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Perspective

Critical Minerals and REEs in Coal And Coal Wastes: Extraction and Environmental Perspectives

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Abstract

Research studies have demonstrated the technical feasibility of extracting high-purity critical minerals and separating high-purity REEs from low concentrates in coal and coal wastes. The low concentrations of REEs require extremely high volumes of coal/coal wastes for economic recovery. Extraction studies through bench-scale experiments rely on chemically intensive processes with the potential production of huge volumes of extraction wastes in future large-scale economic commercialization. Attempts to reduce the use of chemicals such as acid leaching for separation of REEs from REE-bearing coal wastes have been demonstrated in pilot-scale projects funded by the United States Department of Energy authorized by the 2020 Energy Act. These applied research projects are enabled to demonstrate environmentally sustainable pathways to reduce cost of commercial-scale production of high-purity REEs, which they have accomplished to an acceptable degree. The push for large-scale production and commercialization of REEs is strongly motivated by protection of national security and indispensable applications to advanced technologies like computer hard disks and monitors, smartphones, digital cameras, flat screen televisions, magnets in wind turbines, etc. Thus, this government-funded applied, scientific, and technical research deserves recognition as they drive innovation, solve real-world problems, and improve society.

Introduction

The United States 2020 Energy Act defines a critical mineral as any non-fuel mineral, element, substance, or material that is determined to be at high risk of supply chain disruption and serves an essential function in one or more energy technologies including industries that produce, transmit, store, and conserve energy [1]. Coal has relatively low concentrations of critical minerals, which also include critical elements called Rare Earth Elements (REEs) because they are rarely found in the Earth's crust (Table 1), [2,3]. However, many studies have demonstrated that REEs in coals have equal to even higher concentrations than ore-bearing rocks [3,4, among others]. The presence of critical minerals, including REEs, in coal is controlled by rank, groundwater infiltration, origin through magmatic or hydrothermal processes, the original depositional environments, and source-rock provenance, among others [2]. Conventional mining of critical minerals with low concentrations, such as in coal, is challenging and less economically viable to recover compared to mining ore-bearing source rocks. However, the concentration of critical minerals and REEs can be upgraded volume-wise during coal utilization and mining as well as during coal preparation (e.g., cleaning/sizing/washing) for utilization. These types of coal operations can produce large legacy coal wastes that are stockpiled (see Figure 1). Coal wastes are either solid consisting of various fragments and sizes of floor/roof rocks and other coal organic/inorganic materials or liquid/slurry wastes. These coal wastes are stored in landfills, impoundments, ponds, and reclaimed surface mine fills (see Figure 1) [5].

**Table 1:** Groups of rare 17 earth elements (REEs) including Yttrium in coal and coal wastes based on atomic number (weight), crustal abundance in parts per million (ppm), and criticality of assessment. Industrial and technology applications for each element are included in the last column. Adopted from [2].

Element	Symbol	Atomic Number	Crustal Concentration (ppm)	Criticality Assessment (Supply Risk versus Importance to clean Energy)	Industrial and Technology Applications
Light REEs					
Lanthanum	La	57	39	Near critical	Hybrid engines, Metal alloys, Optical glass (refractive index), Camera lenses, Catalyst for petroleum refining, Hydride batteries (hybrid cars), Phosphors (cathode ray tube, flat panel display screens, incandescent, fluorescent, light-emitting diode lighting), Pigments for ceramics
Cerium	Ce	58	66.5	Near critical	Catalysts (automotive catalytic converters), Metal alloys, Flat panel display screen, Decolorize glass, Hydride batteries (hybrid cars), Phosphors (cathode ray tube, flat panel display screens, incandescent, fluorescent, light-emitting diode lighting), Pigments for ceramics
Praseodymium	Pr	59	9.2	Not critical	Permanent magnets (hard disk drives, cell phones, electric motors for hybrid vehicles, windmills, actuators in aircraft), Colorant (filter and glare reduction), Catalysts (catalytic converters), Hydride batteries (hybrid cars), Pigments for ceramics
Neodymium	Nd	60	41.5	Critical	Catalysts (catalytic converters), Hard drives in laptops, headphones, Permanent magnets (hard disk drives, cell phones, electric motors, Hydride batteries for hybrid vehicles, windmills, actuators in aircraft), Colorant (filter and glare reduction), Pigments for ceramics, Fertilizer
Promethium	Pm	61	<0.001	Not critical	Watches, Pacemakers
Samarium	Sm	62	7.05	Not critical	Permanent magnets (hard disk drives, cell phones, electric motors for hybrid vehicles, windmills, actuators in aircraft), Hydride batteries (hybrid cars)
Europium	Eu	63	2.0	Critical	Red color for television, Computer screens, Magnets for wind turbines and electric vehicles, Dopant for optics fibers, Phosphors (cathode ray tube, flat panel display screens)
Gadolinium	Gd	64	6.2	Not critical	Permanent magnets (hard disk drives, cell phones, electric motors for hybrid vehicles, windmills, actuators in aircraft)
Scandium	Sc	21	22	Critical	Television, Fluorescent, Energy-saving lamps
Heavy REEs					
Terbium	Tb	65	1.2	Critical	Permanent magnets (hard disk drives, cell phones, electric motors for hybrid vehicles, windmills, actuators in aircraft), Phosphors (cathode ray tube, flat panel display screens, incandescent, fluorescent, light-emitting diode lighting)
Dysprosium	Dy	66	5.2	Critical	Permanent magnets for (hard disk drives, cell phones, electric motors for hybrid vehicles, windmills, actuators in aircraft), Electric vehicles, Hybrid engines
Holmium	Ho	67	1.3	Not critical	Glass coloring (filter and glare reduction), Lasers
Erbium	Er	68	3.5	Critical	Phosphors (cathode ray tube, flat panel display screens incandescent, fluorescent, light-emitting diode lighting), Colorant (filter and glare reduction)
Thulium	Tm	69	0.52	Not critical	Medical x-ray units
Ytterbium	Yb	70	3.2	Not critical	Lasers, Steel alloys, Colorant (filter and glare reduction), Catalysts (catalytic converters)
Lutetium	Lu	71	0.8	Not critical	Petroleum catalysts, Optical glass (refractive index), immersion lithography
Yttrium	Y	39	33	Critical	Permanent magnets (hard disk drives, cell phones, electric motors for hybrid vehicles, windmills, actuators in aircraft), Colorant (filter and glare reduction), Phosphors (cathode ray tube, flat panel display screens, incandescent, fluorescent, light-emitting diode lighting)



Figure 1: Aerial view of a coal mine waste slurry impoundment. The embankment across the valley is built with coarse coal wastes from mining to create a fine coal waste collection pond. Water from the preparation plant near the coal silos on the hill transporting fine slurry is decanted in the pond below. Adopted from [5].

For example, U.S. Department of Energy studies of samples from coal preparation plants in the United States found that REEs are partitioned during processing using washability (float-sink) tests [6]. Float-sink tests are performed to determine which coal requires washing and how much coal can be separated from rock and minerals in fluids of different densities [7]. This cleaning process produces various gravity samples analyzed for heating values, ash, and sulfur contents. The analysis of coarse coal refuse shows the total concentration of REEs for each of the specific gravity fractions that were used to evaluate clean coal from mineral matter content (see Figure 2) [6]. Concentration of REEs increases from more than 50 parts per million (ppm), on a whole coal basis) to more than 250 ppm between 1.6 and 2.1 x 2.2 float fractions. Concentration of REEs is then stabilized at about 275 ppm between 2.2 x 2.1 and 2.68 x 2.9 float fractions until a significant drop at 2.9 sink. Furthermore, analysis of ash content of coal samples from 19 preparation plants suggests that coal preparation can be used to partition light REEs from heavy REEs (see Figure 3) [6]. While a precise, definitive global amount of total coal wastes is difficult to pinpoint, it's estimated that coal mining and power generation annually produce a significant amount of wastes, with mine waste accounting for a substantial portion of the overall wastes. For example, the United States produces more than 4.4 billion short tons composed of coal wastes (coal ash and other power plant byproducts) stored at over 1,000 sites scattered across the country [8]. Of this total about 2 billion short tons comprises coal ash wastes (fly and bottom ashes) produced from coal combustion. For instance, about 115 million metric tons (117,647 million short tons) of coal combustion wastes are generated annually, which includes 45 million metric tons (46,035 million short tons) of coal fly ash [9]. The fly ash waste contains REEs [e.g., Neodymium (Nd), Europium (Eu), Terbium (Tb), Dysprosium (Dy), and Erbium (Er) (see Table 1)] in concentrations that are more than those of conventional ore deposits [9]. At the projected level, REEs in coal wastes may be of economic value.

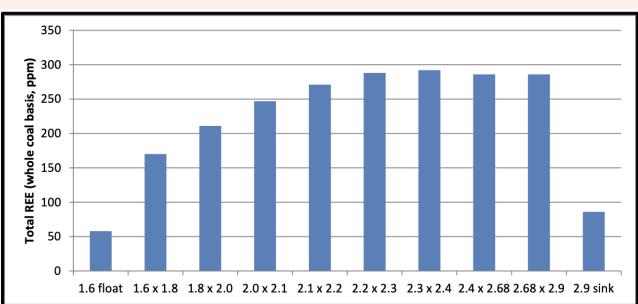


Figure 2: A histogram chart showing total REE concentrations in coarse refuse for each specific gravity fraction that was used in evaluating the separation of clean coal from mineral matter. The average concentration of REEs increase successively from 1.6 to 2.68 x 2.9 float fraction until a significant drop at 2.9 sink fraction. This analysis suggests partition of total concentrations of REEs by float-sink (or washability) tests. Adopted from [6].

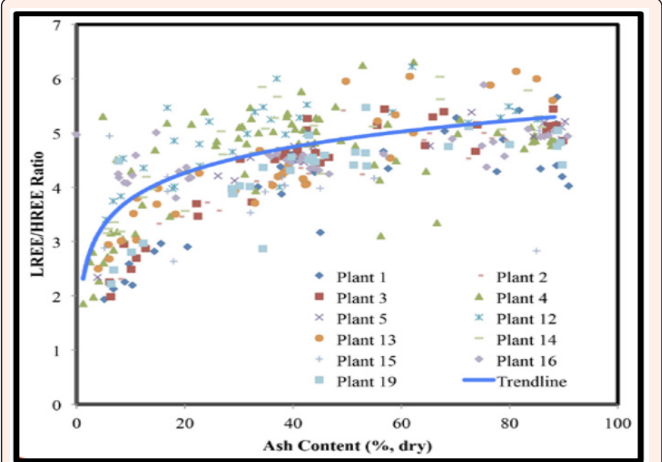


Figure 3: A chart showing the ratio of light and heavy rare earth elements (LREE/HREE) versus coal ash content analyzed from samples from 11 U.S. coal preparation plants. The plot shows higher ash content samples scatter between the 4 and 6 ratio values of the LREE/HREE. Also, lower ash content (<20%) samples are plotted between the 2 and 5 ratio of the LREE/HREE. This suggests that coal preparation processes partition LREE from HREE. Adopted from [6].

Historically, the United States faces a shortage of critical minerals and elements due to a decline in domestic mining, insufficient reserves, and refining capacity. Reliance on imports of these critical minerals mainly from China presents geopolitical risks associated with supply chain vulnerabilities endangering national security. The U.S. Department of Energy identified five REEs as critical, with the supply of these REEs in shortage between the present and year 2035 (see Figure 4). As a top coal consumer globally, the development of critical minerals and associated REEs from huge coal wastes in the United States offers a promising alternative supply and at the same time helps remediate and reclaim legacy coal wastes. Also, this innovative development model may apply globally to coal-producing/consuming countries.

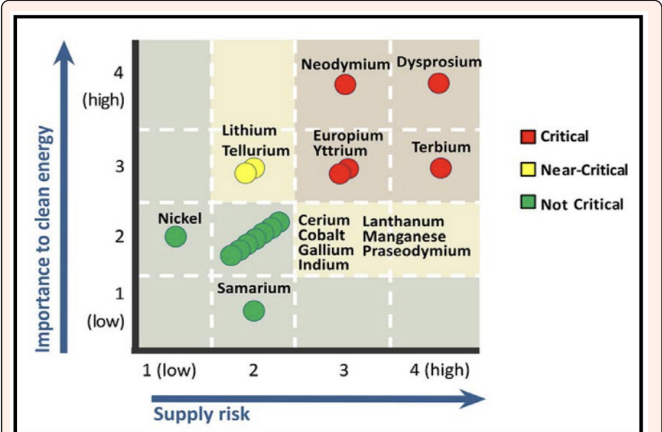


Figure 4: A chart showing the criticality matrix of REEs according to their importance to clean energy and supply risk. REEs including Dysprosium, Europium, Neodymium, Terbium, and Yttrium were identified as critical rare earth elements by the U.S. Department of Energy. Adopted from [6].

Perspectives

Extracting critical minerals to recover REEs is not a new idea, but development from coal wastes requires inventive processes to resolve supply shortages of strategic minerals while remediating legacy coal wastes. Extraction of critical minerals including REEs from coal wastes potentially eliminates the source of toxic metals and elements like mercury and arsenic that leach and infiltrate surface water and groundwater posing a serious threat to an already compromised drinking water supply [2]. There

are more than 250 different minerals (e.g., clays, crandallite, hematite, bastnaesite, monazite, mullite, xenotime, zircon, etc.) in coal, which are major hosts of 17 REEs including Yttrium each either toxic or nontoxic [10,11]. Figure 5 shows scanning electron microscope images of REE-bearing critical minerals consisting of bastnaesite (left) bastnaesite and (right) monazite extracted from the fly ash of the Fireclay coal seam [6]. These critical minerals represent the main REE-bearing minerals in coal. REEs are classified as light or heavy elements, atomic number, crustal concentration, and criticality of assessment (see Table 1). REEs are vital to components in advanced technologies due to their unique properties (magnetic, luminescent, catalytic) making them indispensable for smartphones, digital cameras, computer monitors, televisions, wind-turbine magnets, etc. (see Table 1).

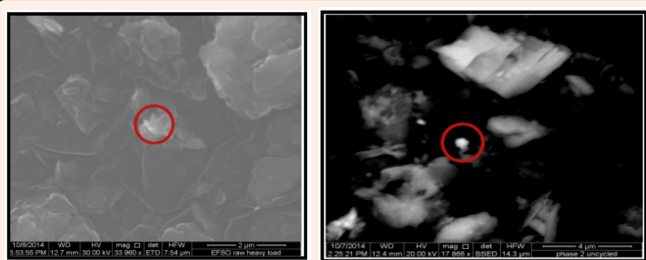


Figure 5: Scanning electron microscope images of critical minerals including REEs. (Left) bastnaesite and (right) monazite are the chief REE-rich critical minerals in coal. Adopted from [6].

Figure 6 is an ideal schematic diagram showing the value chain of opportunities for recovery of critical minerals and associated REEs spanning from mining production to coal processing and utilization [6]. For example, the value chain of REEs recovery increases from coal mining pit cleanings to coal separation and grinding, to energy conversion byproducts such as fly and bottom ashes. Thus, concentrations of critical minerals and REEs generated by production, processing, and utilization highly vary between specific coal operations. That is, upgrade of these critical materials depends on how much raw coal is prepared and cleaned, creating more total volume of coal wastes from coal mining to combustion.

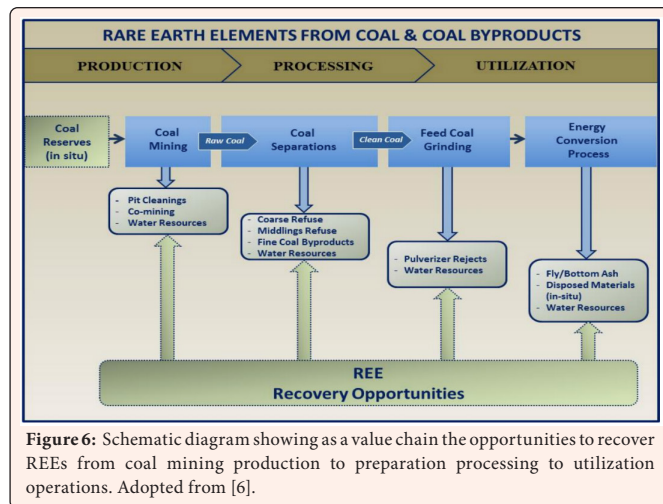


Figure 6: Schematic diagram showing as a value chain the opportunities to recover REEs from coal mining production to preparation processing to utilization operations. Adopted from [6].

Extraction in Coal Wastes

While technically possible, extracting the critical minerals and separating the REEs is challenging and complex due to their low concentrations in coal and coal wastes. The challenge is to invent physical, chemical, and biological processes/treatments that would concentrate REEs to economic levels. Scientific researchers are actively assessing these methods from bench-scale experiments to feasibility pilot-scale studies with limited but encouraging successes. Different methodologies are being trialed including physical beneficiation, thermal pretreatment, alkali pretreatment, acid leaching, ion-exchange leaching, bioleaching, solvent extraction, and other recovery technologies [12]. Successes are highly dependent on the properties of the coal and minerals (e.g., rank, composition, crystallinity/glassy, size, density, magnetism, etc.). Selected effective extraction techniques of critical minerals and REEs

are highlighted below according to types of coal wastes and special methodology. For instance, in coal fly ash critical-minerals extraction benefits from size fractionation with small fractions yielding REE concentrates [12,13]. Also, physical beneficiation of critical minerals in coal fly ash uses float/sink tests that separate minerals by density at 1.8 specific gravity. Critical minerals that sink contain 521 ppm REEs compared to those that float, which contain 376 ppm REEs [12,14]. Critical minerals can be separated using magnetism with non-magnetic minerals yielding more REEs [15-17]. The amounts of critical metals and REEs from coal ash wastes in the United States are shown in Table 2 [8]. The advantage of these physical methods is a reduction of the use of chemicals for extraction.

Table 2: Coal ash provides the best potential supply of critical metals and Rare Earth Elements (REEs) from United States legacy coal wastes. The estimated mass varies from 40 to 288,000 tons (U.S.) and years of potential supply from 1.1 to 3,900 years, respectively. Adopted from [8].

Critical Metals and REEs	Estimated Mass (Tons)	Potential Supply (Years)
Neodymium (Nd)	172,000	40
Dysprosium (Dy)	62,000	14
Lithium (Li)	288,000	130
Cobalt (Co)	110,000	15
Nickel (Ni)	252,000	1.1
Iridium (Ir)	40	15
Platinum (Pt)	600	15
Gallium (Ga)	20,000	1,100
Germanium (Ge)	30,000	3,900

Extraction of REEs can be enhanced by using alkali and thermal pretreatments of coal wastes to improve recovery by acid leaching [12]. For example, sodium hydroxide (NaOH) pretreatment of fine-grained coal wastes roasted at 75 °C temperature roasted for 2 hours increased REE concentrations by more than 50% after acid leaching treatment. Similarly, fine-grained coal ash pretreated by sodium hydroxide (NaOH) at 190 °C roasted for 30 minutes followed by hydrogen chloride treatment increased recovery of heavy REEs by 97% and light REEs by 76% [12]. Acid leaching tests on coal bottom ash using hydrochloric (HCl), nitric (HNO₃), and sulfuric (H₂SO₄) acids under similar conditions found the order of REE recoveries as HCl > HNO₃ > H₂SO₄ [12]. These studies show that REE recoveries from coal ash and wastes are improved by alkali-additive and roasting-temperature pretreatments prior to acid leaching treatment.

The efficiency of different chemical additives prior to acid leaching was tested at various roasting temperatures on coal fly ash from various coal ranks collected from power plants in the United States and China [9,12,18]. The optimum recovery by HNO₃ leaching for all of coal fly ash (Appalachian/Illinois Basins) except for the fly ash of the Powder River Basin was more than 44%. Many recoveries were significantly lower although leached by more concentrated HNO₃ and heated from 85 °C to 90 °C. Pretreatment of the fly ash with sodium peroxide (Na₂O₂) and leached with ~4 M HNO₃ increased the leaching efficiency (>80% relative to hydrofluoric acid digestion). In contrast, sodium carbonate (Na₂CO₃) additive for coal fly ash from China at roasting temperature of 860 °C for 2 hours yielded total REEs recovery of about 73% leached with HCl [18].

Extraction in Acid Mine Drainage Wastes

Acid Mine Drainage (AMD) is an outflow of acidic water from coal mines and wastes from (1) abandoned or active underground/surface mines, (2) preparation plants where wastewater is discharged as coal thickener, (3) rainwater flowing through the coal mine rock piles, and (4) groundwater infiltration in coal mine shafts and floor/roof rocks [2,19]. Water is acidified by oxidation of iron sulfide (pyrite) in the coal and bedrock, which in turn is leached, dissolved, and releases REEs from critical minerals under low pH conditions [20-22]. The REEs fractionated during acid leaching and dissolution of critical minerals are enriched with light and heavy REEs. Figure 7 shows a stream impacted by AMD from an abandoned coal mine in the Mid-Atlantic

region of the United States [23]. The reddish color of the stream water affected by AMD is caused by the oxidation of iron (Fe) from the pyrite (iron sulfide) into ferric iron (Fe_2O_3), which precipitates as rust-like sediments.

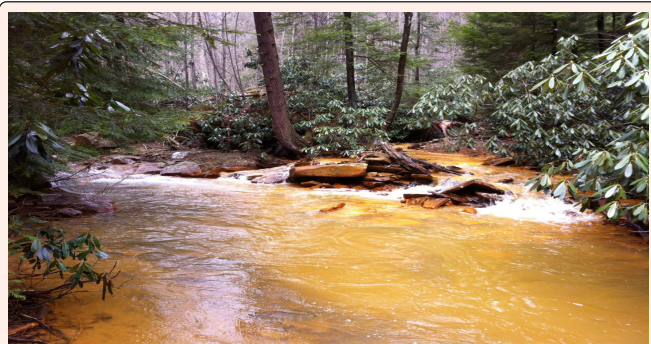


Figure 7: Photograph of a stream from an abandoned coal mine site in the Mid-Atlantic region of the United States, which is impacted by acid mine drainage. The reddish color of the stream water is due to the oxidation of iron (Fe) from the pyrite (iron sulfide) into ferric iron (Fe_2O_3) that precipitates as rust-like sediments. Adopted from [23].

Extraction by acid leaching of mine coals, bedrocks, aqueous effluents, and sludge were tested indicating extremely high recoveries (>90%) of REEs [24-27]. Acid leaching of raw coals significantly varies from 30% to 70% for Cesium (Ce) [24]. The leachability and characteristics of REEs in raw floor/roof rocks adjacent to coals were analyzed for recovery of light and heavy REEs as well as medium REEs as defined by [25]. The recovery values of these REEs vary between raw materials with middle REEs [Europium (Eu), Samarium (Sm), and Gadolinium (Gd)] higher than the light and heavy REEs. The highest recovery values are from the light REEs [(Ce, Lanthanum (La), Praseodymium (Pr), and Neodymium (Nd))] with the floor rocks yielding 47%-65% recovery, which were leached for 5 minutes. Extended reaction time to 120 minutes increased the recoveries to 75%. The heavy REEs recovery values from roof and floor materials were less than 25%. Extraction of REEs from AMD sludge is simpler than extraction from raw materials (coal and bedrocks) because sludge is predigested with acid leaching [26]. For wet and dry sludges, the total REEs recovery (at a pH of 2) ranges from 65% to >95% with heavy REEs concentrations higher than light REEs. AMD outflows generate a total of 538 metric tons of REEs including Yttrium per year in a dissolved state, with 83% of that from low pH (<5) discharges [27].

Environmental Concerns

The focus of the bench-scale studies to digest, extract, and purify critical minerals and REEs from coal and coal wastes is to demonstrate technical feasibility of chemical-processing systems. Paucity of parallel studies on the environmental impacts of these chemical-assisted processes and environmentally friendly extraction raises environmental concerns [28-31]. The major concern is the potentially high volume of acids and byproducts to be created during future large-scale commercialization production. Release of extraction wastes consisting of toxic acids, radionuclides, and heavy-metal residues to the environment is a serious threat to the soils, surface water, groundwater, drinking water, and associated biological ecosystems as well as human health. Addressing these concerns requires developing sustainable environmentally friendly, extraction-benign technologies and acceptable waste management plans. The extraction wastes must be carefully managed according to environmental and occupational health standards and risks, while not unnecessarily adding more operational costs.

As a part of the United States 2020 Energy Act, the Department of Energy (USDOE) has performed assessments and analyses of the feasibility of economically recoverable critical minerals, especially REEs, in coal and coal byproducts in selected eastern and western States endowed with large recoverable coal reserves [31,32]. The government has funded demonstration pilot-scale projects in collaboration with researchers in state universities, national laboratories, and government agencies to integrate new extraction technologies and innovative developmental designs to reduce production costs while maintaining environmentally sustainable processes.

These USDOE projects are typified by the University of Kentucky pilot project, which uses an innovative technique consisting of microbubble-assisted solvent extraction to separate and concentrate REEs [33]. This environmentally friendly method separates substances based on differing solubilities in immiscible liquids (water and organic solvent). The microbubbles improve the extraction process by increasing interfacial area, improving mass transfer, and promoting better mixing of the two liquid phases. Another USDOE-funded project in cooperation with researchers from Sandia National Laboratories [29] developed a groundbreaking method for extraction of REEs from coal ash using water, supercritical carbon dioxide, and food-grade citric acid, which are environmentally benign ingredients. The method achieved 42% extraction efficiency and can preferentially recover the most critical REEs and heavy metals. These environmentally friendly projects funded by the previous U.S. Government administration (i.e., pre-January 2025) may suffer from cost-cutting policies of the present administration.

Bench-scale experiments on bioleaching by fungi and bacteria have been proven to be feasible to extract REEs in ore-bearing rocks and coals. For instance, a solubilizing fungus bioleached monazite, a critical mineral found in ore-bearing rocks and coals and released rare earth cations into solution [28]. Also, extraction of REEs using bacteria by consuming sulfides from pyrite in coals generated sulfuric acid and ferric sulfate, which enabled separation of REEs [34]. This environmentally friendly process is an important alternative to chemical-intensive techniques and requires investigation in small pilot-scale projects to demonstrate feasibility for economic large-scale REEs production. The low efficiency of bioleaching and high cost of cell cultures are challenges that need to be addressed to move toward feasibility. The directions and opportunities of bioleaching are extensively discussed and explained in review papers [35,36].

Conclusion

The research efforts on extraction of critical minerals, including REEs, from coal and coal wastes are driven by their vital importance in advanced technologies and thus, also national security. The low concentrations of critical minerals in general have led to studies to find extraction and separation techniques heavily focused on chemical-intensive procedures. To increase the efficiency of extraction/separation, acidification processes have been further intensified to achieve higher and purer REE concentrates. However, these studies being bench-scale experiments often lose sight of the effects of acid-leaching products on the environment and how extraction wastes may impact future large-scale commercial production of REEs. Countervailing parallel studies to understand the environmental management of these extraction wastes have been lacking and infrequent. On-going bench-scale studies of bioleaching processes to extract REEs may alleviate these environmental concerns.

However, "big-ticket" government-funded research can be mandated to find environmentally friendly extraction techniques. Applied research pilot-scale projects in collaboration with universities, national laboratories, and the U.S. Department of Energy are driven by the United States 2020 Energy Act. These worthy projects are directed to demonstrate environmentally sustainable pathways to commercial production of high-purity critical minerals, especially REEs. Efforts have been made to reduce the use of toxic chemical-assisted processes in favor of using environmentally benign ingredients to demonstrate the technical feasibility of economic commercialization of REEs. Thus, these ongoing pilot-scale projects may reduce the costs of commercialization as well as the environmental footprint of extraction and at the same time as remediate and reclaim legacy coal waste sites. This applied, scientific, and technical research deserves government funding, recognition, and support to drive innovation, solve real-world problems, and improve society.

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