

# The evolution of mineral processing in extraction of rare earth elements using solid-liquid extraction over liquid-liquid extraction: A review



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## ABSTRACT

This review paper summarises the fundamental in the production of rare earth elements (REE) specifically on the extraction of REE. Liquid-liquid extraction (LLE) is known to be the most common method employed in the extraction of REE. However, it possesses a few disadvantages by having poor contact area and the formation of third phase during the extraction process. Solid-liquid extraction (SLE) compensates most of the disadvantages in LLE such as the formation of third phase and poor contact between extractant and desired elements. The focus of this paper is to review the evolution of REE extraction and discovers the potential of REE extraction through SLE. Extractants are available widely but when assisted by supporting material via immobilisation, theoretically it elevates the contact area between extractant and desired REE and this concept is known as extractant immobilised material (EIM). The graphical abstract illustrates the concept of EIM between extractant immobilised supporting materials which increases the potential of REE being extracted from aqueous phase. The material is not limited to polymeric resin, silica and membrane, but also microorganism, bio-derived and hybrid materials. EIM is expected to enhance the contact surface area, avoid third phase formation, and reduce the use of chemicals thus increasing the extraction and selectivity of REE. Also, EIM in SLE has the potential to surpass the conventional method in LLE in terms of quantity and quality.

## 1. Fundamental in production of REE

The production of REE mainly starts with mining the mineral bearing rare earth elements (REE). Minerals bearing REE are available in mineral rocks mined from dry lands or placer deposit dug from watery reserves (Jordens et al., 2013; Rezaee Ebrahim Sarae et al., 2009; Sultan and Shazili, 2009). However, the concentration of REE in most of rock forming minerals are not economically efficient for commercial refining. The properties of minerals formed in different locations are varied in terms of its compositions. Apart from REE, other elements with similar properties are found in the same minerals as REE (Li and Yang, 2014).

Based on geological survey, monazite, bastnasite, and xenotime are the three major minerals enriched with REE and feasible for commercialisation. Mining and refining these minerals are practicable

considering cerium (Ce), lanthanum (La), and neodymium (Nd) are enriched in batnasite and monazite. While, xenotime bears mostly yttrium (Y), dysprosium (Dy), erbium (Er), ytterbium (Yb), and holmium (Ho) (Walters et al., 2011). Monazite is classified as phosphate minerals with a mixture composition of Ce, La, praseodymium (Pr), and Nd. In addition, monazite has a mix of radioactive elements such as thorium (Th) and uranium (U) around 4–12% composition (Jordens et al., 2013). Bastnasite is a fluorocarbonate mineral and xenotime is yttrium phosphate mineral (YPO<sub>4</sub>). Apart from bastnasite, monazite, and xenotime, ion-adsorbed clays is another source of REE that normally enriched with 60% of rare earth oxides (REO) (Haque et al., 2014). These mixed up elements in minerals require a long chain of process for high purity production of REE.

The production of REE requires most practices in metallurgy. Hydrometallurgy, pyrometallurgy, ionometallurgy, electrometallurgy,

**Abbreviations:** CHON, Carbon, hydrogen, oxygen, and nitrogen; CMPO, Octyl(phenyl)-N,N-diisobutylcarbamoylmethylphosphine oxide; COOH, Carboxylic acid; Cr, Chromium; CREE, Critical rare earth elements; D2EHPA, Di-(2-ethylhexyl)phosphoric acid; DHOA, N,N-di-n-hexyl octanamide; DODGAA, N,N-dioctyldiglycol amic acid; EDASIDGA, 3-(ethylenediamino) propyl silica gel; EGDMA, Ethylene Glycol Dimethacrylate Copolymers; EIM, Extraction immobilised material; HFSLM, Hollow fibre supported liquid membrane; HLW, High level liquid wastes; HRE, Heavy rare earth element; HTTA, Thenoyltrifluoro-acetone; IIL, Imprinted ionic liquid; IL, Ionic liquid; LAMP, Lynas Advanced Material Plant; LLE, Liquid-liquid extraction; LRE, Light rare earth elements; MAH, Maleic anhydride; MRE, Medium rare earth; NPH, Normal paraffin hydrocarbon; P204, Bis(2-ethylhexyl) phosphate; REE, Rare earth elements; REO, Rare earth oxides; RNO3, Resin nitrate; RPF6, Resin hexafluorophosphate; SE, Synergist extractant; SHLW, Simulated high-level waste; SIL, Supported ionic liquid; SIR, Solvent immobilised resin; SLE, Solid liquid extraction; SLM, Supported liquid membrane; TBP, Tributyl phosphate; TEOS, Tetraethoxysilane; TODGA, N,N,N',N'-Tetraoctyl Diglycolamide

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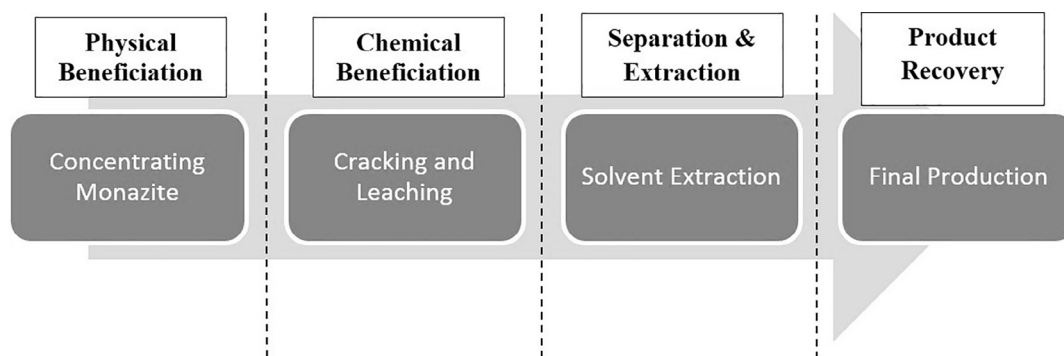


Fig. 1. Example of simplified process flow based on REE plant.

and beneficiation are examples of standard practices in production and purification of metals. Even though the characteristics and applications of REE are slightly different from other metals, these practices are feasible for REE processing. Fig. 1 shows a simplified process flow of common practice in REE industrial production. The examples of process were adapted from Lynas Advanced Materials Plant (LAMP) process flow (Walters et al., 2011; Earth and Processing, 2016; Chu, 2011; Lynas, 2016). Lynas is a corporation from Australia that mining rare earth bearing minerals from Mt Weld, Western Australia and then shipped to LAMP for refining and concentration operation. The first phase is physical beneficiation to concentrate the monazite and followed by chemical beneficiation via cracking and roasting which produces leachate. The leachate will undergo the separation and extraction phase to segregate the desired elements before going through the final phase of REE product recovery. Each of the phase has a sequences of other subsidiary processes such as solvent extraction, leaching, precipitation, floatation, and wet processing. Most of the processes were combined for a complete production of REE which resulted into thousands of stages (Jordens et al., 2013; Haque et al., 2014; Abbott and Frisch, 2013; Gupta, 1981; Krishnamurthy and Gupta, 2004).

### 1.1. Physical and chemical beneficiations

Beneficiation is a process commonly applied in metallurgy sector and it is divided into physical and chemical beneficiations. The main purpose of beneficiation is to remove gangue minerals from raw ores to ensure high concentration of REE output solution. The route flow in beneficiation processes varies corresponding to the type of input of REE. The input can be from raw rock minerals, placer deposits, recycle materials, or heavy metal waste that expected to be rich in REE. Physical beneficiation is a treatment conducted on excavated hard rocks minerals by breaking down the solid into fine powder (Walters et al., 2011). On the other hand, chemical beneficiation or also known as pyrometallurgy is a process that breaks down the chemical bonding between REE and other gangue elements to form highly concentrated REE mixtures (Lynas, 2016; Gupta, 1981; Krishnamurthy and Gupta, 2004).

Physical beneficiation is applied according to the type of ores. Normally, the dried mined ores are crushed and ground to produce solids with high particle surface area and further processed in the next subsequent phase (Lee Yee et al., 2011). However, mineral ores from placer deposits have different physical treatment considering the watery feed. Placer deposit is collected from watery reserves such as river, stream, and marine environment (Rezaee Ebrahim Saraee et al., 2009; Sultan and Shazili, 2009). Gravitational separation and floatation are the additional processes for watery feed to separate water from ores before the drying process. Magnetic separation, electrostatic separation, and screening are usually the supporting practices used in the physical beneficiation (Gupta, 1981). However, not all ores require physical

treatment. Feeds such as recycled materials and ion adsorbed clays are going straight to the chemical processing since these kind of feeds do not require physical beneficiation treatment.

Chemical beneficiation produces highly concentrated REE leachate mixture in a composition that compromise with water phase which increase the separation and extraction of minerals in organic phase. (Lynas, 2016; Gupta, 1981; Krishnamurthy and Gupta, 2004). First, chemical beneficiation uses high temperature to break down the bond between REE and gangue elements. The temperature must be at least between 300 and 1000 °C and processed in 60-m long gas fired kilns (Haque et al., 2014; Krishnamurthy and Gupta, 2004). This high temperature process is also known as roasting or cracking, with the purpose to ease water introduction to the rare earth ores (Lynas, 2016; Lee Yee et al., 2011; Xie et al., 2014). Then, high concentrations of chemicals such as hydrochloric, sulphuric, and nitric acids are introduced into the roasted or cracked rare earth ores. Lastly, ore slurry is produced from the chemical beneficiation phase.

Approximately 30–35% of ore slurry is produced that is then treated with froth flotation and pressed to get high concentrated leachate (Walters et al., 2011) and followed by treatment process known as leaching. Leaching is a process that separates ore slurry into leachate that contains valuable REE metals and undesired compounds for disposal. Leachate consists of at least 60% of REO mixture and 40% impurities (Walters et al., 2011). This leachate will be an input for the next process which is separation and extraction to slim down the elements into desired metal compositions.

High operating temperature, high chemical concentration, and large quantity of chemicals consumed produces a severe toxic waste that harms the environment (Liao et al., 2013). Environmental activists are really against the production of REE mainly due to physical and chemical beneficiation phases. Apart from having to deal with the existence of radioactive elements during the process, the waste from the production of REE is also difficult to manage and dispose. Though, studies have been conducted to find alternatives to REE production and introduce greener methods.

### 1.2. Separation and extraction

Separation and extraction is the most important phase to ensure high purity REE is obtained. Separation in REE industry is defined as classification process of desired compounds (REE) and undesired compounds in a mixture of leachate. Meanwhile, extraction is a selective action taken on desired elements (REE) from aqueous phase (leachate). For example, a high level liquid wastes (HLW) is separated from a mixture of minor actinides (MA) and lanthanides. Then, La is extracted from MA using N,N,N',N'-tetrakis(4-propenyloxy-2-pyridylmethyl) ethylene-diamine (TPEN) that acts as an extraction solvent (Takeshita, 2010; Takeshita et al., 2008).

Separation and extraction phase is complex and sensitive. It is crucial to ensure a high efficiency of individual REE separation and able

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