A critical review of bioleaching of rare earth elements: The mechanisms and effect of process parameters

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Table of contents

Abstract

Rare earth elements (REEs) are important high-tech materials with widespread industrial applications and a high risk of supply disruption. Current physico-chemical methods for REE recovery are high energy intensive, complex and costly. Thus, it is crucial to undertake measures to secure future REE demand and protect the environment as well. Bioleaching as an alternative technology for the recovery and recycling of REEs from a variety of primary and secondary sources has been increasingly explored. The present review provides an overview on REE content of various primary and secondary sources, applied bioleaching microorganisms and methods, reactor setups, as well as a detailed description of known REE leaching mechanisms. The effects of different process parameters including temperature, pH, medium composition, pulp density and particle size on efficiency of REE leaching are also evaluated in detail.

Key words: Rare earth elements; Biohydrometallurgy; Heterotrophs; Bioreactor; Metal leaching

1. Introduction

Rare earth elements (REEs) comprising 15 lanthanides as well as yttrium and scandium are classified into the sub-categories of heavy REEs (HREEs) and light REEs (LREEs) depending on their physical and chemical properties. LREEs, also known as the cerium group, consist of Sc, La, Ce, Pr, Nd, Pm, Sm, Eu and Gd while HREEs, also called the yttrium group, include Y, Tb, Dy, Ho, Er, Tm, Yb and Lu (Royen, Fortkamp, & IVL, 2016). Because of their unique properties of luminescence, ferromagnetism and superconductivity, REEs are widely used in many electronic devices including cell phones, hard disc drives and computer screens as well as in a variety of low-carbon technologies such as electric vehicles and wind turbine generators (Barteková & Kemp, 2016). The rising imbalance between the increasing REE demand and supply has resulted in growing concerns about their criticality and the related economic and political issues. REEs are materials with the highest supply risk scores among all the critical raw materials assessed by the European Commission, having both high risk of supply disruption and high economic importance to the industry (Barteková & Kemp, 2016; European Commission, 2017). In order to tackle the REE supply challenge, three different strategies can be proposed: 1) substitution of REEs by less critical metals, which however seems currently challenging without experiencing considerable losses in performance; 2) mitigation of supply risk through investment in old and new primary REE deposits; and 3) investment in REE recycling from secondary sources (waste materials), as many countries do not have any primary REE deposits (Binnemans, Jones, Blanpain, Van Gerven, & Pontikes, 2015; Innocenzi, Ippolito, De Michelis, Medici, & Vegliò, 2016; Johansson, Krook, Eklund, & Berglund, 2013). Although several countries have made some efforts, the recycling rates of REEs are still very low due to technological difficulties and lack of incentives (Binnemans & Jones, 2014; Y. Wu, Yin, Zhang, Wang, & Mu, 2014). As of right now there is no incentive via REEs price, enacting governmental policies on collection systems complementing with financial backing programs are needed for development of long-term sustainable recycling processes (Omodara et al., 2019).

REE recovery typically requires several sequential extraction steps and further processing and refining. Pyrometallurgy and hydrometallurgy are the main current methods for extraction of REEs (Haque, Hughes, Lim, & Vernon, 2014). Depending on the mineralogy of the REE containing material, the extraction step may involve roasting at different temperatures and utilization of various acidic or alkaline leaching solutions (Haque et al., 2014; Innocenzi & Vegliò, 2012). Other physicochemical methods used for recovery and separation of REEs include electrowinning, ion exchange, fractional crystallization and solvent extraction (Kronholm, Anderson, & Taylor, 2013). Despite their extensive use, there are several disadvantages with these physicochemical methods including high acid/base consumption, long residence times, requirement for high temperatures and process complexity leading to high costs (Rao et al., 2016; Walawalkar, Nichol, & Azimi, 2016b). Thus, there is a need to develop alternative REE recovery technologies in order to secure the future demand and to protect the environment either. Biological methods can be more cost-efficient and environmentally friendly routes for recovery of metals from primary and secondary sources compared to their conventional chemical counterparts (Potysz et al., 2015; Thompson et al., 2018; Willner, Kadukova, Fornalczyk, & Saternus, 2015). Bioleaching facilitates metal mobilization from solid sources via different biologically catalyzed reactions mediated by different microbial leaching agents such as (in)organic acids. A wide range of microorganisms such as chemolithoautotrophic bacteria and archaea, and chemoorganoheterotrophic bacteria, archea and fungi have been applied for bioleaching of metals including REEs from different solid materials (Hopfe et al., 2018; Kaksonen et al., 2018; Pathak, Morrison, & Gerard, 2017).

Recent reviews on bioleaching of REEs have focused on biotechnological strategies for recovery of critical raw materials from waste electrical and electronic equipment (WEEE) and the role of phosphate solubilizing microorganisms (Barmettler, Castelberg, Fabbri, & Brandl, 2016; Fathollahzadeh, Eksteen, Kaksonen, & Watkin, 2019; Işıldar et al., 2019). However, the effects of operational parameters on REE leaching efficiency have not been summarized in detail in any previous review. The aim of the present review is to provide an overview of the current status of REE bioleaching from various primary and secondary resources via evaluating the effects of different process parameters. Furthermore, different applied bioreactor types and the REE bioleaching mechanisms are discussed.

2. Primary REE resources

Altogether over 200 different distinct REE containing minerals i.e. primary REE resources, which differ in respect of their geology, mineralogy, and REE content, are known (Golev, Scott, Erskine, Ali, & Ballantyne, 2014; Kanazawa & Kamitani, 2006; Q. Wang et al., 2010). However, economically exploitable deposits are scarce and difficult to find (Massari & Ruberti, 2013). Majority of the global REE reserves are located in only few countries (Figure 1a). China is the dominant world REE producer, responsible for almost 81% of the global production quota (US Geological Survey, 2018) (Figure 1b and 2). However, several exploration efforts in order to develop REE extraction projects are also underway e.g. in Finland, Kyrgyzstan, Madagascar, Mozambique, Namibia, Sweden, Tanzania and Turkey (US Geological Survey, 2018).

REEs can be extracted as the main product or as co-product alongside other metals such as iron, zirconium, titanium, niobium, thorium and uranium (Gupta & Krishnamurthy, 1992; Kumari, Panda, Jha, Kumar, & Lee, 2015). In China, REEs are largely extracted from tailings of iron ore production, but the complexity of the required technology and limitations regarding the scale of production, prevent widespread application of this practice (Golev et al., 2014; Jordens, Cheng, & Waters, 2013). Currently, REEs are commercially extracted from six major primary resources including: bastnaesite [$(Ce,La)(CO_3)F$], monazite [$(Ce,La,Nd)PO_4$)], xenotime (YPO_4), apatite [$(Ca,REE,Sr,Na,K)_3Ca_2(PO_4)_3(F,OH)$], loparite [$(Ce,Na,Ca)(Ti,Nb)O_3$] and ion-adsorption clays (Golev et al., 2014; Kumari et al., 2015). Of these bastnaesite, monazite and xenotime, form nearly 95% of world's known primary REE reserves, and are therefore the most important primary REE sources (Gupta & Krishnamurthy, 1992; Jordens et al., 2013). Bastnaesite, monazite, apatite and loparite mainly consist of LREEs, while xenotime and ion-adsorption clays contain a higher share of HREEs (Golev et al., 2014).Different soil types as potential low-grade REEs sources also contain variable concentrations of REEs (1-1030 mg/kg) depending on the parent material and the location (Mihajlovic & Rinklebe, 2018).

Bastnaesite is the most abundant primary source of REEs (L. Wang et al., 2017). It is a fluorocarbonate mineral containing approximately 60-70 wt% rare earth oxides (REO), including 33 wt% La with considerable amounts of Ce, Nd, Pr, Sm and Gd (Perämäki, 2014; Sinha, Abhilash, Meshram, & Pandey, 2016; L. Wang et al., 2017). Monazite is a phosphate mineral present in acidic igneous rocks and vein deposits containing mainly LREEs including 10-40 wt% La, 20-30 wt% Ce along with remarkable amounts of Sm, Pr and Nd, but deposits often contain also 4-12 wt% Th and variable quantities of U (Kumari et al., 2015; Sinha et al., 2016). Typically, bastnaesite contains more La and less Nd, HREEs and Th (0.2-0.3 wt%) in comparison to monazite (Perämäki, 2014; Sinha et al., 2016; L. Wang et al., 2017). Xenotime is an yttrium phosphate that contains approximately 67 wt% REO and significant amounts of HREEs, which increases its importance as a primary REE resource as HREEs are less available in comparison to LREEs (Perämäki, 2014). Apatite, utilized in phosphoric acid production, contains typically only 0.1-1 wt% REO, but its widespread occurrence makes it an important REE resource (Ogata et al., 2016). Loparite, is an oxide mineral that is no longer widely used for REE extraction, because of its high content of radioactive thorium (Massari & Ruberti, 2013). Ion adsorption clays are a quite newly recognized low-grade REE resource, known for their very low thorium and uranium content (Haschke, Ahmadian, Zeidler, & Hubrig, 2016; Peelman, Sun, Sietsma, & Yang, 2015). Unlike ores, these clays contain 0.05-0.35 wt% REEs in an easily extractable ionic state and do not require pre-processing steps required for hard rock deposits, as they are already cracked (Dutta et al., 2016; Haschke et al., 2016). Ion adsorption clays are currently the most important source of HREEs such as Dy and Tb (Haschke et al., 2016).

3. Secondary REE resources

Secondary REE sources include End-of-Life (EoL) products, new and stockpiled or landfilled industrial residues and pre-consumer scraps (Golev et al., 2014; Sethurajan et al., 2019). Recycling and reuse of these secondary sources using environmental friendly processes is crucial as they contain considerable amounts of REEs (Figure 3a and 3b) and other valuable compounds. Nickel-metal hydride (NiMH) batteries, permanent magnets, and lamp phosphors constituting more than 80% of REE market in terms of value, exist in a variety of EoL products such as electric vehicles, hard disc drives, and fluorescent lamps (Binnemans et al., 2013). Other REE containing EoL products include fluid catalytic cracking (FCC) catalysts, cathode-ray tube (CRT) phosphors, glass polishing powders, and optical glasses (Binnemans et al., 2013).

In NiMH batteries, REEs are found in intermetallic compounds which are used as anode material. The most common intermetallic compounds used are so-called AB_5 compounds where A represents a mixture of REEs such as La, Ce, Nd and Pr, while B refers to Ni, Co, Al and Mn (Innocenzi et al., 2017). NiMH batteries are mainly used in (hybrid) electric vehicles (Innocenzi et al., 2017). The most important elements in spent NiMH batteries include 36-42 wt% of Ni, 3-4 wt% of Co and 8-10 wt% of REEs in the form of hydroxides, oxides and pure metals (Müller & Friedrich, 2006; X. Yang, Zhang, & Fang, 2014).

Permanent magnets that contain REEs can be divided into two types of NdFeB magnets and SmCo magnets, having an REE content of 27 wt% and 23-33 wt% respectively (Peelman et al., 2015). NdFeB magnets are more common as they are cheaper and stronger compared to SmCo magnets which make up less than 2% of the share of REE magnet market (Binnemans et al., 2013; Peelman et al., 2015). NdFeB magnets consist of an $Nd_2Fe_{14}B$ matrix phase, separated by a thin Nd-rich intergranular phase, with small admixtures of Pr, Gd, Tb and Dy (Binnemans et al. 2013; Gutfleisch et al. 2011).

Recycled lamp phosphors are chemically very complex and their composition varies largely between different countries and recycling companies (Binnemans et al., 2013). Typical dyes in recycled fluorescent phosphors (FP) include: 1) red dye Y_2O_3:Eu^{3+} (YOX); 2) green dye comprised of $LaPO_4$:Ce^{3+},Tb^{3+}(LAP), $(Gd,Mg)B_5O_{12}$:Ce^{3+},Tb^{3+} (CBT) and $(Ce,Tb)MgAl_{11}O_{19}$ (CAT); 3) blue dye $BaMgAl_{10}O_{17}$:Eu^{3+} (BAM); and 4) white dye halophosphate $(Sr,Ca)_{10}(PO_4)(Cl,F)_2$ (Hopfe et al., 2017; Peelman et al., 2015). YOX is the most easily extractable REE constituent as it contains REEs in their oxide form, while REEs in the other dyes have much stronger chemical bonds (Peelman et al., 2015; F. Yang, Kubota, Baba, Kamiya, & Goto, 2013). A typical FP powder contains 10% REO, mainly Y, Ce, Eu, Tb and Gd (Hopfe et al., 2017).

FCC catalysts, containing almost 3.5 wt% of REO with mainly La with smaller amounts of Ce, Pr and Nd, are extensively utilized in petrochemical industry (Z. S. Yu & Chen, 1995). Special optical glasses used in binoculars, microscopes and cameras lenses can contain more than 40 wt% of La_2O_3 as well as some Y_2O_3 and Gd_2O_3 in certain cases (Binnemans et al., 2013). The use of CRT phosphors in color TV sets and computer monitors is declining and the only REE they contain is europium in their red dye as Y_2O_2S:Eu^{3+} (Bredol, Kynast, & Ronda, 1994).

Certain industrial residues such as red mud and phosphogypsum are also valuable secondary REE resources. Red mud is a waste product of the alkaline extraction of alumina from bauxite ore and can contain 0.05-0.17 wt% of REEs (Peelman et al., 2015). Phosphogypsum is a residue of phosphoric acid production during which approximately 70-85% of the REE content of phosphate rock (0.04-1.57 wt%) is precipitated with phosphogypsum (Walawalkar, Nichol, & Azimi, 2016a). Despite the relatively low REE content of stockpiles and tailing waste streams (<1%), due to considerably high production volumes, they are recognized as a large ever growing and readily accessible REE source (Golev et al., 2014; Walawalkar et al., 2016a).

4. Bioleaching mechanisms and methods

4.1. Mechanisms of REE bioleaching

Bioleaching is the mobilization of metal cations from often hardly soluble substrates by biological mediated complexation and oxidation processes (Vera, Schippers, & Sand, 2013). Mineral dissolution may proceed via one or several mechanisms, which include (a) complexation promoted dissolution (complexolysis), (b) proton promoted dissolution (acidolysis), and (c) redox reactions (redoxolysis) (Glombitza & Reichel, 2013). In the following complexolysis and acidolysis will be discussed. Redoxolysis will not be discussed since it is typically related to acidophilic bioleaching and more information can e.g. found in the review by Rohwerder, Gehrke, Kinzler, & Sand (2003). Microorganisms can produce metabolites and change the pH of their surroundings. The formed metabolites might form complexes with metals and therefore lead to their mobilization from solids (complexolysis). Similarly, a reduced pH might entail the dissolution of solids (acidolysis) (Figure 4). Complexolysis and acidolysis involve the formation of surface complexes (Furrer & Stumm, 1986), but also changes in the solution speciation and the saturation state of the solution with respect to the mineral have been suggested as possible reaction steps in mineral dissolution (Drever & Stillings, 1997). Recently, it has been shown that surface lanthanide ions in monazite are missing one or two of their nine oxygen atoms which allows them to bond to water molecules and other dissolved species to restore their nine fold coordination (Geneyton, Filippov, Renard, Mallet, & Menad, 2019). Furthermore, it has been suggested that surface lanthanide ions undergo direct hydrolysis rather than (re)adsorption of dissolved lanthanide hydroxides to the mineral surface (Geneyton et al., 2019). Similar to simple metal oxide surfaces, these surface hydroxyl groups could be subject to protonation or ion exchange reactions entailing similar dissolution mechanisms (Dzombak & Morel, 1990). Proton adsorption to surface hydroxyl groups is a fast process, followed by the rate-determining detachment step of the metal ion (Furrer & Stumm, 1986). As this mechanism is dependent on surface protonation, a low pH is favorable due to high proton availability. At the same time, protonation of (in)organic ligands reduces their coordination to the mineral surface and, thus, suppresses ligand promoted dissolution at acidic pH (Furrer & Stumm, 1986).

For metal oxides it has been shown that complex stability correlates with the ability of the ligand to promote the dissolution reaction (Ludwig, Casey, & Rock, 1995). Lanthanides are hard Lewis acids and prefer binding to hard donor atoms such as oxygen (Cotton, 2006). This is reflected in both their inorganic and organic complexation behavior. Generally, complex stabilities of the lanthanides increase with increasing atomic number due to the increased charge density of Lu^{3+} compared to La^{3+}, which causes stronger electrostatic attraction of the ligand (Cotton, 2006). Inorganic ligands relevant to leaching environments include carbonate, sulfate, phosphate, fluoride, chloride, and nitrate. Inorganic speciation of REEs is dominated by carbonate complexation in neutral and alkaline conditions (Johannesson, Kevin & Xiaoping, 1997). Furthermore, carbonate complexation of REEs inhibits CeO_2 precipitation since the

Ce^{4+} ion is stabilized as $Ce(CO_3)_5^{6-}$ complex at alkaline conditions (Möller & Bau, 1993). At acidic conditions, REEs are mainly complexed by sulfate or exist as free aquo ions (Johannesson & Lyons, 1995; Serrano, Sanz, & Nordstrom, 2000; Verplanck, Nordstrom, Taylor, & Kimball, 2004). Mg^{2+} and Ca^{2+} are competing for the sulfate ligands, which increases subsequently the proportion of the REE aquo ions in solution (Johannesson, Lyons, Yelken, Gaudette, & Stetzenbach, 1996). Furthermore, when sulfate and phosphate ions coexist, the formation of sulfate complexes prevents precipitation of the respective REE-phosphates (E. Kim & Osseo-Asare, 2012) as discussed also in section 5.2. Verplanck et al. (2004) also showed that sulfate complexation is reducing REE sorption to Fe-(hydr)oxides up to pH 5. In natural acidic systems phosphate is usually not an important ligand for REEs due to its low concentration (Serrano et al., 2000). However, phosphate complexation can be an important parameter at circumneutral conditions when the molar ratio of HPO_4^{2-}/HCO_3^- is greater than 0.001 (Byrne, Lee, & Bingler, 1991) and when the respective solubility products are not exceeded. Additionally, fluoride has been shown to be an important REE ligand at acidic conditions in the absence of Al^{3+} (Serrano et al., 2000). Complexation with chloride and nitrate will only occur at higher concentrations of these ligands due to the relatively low stability of their complexes (Cotton, 2006). This complexation behavior is in accordance with inorganic acid leaching studies from REE bearing phosphates showing that sulfuric, perchloric, hydrochloric and nitric acid cause high release of REEs, whereas phosphoric acid releases high amounts of calcium but minimal REE concentrations, since insoluble REE phosphates are formed (Bandara & Senanayake, 2019; Battsengel et al., 2018; Stone, Bandara, Senanayake, & Jayasekera, 2016). Thus, it has been suggested that phosphoric acid could be used in pre-leaching to remove e.g. Ca, Fe, Al impurities (Stone et al., 2016).

In bioleaching environments, organic molecules can also induce metal solubilization processes. Such molecules can create a low pH environment, promoting acidolysis, but can also promote mineral dissolution via complexolysis. Thus, their effectiveness in furthering mineral dissolution depends on their complex formation abilities either on whether they can generate protons (Table 1) (Ilyas & Lee, 2014; Shin, Kim, Kim, Jeong, & Lee, 2015). In general, complex stability increases with the denticity of the (in)organic ligand (chelate effect) (Janiak, Meyer, Gudat, Alsfasser, & Meyer, 2012). Additional stability can result from the so-called macrocyclic effect i.e. that cyclic ligands form more stable complexes than their open chain analogues (Cabbiness & Margerum, 1969; Hinz & Margerum, 1974). This can be observed e.g. when comparing stabilities of Fe^{3+}-desferrioxamine E (DFOE) and Fe^{3+}-desferrioxamine B

(DFOB) complexes (Hider & Kong, 2010). Whether acidolysis or complexolysis dominates, depends on solution pH, acid dissociation constant (pK_a) of the functional groups and stability of the formed complexes (expressed as the respective complex stability constant logK). At pH values below the pK_a of the acids, protonation of the acid reduces the stability of the formed (surface)-complexes (Furrer & Stumm, 1986) and acidolysis is expected to dominate, whereas at higher pH, complexolysis dominates.

Brisson, Zhuang, & Alvarez-Cohen (2016) investigated monazite dissolution at pH 1.8 – 4.0 by HCl and various organic acids and demonstrated that the REE extraction has inversely linear correlation with pH suggesting a proton-promoted dissolution. Only in the case of citric acid dissolution proceeded via acidolysis and complexolysis (Brisson et al., 2016). At $pH > 5$, it was shown that aliphatic acids were more effective than aromatic acids in REE extraction from monazite and apatite (Goyne, Brantley, & Chorover, 2010). Differences in extraction patterns of REEs from apatite and monazite were attributed to differences in crystal structure rather than the solution chemistry (Goyne et al., 2010). In case of hydroxyapatite, it was observed that lower crystallinity correlates with an increased adsorption of organic acids and leads to an increased dissolution of REEs – especially at $pH < pH_{pzc}$ (pH at point of zero charge) of the mineral, when adsorption is more favorable by electrostatic attraction among the positively charged mineral and the negatively charged organic acid (Wei, Zhang, Cui, & Wei, 2011). For example, Wei et al. (2011) observed comparatively high adsorption of oxalic acid and explained that by a strong coordination to calcium atoms on the mineral surface. It has also been suggested that organic ligands, which are able to form multi-dentate mononuclear surface complexes, are especially efficient in promoting metal dissolution (Bartlett et al., 2018; Duckworth & Martin, 2001; Furrer & Stumm, 1986). Apatite dissolution also depends on the type of apatite due to different composition of surface layers and formation of secondary minerals (Bengtsson & Sjöberg, 2009). Bioleaching studies with bastnaesite show a limited effect of proton promoted dissolution at $pH > 4$ and the correlation of REE extraction with complex stability (Zhang et al., 2018).

Besides organic acids, siderophores can enhance REE mobilization. Siderophores are molecules produced by microorganisms to specifically scavenge Fe^{3+} under iron limiting conditions (Akafia, Harrington, Bargar, & Duckworth, 2014). A huge variety of siderophores are known, typically showing tetra- or hexadentate coordination of the central atom (Hider & Kong, 2010). Complexation often takes place via hydroxamate, catecholate, and α-hydroxycarboxylate groups (Hider & Kong, 2010). Despite this structural diversity most research is focused on DFOB, which is well known to complex a variety of other metals besides Fe^{3+} such as Pb^{2+}, Zn^{2+}, Cd^{2+} (Karimzadeh, Nair, & Merkel, 2013), Cu^{2+} (Barthen et al., 2018; Karimzadeh, Lippmann-Pipke, Franke, & Lippold, 2017), Ga^{3+} (R. Jain et al., 2019) and REEs (Christenson & Schijf, 2011). It was reported that DFOB solubilizes primarily heavy REEs, however, it also promotes a selective enrichment of Ce in the leachate (Bau, Tepe, & Mohwinkel, 2013; Kraemer, Kopf, & Bau, 2015). This is caused by the higher stability of the Ce^{4+}-DFOB complex in comparison to the REE^{3+}-DFOB complexes. Lee at al. (2014) showed that complexation of Ce^{3+} by DFOB and synthetic hydroxamate type ligands leads to a reduction in the redox potential of Ce^{3+}/Ce^{4+} so that Ce^{3+} can be oxidized by O_2 (Lee, Bogart, Carroll, & Schelter, 2014). A similar behavior has been described for Ce complexes with catechol (Cervini-Silva, Gilbert, Fakra, Friedlich, & Banfield, 2008), a typical structural feature of siderophores, and ascorbic acid (Cervini-silva, Fowle, & Banfield, 2005). However, Ce^{4+} can also precipitate as CeO_2 (Cervini-silva et al., 2005) when the resulting Ce^{4+}-complex is instable in solution. Furthermore, Ce^{3+} oxidation is catalyzed by surface reactions with Mn-(hydr)oxides leading to lower Ce concentrations in the liquid phase due to increased Ce sorption (Kraemer, Tepe, Pourret, & Bau, 2017). Kraemer et al. (2017) argued that DFOB complexation is counter acting surface mediated oxidation causing Ce^{4+} enrichment in the liquid phase. Similarly, it was shown that humic and citric acid complexation is reducing surface mediated oxidation by either shielding Ce^{3+} from the surface in ternary surface complexes or due to reduced sorption of the Ce^{3+}-complexes (Davranche et al., 2008; Q. Yu et al., 2016). Yoshida, Ozaki, Ohnuki, & Francis (2004) showed that DFOB complexation is also reducing (re)sorption of Ce to bacterial cell walls compared to trivalent REEs thus maintaining the Ce enrichment in solution. This indicates that complexes which exhibit a strong affinity for Ce^{4+} could be used to selectively extract Ce over other REEs that are not redox active and reduce (re)adsorption to surfaces. However, Osman, Gebreil, Mowafy, Anan, & Hamed (2019) reported the preferential extraction of LREE by the siderophore ferrichrome produced by *Aspergillus niger* from phosphorites without any selectivity for Ce. Additionally, siderophore production is suppressed when microbes are cultivated in the presence of the REE-bearing substrate due to high iron content as observed in bioleaching of bastnaesite (Zhang et al., 2018).

More recently, biosurfactants, such as rhamnolipids and saponins, have been investigated with regard to REE extraction. It has been shown that REE-rhamnolipid complexes show higher stabilities than Fe^{3+} or divalent transition and alkaline earth metal complexes but lower stabilities than the respective Al^{3+} and UO_2^{2+} complex (Hogan, Curry, Pemberton, & Maier, 2017). This could make them useful for REE extraction from REE sources containing transition or alkaline earth metals but probably not for REE extraction from bauxite or red mud. Zhou, Li, Luo, & Su (2017) showed that plant derived saponins were more effective in leaching La, Ce, Y and Eu from soils than rhamnolipids, produced e.g. by *P. aeruginosa* (Toribio, Escalante, & Soberón-Chávez, 2010; Zhou et al., 2017). However, knowledge of biosurfactant interactions with REE is still limited and further research is needed.

4.2. Bioleaching methods

Bioleaching of REEs can be conducted in different ways: contact bioleaching processes, which include one-step and two-step methods, and non-contact bioleaching processes, also known as spent-medium bioleaching (Barnett, Palumbo-Roe, & Gregory, 2018; Brisson et al., 2016; Golev et al., 2014; Hassanien, Desouky, & Hussien, 2014; Qu & Lian, 2013; Thompson et al., 2018; Zhang et al., 2018). In one-step bioleaching, the microbial culture and the REE bearing material are simultaneously added to the vessel and incubated in the same medium (Qu & Lian, 2013). In two-step bioleaching, the REE containing material is added after pre-cultivation of the microbial culture (Qu & Lian, 2013) to ensure optimal growth and lixiviant production. In non-contact bioleaching, the microorganisms are first cultivated in absence of the REE-containing material to produce the maximum amount of leaching agents, and then the REE-containing material is brought into contact with the cell-free medium containing the biologically produced leaching agents (Qu & Lian, 2013; Rasoulnia, Mousavi, Rastegar, & Azargoshasb, 2016; Zhang et al., 2018). Each of these methods have their advantages and disadvantages. In contact bioleaching, microbial growth and metabolic activity might be retarded because of the toxicity of the extracted metals and other toxic compounds present in the REE containing material (e.g. fluoride present in bastnaesite). However, the presence of microorganisms may promote leaching effectiveness through: (a) adsorption to the surface of solid material, thus decreasing mass-transfer limitations by production of biolixiviants directly on the surface of the REE-containing material (Thompson et al., 2018), (b) altering metal equilibrium in the culture due to metal uptake, adsorption or complexation by excreted compounds (Hassanien et al., 2014), and (c) consumption of constituents released from the solid REE sources (e.g. phosphate in monazite) by the microorganisms reducing precipitation of solubilized REEs (Brisson et al., 2016). Non-contact bioleaching enables separate optimization of microbial leaching agent production and REE leaching steps, regardless of the toxicity of the REE containing material (Hassanien et al., 2014). Spatial separation of biolixiviant production and REE leaching might be helpful to decrease leaching times, when duration of one-step bioleaching is extended due to retarded growth of the microorganisms in the presence of the REE material (Barnett et al., 2018). Growing biofilm embedded, surface attached microorganisms enables production of the leaching lixiviant in a separate bioreactor. This has been shown to be a feasible option for copper bioleaching using iron-oxidizing microorganisms (Pakostova, Grail, & Johnson, 2017). Hekmat, Bauer, & Neff (2007) have demonstrated that immobilized *Gluconobacter oxydans* cells can be used for the production of dihydroxyacetone in bioreactors. Such a set-up would allow for easy external lixiviant production which could be continuously fed into the bioleaching tank, column or heap without the need for a harvesting step for the removal of the microorganisms.

5. Parameters influencing REE bioleaching

Several physicochemical and microbiological factors that affect the efficiency of REE bioleaching have to be considered to develop an efficient and economic process. Elemental composition and the leachability of REE compounds, pretreatment of the solid matrix, type of microorganism(s) and the excreted metabolites, medium composition, pH, temperature, pulp density, particle size, stirring, aeration rate, cultivation and leaching duration are among the influencing parameters. Current knowledge on the specific effects of some typical environmental parameters and chemical constituents affecting REE leaching are discussed in more details in the following subsections.

5.1. Bioleaching microorganisms and medium composition

A large variety of microorganisms have been studied with respect to their application in bioleaching processes. Acidophilic iron and sulfur oxidizing microorganisms are commercially utilized for recovery of metals such as Cu, Au, U, Ni, Co, and Zn from sulfide ores (Brierley & Brierley, 2013; Kaksonen et al., 2018; Karthikeyan, Rajasekar, & Balasubramanian, 2015). Among these acidophiles (preferred growth pH ≤2.0) especially *Acidithiobacillus ferrooxidans* and *Acidithiobacillus thiooxidans* have shown good potential also for REE mobilization (Fathollahzadeh, Hackett, et al., 2018; Funari et al., 2017; Ibrahim & El-Sheikh, 2011; Marra, Cesaro, Rene, Belgiorno, & Lens, 2018; Muravyov, Bulaev, Melamud, & Kondrat'eva, 2015; Tsaplina, Panyushkina, Grigor'eva, Bulaev, & Kondrat'eva, 2015). Chemolithoautotrophic bacteria utilize CO_2 from the atmosphere as their carbon source and oxidize iron and/or sulfur to obtain energy, producing sulfuric acid and ferric iron which lead to dissolution of sulfidic minerals (Ijaz et al., 2017; Klaus & Bosecker, 1997; Rawlings, Dew, & Plessis, 2003). Application of acidophilic Fe/S-oxidizing bacteria for bioleaching of REE containing non-sulfidic sources is possible. However, addition of sulfur or pyrite as well as acidification of the cultivation medium is required for optimal microbial growth and metal solubilization (Fathollahzadeh, Hackett, et al., 2018; N. Jain & Sharma, 2004; Pollmann et al., 2018; Tsaplina et al., 2015). Elemental sulfur from gas and oil purification of sulfurous contaminants is a readily available, cheap growth substrate (Muravyov et al., 2015). It has been shown to be more efficiently utilized by microbial communities (stronger decrease of pH) compared to thiosulfate and tetrathionate (Muravyov et al., 2015). Pyrite as an iron and sulfur containing growth substrate has been successfully used to enable acidophilic bioleaching of metals from low-grade electronic wastes (Bas, Deveci, & Yazici, 2013; Bryan et al., 2015). Although Fe^{2+} is a growth substrate for Fe-oxidizing bacteria and a strong leaching agent in hydrometallurgical processes when oxidized to Fe^{3+}(Alemzadeh, Kahrizi, & Vossoughi, 2009; Sugio, Domatsu, & Tano, 1984), high concentrations of Fe^{3+}can be toxic for Fe/S-oxidizing bacteria. It has been reported that *A. ferrooxidans* growth is inhibited in presence of more than 36 mM Fe^{3+}, while a concentration of 358 mM Fe^{3+}can inhibit oxidation of Fe^{2+} and lead to cell lysis (Dopson, Baker-Austin, Koppineedi, & Bond, 2003). Assessment of the effect of Fe^{2+}addition on biological activity of a mixed culture of Fe/S oxidizing bacteria and yttrium extraction from CRT powders confirmed the key role of Fe/S-oxidizing bacteria and Fe^{2+} as promoters of Y mobilization (Beolchini, Fonti, Dell'Anno, Rocchetti, & Vegliò, 2012). However, Y dissolution was mostly associated to bacterial activity rather than oxidative effects of Fe^{3+}(Beolchini et al., 2012). Furthermore, for acidophilic bioleaching of REEs from non-sulfidic materials with alkaline nature, addition of sulfuric acid within the main period of REE leaching is required, indicating that H_2SO_4 production rate is the dominant limiting factor (Muravyov et al., 2015). In these cases, for industrial application of the process, designing two reactor systems has been suggested; one optimized for biological production of H_2SO_4 from elemental sulfur in absence of REE-containing material, and the second for chemical leaching of the elements using bio-based lixiviants (Marra et al., 2018; Muravyov et al., 2015).

Certain chemoorganoheterotrophic microorganisms can be utilized for bioleaching of non-sulfidic REE resources containing e.g. phosphates, carbonates, oxides and silicates without the need to maintain low pH and to add sulfur and/or iron to the system. These microorganisms utilize organic carbon sources such as glucose for growth and production of a variety of metabolites including organic acids, exopolysaccharides, amino acids and proteins (Hopfe et al., 2017; Pollmann et al., 2018). The fungi *Aspergillus* and *Penicillium* spp. are the most widely used heterotrophic microorganisms for bioleaching of REEs and other valuable metals from a variety of primary and secondary sources (Barnett et al., 2018; Brisson et al., 2016; Keekan, Jalondhara, & Abhilash, 2017; Qu et al., 2015; Qu & Lian, 2013; Rasoulnia & Mousavi, 2016a). Several other fungi, bacteria and yeasts have been also considered for REE leaching from different sources (Tables 2, 3 and 4). *G. oxydans* that has demonstrated good potential at recovering REEs e.g. from FCC materials (Reed, Fujita, Daubaras, Jiao, & Thompson, 2016; Thompson et al., 2018) has been suggested as an attractive candidate for genetic engineering in order to enhance organic acid production rate and REE leaching efficiencies, because of the availability of its genome sequence and a recently developed metabolic network model (Prust et al., 2005; Reed et al., 2016; X. Wu, Wang, & Lu, 2014). *Candida bombicola*, a non-pathogenic yeast strain capable to produce biosurfactants such as sophorolipids, was recently identified as another potent microorganism for bioleaching of REEs from coal fly ash (Park & Liang, 2019). Phosphate solubilizing microorganisms (PSMs) are promising candidates for REE solubilization from REE-phosphates. However, their ability to solubilize metal phosphates seems to be dependent on the phosphate compound. While Shin et al. (2015) reported that Ca-phosphate solubilizing *Acetobacter aceti* was also able to solubilize REE-phosphates this was not observed for other bacteria such as *Klebsiella pneumoniae* and *Klebsiella oxytoca* (Corbett, Eksteen, Niu, Croue, & Watkin, 2017).

Utilization of mixed cultures can sometimes result in more efficient REE bioleaching than pure cultures (Fathollahzadeh, Hackett, et al., 2018; Johnson, 2001). Fathollahzadeh, Hackett, et al., (2018) reported that synergic interactions of heterotrophic organic acid producing *Enterobacter aerogenes* and autotrophic sulfuric acid producing *A. ferrooxidans* in a synthetic co-culture improved REE bioleaching from monazite compared to their pure cultures. Due to their differing pH requirements a two-step process was utilized which allowed for pH reduction by *E. aerogenes* before *A. ferrooxidans* was inoculated. Co-cultivation of indigenous species of monazite concentrate with introduced PSMs also revealed synergetic cooperation between the microbial populations, suggesting more efficient REE bioleaching from non-sterilized ore (Corbett, Eksteen, Niu, & Watkin, 2018). Similarly, *A. ferrooxidans* demonstrated a better performance for bioleaching of REEs from non-sterilized gibbsite ore in comparison to the sterilized sample, confirming presence of a synergic effect between indigenous and inoculated microorganisms (Corbett et al., 2018; Ibrahim & El-Sheikh, 2011). Application of acidophilic mixed cultures is also often more effective than using pure cultures (Ijaz et al., 2017).

As different microorganisms have different growth requirements and need well adapted growth media, comparison of results from different bioleaching studies is difficult. Selection of the most active microbial strains and defining optimum growth conditions including medium composition is critical for development of efficient bioleaching procedures (Hopfe et al., 2018). A limited number of studies have focused on medium optimization for bioleaching of REEs in terms of both microbial performance and process cost-efficiency (Brisson et al., 2016; Thompson et al., 2018). While variations in medium composition and carbon source can have significant effects on leaching agent production and bioleaching efficiency, nitrogen source effects are typically minor (Nautiyal, 1999; Thompson et al., 2018). Furthermore, growth medium constituents can prevent production of leaching agents such as siderophores that are only produced under iron starvation (Bau et al., 2013; W. Wang, Qiu, Tan, & Cao, 2014).

Providing organic carbon source accounts for remarkable percentage of the costs of REE bioleaching in case of heterotrophs (Thompson et al., 2018). Economic evaluations of bioleaching of REEs from FCC catalysts using *G. oxydans* showed that glucose as a carbon source constituted almost 44% of total costs and about 98% of medium component costs (Thompson et al., 2018). Therefore, it is crucial to avoid addition of surpluses of organic carbon sources and investigate the use of alternative inexpensive, waste-derived organic carbon substrates such as food processing and municipal and agricultural wastes for an economical REE bioleaching process (Reed et al., 2016; Thompson et al., 2018). Molasses and glycerin are examples of cheap alternative carbon sources to use in bioleaching of REEs (Hopfe et al., 2018). Addition of glycerin to the medium can positively influence microbial growth and improve REE mobilization due to increased acid production (Hopfe et al., 2018). Inoculum density and adaptation of microorganisms through repeated sub-culturing with increased metal concentrations in the medium are also important aspects to consider, as they can have significant effect on REE leaching capability of the microorganisms (N. Jain & Sharma, 2004).

5.2. REE solubilities and effect of temperature

Temperature is an important parameter in bioleaching, as it affects metal solubility, rate of chemical reactions and the growth and metabolism of microorganisms. Consequently, reported effects of temperature on bioleaching efficiency are always the sum of these processes. In order to identify the influence of each process, they have to be studied separately.

Metal solubilities are important regarding the different leaching agents used such as sulfuric or oxalic acid since they react with extracted REEs and may form insoluble salts (Chi & Xu, 1999; Han, 2019; Porvali, Wilson, & Lundström, 2018). Salts of REEs potentially encountered in leaching environments include phosphates, sulfates, carbonates, chlorides, fluorides, hydroxides, and oxalates. Solubilities of REE salts show a great variety encompassing 15 orders of magnitude (Table 5). Nitrate, chloride and sulfate salts show the highest solubilities, whereas fluorides, oxalates, carbonates, and hydroxides show intermediate solubility. Phosphate salts have distinctively lower solubilities than all other REE salts. However, in presence of sodium and sulfate ions REEs tend to form double salts of the form $REE_2Na_2(SO_4)_4$ which show substantially lower solubilities than the pure REE sulfates especially in the case of Pr (Das et al., 2019; Lokshin, Tareeva, & Kashulina, 2008). This is restricting the accumulation of lanthanides in sulfuric-phosphoric acid solutions forming e.g. during sulfuric acid leaching of phosphogypsum (Lokshin, Tareeva, Ivlev, & Kashulina, 2005). Interestingly, the effect of potassium on REE sulfate solubility is less pronounced (Lokshin et al., 2005). The presence of calcium in sulfuric acid systems can lead to reduced REE recovery due to isomorphous substitution of Ca^{2+} by REE ions in $CaSO_4$ precipitates (Todorovsky, Milanova, Minkova, & Balarev, 1993). Due to its low solubility, phosphate is the solubility-determining factor in non-complexing media. The presence of phosphate has to be considered especially in bioleaching applications since it is an important microbial nutrient. Fujita et al. (2015) showed that below pH 8 Y and Eu solubility in a microbial growth medium was controlled by the formation of phosphate phases. At higher pH, $Y(OH)_3$ and $Eu(OH)_3$ were the solubility controlling phases. In addition, chemical reactions such as complexation with e.g. acid anions generally increase solubility of REE salts (Antonick et al., 2019; Han, 2019; E. Kim & Osseo-Asare, 2012; Lokshin et al., 2008). At ambient temperatures the light $REE_2(CO_3)_3$, and at 100 °C all $REE_2(CO_3)_3$, have been shown to undergo hydrolysis and transform into their respective hydroxycarbonates which are generally more soluble as reviewed by P. Kim, Anderko, Navrotsky, & Riman (2018). The low solubility of REE oxalates may impede efficient REE extraction (Brisson et al., 2016; Lazo, Dyer, Alorro, & Browner, 2017). However, the formed REE oxalate salts are amenable to organic acid leaching and thus selective REE precipitation by oxalic acid can be used as a purification step (Chi & Xu, 1999; Lazo, Dyer, Alorro, & Browner, 2018). Mihajlovic, Giani, Stark, & Rinklebe, (2014) reported a fractionated solubility behavior for REEs via investigating sequential extraction of REEs from two types of soils, where evaluation of different fractions revealed that HREEs tend to be more leachable as they form easier soluble complexes.

Moreover, the solubility of REE salts is dependent on temperature. An increase in temperature decreases solubility of REE sulfates (Das et al., 2019), phosphates (Cetiner, Wood, & Gammons, 2005; Firsching & Brune, 2005), fluorides (Migdisov, Williams-Jones, & Wagner, 2009; Mioduski, Gumiński, Zeng, et al., 2014) and hydroxides which can be estimated from the behavior of La- and Gd-hydroxides (Deberdt, Castet, Dandurand, Harrichoury, & Louiset, 1998) due to the similar chemistry of the lanthanides. Only the solubility of REE chlorides (Das, Lencka, Eslamimanesh, Anderko, & Riman, 2017; Mioduski, Gumiński, & Zeng, 2008) and REE nitrates (Siekierski, Mioduski, & Salomon, 1983) increases with temperature. For the temperature dependence of REE oxalate solubility, no data is currently available.

The rate of chemical reactions increase with increasing temperature (Franzmann, Haddad, Hawkes, Robertson, & Plumb, 2005). This relationship can be described by the Arrhenius equation

$$k = Ae^{\frac{-E_a}{RT}} \tag{1}$$

where k is the rate constant, T is the absolute temperature, E_a is the activation energy, R is the universal gas constant (8.314 J K^{-1} mol^{-1}), and A is a "pre-exponential factor" related to the frequency of correctly oriented collisions of the reactants. Numerous studies have reported a positive correlation between increasing temperature and REE extraction efficiency irrespective of REE source and the used leaching agent (Behera & Parhi, 2016; Lazo et al., 2017; Oelkers & Poitrasson, 2002; Walawalkar et al., 2016b; Whitty-Léveillé, Reynier, & Larivière, 2018; Xiao et al., 2015; F. Yang et al., 2013; Yoon et al., 2014). This is in contrast to the reduced solubilities of most REE salts at increasing temperatures. In fact, reduced La extraction with sulfuric acid at higher temperatures was attributed to the decreased solubility of $La_2(SO_4)_3$ (Abhilash, Sinha, Sinha, & Pandey, 2014; Meshram, Pandey, & Mankhand, 2016). However, $La_2(SO_4)_3$ solubility is not solely dependent on temperature but also on sulfuric acid concentration (Das et al., 2019) and higher temperatures can be used at low sulfuric acid concentrations (C. J. Kim et al., 2014). REE extraction at higher temperatures raises the question as to whether the reaction has to be heated externally or whether the reaction is exothermic enough to generate the needed heat intrinsically as known for sulfidic ores (Olson, Brierley, & Brierley, 2003). When additional heating is needed, the required energy costs have to be considered as well. Furthermore, depending on the used microorganism this may also require the separation of lixiviant production and leaching reaction. Moreover, the solubility of O_2 (Battino & Clever, 1966) and CO_2 (Carroll, Slupsky, & Mather, 1991) decreases with increasing temperature adding further operational costs to bioleaching processes (Guezennec et al., 2017).

In bioleaching studies, the choice of temperature is dependent on the temperature tolerance of the used microorganism(s). The dependency of the microbial growth rate on temperature can be described with the Ratkowsky equation (Ratkowsky, Lowry, McMeekin, Stokes, & Chandler, 1983; Ratkowsky, Olley, McMeekin, & Ball, 1982) which in contrast to the Arrhenius equation takes reduced reaction rates due to enzyme degradation at higher temperatures into account (Franzmann et al., 2005).

$$\sqrt{\frac{1}{t}} = b\,(T - T_{Min})(1 - e^{(c(T-T_{Max}))}) \tag{2}$$

In this equation t is the doubling time, T is the temperature, T_{Min} and T_{Max} are the theoretical minimum and maximum temperatures for growth, and b and c are fitting parameters. Franzmann et al. (2005) applied this equation to the temperature dependence of Fe^{2+} and sulfur oxidation by acidophilic microorganisms and could successfully describe the temperature dependence of both processes. The highest iron and sulfur oxidation rates were found close to the reported optimal growth temperatures of the investigated microorganisms. Consequently, bioleaching studies are mostly done at the optimal growth conditions of the organism(s) which are generally in the mesophilic range (see Tables 2 and 3). However, in some cases the effect of temperature on REE bioleaching has been studied in detail. Amin et al. (2014) and Hassanien et al. (2014) showed that REE extraction by *Aspergillus* species and *Pseudomonas aeruginosa* from shale and monazite was optimal at 30 °C and 35 °C, respectively. These are also the optimal growth temperatures reported for these species (Kück, Nowrousian, Hoff, & Engh, 2009; LaBauve & Wargo, 2012). This is indicating that the positive temperature effect on the dissolution rate is not compensating the reduced lixiviant production efficiency by the microorganisms. However, this might be very specific for each combination of REE resource and microorganism and cannot be easily transferred to other combinations. Interestingly, there has been no study using psychrophilic or psychrotolerant microorganisms such as *Acidithiobacillus ferrivorans* for REE bioleaching. These could be interesting for bioleaching operations in colder climates. It should also be noted that temperature effects in bioleaching studies using mixed cultures are difficult to assess since the community composition will change with temperature (Quatrini & Johnson, 2018) introducing variations not directly related to the temperature variation.

Whether a change of temperature is beneficial to a bioleaching process depends thus on several factors. REE solubility, leaching rates, and temperature preferences of the microorganism(s) have to be considered. Increased temperature can lead to an increase in mineral dissolution, which compensates the decreased solubility of REEs. However, an increase in temperature beyond the growth capabilities of the employed microorganisms is detrimental.

5.3. Effect of pH

In bioleaching, pH acts as an important factor affecting both microbial activity and metal leaching efficiency as discussed above (section 5.2). Optimizing pH for microbial growth enables maximum amount of bio-produced leaching agents in the medium. In contact bioleaching, variations in medium pH are a result of both microbial activity and the REE containing material. Since different microorganisms have different optimum growth pH, it is important to choose an appropriate microorganism based on the nature of the REE source (acidic or alkaline) (Hopfe et al., 2017; Pradhan, Nathsarma, Srinivasa Rao, Sukla, & Mishra, 2008). For example, FP have been shown to buffer the medium pH in a range (4.5-7) that inhibits the growth of acidophiles, impeding their use for bioleaching of FP (Hopfe et al., 2017). Bioleaching microorganisms typically reduce medium pH as they grow due to production of acidic metabolites (Hassanien et al., 2014). However, in contact bioleaching, the medium pH may increase as result of biomass lysis and consequent release of alkaline intracellular metabolites caused by toxicity of the REE source, or as a consequence of proton consumption (Bhardwaj et al., 2007; Hassanien et al., 2014; Rasoulnia et al., 2016).

It is important to identify pH conditions that propel microbial cultures towards production of agents that are most beneficial for REE leaching. For instance, *A. niger* is known to produce a variety of different organic acids (Brisson et al., 2016; Rasoulnia & Mousavi, 2016b). It has been shown that at low pH values (≤ 2) *A. niger* produces mainly citric acid, while at higher pH values (≥ 4) it tends to produce gluconic and oxalic acids (Brisson et al., 2016; Ramachandran Sumitra, Fontanille Pierre, 2006; Rasoulnia & Mousavi, 2016b). REE-oxalates are known to have a low solubility, meaning that higher concentrations of oxalic acid typically correspond with lower REE concentrations in the leachate (Brisson et al., 2016). Therefore, for mobilization of REEs using *A. niger*, it is crucial to maintain a low pH to ensure citric acid production which is more beneficial for REE leaching.

The highest REE leaching efficiencies usually occur when acid production is high and the pH is decreased (Barnett et al., 2018; Brisson et al., 2016; Fathollahzadeh, Becker, Eksteen, Kaksonen, & Watkin, 2018; Qu & Lian, 2013). Mihajlovic, Stark, & Rinklebe, (2017) studying a German soil, revealed that the decreased pH resulting from increased redox-conditions, led to enhancement of REEs release under acidic conditions. Increase in Eh was accompanied by a decrease in pH probably due to proton formation caused by oxidation reactions.

However, in case of samples with highly alkaline nature, during bioleaching pH might not decrease or it might even increase due to proton consumption and/or release of alkaline elements. At weakly acidic or neutral conditions, REEs may also be extracted due to production of other metabolites such as siderophores (Amin et al., 2014; Zhang et al., 2018). It has been shown that in a pH range of 7.0-7.7 (incubation temperature and time of 20 °C and 72 h, respectively), REE mobilization from volcanic ash particles is significantly enhanced in presence of DFOB (Bau et al., 2013). As discussed in section 4.1, pH also affects the dominant REE mobilization mechanisms that take place during bioleaching.

When performing bioleaching, there is also a possibility of underestimation of the actual attainable REE recovery because of REE absorption to biomass or surfaces of remaining waste/ore particles (Hopfe et al., 2017). REE sorption by various microorganisms typically increases with increasing pH (Tsuruta, 2007). Although lower pH conditions might seem beneficial to avoid REE absorption during bioleaching, the absorption and desorption rate of various REEs as a function of pH may differ considerably (Bonificio & Clarke, 2016). This indicates that depending on the REEs present in the solid source, various pH ranges can be suitable for conducting bioleaching so that all leached REEs remain dissolved in the solution. Another phenomenon likely to cause underestimation of REE bioleaching is possibility of REE precipitation at different pH values. For instance, precipitation of Nd has been reported to occur at pH values ≥ 8 (Armstrong & Wood, 2012).

5.4. Pulp density and particle size

Pulp density is an important parameter commonly reported for shake flask and stirred tank reactor bioleaching. In contact bioleaching, microbial leaching performance is usually inhibited at elevated pulp densities, due to release of toxic compounds, insufficient agitation, and/or mass-transfer limitations (Ilyas, Kim, Lee, Jabeen, & Bhatti, 2017; Marra et al., 2018; Meshram et al., 2016). However, at low ranges (1-2 %w/v), increase in pulp density may promote the microbial growth due to the increase in presence of essential metal ions in the leaching material required for microbial growth and activity (Hopfe et al., 2018; Qu & Lian, 2013). It has been shown that REEs can have a stimulating effect on microbial growth and enzyme activity, because they act as essential cofactors for certain enzymes (Barmettler et al., 2016; d'Aquino et al., 2009). Most leaching materials such as spent catalysts, fly ashes and electronic scraps have been reported to have a toxic nature, hindering microbial growth (Amiri, Yaghmaei, & Mousavi, 2011; Brandl et al., 2001; H. Y. Wu & Ting, 2006). Nevertheless, bioleaching of REEs from red mud using *Penicillium tricolor* showed that at 2% pulp density, the fungus did not undergo intense toxic effect on its growth, but even induced a stimulating effect as the biomass growth and REE leaching efficiency at 2% (w/v) pulp density were higher than those obtained at lower and higher pulp densities (Hopfe et al., 2018; Qu & Lian, 2013). Similar beneficial effects of red mud on growth of the microalga *Desmodesmus quadricauda* has been reported (Čížková et al., 2019).

In non-contact bioleaching, because of absence of microorganisms in the leaching solution, higher pulp densities than in contact bioleaching can be applied (Hopfe et al., 2018; Rasoulnia & Mousavi, 2016a; Thompson et al., 2018). However, reduced REE leaching yield at increasing solid loadings is still inevitable. Here, the reduced yield can be attributed to the poor mixing efficiencies and insufficient amount of leaching agents in the solution (Rasoulnia & Mousavi, 2016a; Thompson et al., 2018). As pulp densities increase, providing adequate mixing for the bioleaching becomes more costly and difficult (Thompson et al., 2018). However, in some cases, a higher pulp density with a lower leaching yield might be preferred in order to gain more profit (Gao & Owens, 2012; Thompson et al., 2018). Techno-economic analysis on the size of bioreactors required to produce essential biolixiviants for non-contact bioleaching of REEs from FCC catalysts revealed that applying 50% pulp density which required a bioreactor volume of 193 m^3 was more economical than applying lower pulp densities of 18% and 1.5% requiring much larger bioreactor volumes of 536 and 6436 m^3, respectively (Thompson et al., 2018). Thus, the knowledge of the optimum relationship between the applied pulp density, obtained REE leaching yield, costs and the attainable profit is crucial for designing and operating an economically viable process at industrial-scale.

An effective strategy to enhance reactivity of the solid materials can be the introduction of physicochemical transformations including changes in specific surface area, particle size, crystal lattice and chemical composition (Van Loy, Binnemans, & Van Gerven, 2018). Smaller particles, having a greater accessible surface area, allow for increased number of surface interactions of the leaching agent and more colonization and microbial attachment, resulting in release of a greater portion of REEs into the leachate (Corbett et al., 2017). However, intensive grinding may cause agglomeration of small particles to bigger aggregates (Gelencsér et al., 2011), which is unfavorable for the leaching. In addition, grinding is an energy consuming operation that strongly affects the economy of the whole bioleaching process (Gentina & Acevedo, 2013).

Recent findings demonstrate that mechanical activation of waste phosphors using planetary ball-mills improves REE leaching rate and yield not only via reducing particle size, but also through compound decomposition and changing crystal structure of the particles (Tan, Deng, & Li, 2016, 2017; Van Loy et al., 2018). Mechanochemical-assisted leaching of REEs from lamp phosphors showed that ball milling for up to 15 minutes caused a sharp increase in REE leaching, which was attributed to a change from monocrystalline to polycrystalline morphologies (Tan et al., 2017; Van Loy et al., 2018). Increased milling times (>15 min) did not have a further positive effect on the leaching efficiency presumably due to particle aggregation (Tan et al., 2016; Van Loy et al., 2018). Reduction of the crystallite size to the nanoscale in a polycrystalline material leads to enhancement of defect-rich grain boundaries in the crystals and consequently improves the leaching process (Van Loy et al., 2018). Furthermore, high-intensity ball-milling has been shown to cause oxidation of common trivalent cerium to a tetravalent state (Van Loy et al., 2018). This can be useful in downstream processing (Um & Hirato, 2016) by e.g. exploiting the different solubility of Ce^{4+} compared to the trivalent REEs (Holleman, Wiberg, & Wiberg, 2007).

6. Bioleaching process designs

As different REE containing materials require their own specific microorganism(s) and conditions for efficient REE leaching, most bioleaching studies have been conducted in small batch systems with the aim of screening REE leaching capability of different types of microorganisms from various primary and secondary sources (Table 4). Especially autotrophic bacterium *A. ferroxidans* and heterotrophic bacteria and fungi *G. oxydans*, *Komatogateibacter xylinus* and *A. niger,* as well as recently investigated yeast strain of *Candida bombicola* have demonstrated promising capabilities for REE leaching (Tables 2 and 3). However, the leaching capability of microorganisms is highly dependent on the bonding of REEs within the REE-containing solid material. Phosphate and carbonate bound REEs sources such as monazite and bastnaesite are harder to leach, while REE oxide containing materials such as WEEE shredding dust and municipal solid waste incineration fly ash (MSW FA) as well as ion-adsorption clays, in which REEs occur as ions (Golev et al., 2014), are easier to leach (Table 2 and 3). Nevertheless, there are some inconsistencies between the obtained REE leaching efficiencies of different and even same REE mineral or waste type, likely due to the differences in the composition, growth medium constituents, applied microorganisms and/or bioleaching methods (Brisson et al., 2016).

Bioreactors and columns enable batch-wise, semi-continuous or continuous addition of the substrate and removal of the leached metals, providing a more comprehensive understanding of bioleaching processes that could be utilized at industrial scale. Bioreactors can be operated in contact or non-contact mode (Gu, Rastegar, Mousavi, Li, & Zhou, 2018; Thompson et al., 2018) and the bioreactor types used in REE bioleaching studies include columns, airlift percolators and stirred tank reactors. Each of these bioreactor setups has its own advantages and disadvantages. Stirred tank bioreactor consists of a vessel with mechanical agitator or impeller, spargers for air addition and baffles closed to the wall of the vessel for enhanced mixing. Stirred tank reactors enable good aeration for the microorganisms which shortens the required reaction times (Gu et al., 2018). However, they typically have high capital costs and suffer from the requirement for fine particle sizes and high energy consumption for mixing at high pulp densities (Borja et al., 2016; Klaus & Bosecker, 1997; Thompson et al., 2018). Airlift percolators, in which the particles are suspended in a leaching solution that is circulated to the top of the column by compressed air (Klaus & Bosecker, 1997; Muravyov et al., 2015), can suffer from slow mass transfer and low dissolved oxygen concentrations likely due to lack of optimal mixing or aeration (Gu et al., 2018). Unlike in airlift percolators, in column bioleaching the leaching solution passes through a column packed with solid particles (ore/waste) and is recirculated back to the top with pumps (Gu et al., 2018; Klaus & Bosecker, 1997). Column reactors are typically used to simulate heap or dump bioleaching, and suffer from incomplete or unequal contact of packed ore/waste with the leaching solution and long reaction times required for efficient leaching (Ghorbani, Franzidis, & Petersen, 2016). However, the capital costs of columns are typically lower than those of stirred tanks (Gu et al., 2018; Rawlings, 2002).

So far only few studies on REE bioleaching have been conducted in bioreactors and columns (Table 3). Column bioleaching of REEs from quartz-pebble conglomerate resulted in extraction of 45% of REEs after 52 weeks by use of acidophilic autotrophic bacteria (Sapsford, Bowell, Geroni, Penman, & Dey, 2012). In a similar way, 67% of REEs were extracted from a gibbsite ore in an undefined amount of time by recirculating the leaching solution 35 times (Ibrahim & El-Sheikh, 2011). Column bioleaching of spent FCC catalysts using filtered biolixiviants produced by *G. oxydans* resulted in 46% REE leaching efficiency in 24 h (Thompson et al., 2018). Batch and continuous stirred tank bioleaching of the same FCC catalysts resulted in slightly higher REE leaching efficiencies (49% at 1.5% pulp density) (Thompson et al., 2018) likely due to better aeration. Similarly, REE leaching from ash slag wastes in stirred tank leaching resulted in higher REE extraction efficiency (22-59%) than shake flasks (15-30%) and airlift percolators (30-33%). Furthermore, leaching durations were shorter in both stirred tank and shake flask leaching (10 days) than in airlift percolator leaching (60 days) (Muravyov et al. 2015). Funari et al (2017) used acidophilic mixed cultures in stirred tank bioleaching applying a two-step mode for REE extraction from MSW FA. The highest and lowest leaching efficiencies were obtained for Nd (76%) and Sm (9%). Continuous stirred tank bioleaching of red mud by *A. niger* led to REE leaching yields of 27-62% (Qu et al., 2015). Although, the results seem to indicate that stirred tank reactors might be advantageous for REE extraction, the low number of studies does not allow for a definitive statement.

7. REE bioleaching in comparison to chemical leaching

Bioleaching usually suffers from lower leaching yields and rates when compared to chemical leaching using strong (in)organic acids or complexants (Pant, Joshi, Upreti, & Kotnala, 2012). Nevertheless, bioleaching has shown some advantages over chemical leaching in case of certain low-grade REE containing materials such as FCC catalysts, MSW FA and ion adsorption clays (Barnett et al., 2018; Funari et al., 2017; Reed et al., 2016). Bioleaching of FCC catalysts using *G. oxydans* spent-medium containing 10-15 mM gluconic acid was more effective compared to chemical leaching using pure organic acids at even higher concentrations, indicating that other microbial metabolites present in supernatants contribute to REE leaching (Reed et al., 2016). Sulfuric acid is often preferred to other inorganic acids as it dissolves fewer impurities like Ca and Sr (Peelman et al., 2015). Funari et al. (2017) demonstrated that bioleaching of MSW FA using acidophilic mixed culture improved Nd and La leaching yields and resulted in solubilization of lower amount of unnecessary elements, acting more selective in REE extraction compared to chemical leaching by H_2SO_4. Barnet et al. (2018) compared bioleaching of REEs from an ion-adsorption clay using *Aspergillus* and *Bacillus* cultures, to leaching with 0.5 M ammonium sulfate, demonstrating that bioleaching improved Ce extraction. The improved Ce leaching yield was attributed to the occurrence of Ce as Ce^{4+} due to oxidization of Ce^{3+} during rock weathering and soil formation, making it unavailable for simple ion-exchange extraction (Sanematsu & Watanabe, 2016). During the first three days of salt leaching a rapid release of REEs (except Ce) was observed. After that REE leaching yields did not increase further. However, in bioleaching maximum REE yields were observed between 10 and 60 days of leaching, presumably caused by the differing solubilization mechanisms (Barnett et al., 2018).

8. Conclusions

Bioleaching has been increasingly explored for extraction of REEs from most primary and secondary resources. Due to limited availability of primary resources an increase in REE recycling is urgently needed to expand REE supply and reduce waste generation and environmental pollution caused by mining activities. In order to improve biohydrometallurgical recovery of REEs, different factors can be optimized. Two of the most important factors for REE bioleaching are pH and temperature since they control, among others, the leaching mechanism, microbial activity, and REE solubility. Thus, careful adjustment of these parameters is crucial to ensure successful REE extraction. Owing to the great variety of possible combinations of REE resources and bioleaching systems, a case by case assessment and optimization has to be done. Using geochemical modeling tools can help to predict REE mobilization behavior and thus improve the bioleaching process. Additionally, an improvement in bioreactor design and an increased number of bioreactor studies is necessary to understand and optimize REE recovery for future large-scale applications. Techno-economical analyses are also essential to design a cost-effective process e.g. by utilization of waste products as carbon source for microbial lixiviant production.

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Figures and tables captions

Figure 1: World quota of **(a)** known primary REE reserves and **(b)** Mine REE production in 2017; data obtained from US Geological Survey (US Geological Survey, 2018).

Figure 2: Trend of REE production of major producers and world overall, 2010-2017; data obtained from US Geological Survey (2010-2018).

Figure 3: (a) REE content of most important secondary resources and **(b)** The individual REE share per total REE content; data obtained from (Binnemans et al., 2013; Innocenzi et al., 2017; Peelman et al., 2015).

Figure 4: Schematic representation of possible mechanisms and reactions in lanthanide (Ln^{3+}) bioleaching systems: a) Secretion of ligand (L), b) dissociation of ligand, c) proton promoted dissolution due to reaction with Ln^{3+}, d) reaction of Ln^{3+} (aq) with protonated ligand (HL), e) proton promoted dissolution due to reaction with arbitrary anion (X^{3-}), f) uptake of X^{3-} by microorganism, and g) ligand promoted dissolution.

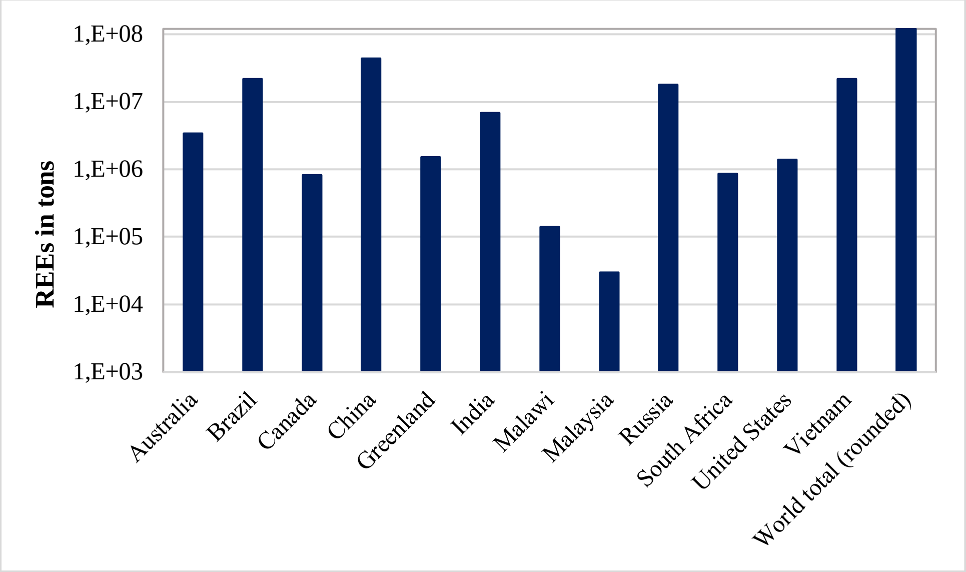
Table 1: REE-complexation of important leaching agents produced by various microorganisms.

Table 2: Overview of bioleaching studies from primary REE resources.

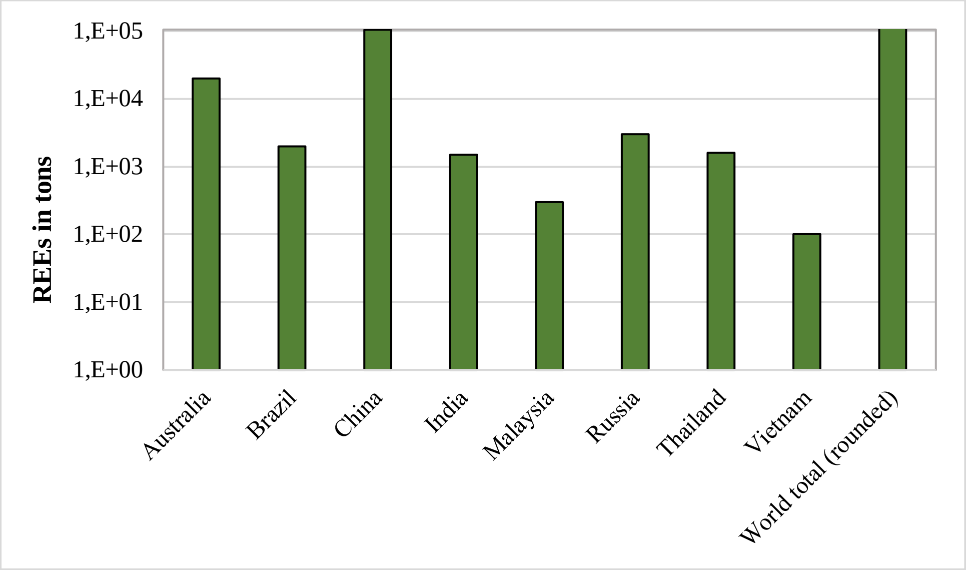
Table 3: Overview of bioleaching studies from secondary REE resources.

Table 4: Screened microorganisms for their REE capability on various primary and secondary sources.

Table 5: Overview and comparison of solubilities of REE salts at 25° C.


1,E+08
1,E+07
1,E+06
1,E+05
1,E+04
1,E+03
REEs in tons
Australia
Brazil
Canada
China
Greenland
India
Malawi
Malaysia
Russia
South Africa
United States
Vietnam
World total (rounded)

(a)


1,E+05
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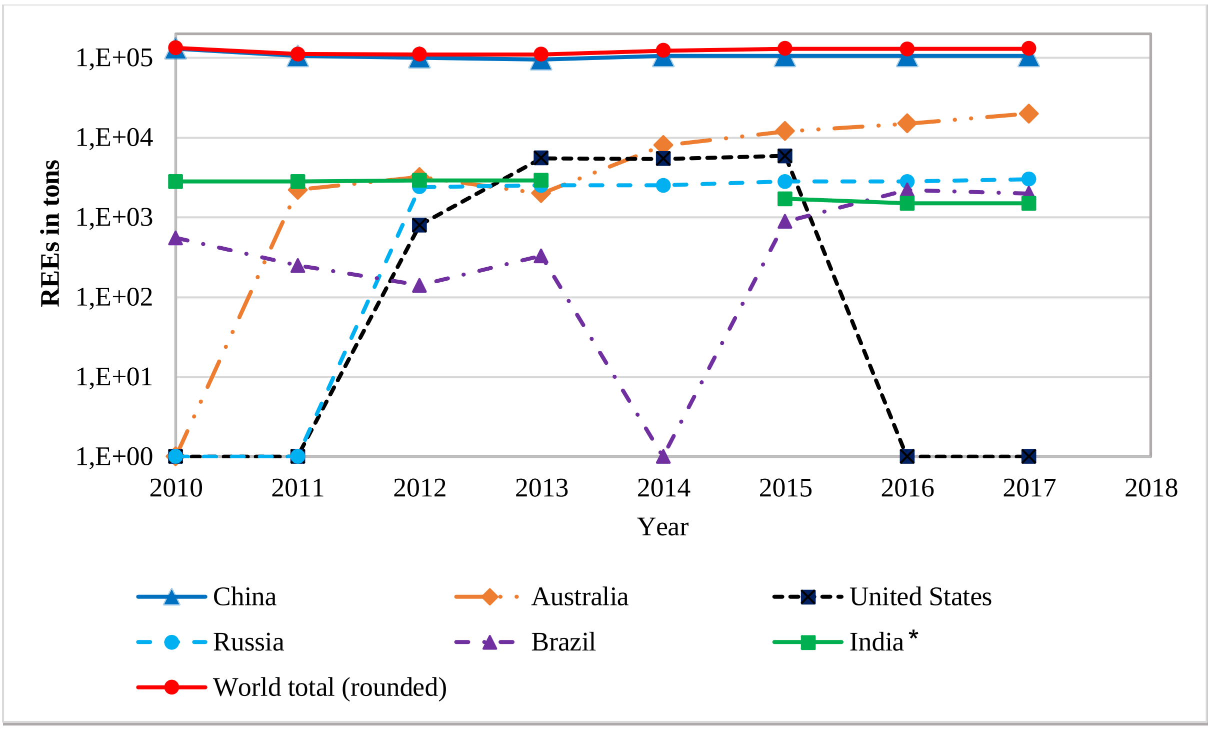
1,E+01
1,E+00
REEs in tons
Australia
Brazil
China
India
Malaysia
Russia
Thailand
Vietnam
World total (rounded)

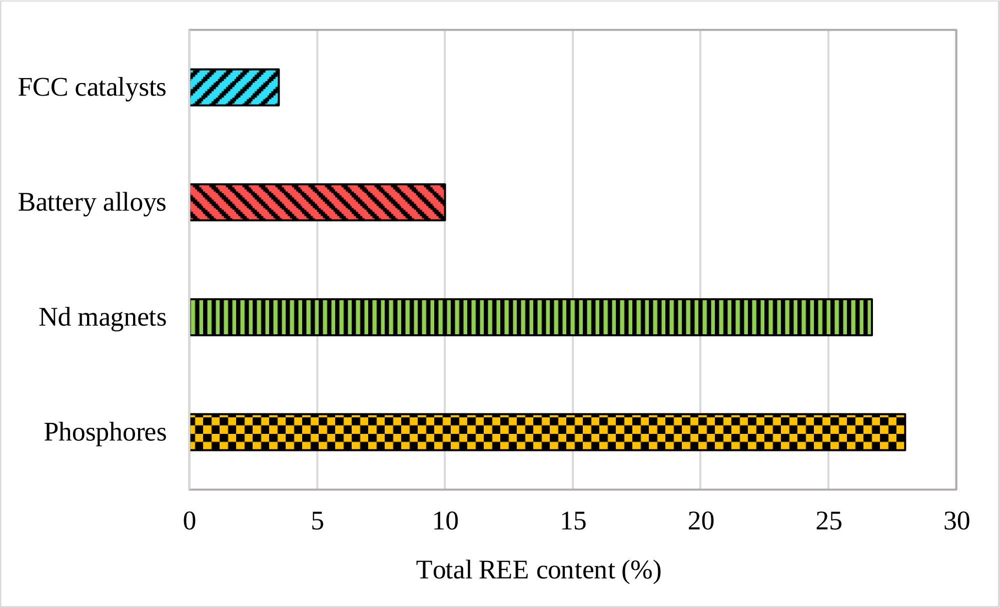
(b)

Figure 1

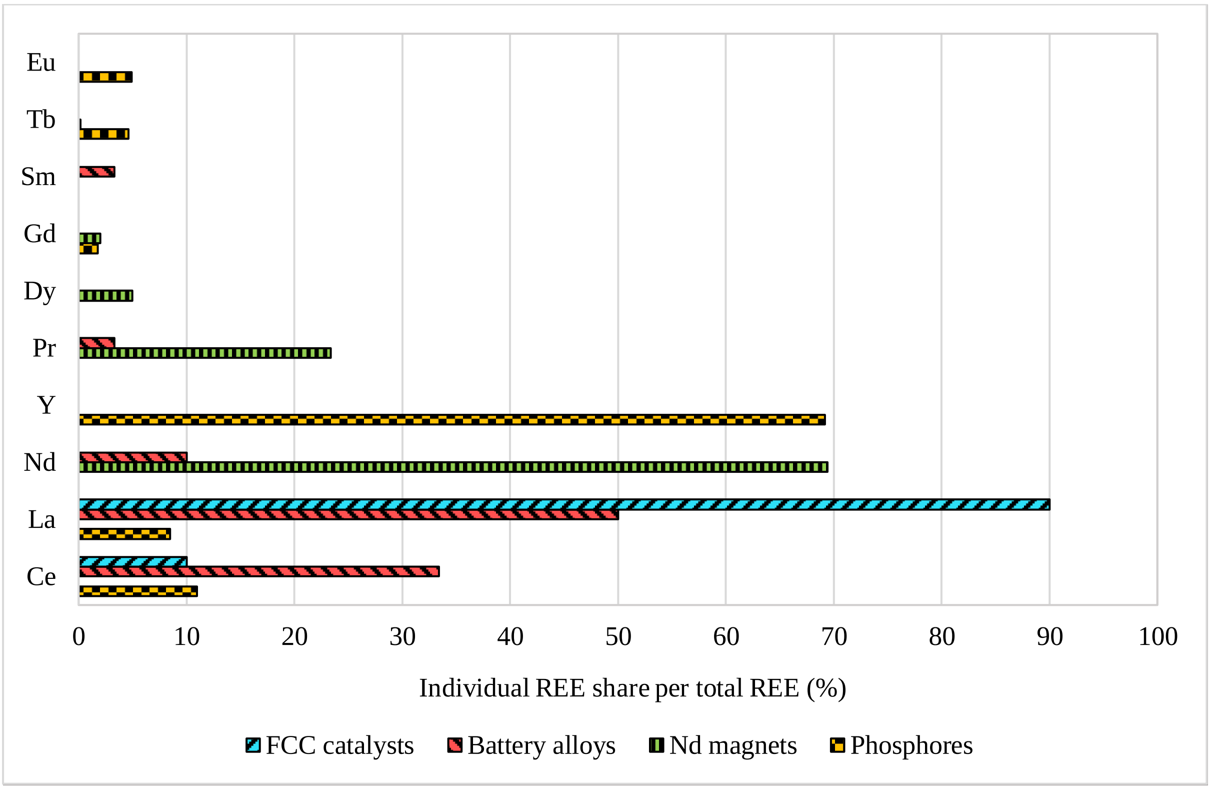


* Data not available for the year 2014

Figure 2


FCC catalysts
Battery alloys
Nd magnets
Phosphores
0
5
10
15
20
25
30
Total REE content (%)

(a)


Eu
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Dy
Pr
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10
20
30
40
50
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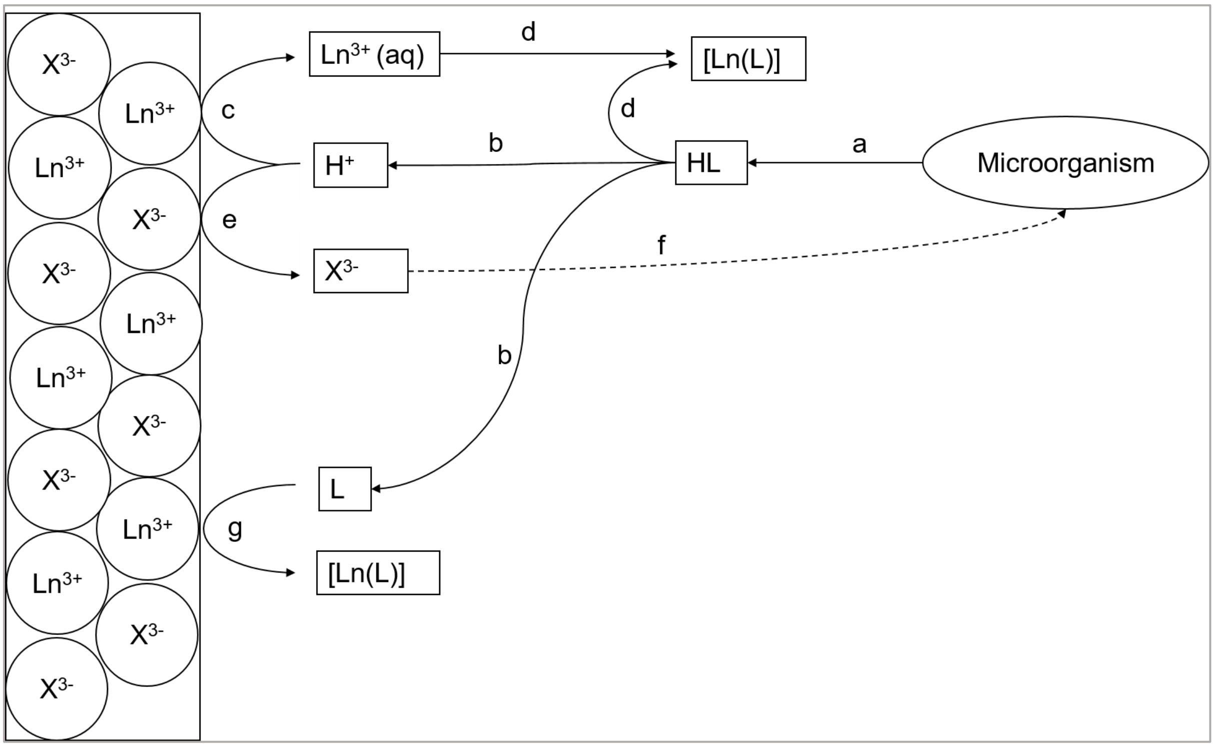
90
100
Individual REE share per total REE (%)
FCC catalysts
Battery alloys
Nd magnets
Phosphores

(b)

Figure 3


X3-
Ln3+
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Ln3+
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X3-
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X3-
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X3-
X3-
c
e
g
Ln3+ (aq)
d
[Ln(L)]
d
H+
b
HL
a
Microorganism
X3-
f
b
L
[Ln(L)]

Figure 4

Table 1

Leaching agent	Producing microorganisms	Production process	REE complexation reactions	pKa and logK values*^	References for pKa and logK values
Sulfuric acid	Autotrophic sulfur oxidizing bacteria i.e. *Acidithiobacillus ferrooxidans*	Oxidization of sulfur or sulfides to sulfuric acid by bacteria	$H_2SO_4 \leftrightarrow HSO_4^- + H^+$ $HSO_4 \leftrightarrow H^+$ $REE^{3+} + HSO_4^- \leftrightarrow REE(HSO_4)^{2+}$ $REE^{3+} + 2\ HSO_4^- \leftrightarrow REE(HSO_4)_2^+$	$pK_a = 1.99$ $pK_a = -1.99$ logK = 3.4 – 3.3 logK = 5.1 – 5.3	Marshall and Jones (1966); Smith and Martell (1976); Spahiu and Bruno (1995)
Nitric acid	Nitrifying MO e.g. *Nitrosomonas* spp. and *Nitrobacter* spp.	Oxidization of ammonia via nitrite to nitrate by archaea or bacteria	$HNO_3 \leftrightarrow NO_3^- + H^+$ $REE^{3+} + NO_3^- \leftrightarrow REE(NO_3)^{2+}$	$pK_a = 3.15$ logK = 0.8 – 0.6	Smith and Martell (1976); Spahiu and Bruno (1995)
Gluconic acid	Heterotrophic MO e.g. *Gluconobacter oxydans*, *Aspergillus niger*, *Penicillium simplicissimum*	Oxidization of Glucose or glucose-containing substances to gluconic acid under aerobic conditions	$C_6H_{12}O_7 \leftrightarrow (C_6\ H_{11}O_7)^- + H^+$ $REE^{3+} + (C_6H_{11}O_7)^- \leftrightarrow REE(C_6H_{11}O_7)^{2+}$ $REE^{3+} + 2\ (C_6H_{11}O_7)^- \leftrightarrow REE(C_6H_{11}O_7)_2^+$	$pK_a = 3.66$ logK = 2.32 – 2.85 logK = 4.25 – 4.78	Martell and Smith (1977)
Citric acid	Heterotrophic MO e.g. *Yarrowia lipolytica*, *Aspergillus niger*	An intermediate of the TCA cycle, occurring in metabolism of aerobic MO	$C_6H_8O_7 \leftrightarrow (C_6H_7O_7)^- + H^+$ $(C_6H_7O_7)^- \leftrightarrow (C_6H_6O_7)^{2-} + H^+$ $(C_6H_6O_7)^{2-} \leftrightarrow (C_6H_5O_7)^{3-} + H^+$ $REE^{3+} + (C_6H_5O_7)^{3-} \leftrightarrow REE(C_6H_5O_7)$	$pK_a = 6.40$ $pK_a = 4.76$ $pK_a = 3.13$ logK = 7.1 – 8.07	Smith and Martell (1989)
Oxalic acid	Heterotrophic MO e.g. *Aspergillus niger*	An intermediate product TCA cycle; Excreted under alkaline conditions	$C_2H_2O_4 \leftrightarrow (C_2HO_4)^- + H^+$ $(C_2HO_4)^- \leftrightarrow (C_2O_4)^{2-} + H^+$ $REE^{3+} +(C_2O_4)^{2-} \leftrightarrow REE(C_2O_4)^+$ $REE^{3+} + 2\ (C_2O_4)^{2-} \leftrightarrow REE(C_2O_4)_2^-$	$pK_a = 4.20$ $pK_a = 1.20$ logK = 4.71 – 5.60** logK = 7.83 – 9.52**	Martell and Smith (1977)
Acetic acid	Acetic acid bacteria e.g. *Acetobacter aceti*	Oxidization of sugars or ethanol to acetic acid by bacteria	$C_2H_4O_2 \leftrightarrow (C_2H_3O_2)^- + H^+$ $REE^{3+} + (C_2H_3O_2)^- \leftrightarrow REE(C_2H_3O_2)^{2+}$ $REE^{3+} + 2\ (C_2H_3O_2)^- \leftrightarrow REE(C_2H_3O_2)_2^+$ $REE^{3+} + 3\ (C_2H_3O_2)^- \leftrightarrow REE(C_2H_3O_2)_3$	$pK_a = 4.70$ logK = 1.82 – 1.85 logK = 2.82 – 3.16 logK = 3.53 – 4.02	Martell and Smith (1977)
Malic acid	Heterotrophic MO e.g. *Pseudomonas rhizosphaerae*, *Aspergillus niger*	An intermediate of the TCA cycle	$C_4H_6O_5 \leftrightarrow (C_4H_5O_5)^- + H^+$ $(C_4H_5O_5)^- \leftrightarrow (C_4H_4O_5)^{2-} + H^+$ $REE^{3+} + (C_4H_4O_5)^{2-} \leftrightarrow REE(C_4H_4O_5)^-$ $REE^{3+} + 2\ (C_4H_4O_5)^{2-} \leftrightarrow REE(C_4H_4O_5)_2^+$	$pK_a = 5.11$ $pK_a = 3.40$ logK = 4.37 – 5.08 logK = 7.16 – 8.67	Martell and Smith (1977)
DFOB	Heterotrophic MO e.g. *Streptomyces pilosus*	Ferric iron scavenging	$(C_{25}H_{45}N_6O_8) \leftrightarrow (C_{25}H_{45}N_6O_8)^- + H^+$ $(C_{25}H_{45}N_6O_8)^- \leftrightarrow (C_{25}H_{45}N_6O_8)^{2-} + H^+$ $(C_{25}H_{45}N_6O_8)^{2-} \leftrightarrow (C_{25}H_{45}N_6O_8)^{3-} + H^+$ $(C_{25}H_{45}N_6O_8)^{3-} \leftrightarrow (C_{25}H_{45}N_6O_8)^{4-} + H^+$ $REE^{3+} + (C_{25}H_{45}N_6O_8)^{3-} \leftrightarrow REE(C_{25}H_{48}N_6O_8)$	$pK_a = 8.54^{\#}$ $pK_a = 9.06^{\#}$ $pK_a = 9.70^{\#}$ $pK_a = 10.89^{\#}$ logK = 10.09 – 15.19#	Christenson and Schijf (2011)
Succinic acid	Heterotrophic MO e.g *Aspergillus niger*	An intermediate of TCA cycle	$C_4H_6O_4 \leftrightarrow (C_4H_5O_4)^- + H^+$ $(C_4H_5O_4)^- \leftrightarrow (C_4H_4O_4)^{2-} + H^+$ $REE^{3+} + (\ C_4H_5O_4)^- \leftrightarrow REE(C_4H_5O_4)^{2+}$ $REE^{3+} + 2\ (C_4H_5O_4)^- \leftrightarrow REE(C_4H_5O_4)^+$	$pk_a = 5.64$ $pk_a = 4.21$ logK = 1.48 – 1.76 logK = 2.7 – 3.1	Martell and Smith (1997)

Me = Metal, MO = Microorganism, TCA = tricarboxylic acid, * logK values given for La^{3+} – Lu^{3+} except were specified otherwise, **Data for La^{3+} - Tb^{3+}, ^ = pKa values at 25 °C, *I* = 0, logK values are given for 25 °C and *I* = 0.1 except for malic acid where logK values are given for 20 °C and *I* = 0, # = Values are given for 25 °C, *I* = 0.7

Table 2

Primary source	**REEs present**	**Microorganism used for leaching**	**Experiment type**	**Identified Leaching agents (acids)**	**Pulp density (% w/v)**	**Time (d)**	**Temp (°C)**	**Total REE leaching efficiency (%)**	**Reference**	**Optimum growth temp (°C)**	**Reference**
Bastnaesite-bearing rock	Ce, La, Y, Pr, Nd, Sm, Eu, Gd, Tb	*Streptomyces sp*. FXJ1.172	batch, shake flask, spent medium	pyruvic, lactic, oxalic, siderophores possibly DFOE	0.5	20	25-28	0.08	Zhang et al. (2018)	n.a	
Bastnaesite (Pure)	Ce, La, Y, Pr, Nd, Sm, Eu, Gd, Tb	*Micromonospora sp*. R171	batch, shake flask, one-step	pyruvic, lactic, oxalic	0.5	15	25-28	0.1	Zhang et al. (2018)	20-40	Genilloud (2015)
Carbonaceous shales	n.a	*Aspergillus niger*	batch, shake flask, one step	citric, oxalic	1	7	30	86	Amin et al. (2014)	35 - 37	Kück et al. (2009)
Gibbsite ore	n.a	*Acidithiobacillus ferrooxidans*	column bioleaching	sulfuric acid	n.a	n.a	30	67.58	Ibrahim & El-Sheikh (2011)	30–35	Kelly &Wood (2015)
Ion-adsorption clay	La Ce Dy Lu	*Aspergillus sp.*	batch, shake flask, two step	gluconic, citric, oxalic	2	$^{a}2 + {}^{b}3$	25	La(72.6), Dy(62.2), Lu(64.8), Ce(15.3)	Barnett et al. (2018)	n.a	
Ion-adsorption clay	La Ce Dy Lu	*Bacillus sp.*	batch, shake flask, two step	gluconic, citric, oxalic	2	$^{a}2 + {}^{b}3$	25	La(64.8), Dy(51.2), Lu(50.8) Ce(14.0)	Barnet et al. (2018)	n.a	
Monazite	Ce, La, Nd, Pr	*Aspergillus terreus* strain ML3-1	batch, shake flask, one step	itaconic, succinic	1	6	25-28	3-5	Brisson et al. (2016)	n.a	
Monazite	Ce, La, Nd, Pr	*Paecilomyces spp*. strain WE3-F	batch, shake flask, one step	acetic, gluconic, succinic	1	6	25-28	3-5	Brisson et al. (2016)	n.a	
Monazite	Ce	*Aspergillus niger*	batch, shake flask, one step	oxalic, acetic, citric, succinic, kojic, itaconic	2	30	30	Ce(100)	Keekan et al. (2017)	35 - 37	Kück et al. (2009)
Monazite	n.a	*Aspergillus ficuum*	batch, shake flask, one-step	citric, oxalic	0.6	9	30	75.4	Hassanien et al. (2014)	n.a	
Monazite	n.a	*Pseudomonas aeruginosa*	batch, shake flask, one-step	2-ketogluconic	0.6	8	35	63.5	Hassanien et al. (2014)	37	Palleroni (2015)
Monazite	Ce, La, Nd, Pr,	*Acetobacter aceti*	batch, shake flask, one-step	citric, malic, tartaric, acetic	16.7	9	30	Ce (0.13) La (0.11)	Shin et al. (2015)	30	Sievers & Swings (2015a)
Monazite	Ce La Nd Y Pr	*Penicillium sp*.CF1	batch, shake flask, one step	oxalic, citric, phosphatases	0.5	8	30-37	0.79	Corbett et al. (2018)	n.a	

Table 2. Continued.

Monazite	Ce La Nd Y Pr	*Penicillium sp*.CF1 + indigenous consortia	batch, shake flask, one step	oxalic, citric, phosphatases	0.5	8	30-37	1.52	Corbett et al. (2018)	n.a	
Monazite	Ce, La, Nd, Pr, Y	Enterobacter aerogenes + *Acidithiobacillus ferrooxidans*	batch, shake flask, one step	gluconic, acetic, malic + sulfuric	1	12	30	1.28	Fathollahzadeh, Hackett, et al (2018)	n.a	
Quartz-pebble conglomerate ores	LREEs (La,Ce, Pr,Nd,Sm,Eu, Gd) HREEs (Tb,Dy, Ho,Er,Tm,Yb, Lu)	Environmental consortium isolated from Elliot Lake mine waters	column bioleaching	n.a	n.a	210	n.a	HREEs (29-54) LREEs (6-23)	Williamson (2014)	n.a	
Quartz-pebble conglomerate ores	Y, Ce,Pr, La,Nd,Yb, Dy,Sm	Endogenous acidophiles	column bioleaching	n.a	n.a	364	n.a	45	Sapsford et al. (2012)	n.a	
Soil	n.a	*Streptomyces fungicidicus* YH04	batch,shake flask, one step	n.a	1	2	30	37.13	Hewedy, Rushdy & Kamal (2013)	n.a	
Th-U concentrate	La, Ce, Y	*Aspergillus ficuum*	batch, shake flask, Spent medium	citric, oxalic	0.75	$^{a}10 + {}^{b}1$	$^{a}28, {}^{b}25$	La(20), Ce(33), Y(2.51)	Desouky et al. (2016)	n.a	
Th-U concentrate	La, Ce, Y	*Pseudomonas aeruginosa*	batch, shake flask, Spent medium	siderophores	1.20	$^{a}8 + {}^{b}1$	$^{a}35, {}^{b}25$	La(4.3),Ce(5.4), Y(1.2)	Desouky et al. (2016)	37	Palleroni (2015)
Th- U concentrate	n.a	*Aspergillus ficuum*	batch,shake flask, one-step	citric, oxalic	1.20	9	30	63.8	Hassanien et al. (2014)	n.a	
Th- U concentrate	n.a	*Pseudomonas aeruginosa*	batch,shake flask, one-step	2-ketogluconic	1.20	8	35	52.6	Hassanien et al. (2014)	37	Palleroni (2015)
Zircon	Y, La, Ce, Pr, Nd, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb, Lu	*Acetobacter methanolicus*	batch,shake flask, one step	gluconic	1	10	32	67	Glombitza, Iske, & Bullmann (1988)	30-32	Uhlig, Karbaum & Steudel (1986)
Zircon	Y, La, Ce, Pr, Nd, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb, Lu	*Acidithiobacillus ferrooxidans*	batch,shake flask, one step	sulfuric	1	10	32	80	Glombitza, Iske, & Bullmann (1988)	30–35	Kelly & Wood (2015)

n.a = not available ^{a} = microorganism growth

^{b} = leaching using biolixiviants d = day

Table 3

Secondary source	**REE**	**Microorganism used for leaching**	**Experiment type**	**Identified Leaching agents (acids)**	**Pulp density (% w/v)**	**Time (d)**	**Temp (°C)**	**Total REE leaching efficiency (%)**	**Reference**	**Optim. growth temp (°C)**	**Reference**
Ash slag waste	Sc, Y, La, Nd, Sm,Gd,Ce,Pr,Dy, Er, Eu	Acidophilic chemolithoautotrophs	batch,shake flask, one step	sulfuric	3	10	45	15-30	Tsaplina et al. (2015)	n.a	
Ash slag waste	Y, Sc, La, Nd, Gd, Sm	Acidophilic chemolithoautotrophs	batch, stirred bioreactor, one step	sulfuric	10	10	45	Sm(26.5), Gd(27.6) Sc(52), Y(52.6), La(59.5), Nd (22.1),	Muravyov et al. (2015)	n.a	
Ash slag waste	Y, Sc, La	Acidophilic chemolithoautotrophs	batch, airlift percolators	sulfuric	n.a	60	20-22	Y(31), Sc(30), La(33)	Muravyov et al. (2015)	n.a	
Coal fly ash	Dy,Y,Sc,Er,Yb, Gd,Eu,Sm,La, Ce,Pr,Nd	*Candida bombicola*	batch, shake flask, spent medium	acidic sophorolipids	1	3 + 6 h*	28	Dy,Y,Sc,Er,Yb (60-67) Gd(52), Eu(48), Sm(42) La,Ce,Pr,Nd (27-30)	Park & Liang (2019)	n.a	
CRT fluorescent powders	Y	Acidophilic mixed culture	batch,shake flask, one step	sulfuric	10	16	30	Y (70)	Beolchini et al. (2012)	n.a	
FCC catalyst	Y,La,Ce,Pr,Nd, Sm,Eu,Gd,Tb, Ho,Yb,Lu,,Dy, Er,Tm	*Gluconobacter oxydans*	batch,shake flask, spent medium	Gluconic	1.5	^{a}4 + ^{b}1	30	49	Reed et al. (2016)	25-30	Sievers & Swings (2015b)
FCC catalyst	Y,La,Ce,Pr,Nd, Sm,Eu,Gd,Tb, Ho,Yb,Lu,,Dy, Er,Tm	*Gluconobacter oxydans*	batch, stirred bioreactor, spent medium	Gluconic	1.5	^{a}36-40 h + ^{b}1	30	56	Thompson et al. (2018)	25-30	Sievers & Swings (2015b)
FCC catalyst	Y,La,Ce,Pr,Nd, Sm,Eu,Gd,Tb, Ho,Yb,Lu,,Dy, Er,Tm	*Gluconobacter oxydans*	continuous, stirred bioreactor, spent medium	Gluconic	1.5	^{a}100 h* + ^{b}1	30	51	Thompson et al. (2018)	25-30	Sievers & Swings (2015b)
FCC catalyst	Y,La,Ce,Pr,Nd, Sm,Eu,Gd,Tb, Ho,Yb,Lu,,Dy, Er,Tm	*Gluconobacter oxydans*	column bioleaching, spent medium	Gluconic	1.5	1	30	46	Thompson et al. (2018)	25-30	Sievers & Swings (2015b)
Fluorescent phosphors	Y,La,Ce,Pr,Nd, Sm,Eu,Gd,Tb, Ho,Yb,Lu,,Dy, Er,Tm	*Gluconobacter oxydans*	batch, shake flask, spent medium	Gluconic	1.5	^{a}4 + ^{b}1	30	2	Reed et al. (2016)	25-30	Sievers & Swings (2015b)

Table 3. Continued.

Fluorescent phosphors	Y, La, Ce, Eu, Gd, Tb	Symbiotic mixed culture of *Kombucha*	batch, shake flask, one step	acetic, gluconic	2.85	14	25	6.5	Hopfe et al. (2017)	n.a	
Fluorescent phosphors	Y, La, Ce, Eu, Gd, Tb	*Lactobacillus casei*	batch, shake flask, one step	Lactic	2.85	14	25	6.1	Hopfe et al. (2018)	30-40	Hammes & Hertel (2015)
Fluorescent phosphors	Y, La, Ce, Eu, Gd, Tb	*Komagataeibacter xylinus*	batch, shake flask, one step	citric, Tartaric, Gluconic	2.85	14	25	12.6	Hopfe et al. (2018)	n.a	
Fluorescent phosphors	Y, La, Ce, Eu, Gd, Tb	*Yarrowia lipolytica*	batch, shake flask, one step	citric	2.85	14	25	10.6	Hopfe et al. (2018)	n.a	
Municipal solid waste incinerator fly ash	Nd, Ce, Sm, La	Acidophilic mixed culture	batch, stirred bioreactor, two step	sulfuric	5	$^{a}4 + ^{b}21$	30	Nd(76), Ce(48), Sm(9), La(30)	Funari et al. (2017)	n.a	
Phosphogypsum	Y	*Desulfovibrio desulfuricans*	column bioleaching	n.a	n.a	n.a	32	Y(77)	Dudeney (1993)	25–35	Niel et al. (2015)
Red mud	La, Ce, Pr, Nd, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb, Lu, Y, Sc	*Penicillium tricolor*	batch, shake flask, one step	oxalic, citric, gluconic	2	50	30	LREEs (~40) HREEs (~65)	Qu & Lian (2013)	n.a	
Red mud	Sc, La, Eu, Yb	*Aspergillus niger*	batch, shake flask, spent medium	citric, oxalic, gluconic	2	30	30	Yb(59), Sc (44), Eu(>30), La(>30)	Qu & Lian (2015)	35 - 37	Kück et al. (2009)
Red mud	Sc, La, Eu, Yb	*Aspergillus niger*	continuous, stirred bioreactor, one step	citric, oxalic, gluconic	2	$^{a}60\ h^{*} + ^{b}4$	30	Yb(62), Sc(45), Eu (30), La (27)	Qu et al. (2015)	35 - 37	Kück et al. (2009)
WEEE Shredding dust	La,Nd ,Y,Ce,Eu	*Acidithiobacillus thiooxidans*	batch, shake flask, two-step	sulfuric	1	8	30	Ce, Eu, Nd (>99) La, Y (80)	Marra et al. (2018)	28-30	Kelly & Wood (2015)

n.a = not available ^{a} = microorganism growth

^{b} = leaching using biolixiviants d = day

h = hour

Table 4

Screened microorganisms	Most efficient microorganism	REE containing source	Reference
Penicillium sp, *Aspergillus niger*, *Aspergillus tubigensis*, *Pantoea agglomerans*, *Enterobacter aerogenes*, *Pseudomonas aeruginosa*, *Pseudomonas putida*, *Klebsiella pneumoniae*, *Klebsiella oxytoca*, and *Bacillus megaterium*	*Penicillium sp*	Monazite	Corbett et al. (2017)
Aspergillus niger, *Aspergillus flavus*, *Aspergillus ficuum*, *Aspergillus terreus*, *Pseudomonas aeruginosa*, *Penicillium cyclopium*, *Penicillium diversum*, and *Penicillium oxalicum*	*Aspergillus niger* > *Aspergillus flavus*	Carbonaceous shales	Amin et al. (2014)
Aspergillus niger and *Bacillus sp.* isolates	*Aspergillus niger*	Ion-adsorption clay	Barnett et al. (2018)
Eight isolated actinomycetes strains including *Streptomyces fungicidicus*, *Streptomyces aureofaciens* and reference isolate of *Streptomyces chibaensis*	*Streptomyces fungicidicus*	Low grade ore	Hewedy et al. (2013)
100 isolates including *Pseudomonas fluorescens*, *Pseudomonas frederiksbergensis*, *Yersinia enterocolitica*, *Acinetobacter calcoaceticus*, *Talaromyces purpureogenus*,	*Acinetobacter calcoaceticus* > *Talaromyces purpureogenus*	Fluidized cracking catalysts (FCC)	Reed, Fujita, Daubaras, Bruhn, Reiss & Thompson (2016)
Acinetobacter calcoaceticus, *Talaromyces purpureogenus*, *Pseudomonas frederiksbergensis*, *Gluconobacter oxydans*	*Gluconobacter oxydans*> *Acinetobacter calcoaceticus*> *Talaromyces purpureogenus*> *Pseudomonas frederiksbergensis*	FCC and retorted phosphor powder (RPP)	Reed et al. (2016)
A wide spectrum of microorganisms including: *Acidithiobacillus ferrooxidans, Acidithiobacillus thiooxidans, Bacillus licheniformis, Burkholderia glumae, Corynebacterium callunae, Corynebacterium stationis, Komatogateibacter xylinus, Lactobacillus casei, Priceomyces haplophilus, Yarrowia lipolytica, Pseudomonas fluorescens, Streptomyces acidiscabies, Lysinibacillus sphaericus*	*Komatogateibacter xylinus*> *Lactobacillus casei*> *Yarrowia lipolytica*	Fluorescent phosphor	Hopfe et al. (2018)
The tea fungus *Kambucha*, *Komatogateibacter xylinus, Lactobacillus casei, Yarrowia lipolytica*	*Komatogateibacter xylinus* > *Lactobacillus casei* ≥ *Kambucha* > *Yarrowia lipolytica*	Fluorescent phosphor	Hopfe et al. (2017, 2018)
Azospirillum brasilense, Mesorhizobium cicero, Azospirillum lipoferum, Pseudomonas rhizosphaerae, Acetobacter aceti	*Acetobacter aceti*	Monazite	Shin et al. (2015)
Phanerochaete chrysosporium, *Candida bombicola, Cryptococcus curvatus*	*Candida bombicola* > *Cryptococcus curvatus* > *Phanerochaete chrysosporium*	Coal fly ash	Park & Liang (2019)

Table 5

Salt	Solubility [mol/kg]	Reference
$(REE)(NO_3)_3$	6.8 – 4.2	Rard and Spedding (1975)
$(REE)Cl_3$	4.2 – 3.6	Das et al. (2017), Mioduski, Guminski & Zeng (2008), Mioduski, Guminski & Zeng (2009a), Mioduski, Guminski & Zeng (2009b)
$(REE)_2(SO_4)_3$	$10^{-1} – 10^{-2}$	Spedding & Jaffe (1954)* Das et al. (2019)
$(REE)_2Na_2(SO_4)_4$	$10^{-1} – 10^{-4}$	Das et al. (2019)+
$(REE)F_3$	$10^{-5} – 10^{-6}$	Migdisov et al. (2009)*, Mioduski, Guminski & Zeng (2014a), Mioduski, Guminski & Zeng (2014b), Mioduski, Guminski & Zeng (2015)
$(REE)_2(Oxalate)_3$	$\sim10^{-7}$	Chung, Kim, Lee & Yoo (1998)*
$(REE)(OH)(CO_3)$	$10^{-5} – 10^{-7}$	Kim et al. (2018)*
$(REE)(OH)_3$	$10^{-6} – 10^{-8}$	Diakonov, Ragnarsdottir and Tagirov (1998)*
$(REE)_2(CO_3)_3$	$10^{-7} – 10^{-8}$	Kim et al. (2018)*
$(REE)(PO_4)$	$10^{-13} – 10^{-14}$	Liu and Byrne (1997)*

* Solubilities were calculated from solubility products given in the references

\+ Solubilties of $(REE)_2Na_2(SO_4)_4$ were estimated from figure 39 in the paper by Das et al. (Das et al., 2019)