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Review

Potential Future Alternative Resources for Rare Earth Elements: Opportunities and Challenges

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Abstract: Currently, there is increasing industrial demand for rare earth elements (REE) as these elements are now integral to the manufacture of many carbon-neutral technologies. The depleting REE ores and increasing mining costs are prompting to look for alternative sources for these valuable metals, particularly from waste streams. Although REE concentrations in most of the alternate resources are lower than current REE ores, some sources such as marine sediments, coal ash, and industrial wastes like red mud are looking promising with significant concentrations of REE in them. This review focuses on the alternative resources for REE such as ocean bottom sediments, continental shelf sediments, river sediments, stream sediments, lake sediments, phosphorites deposits, industrial waste products like red mud, and phosphogypsum, coal, coal fly ash, and related materials, waste rock sources from old and closed mines, acid mine drainage, and recycling of e-waste. Possible future Moon exploration and mining for REE and other valuable minerals are also discussed. It is evident that REE extractions from both primary and secondary ores alone are not adequate to meet the current demand, and sustainable REE recovery from the alternative resources described here is also necessary to meet the growing REE demand. An attempt is made to identify the potential of these alternative resources and sustainability challenges, benefits, and possible environmental hazards to meet the growing challenges in meeting the future REE requirements.

Keywords: rare earth elements; deposits; alternative sources; marine sediments; river sediments; phosphorites; red mud; fly ash; acid mine drainage; e-waste; extra-terrestrial

1. Introduction

Seventeen elements in the periodic table including fifteen lanthanides (La, Ce, Pr, Nd, Pm, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb, and Lu) plus Sc, and Y are often collectively referred to as rare earth elements (REE). In nature, REE do not occur as single native metals, such as gold or silver, instead because of their similar physical and chemical properties very often they occur together in several geologic formations in many ores or minerals as minor or major constituents. Due to their unique physical, chemical, mechanical, electronic, magnetic, luminescent, phosphorescent, and catalytic properties, these elements have become exceptionally important during the last couple of decades, and there has been an explosion in the industrial application of these elements in different high technology devices such as smartphones, computers, televisions, light emitting diodes, catalysts for fuel cells, corrosion inhibitors, hard drives, and magnets for wind turbines and other power generating systems. They are also crucial for national security as they are extensively used in several military defence systems [1-3]. Their important medical applications include metallic implants, lasers, and magnetic resonance imaging (MRI) measurements [4]. REE are vital even to the space industry as they are used in everything from launch vehicles to national defence and commercial communications and observation satellites. REE are also used in making catalysts in several industrial processes as well as in the fabrication of autocatalytic catalytic converters in transport vehicles [5,6]. Although REE deposits are found practically in all parts of the world, currently there is a global shortage, due to the decreasing number of economically profitable deposits. Also, our reliance on

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REE in our high-tech gadget-hungry world is growing with time, but their supply is far from secure. Unfortunately, the conventional ore resources are becoming depleted, as the demand for REE increases, and the industry is paying attention to unconventional resources like coal, recycling, and marine sediments as alternative sources for these elements. It is also very important to maintain a proper supply chain to meet the demand for the development of highly advanced technologies. Even the substitution of REE by other cheaper metals is difficult for most applications, though powerful permanent magnets made out of iron and nitrogen (iron nitride, $Fe_{16}N_2$) are helping the automotive and wind energy industries in a small way [7]. Pavel et al. [8] studied the possibilities of substitution of REE in wind turbines in order to reduce the dependency on REE, but later it was found that these claims were impossible without neodymium. As a result, recycling is being scaled up in several countries at present. Many approaches such as physical, chemical, and biological procedures such as pyrometallurgy, solvent extraction, and membrane separation are available for the efficient extraction of REE from these resources [9]. One very important aspect is that currently extracted ore grades, with an average of 5% REE and running as high as 15%, have far higher concentrations than most of the discussed alternative sources [10]. Since only a few countries are producing these elements on a commercial scale, meeting the growing demand for these elements is becoming exceedingly difficult in several other countries. As the demand for REE is growing and the conventional ores are also getting depleted, attention is turning to unconventional and alternative REE resources. Therefore, it is very important to understand the global REE resources including alternative resources, and their production scenario and take measures against future prospects. Therefore, this study which is the first of its kind brings out a summary of the primary, and secondary, REE deposits, and a comprehensive report on alternative REE resources and their potential to meet the current and future demand. This article also serves as technical information for alternative resources for REE other than the primary and secondary resources to policymakers and other stakeholders who are concerned. Before knowing more about alternative REE resources, a short review of the major REE primary and secondary deposits is presented below:

2. REE Deposits

Although REE comprise significant amounts of a wide range of minerals including oxides, silicates, carbonates, phosphates, and halides, almost all production comes from less than ten minerals such as apatite, monazite, xenotime, allanite, and bastnasite [11]. Currently, these deposits are mainly located in China, Brazil, Vietnam, Russia, and India. Though China has been dominating in this area, several other countries such as the USA, Australia, Turkey and Sweden have successfully discovered new REE deposits recently. REE deposits are divided into primary (formed by magmatic, hydrothermal and/or metamorphic processes) and secondary deposits (formed by weathering and sedimentary transport) depending on their form of occurrence, genetic associations, and mineralogy [12]. Figure 1 Illustrative view of different types of REE resources including the potential alternate future resources [13]

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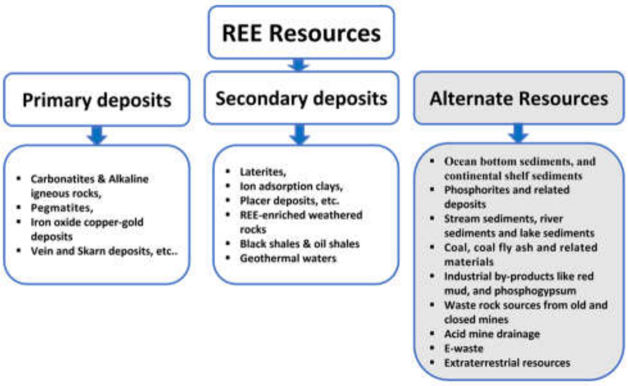

Figure 1. Illustrative view of different types of REE resources including the potential alternate future resources in light shaded, modified after [13].

2.1. Primary Deposits

Primary deposits of REE are commonly associated with carbonatites and alkaline igneous rocks, pegmatites, iron oxide copper-gold deposits, and vein and skarn deposits. Of these carbonatites and peralkaline silicate rocks with appreciable concentrations of REE are the most important REE resources [13, 14]. These REE deposits in igneous rocks have played an important role in meeting the industrial demand for decades. These igneous mineral deposits can be divided into five distinct categories depending on the provenance, the evolution of the magma, and the rock types hosting mineralization: i) carbonatites, ii) peralkaline silica undersaturated rocks, iii) peralkaline granites and pegmatites, iv) pegmatites associated with sub- to metaluminous granites, and v) Fe oxide–phosphate deposits [15]. More insights on these primary REE deposits were presented by Weng et al. [16] and Dushyantha et al. [12]. Skarn deposits of REE are important in sustainable economic development across the world [17]. In Fe oxide–phosphate deposits, these elements are concentrated in apatite minerals [18].

2.2. Secondary Deposits

Some of the important secondary deposits of REE are laterites, ion adsorption clays, and placer deposits. Ion-adsorption deposits, which are formed due to the erosion and weathering of primary deposits from within weathering crusts which supply more than 95% of the global HREE demand. Intensive lateritic weathering of bedrocks under tropical or sub-tropical climatic conditions can form a variety of secondary deposits which may range in composition from aluminous bauxites to iron and niobium, and REE [19]. Recently Jo and Shin [20] reported REE-enriched weathered anorthosite rocks with ΣREE range 242 – 857 μg/g. Ionic clays, formed by the natural weathering process of REE-minerals and the adsorption of the resulting liberated REE ions onto the clay surface, are an important resource for critical REE. These ion-adsorption clay deposits are most suitable for in situ leaching mining, and supply > 95% of the global HREE demand [21]. Ion-adsorption REE deposits are mostly developed due to weathering of REE-rich granites and REE ions are mainly adsorbed on clay minerals [22]. Currently, regolith-hosted REE deposits from areas with abundant REE deposits have become one of the major sources of global REE. Regolith-hosted deposits with abundant REE around Meizhou City, Guangdong Province, South China with the highest ΣREE of 1162 μg/g were reported recently by Lin et al. [23]. The REE concentrations in the regolith-hosted REE deposits in Chilean coastal range of the central Andes were found to be up to 2000 μg/g [24]. Beach sands are the products of a combination of weathering, fragmentation and degradation and are well-known for their economic concentration of heavy minerals like monazite, illuminate, zircon, rutile, allanite, sillimanite and garnet. REE are abundant in some of these minerals, particularly in monazite and allanite [25]. Black shales can also be considered as secondary resource for these valuable metals. Recently, El-Anwar et al. [26] reported REE enrichment (average 255.3–325.3 μg/g) in black shales of the Safaga-Qussier sector, Egypt. Another such study is reported from the rock-soil-moss system in the black shale area in China with an average concentration of ∑REE around 245 µg/g [27]. These recent studies together with the industry standard of ~ 300 µg/g cut-off grade for mining, prompt more studies on black shale formations worldwide before considering the black shale deposits as a source of REE. Ketris and Yudovich [28] reported an average worldwide REE concentration of 134.19 in black shales. Geothermal fluids are potentially significant sources of valuable minerals and metals. The total REE in geothermal waters in places like Iceland can go up to 21.76 µg/g [29]. Kurzawa et al. [30] in an attempt to understand the REE concentrations in the mineral and thermal waters in the Polish Lowlands, found that high temperature ($T > 60$ °C) favoured the release of the more easily soluble REE from rocks into the water. Wei et al. [31] recently reported the total REE content of the geothermal water in the range of 0.059–0.547 ng/ml, in the Ganzi–Litang fault, western Sichuan, China. The REE contents in geothermal waters are related to the reservoir lithology and are significantly influenced by pH and HCO_3^-, Na^+, and Mn minerals. Though the REE content in geothermal waters is not considerable, if the abundant geothermal waters in places such as Iceland are taken into consideration, this source could be one of the most promising ones for REE with an efficient extraction technology. Li et al. [32] reported moderately high concentrations of REE ranging from105 to 195 µg/g at an average concentration of 151µg/g in oil shale samples in Tongchuan City, Southern Ordos Basin, China prompting the necessity for further investigations to understand the REE potential of these rocks.

2.3. Different Types Potential Alternative Deposits

So far, the identified primary and secondary REE resources are not able to meet the global demand and requirements. Finding out and establishing alternative resources for REE has been a topic of high interest for the past decade. Hence, lot of studies are going on for the identification of the potential alternative deposits to meet the growing demand world over. Powerful analytical techniques such as inductively coupled plasma mass spectroscopy (ICP-MS), inductively coupled plasma optical emission spectrometry (ICP-AES), scanning electron microscopy with energy dispersive X-ray spectrometry (SEM-EDX), x-ray fluorescence spectrometry (XRF), x-ray diffractometry (XRD), laser induced breakdown spectroscopy (LIBS), and TESCAN, integrated mineral analysis (TIMA), are being utilised to understand the future potential of various alternative REE resources [33-36]. Following are some of the most important alternative sources for REE.

2.3.1. Ocean Bottom Sediments, and Continental Shelf Sediments

Rifting of the continents has formed widespread sedimentary basins as rifting processes produce greater subsidence and lower basal heat flow, enlarging the depth extent of hydrothermal circulation and favouring the formation of giant deposits of elements such as copper, lead, zinc and nickel [37]. Several studies in recent times revealed that marine sediments such as ferromanganese crusts (cobalt crusts), manganese nodules, and marine mud from different parts of deep oceans are found to contain significant concentrations of REE and Y [38-41]. REE concentration ranges (µg/g) in some marine sediments from different oceans across the globe are presented in Table 1. Recent studies suggested that the continental shelf sediments of the Atlantic Ocean and Northern South China could potentially become resources for REE, similar to metalliferous deep-sea sediments [42,43]. Sediment samples from the northern South China Sea continental shelf reported concentrations of REE ranging from 32.97 µg/g to 349.07 µg/g, with an average concentration of 192.94 µg/g. These elevated concentrations of REE in the bottom sediments of Siberian seas in the Arctic were found to be mainly controlled by the characteristics of the original rocks [44]. Thus, deep-sea sediments because of their vast amounts of REE and a remarkably high proportion of HREE have attracted interest as a future REE resource. However, at the moment the regulatory authorities are not able to take decisions on future seabed mining because of the possible environmental effects of seabed mining [45].

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Table 1. Overview of ΣREE range (µg/g) in marine sediments from different oceans (modified after Balaram, 2019).

Ocean.	Matrix	ΣREE range (µg/g)	Reference
East Siberian Arctic Shelf	Bottom sediments	104 to 220	[44]
Central North Pacific Ocean	Siliceous sediments	810.4	[46]
Afanasy Niktin Seamount (ANS) in the Eastern Equatorial Indian Ocean.	Cobalt crust	1727 – 2511	[38,39]
Mid-Pacific seamount	Cobalt-rich crusts	2085	[47]
Indian Ocean	Ferromanganese crust	928 - 1570	[48]
Scotia Sea	Ferromanganese crust	3400	[49]
Eastern South Pacific	Deep sea mud	1000 – 2230	[50]
North Pacific (east & west of Hawaiian Islands)	Deep sea mud	400 – 1000	
Minamitorishima Island in the Western North Pacific	REE-Rich Mud	>1446.2 (REE+Y)	[51]
South China Sea	Ferromanganese nodule deposits	1460 (avg)	[52]
Indian Ocean	REY-rich mud	> 400	[53]
	Marine sediments	585 - 920	
Andaman Sea, Indian Ocean	Ferromanganese crust, summit of southern seamount	1139	[54]
	Ferromanganese crust within the two peaks of the same seamount.	2285	
Lakshadweep Sea, Indian Ocean	Ferromanganese crust,	La (200) & Y (150)	[55]
West Sewell Ridge, Andaman Sea, Indian Ocean	Ferromanganese crust	1600	[56]
	Manganese nodules	1186	
Clarion-Clipperton Fracture Zone, North-eastern Pacific Ocean	Deep-sea sediments	>700	[57]
West Clarion–Clipperton Zone, Pacific Ocean	Marine sediments	454.7 (REE+Y)	[58]
North Pacific Ocean near Minamitorishima Island, Japan	Deep-sea mud	> 5,000 (REE+Y)	[60]
Mid Pacific Ocean	Fe-Mn nodules	1178 - 1434	[60]
Pacific Ocean	Deep nodules	1326	[61]
	Shallow nodules	1398	
Pacific Ocean	ocean-floor sediments	22,000	[62]

2.3.2. Phosphorites and Related Deposits

Phosphate ores are divided into two main types on the basis of their origin: sedimentary and igneous phosphate rocks, about 80 % of which are derived from marine origin, and around 17 % are derived from igneous rocks, and the remaining deposits come from the residual sedimentary and guano-type deposits [63]. These rocks contain up to 1% ∑REE, and phosphate rock ores both igneous and sedimentary are, therefore, potential future resources of REE. Igneous phosphate rocks (e.g., Kola Peninsula, Russia, and Brazil), and sedimentary phosphate rocks which are formed by the deposition of phosphate-rich materials in marine environments and are also known as phosphorites are found in Florida, US, Morocco, and the Middle East [64]. Marine phosphorites concentrate REE including Y during the early diagenetic process. The REE content in seamount phosphorites is far greater than in continental margin phosphorites [39]. Morocco has the largest phosphate ore reserves in the world with 50 billion tons, followed by China and Egypt with 3.2 and 2.8 billion tons of phosphate ores respectively [65]. Based on the REE concentration data presented in Table 2. phosphorite deposits can be considered promising and could represent a profitable alternative source for REE in the future. Buccione et al. [66] made an attempt for understanding the economic potential of Northern African phosphorite deposits as alternative REE resources. The very high concentrations of REE (up to 1759 μg/g) in the Northern African phosphorites revealed that they can be considered promising alternative REE resources.

Table 2. Concentrations of REE in marine phosphorites and related rocks from different regions across the world.

Ocean	Phosphorites	Average Concentration (μg/g)	Reference
Pacific and northeast Atlantic	Seamount phosphorites	727 (∑REE+ Y)	[40]
	Continental margin phosphorites	161 (∑REE+ Y)	
Doushantou Formation, South China	Danzhai phosphorite deposit	21 to 447 (∑REE)	[67]
Meishucun excavation sites, South China	Cambrian phosphorites	99.1 - 709.7 (∑REE)	[68]
Sedimentary Abu Tartur phosphate ore, Egypt	Phosphate ore	0.05-0.20 wt. % (∑REE)	[65]
Mississippian phosphorites, USA	Phosphorite ore	18,000 (∑REE)	[69]
Mountain Pass phosphorites, USA	Phosphorite ore		
Chinese clay-type Phosphorite deposits	Phosphorite ore	500 to 2000 (∑REE)	[70]
Hazm Al-Jalamis Phosphorites, Saudi Arabia	Phosphorites	<121.8 (∑REE+ Y)	[71]
Pabdeh Formation, Khormuj anticline, SW of Iran	Phosphorites	48 to 682 ∑REE	[72]
Northern African phosphorite deposits (Morocco, Algeria and Tunisia)		39.2 to 1759.4 ∑REE	[66]
South China	Phosphorus-bearing dolomites	330 ∑REY	[73]
	Phosphorus dolomites	676 ∑REY	
	Phosphorites	1477 ∑REY	

2.3.3. Stream Sediments, River Sediments and Lake Sediments

If REE-bearing source rocks are present in the catchment area of a river or a lake or a stream, the corresponding sediments continuously accumulate transported material and get enriched with respect to REE due to the physical and chemical weathering and erosion of source rocks. An increasing trend in the concentration of REE is seen in the sediments starting from river sediments to the estuary and adjacent bay in the case of Yellow River (Table 3). In sediments, Fe minerals and clay minerals usually promote the accumulation of REE. Recently, Klein et al. [74] undertook a study of the occurrence and spatial distribution of REE along with a few other elements such as Ga, Ge, Nb, In, Te, and Ta in the Rhine River sediments and its tributaries in Europe. The Rhine River represents one of the most important waterways in Europe accommodating a large number of industries with a catchment area of around 60 million inhabitants. The ∑REE concentrations show a steady increase along the Rhine River (Table 3) inferring that. the sources for these elements could be from natural as well as anthropogenic activities as the river sediments pose an important sink for anthropogenically introduced REE. Ramesh et al. [75] presented the distribution pattern of REE and several other trace elements in the sediments of the Himalayan rivers (Table 3). Since this data is more than two decades, the current concentrations can be considerably higher due to increased anthropogenic inputs as well as natural inputs. The Lake sediments in Sri Lanka recorded 1101 µg/g of REE (Table 3) because it is in the proximity of the Eppawala phosphate deposit [76]. Either REE containing source rocks or anthropogenic inputs, or both can be contribute for REE in all these sediments.

Table 3. REE concentrations in stream sediments, river sediments, and lake sediments in different parts of the world.

Country	Type of sediment	∑REE	Reference
Indigirka River, in the Laptev Sea	River sediments	124 to 197	[44]
Rhine river sediments, Europe	Upper Rhine	136.07	[74]
	Middle Rhine	215.32	
	Lower Rhine	340.45	
	Tributaries	291.39	
Himalayan river system sediments	Brahmaputra	95	[75]
	Ganges	97	
	Megna	107	
	Padma	131	
	Jamuna	152	
	Yamuna	100	
Rivers of the east coast of India	Cauvery	171	[77]
	Pennar	203	
	Krishna	131	
	Godavari-Vasista	194	
	Godavari-Gauthami	290	
	Mahanadi	270	
	Hooghly	167	
South America	Amazon sediments	217	[78]
The Mgoua watershed, Cameroon, Africa	Sediments	282 to 728 Average 550	[79]
China	Stream sediments	212	[80]
	Catchment sediments	187	
	Soils	190	
Sri Lanka	Lake sediments*	1011	[74]
Yellow River, China	River sediment	149	[81]
	Estuary	165	
	Laizhou Bay	173	

* Lake sediments in the proximity of the Eppawala phosphate deposit in Sri Lanka. Sediment fraction (<63 µm) of each sample.

2.3.4. Coal, Coal Fly Ash and Related Materials

Coal contains trace amounts of REE either in the form of discrete minerals or chemically bound to organic matter. Several investigations have demonstrated that coal ash may represent a promising alternative resource of REE as the demand for these metals is increasing. Some investigations revealed that the total REE concentrations in the organic and inorganic phases are 31 µg/g and 1,141 µg/g, respectively [82]. Coal fly ash is one of the largest industrial waste streams in the world which contains significant concentrations of REE and has the potential to become an important alternative resource for REE. Until now, this is being used as a building material for construction, concrete and road base, ceramics, etc. A of studies are going on the development of economically feasible procedures for the extraction of REE from coal and coal fly ash as there are large global reserves of coal and coal fly ash the world over. Yesenchak et al. [83] made a study of West Virginia's coal deposits for understanding their economic potential on an ash basis with respect to REE and also to gain insight into elemental modes of occurrence and possible enrichment mechanisms of these elements. Dai et al. [84] have provided an excellent review of anomalous REE and yttrium concentrations in coal. Table 4 presents the potential of these alternative REE resources. A big country like China uses about 4 billion tons of coal for its power plants to generate electricity which produces about 500–550 million tons of fly ash [85]. Coal and coal combustion products such as fly ash, bottom ash, and incinerator ash are found to contain significant amounts of REE as most coal samples contain minor amounts of rhabdophane, Nd, Ce, La $(PO_4).H_2O$, monazite, (Ce, La, Th, Nd)PO_4, and xenotime (Y, Er)PO_4 [86]. Some of the coal resources are enriched in REE due to the contributions from both detrital and hydrothermal sources (Hower et al. [87]. Significant concentrations of REE found in the global reserves of coal (average REE concentrations are estimated at 380–470 µg/g) and its by-products have prompted numerous research studies to understand their economic feasibility as alternative resources for REE and led to the development of several efficient procedures to extract REE from coal ashes [88]. Hence, coal fly ash is considered a potential alternative source for REE in recent years. Recently Lu et al. [89] reported that the REE and Y contents of coals from southwestern China are high, and the coal reserves and their by-products are suitable as potential REE sources. For example, in Poland which is the second biggest coal consumer in the European Union, there can be huge reserves of fly ash which is rich in REE. The concentrations of REE in certain coal deposits such as Pond Creek coalbed, Pike County, Kentucky, US can go up to 1000 µg/g or more. The collection and chemical analysis of over 4,000 samples including acid mine drainage, coal, and coal ash samples, with REE concentrations ranging from ng/ml for raw acid mine drainage materials to thousands of ppm (µg/g) for alternative coal-based resources such as coal and coal fly ash, USGS [90] recently demonstrated the technical feasibility of producing high-purity critical minerals, including REE, from low-grade coal-based materials. For a thorough understanding of the origin of anomalous concentrations of REE in coal, in addition to the influence of diagenetic processes such as temperature, pressure, and time associated with coal-rank advance, it is also necessary to understand a few more interaction mechanisms such as the influence of marine environments, input of hydrothermal fluids, volcanic ashes, and natural waters on peat swamp [83]. In addition, the presence of considerable amounts of radioactive nuclides (^{226}Ra, ^{232}Th, ^{40}K and ^{238}U) in fly ash must be also considered while developing efficient procedures for the extraction of REE, and see that these hazardous radionuclides should not escape in to the environment causing radioactive pollution in the surrounding [91,92].

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Table 4. Concentrations of rare earth elements in coal, and coal fly ash from different sources around the world.

Place & Country	Material	∑REE (µg/g)	Reference
World average	Fly ash	450	[93]
		404	[84]
Poland	Fly ash	101 - 543	[94]
Faer power plant in Guizhou Province, China	Fly ash	240.20 to 520.27	[95]
lignite coal-based thermal power plants, India	Fly ash	2100	[96]
Collie Basin, Western Australia	Fly ash	0.21% **∑REO**	[35]
Pond Creek coalbed, Pike County, Kentucky, US	Coal	<300 to >1000	[87]
World hard coal	Coal	69	[84]
World low-rank coal		65	
World coal		68	
US coal		62	
China		138	
South Korea	Fly ash	267 to 556	[97]
Coal bed, Eastern Kentucky, US	Fire clay	1965–4198	[98]
Qianxi coal-fired power plant, Guizhou province, China	Fly ash	630.51	[73]
Thermal Power Station II (TS II) of Neyveli Lignite Corporation (NLC), Chennai, Tamil Nadu, India	Fly ash	2160 (∑**REE**) 300 (Y)	[99]
Central Appalachian Coal-Related Strata, West Virginia (WV) and Central Pennsylvania (PA), US	WV MKT underclay	**297**	[100]
	WV MKT coarse coal refuse	**345**	
	Central PA LKT underclay	**221**	
	Central PA MKT underclay	**728**	

2.3.5. Industrial by Waste Products Like Red Mud, and Phosphogypsum

Industrial by-products like low-grade bauxite, red mud, phosphogypsum, gem mining waste, and slags are found to contain substantial amounts of REE and are hence these are also considered alternative resources for REE. In fact, mining waste in many cases contain high concentrations of toxic substances and is considered a threat to the environment. However, mining waste discharges produced at the bauxite, phosphate rock and other ore mines may also become valuable resources, especially for REE. For example, in the process of refining titanium dioxide from the mineral ilmenite, REE oxides are produced as co-products. Another example is, during the leaching of bauxite by NaOH by the Bayer process for alumina production, a solid waste known as red mud is generated which contains substantial amounts of REE (Table 5). More than a decade back, the global annual generation of bauxite residue was around 120 million tonnes and 2.7 billion tonnes of this material was stockpiled by then [101]. According to Swain et al. [102], the global production of bauxite and red mud during 2010-19 is 2999 and 1518 million tons respectively. China, Australia, Brazil, India, the US, and Russia produce bauxite waste (red mud) in large quantities. Since the Bayer processes inception, large amounts of red mud are getting stockpiled, and with the increasing awareness of the availability of huge concentrations of REE in this industrial waste material, this would soon become an important alternative resource for REE [103]. Both igneous and sedimentary phosphate rock deposits are used for manufacturing fertilizers. Recent studies revealed that low–grade saprolite ore which is primarily exploited for its gold and copper, also contains significant amounts of REE [104]. The vast amount of gem mining industry tailings of past and present mining activities in Sri Lanka was proven to contain up to 0.3% REE which can become a potential secondary source for many critical raw materials, including REE [105]. Initially, people used red mud to make bricks, and in cement production, but its potential to be an alternative resource for REE has been recently realised. During the production of wet phosphoric acid and phosphate fertilizer from phosphate ores, a solid phosphogypsum by-product is produced to which most of the REE (>80%) in the phosphate ore are transferred [65]. Phosphogypsum can show low levels of radioactivity because of the presence of small amounts of uranium and thorium, but also useful as a valuable REE resource (Table 6). In many instances, industrial metallurgical waste dumps are contaminated with heavy metals posing a huge threat to both humans and the ecosystem. Phosphogypsum ($CaSO_4 \cdot 2H_2O$) which is rich in REE is a waste generated during the production of the phosphoric acid process and the composition of impurities depends on the source of the phosphate. Phosphogypsum and uranium tailings are being used to extract REE in Poland [106]. Such industrial metal wastes can become sources for the extraction of REE. Baron [107] found significant amounts of valuable REE (Table 6) from such waste material in Poland. For example, Arctic Loparite ore tailings in the Kola Peninsula, North-western Russia are being effectively used to extract REE using bioleaching methods [131].

Table 5. REE concentrations (µg/g) of red mud from different countries in comparison with earth average crust composition, modified from [103].

Location	**REE (µg/g)**													
	La	**Ce**	**Pr**	**Nd**	**Sm**	**Eu**	**Gd**	**Tb**	**Dy**	**Ho**	**Er**	**Yb**	**Y**	**Sc**
Average in Earth's crust [108]	39	66	9	41	7	-	6	1	5	1	3	3	33	22
Chinalco, China [109]	416	842	95	341	64	-	56	184	48	25	28	28	266	158
Australia [110]	-	-	-	-	-	-	-	-	-	-	-	-	68	54
Brazil [111]	-	-	-	-	-	-	-	-	-	-	-	-	24	43
India [112]	110	70	0.5	-	-	-	-	-	-	-	-	-	1	5
India [113]	58	98	-	-	-	-	-	-	-	-	-	-	-	48
India [109]	112	191	18	48	9	-	7	-	4	-	1	2	13	58
Jamaica [114]	287	366	74	69	0	-	37	0	37	5	21	16	373	55
Greece [115]	114	386	28	98	21	-	22	-	16	4	13	4	75	121
Alumine de Greece, Greece [116]	130	480	29	107	19	-	22	3	20	4	13	13	108	-
Greece [117]	127	409	28	103	20	-	18	2	19	3	11	13	98	-
Greece [118]	149	418	26	115	29	5.0	23	-	1	4.3.	17	16	91.	
Ajka, Hungary [119]	114	368	-	-	-	-	-	-	-	-	-	-	68	-
Turkey [117]	169	480	47	161	32	-	4	26	23	4	13	14	113	-
Russian Federation [120]	-	-	-	-	-	-	-	-	-	-	-	-	53	25
Russian Federation [109]	-	-	-	-	-	-	-	-	-	-	-	-	-	90
Iran [120]	-	-	-	-	-	-	-	-	-	-	-	-	1	19

Table 6. Concentrations of REE in mine tailings and mine waste of different mines, industrial materials, and industrial waste.

Country	**Industrial waste**	**∑REE**	**Reference**
Poland	Uranium mine tailings, Sudety region	64.9- 109.8 **μg/g**	[106]
Southern Shanxi Province, China	Low-grade bauxite	1539 **μg/g**	[121]
Greece	Bauxite	192 to 1109 (avg. 463) ΣREE + Y+Sc	[122]
Australia	Low–grade saprolite ore	1.14% (∑REE oxides)	[104]
Poland	Metallurgical industry waste.	>140 **μg/g**	[107]
Malaysia	Water Leach Purification (WLP) residue	88367 **μg/g with** Gd as the most abundant element	[123]
Canada	Red Mud	0.03 wt%	[124]
Jamaica	Red Mud	>1303 (REE+Y+Sc)	[114]
Alumine de Greece, Greece	Red Mud	>948 (REE+Y)	[116]
Turkey	Red Mud	> 1086 (REE+Y)	[117]
Sri Lanka	Gem Mine	0.3% (∑REE oxides)	[105]
Bagre-Nechí mining district, Colombia	Mine waste (mostly gold mine residue and monazite waste)	2.19% (Ce, La, Nd & Pr)	[125]
Russia	Different types of red mud	Sc (> 100 **μg/g)**	[126]
Greece	Bauxite residue (Red mud)	0.1% (∑REE+Y+Sc)	[115]
Agios Nikolaos, Greece	Bauxite residue	∑REE 260 & Sc 120	[127]
SARECO LLP, Kazakhstan	Mineral formations (TMF) from the processing of phosphate uranium ores	5%	[127]
Russia	Phosphogypsum	0.43 - 0.52%	[129]
Philippine	Phosphogypsum	266 **μg/g**	[130]

2.3.6. Waste Rock Sources From Old and Closed Mines

Over 100 billion tonnes of solid waste are produced annually through mining operations the world over. The use of mine waste and mine tailings is attractive as this will reduce mining costs because the material is already excavated from the mines which would make it easier to access the minerals.

Recently Abaka-Wood et al. [132] provided a review highlighting the application of the new knowledge for the enhanced REE minerals recovery from selected iron-oxide-silicate-rich tailings and other alternative resources of similar mineralogical composition.

2.3.7. Acid Mine Drainage

Acid mine drainage (AMD) of certain industrial effluents such as coal, REE, and uranium mines, is also considered a potential source of REE though the concentration of ∑REE will be in the ng/ml range in the raw acid mine drainage samples. Table 7 presents concentrations of REE in AMD from different mines across the globe. This is a potential alternative REE resource and the recovery of REE remains largely unexplored though a lot of efficient extraction procedures are being developed. Acid mine drainage is produced by the oxidation of metallic sulphides present in some ores and tailings because of the instability of sulphides, which leads to the generation of sulfuric acid that promotes the leaching of the metals that make up these materials. Depending on the deposit type, acid waters can contain REE in significant concentrations. For example, the acid effluent of a closed uranium mine in Brazil contains 130 μg/ml REE. This acid effluent is treated with lime to produce a precipitate containing 7 % of REE [133], though there are other methods like ion exchange, solvent extraction, and co-precipitation for their recovery. The treatment of acid mine drainage is often carried out by neutralization, oxidation, and metal hydroxide precipitation. The precipitates obtained after the treatment of the Northern Appalachian Coal Basin in the US, the REE concentrations of the precipitates varied from 29 to 1286 μg/g with an average of 517 μg/g [134, 135]. Since REE are present in low concentrations in mining wastewaters and acid mine drainage, adsorbents such as cellulosic materials and ion exchange materials could be more useful as part of extraction/preconcentration and/or purification processes. Pyrgaki et al. [136] in a recent review described mining waste and mine water as promising sources of REE if their extraction is coupled with the simultaneous removal of toxic pollutants. These authors reported very high concentrations of ∑REE+Y of 77 to 1957.7 μg/g in coal combustion ashes, bauxite residue and phosphogypsum, and 1.6 to 24.8 μg/ml in mine discharges from different coal and ore mining areas around the globe

Table 7. Concentrations of REE in acid mine drainage (AMD) from different mines across the globe.

Name and location of the mine	Type	Concentration of ∑REE	Reference
Minas Gerais, Brazil	AMD	130 ng/ml	[133]
Staszic post-mining, Poland	Uranium mine	993.3 μg/ml	[106]
Northern and Central Appalachian Coal Basins, US	Coal mine	282 ng/ml	[134,135]
	Treated precipitate	517 μg/g	
	Coal mine, Treated AMD	724 μg/g	
Central Appalachian AMD source, US	Pregnant leach solution	132.02 μg/g ∑REE+Y+Sc	[137]
Sao Domingo mining complex, Iberian Pyrite Belt, Portugal	AMD	<221.8 ng/ml	[138]
Xingren coalfield, China	AMD	118 to 926 ng/ml	[139]

2.3.7. Recycling of E-Waste such as Magnets, etc.

As the primary and secondary REE ores are getting depleted, alternative sources such as the recycling of e-waste for a substantial recovery of REE gained prominence in recent times [140]. Our dependence on REE for the electronic gadgets required for our daily life and green technologies is increasing, but the required supply of these elements is far from secure. Because of the environmental problems and the escalating cost of mining, the focus is now shifting toward recycling. Recycling REE to some extent can meet this demand. Recycling electronic wastes such as magnets, nickel-metal hydride batteries, hard discs from laptops, desktop computers, mobile phones, loudspeakers, scrap alloy, spent catalysts, and waste light-emitting diodes (LEDs) currently end up in landfills. Instead, these electronic devices can be used to recover REE as a mixture of their oxides which later can be reduced to the production of new magnets. More than 50 million tons of e-waste is generated globally every year [141]. Many economically important metals like REE, Cu, Co, Li, Sn, Ta, Ga, Au, and Ag can be recovered from this kind of electronic waste without affecting the environment [142]. The REE recovery from waste helps sustainable development through the circular economy. Particularly five elements, namely Nd, Eu, Tb, Dy, and Y are extremely important and their demand is expected to grow by 30% in the coming decade. For example, recycling of scrap Nd-Fe-B magnets usually provide substantial amounts of Nd, Pr, Dy, and sometimes Tb. Unfortunately, the recycling activity is not going on in a big way at present mainly because the e-waste contains a very less quantity of these metals. For example, each smartphone contains just 50 mg Nd and 10 mg Pr (Table 8), and in touch screen devices, some of these metals are distributed throughout the material at the molecular scale, though items like wind turbines contain substantial amounts of REE. On the other hand, huge quantities of REE are required for manufacturing certain items such as navy ships, defence aircraft, and permanent magnets. One of the major obstacles in recycling the REE is, the complexity of usages of these elements in a variety of applications with amounts ranging from a few milligrams to a few kilograms [143]. Another critical factor that is affecting the recycling activity of the REE is the lack of cost-effective methods for their extraction from e-waste both collectively and individually. Hence, the efficient recovery of these metals becomes entirely difficult and not cost-effective and as a result, until recently only less than 1% e-waste has been recycled for REE [144]. But nevertheless, both the US and Japan have intensified their research studies on recycling as the dependence on REE is comparatively high in these countries. Rizos et al. [145] reviewed the recycling of REE permanent magnets from end-of-life (EoL) products and opined that if systematically implemented, this could be among the main avenues for meeting the EU’s future REE needs and for mitigating supply risks.

Table 8. Amounts of rare earth elements in different e-waste items.

E-waste item	REE	Concentration per Unit	Reference
9Nickel metal hydride (NiMH) battery	∑REE	5–25%	[146]
Cathode-ray tube (CRT) phosphor (as a coating on the panel.	∑REE	1–7 g	[147]
Fluorescent lamp	∑REE + Y	301.2 mg/1 g powder	[148]
Cathode-ray tube (CRT)		265 mg/1 g powder	
Navy submarines	∑REE	3636 kg	[144]
Navy surface ships	∑REE	1818 kg	
Lockheed-Martin F-35	∑REE	416 kg	
Toyota Prius	∑REE	15 kg	
Air conditioner	∑REE	120 g	
Mobile phone	∑REE	0.5 g	
Wind turbine that generates 3.5 MW Electricity	∑REE	600 Kg	[3]
Fly and bottom ash	∑REE	0.9-1.3%	[10]
$SmCo_5$ magnet	Sm	21.94%	[149]
NdFeB magnet	Nd & Pr	64.5% & 17.32%	[150]

2.3.9. Extraterrestrial

The moon, Earth’s nearest neighbour in space has long been considered a desirable location for space mining operations. Earlier studies have demonstrated that the Moon contains vast amounts of natural resources, like uranium, titanium, silicon, water, nickel, precious metals, REE, and ^{3}He at much higher concentrations than those found on Earth [151]. Robotic technologies will be extensively utilised for future mining on the moon to achieve precise localization, and conceptual mapping of the lunar surface [152]. Consequently, both government organisations from countries like the US and China, and private sectors are engaged in future space mining activities [153]. Recently eight nations signed a US-led Artemis Program for moon exploration and beyond. The path is now clear for those eight nations — Australia, Canada, Japan, Luxembourg, Italy, the UK, the UAE, and the U.S. — to participate in NASA's Artemis program of crewed lunar exploration which aims to establish a sustainable human presence on and around the moon by the end of the decade. In addition, several missions planned for the exploration of the Moon’s surface in search of REE and other valuable minerals by countries such as the USA, China, India, Japan, and Russia [154].

3. Conclusions and Future

Apart from other critical metals like Cu, Co, Ga, and Li, REE are also needed in big quantities for the necessary transition from fossil fuels to green energy solutions for the whole world to deliver net zero by 2050. Hence, there is a great need to intensify the exploration, mining, and extraction efforts for REE not only for the low-carbon energy transition but also for national security and consumer electronics applications. It is also time for looking into alternative resources for REE as the primary and secondary REE resources are dwindling fast and it is becoming difficult for identifying new resources. In addition, there is great uncertainty about the future REE supply from countries like China. The work presented here indicates the range and variety of waste streams with significant REE concentrations and with broader sustainability. Among the alternative resources described here deep-sea sediments, coal fly ash, and industrial waste products such as red mud and phosphogypsum are considered very promising. Future research will focus on finding cheaper and greener ways of extracting REE from these alternative resources. While developing efficient technologies for the extraction of REE from these alternate REE resources, there is a great need to be careful about the possible emissions of toxic metals like Cr, and Hg, radioactive metals like U, Th, and toxic organic compounds into the environment. Some recent studies have established the technical feasibility of a simple, economic, and scalable process for the recovery of REE from most of these alternative REE resources. Also, intense research is going on worldwide for the development of efficient technologies for the recovery of REE from these alternative sources which is expected to show a solution to the present global REE crisis. Some of the recent studies in this direction also suggest that other issues such as environmental, economic, and social factors will strongly influence the development of these alternative REE resources. There are also many questions about the alternative sources to becoming real-world REE sources for which plausible answers can be obtained only in the future.

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