuable metals (Ghosh and Ray, 1991). The major advantage of the leaching process is that it selectively dissolves the metals inherent to the ore, leaving behind most of the impurities. This reduces the processing complications and hence makes the extraction step simpler.

Though leaching is an efficacious technique of REE extraction, it poses a few problems with respect to its usage. The process leaves behind metal depleted materials with residual chemicals, also known as 'leach piles', which are hazardous to the environment. In the absence of certain suitable measures, the leaching process may lead to contamination which might be harmful to life (Hudson et al., 1999). During leaching, some of the undesirable components present in the ore can pass into the solution along with the desired REEs, complicating the subsequent extraction processes (Fulford et al., 1991). The leaching operation involves large volumes of solvent requirement for comparatively small metal outputs, which consumes considerable amount of space and requires laborious handling of large amounts of solvents (Ghosh and Ray, 1991).

From the literature review, it is evident that optimizing the leaching process of REEs is complicated since various parameters/attributes such as raw material selection, leaching agent selection, solvent concentration, leaching temperature, leaching time, agitation rate etc. affect the efficiency of the leaching process simultaneously. Several experiments are required to be conducted to know the precise simultaneous effect of all these parameters on the leaching of REEs, which proves to be costly and time consuming. Hence, techniques such as Multiple Attribute

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Decision Making (MADM) can be applied to this process, in order to obtain optimum parameters and select the most suitable candidate among the available choices for a given application.

The present paper critically reviews the parameters affecting the leaching of REEs. Further, attribute based MADM approach is used to select the pertinent attributes/parameters and evaluate the best candidate leaching system over the available options that would maximize the percentage recovery and minimize the operating costs involved in leaching operation of RE bearing ores. A few conflicting parameters are discussed in the next section to apprise the readers of the difficulties in choosing optimal attributes pertinent to the leaching process.

2. Conflicting attributes in optimization of leaching process

While selecting the leaching agent, one has to review all the relevant influential parameters such as choice of raw material, percentage recovery of REEs, amount of impurities getting dissolved into the solution, cost of leaching agent, corrosive properties of the leaching agent, amount of ore to be processed, leach temperature, concentration of the leaching agent, leaching time, liquid/solid ratio, particle size, capital and operating costs etc. in order to maximize the leaching efficiency.

The choice of acids such as sulfuric, nitric and hydrochloric depends on the selectivity in the separation of REEs, type of gangue minerals in the ore and the type of the reagents to be used in further extraction procedures (Ritcey and Ashbrook, 1979). Sulfuric acid is preferred because of its low cost, ease of availability, effectiveness in reacting with most of the metals and less corrosive properties when compared to HCl and HNO_3 (Bautista, 1974). On the contrary, Habashi (1985) has observed that when the phosphate rock is leached with H_2SO_4, the lanthanides are mostly lost (about 70%) in the gypsum residue but when the rock is leached with HNO_3 or HC1, more than 80% are recovered. Leaching with sulfuric or nitric acids requires high acid concentration whereas hydrochloric acid leaching can be done at diluted concentrations (Yorukoglu et al., 2003). Girgin and Gunduz (1996, 1997) obtained the best results from bastnaesite preconcentrate when it was leached with 2.5 M HCl at 55 °C, with15 M H_2SO_4 at 25 °C and with 8 M HNO_3 at 70 °C. In the above case, HCl and HNO_3 require a higher temperature which increases the operating costs, especially if the leaching operation has to be performed for longer durations. Moreover at higher temperatures, the amount of impurities getting dissolved increases. While H_2SO_4 leaching can be done at room temperature, the acid concentration required is higher and therefore increases the capital cost. Further, the amount of the impurities getting dissolved into the solution will increase when higher concentration of H_2SO_4 is used.

Rintala et al. (2011) emphasized that pretreatment prior to leaching enhances the metal recovery and improves the kinetics of the reaction, but whether the pretreatment operations can be done or not depends on the grade of the ore. As the grade of the ore decreases the energy consumption per unit of metal produced increases during the pretreatment process. Roasting bastnaesite preconcentrate helped in the removal of carbonates and also in oxidizing Ce^{+3} to Ce^{+4} for selective separation from other REEs (Bergmann et al., 1984). However, the fluorine released during this process in gaseous phase causes problems of recovery of value elements and pollution (Bian et al., 2011a). Roasting bastnaesite with sulfuric acid produces hydrogen fluoride (HF) which causes serious air pollution (Huang et al., 2005), whereas roasting bastnaesite ore with sodium carbonate is environment-friendly and offers many benefits by increasing the recovery of RE elements (Chi et al., 2007). Yorukoglu et al. (2003) observed that leaching of roasted bastnaesite preconcentrate with 3 M H_2SO_4 gave 47.4% of dissolution of total REEs which was almost 5.6 times the dissolution percentage (8.5%) of the unroasted preconcentrate. Further, in 3 M H_2SO_4 and 1 M thiourea medium, Ce^{+4} in the roasted preconcentrate had been reduced to Ce^{+3}, hence increasing the dissolution of cerium from 12.2% to 93% and of total rare earth elements from 47.4% to 89% because of the addition of thiourea.

Contrary to the above statements, there are several reported advantages of unroasted bastnaesite. Leaching of unroasted bastnaesite reduces the emission of fluorine and also lowers the cost of energy required (Bian et al., 2011a). By keeping acid concentrations at suitable levels, the leaching rate of RE carbonates can be increased while leaving behind the RE fluorides in the leaching slag which can be used in RE electrolysis or Si–Fe alloy metallurgy directly (Bian et al., 2011a). From the above discussion, it is understood that though roasting of bastnaesite preconcentrate before leaching has increased the percentage recovery of REEs over a greater extent, it has a severe effect on the environment and also involves high costs due to the large amounts of energy required. Hence, one has to make balanced decisions about whether to perform roasting and other pretreatment operations before leaching, by weighing it against factors such as grade of the ore, amount of raw material to be processed, cost of energy and environmental pollution effects.

The design of the leaching reactor is important, since several parameters such as the size of the reactor, the amount of raw materials to be processed, the cost involved in mechanical agitation, the amount of leaching agent to be handled and the capital cost are all conflicting in nature. If the amount of raw material to be processed is large, it requires a huge leaching reactor with mechanical agitators for handling the high amount of solvent and raw material, leading to an increase in the capital cost, even if it may result in increased accrued profits. Zhang et al. (1992) have designed a multistage stirring, leaching and washing tower which not only finished the leaching and washing processes of the RE bearing ore in one physical structure, but also increased the percentage recovery to above 92% and reduced the washing factor to less than 0.06. However, the increase in initial investment cost due to the multistage tower must be borne in mind when selecting this option.

From the examples and references stated, it can be concluded that the parameters discussed here prove to be contradictory to each other while optimizing the leaching process. Because of the ambiguous and conflicting nature of all the above mentioned attributes, optimization of the leaching operation becomes a complex process. For such complex processes, techniques like Multiple Attribute Decision Making (MADM) can be used to choose pertinent attributes among the available options. An expert team can then rate the pertinent attributes relevant to a typical application and the TOPSIS (technique for order preference by similarity to ideal solution) method of evaluation can be employed for ranking the selected based on their priorities. This in turn will give the best suitable candidate which will maximize the percentage recovery of REEs and minimize the operating costs.

3. Multiple Attribute Decision Making (MADM) applied to leaching process

The branch of operation research which deals with decision-making in the presence of various incommensurable criteria is known as Multiple Criteria Decision Making (MCDM), which is further divided into MADM and Multiple Objective Decision Making (MODM) (Triantaphyllou et al., 1998). MADM methods are applied when a product/system needs to be evaluated in the presence of conflicting criteria. It requires the decision-makers to select and rank alternatives which are associated with conflicting attributes pertaining to various candidate alternatives (Jian et al., 2001). MADM techniques include weighted sum method (WSM), weighted product method (WPM), technique for order preference by similarity to ideal solution (TOPSIS), analytical hierarchy process (AHP), preference ranking organization method for enrichment evaluation (PROMETHEE), the elimination and choice translating reality (ELECTRE), compromise programming (CP), multi attribute utility theory (MAUT), etc. (Rao and Baral, 2011). These procedures focus on evaluation of candidate alternatives related to a problem and provide help in decision-making and selection of the best candidate among the available alternatives. MADM/MCDM has been successfully applied in various fields of engineering (Beauchesne