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Assessing the environmental footprint of the production of rare earth metals and alloys via molten salt electrolysis

Ehsan Vahidi[a,b], Fu Zhao[a,b,c,*]

[a] Environmental and Ecological Engineering, Purdue University, West Lafayette, IN 47907, United States
[b] Ecological Science and Engineering Interdisciplinary Graduate Program, Purdue University, West Lafayette, IN 47907, United States
[c] School of Mechanical Engineering, Purdue University, West Lafayette, IN 47907, United States

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ABSTRACT

Much attention has been given in recent years to the rare earth elements, considering their significance in a number of high-tech and clean energy applications. Despite the environmental destructive operations to produce rare earth elements, limited life cycle assessment investigations have yet been carried out. This is specifically true regarding the reduction of rare earth compounds to produce rare earth metals and alloys. In combination with mass/energy balance and stoichiometry, life cycle inventories of molten slat electrolysis process were developed in this study using industrial datasets gathered from different facilities in China and the Ecoinvent v3 database. The results showed that although mining, chemical treatment, and solvent extraction stages are the dominant contributors to the whole neodymium metal production process for most impact categories, molten salt electrolysis significantly impacts ecotoxicity, carcinogenics, non-carcinogenics, and eutrophication categories. Moreover, neodymium fluoride production, electricity consumption, and molybdenum use in cathodes are the dominant contributors in the molten salt electrolysis process. In addition, the neodymium metal production at facilities which produce larger amounts of heavy rare earth metals/alloys demonstrates relatively lower impacts on all impact categories compared to the production of neodymium metal at refining facilities which produce light rare earth metals/alloys.

1. Introduction

Rare Earth Elements (REEs) consist of the lanthanide series in the periodic table of elements beginning from lanthanum (La, 57) to lutetium (Lu, 71) along with yttrium (Y, 39) and scandium (Sc, 21) which have similar chemical and physical properties (Castor and Hedrick, 2006; Haque et al., 2014). Considering the significance of REEs in many green energy applications and advanced technologies such as mobile phones, hybrid cars, wind turbines, liquid crystal screen televisions, and high efficiency lights, REEs have been classified as critical materials by many governments around the world (Bauer et al., 2010; Wübbeke, 2013; Papangelakis and Moldoveanu, 2014; Adibi et al., 2014).

China as the dominant producer of REEs is currently supplying more than 80% of the total global demand (Chu and Majumdar, 2012; Hellman and Duncan, 2014). In seven provinces in southern China, production of rare earth elements from ion adsorption clays ores with 0.02–1% of rare earth oxides (REOs) is carried out via in-situ leaching using ion exchange technique (Vahidi et al., 2016; Schulze et al., 2017). In this processing route, using ammonium bicarbonate or oxalic acid, rare earth ions are precipitated in the form of rare earth carbonate and finally, calcination process is performed to produce mixed REOs. In the end, REOs are separated and then converted to rare earth alloys and metals using solvent extraction and electrolytic/thermal reduction processes, respectively (Navarro and Zhao, 2014; Vahidi and Zhao, 2017; Weng et al., 2016).

In Inner Mongolia region in northern China, monazite and bastnasite ores which contain 4–7% REOs are extracted via open-pit mining (Kanazawa and Kamitani, 2006). Then, following the magnetic separation of iron ores and flotation process in the beneficiation stage, monazite and bastnasite concentrates with 50% REO content are extracted from tailings (Zhang and Edwards, 2012). Next, to produce rare earth chlorides, hydrochloric acid is utilized to leach the roasted ore concentrate. Similar to the processing pathways of the ion adsorption clays deposits, the individual REOs with 99% purity is produced in the solvent extraction operation and subsequently, the concentrated REOs go through the electrolytic/thermal process to produce rare earth alloys

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[*] Corresponding author at: School of Mechanical Engineering, Environmental and Ecological Engineering, Purdue University, West Lafayette, IN 47907, United States.

E-mail address: fzhao@purdue.edu (F. Zhao).

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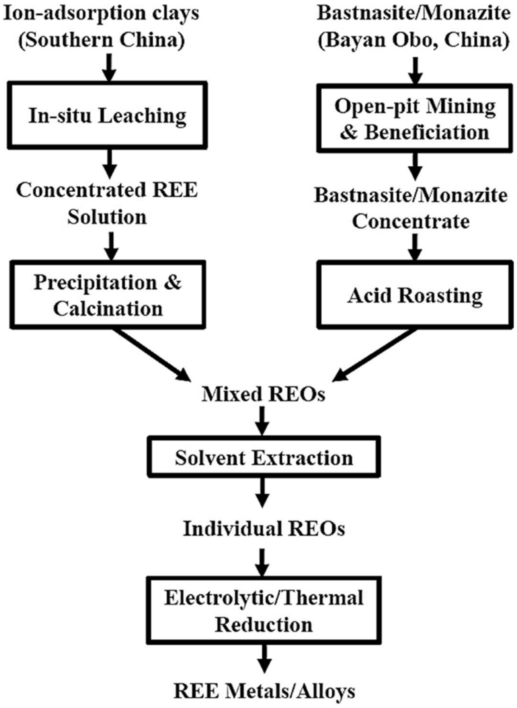

Fig. 1. Major operation stages to produce rare earth alloys and metals from two different rare earth deposits in China.

and metals (Navarro and Zhao, 2014; Vahidi and Zhao, 2016; Schreiber et al., 2016). The main operation stages necessary to produce rare earth alloys and metals from the two major types of rare earth deposits in China are shown in Fig. 1.

All the stages demonstrated in Fig. 1 involve considerable energy and materials use and consequently, significant environmental impacts are incurred in the form of material/energy consumption, waterborne and airborne emissions, along with solid wastes (Vahidi and Zhao, 2017). Furthermore, with continual growth in advanced energy technologies, global production of REEs is expected to increase and therefore, reducing the environmental footprints of rare earth metals becomes critical in the near future. Owing to the intensive environmental concerns raised over the REEs mining and extraction in the United States, the Mountain Pass mine located in California as the dominant supplier of REEs in the world for two decades in the 70 s and 80 s closed in 2002 (Shujing, 2013; Fuerstenau, 2013). In China, the significant material and energy consumption, as well as environmental release in the REEs production process, has also gained significant attention and local governments have cited ecological and environmental damage as one of the major drivers to close heavy polluting facilities and limit production.

Therefore, a holistic and comprehensive evaluation of environmental impacts associated with the rare earth metals and alloys production should be taken into account as the starting point to reduce the environmental destruction and the most widely-used approach is Life Cycle Assessment (LCA) (Evans et al., 2009; Golev et al., 2014; Adibi et al., 2014). U.S. Environmental Protection Agency (U.S. EPA) has defined the LCA as a tool to quantify and compare the environmental footprints affiliated with a product or process in a holistic manner based on energy, material, and emission flows (U.S. EPA, 2006).

The Ecoinvent as the most comprehensive life cycle inventory database covers operations from mining of rare earth deposits in northern China to solvent extraction process to produce high purity individual rare earth oxides while the refining stage has been excluded from the database (Althaus et al., 2007). In addition, there have been a few investigations on the environmental destructions generated by the air or water emissions along with solid wastes from reduction process in rare earth metals and rare earth alloys production (Sprecher et al., 2014; Lee and Wen, 2017). According to the LCA study of neodymium metal production by Sprecher et al. (2014), Nd2O3 is dissolved into fluoride-based molten salt and the solution is subsequently electrolyzed to produce pure liquid metallic neodymium. This is the most common industrial process for neodymium metal production. Sprecher et al. (2014) assumed that the neodymium metal refining process is similar to the Hall-Héroult process utilized for aluminum production and as the result; the life cycle inventory of Hall-Héroult process was modified to assess the life cycle impacts of neodymium metal production process.

In another investigation by Lee and Wen (2017), a comprehensive LCA on the rare earth metals production was conducted. In the mentioned study, a life cycle inventory for molten salt electrolysis of cerium was compiled using process inputs collected from Chinese literature while life cycle inventories of producing gadolinium via calciothermic reduction and samarium via metallothermic reduction were also developed. However, no inventory datasets were presented for neodymium, praseodymium, and dysprosium which are the main rare earth elements utilized in the permanent rare earth magnet manufacturing.

In this research, new life cycle inventories for the production of rare earth metals and alloys which are critical to clean energy applications were developed. In addition to the Ecoinvent v3.0 database, new datasets were created for the production of chemicals and materials employed in the operation to minimize the use of surrogates in the assessment. Energy and materials consumption data were collected from four different REEs refining facilities in China. Taken together, the results of this LCA investigation enhance our knowledge on the environmental footprint of the production of rare earths metals and alloy and help us identify opportunities to develop refining processes with improved environmental performance.

2. Production of rare earth metals/alloys via molten salt electrolysis

As shown in Fig. 1, separation of rare earth oxides via solvent extraction operation is followed by the electrolytic/thermal process to produce rare earth alloys and metals. Due to the strong affinity of rare earth elements to oxygen, REOs are in the class of the most stable oxides and as a result, the removal of oxygen to produce rare earth metals and alloys is very laborious. The literature briefly mentions different techniques such as molten salt electrolysis, calciothermic reduction, and metallothermic reduction (Schüler et al., 2011; Lee and Wen, 2017; Zapp et al., 2018). Calciothermic and metallothermic reduction operations must be performed on a batch basis and the process needs high temperatures and high energy consumption which make the industrial production uneconomic (Vogel and Friedrich, 2015).

Given the lower energy consumption in molten salt electrolysis which can also be conducted as a continuous process, this electrolytic process has become the dominating industrial technique currently used to produce rare earth metal (Stefanidaki et al., 2001; Abbasalizadeh et al., 2015; Vogel et al., 2017). It has been reported that the molten salt electrolysis is an effective method to produce cerium, lanthanum, praseodymium, and neodymium from individual rare earth oxides produced at solvent extraction facilities (Lee and Wen, 2017). Gadolinium, dysprosium, lutetium, holmium, erbium, terbium, and yttrium can be produced using calciothermic reduction. In addition, samarium, thulium, europium, and ytterbium are usually produced by the metallothermic reduction.

In the molten salt electrolysis, the rare earth oxides are converted to rare earth fluorides or rare earth chlorides and then the rare earth halides are reduced to rare earth metals. Fig. 2 shows a simplified molten salt electrolysis cell. While the cell is made of alumina and refractory brick or metal, the two electrodes are dipped into the molten bath where a voltage adequate for the salt reduction is applied to the circuit. In the molten salt electrolysis, rare earth metals/alloys with high purity can be manufactured by using tantalum, tungsten, or