

Sustainable Recovery of Rare Earth Elements from Coal Ash: Circular Economy Perspectives for South Africa

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Abstract—There is an increased need for rare earth elements (REEs) due to their importance in modern technologies such as renewable energy, electronic gadgets and defence equipment. Specifically, the demand for some REEs, including neodymium, dysprosium and terbium, often exceeds their existing global supply. Consequently, researchers are seeking alternative sources of extraction of these elements from secondary sources. This review paper examines the existing studies in this area of research. The specific objectives of this article are to: (1) examine the current extraction methods for REEs from coal ash; (2) investigate the current trends of research in this knowledge area and technological innovations; (3) discuss challenges and future directions for sustainable recovery. An integrative review approach was adopted to discuss the trends of recent research in this field of study. Results reveal that coal ash contains significant REE concentrations, suggesting its potential as a secondary resource within a circular economy framework. The study underscores that integrating environmentally benign extraction methods could reduce waste, support resource recovery, and promote industrial sustainability. The findings contribute to ongoing efforts to transform coal combustion residues from environmental liabilities into strategic assets for green and sustainable mineral supply chains.

Keywords—Rare Earth Elements (REEs), coal, fly ash, bottom ash, mineral extraction, renewable energy, critical minerals.

I. INTRODUCTION

Rare Earth Elements (REEs) are attracting considerable global attention due to their vital roles in clean energy, consumer products, defence and security, among other applications [1]. The growing need for these minerals has ignited efforts to explore and extract them from even unconventional sources. While the significance of REEs is not in doubt, a disparity exists between the demand for these

elements and their production. This demand has significantly increased over the past 15 years and is expected to grow to about 240,000 tonnes by 2030 from 171,300 tonnes in 2010 [2]. Available data has shown that only one country (China), controls approximately 70% of global supply in terms of production. Another factor that contributes to the supply risks is that each REEs are not mined independently: they typically occur together in geologic deposits, which makes mining of individual REEs economically untenable. Thus, the supply of one individual REE depends on multiple factors, including the geology of the deposits, the costs of the technology involved and the price of the basket of rare earths [2]. Studies have, therefore, argued that the future availability of REEs is of concern due to the rapid demand growth, environmentally unsustainable mining practices and the monopolistic supply conditions. Consequently, researchers are now exploring alternative sources of REEs to fill the supply gap. This research aligns with the principles of green chemical engineering by advancing circular approaches to transform coal combustion residues into valuable secondary sources of critical minerals.

Recent studies have explored the potential of coal combustion residue as a source of REEs. This is due to the retention of rare earth elements and yttrium (REY) during coal burning [1]. Coal is a fuel that is currently used to generate up to 39% of global energy output. During coal combustion, many elements are oxidised and volatilized. However, a significant residue, which is the coal ash, remains. This ash can be categorised into three types: fly ash (CFA), bottom ash (BA) and slag. The conventional technologies used for coal utilisation in energy generation produce a large volume of coal ash. For example, in 2014/2015 in South Africa, 119.2 million tons of coal were consumed by the electricity generation plants, and 34.4 million tons of ash, amounting to 28.9% were produced [3]. This ash, when left untreated, may pose a threat to the environment, and the costs for the treatment and disposal of this waste create an extra economic burden. Thus, proper disposal and recycling, if feasible, are important.

Rare earth elements (REEs) are mainly in fly and bottom ash among the three categories of coal residues from coal combustion [3]. Specifically, studies have shown that the burning of coal results in the enrichment of some metals, such as REEs, such that these ashes can be considered as low-grade ores for these metals [4]. Coal ash may contain several REEs depending on the coal's origin. Several studies have shown that the rare earth elements and yttrium (REY) in fly ash are higher than those of the original coal by as much as eight to ten times [4], [5]. This may be as a result of REY's low volatility [5].

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In South Africa, Eskom—the nation's major electricity producer—stated that only about 7% of its ash was sold from 6 of its 13 coal-fired power stations in 2014/2015 [3]. This ash was sold to the construction industry for inclusion in cement or brick making, which is one of the few existing means to economically dispose of the ash. Another application of ash being explored is in the use for backfilling in mines [3]. There are several benefits to dealing with ash in an environmentally responsible way. One, the environmental footprint of coal and its products will be limited. Also, there will be positive impacts from the applications of the waste materials. Therefore, it is essential to explore the potential applications of coal ash. This study reviews the trend in the literature on the recovery of REEs from coal ash (bottom and fly). While extensive research has been conducted on this knowledge area, a comprehensive synthesis of methodologies, efficiencies, and feasibility remains fragmented, which necessitates this review to consolidate insights, identify research gaps and chart a path toward scalable REEs recovery solutions.

The subsequent sections in this article will provide an overview of REEs' composition in coal ash; examine the current methods for characterisation and recovery of REEs from coal ash; discuss the current trends in this knowledge area, and the gaps for future research.

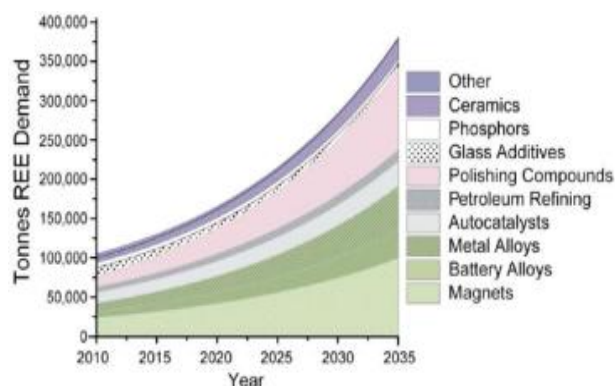


Figure 1. Projected REEs demand from 2010 to 2035 [2]

II. OVERVIEW OF RARE EARTH ELEMENTS IN COAL ASH

Composition and distribution of REEs in coal ash

Rare earth elements are a group of 17 element comprising the 15 lanthanides including lanthanum (La), cerium (Ce), praseodymium (Pr), neodymium (Nd), promethium (Pm), samarium (Sm), europium (Eu), gadolinium (Gd), terbium (Tb), dysprosium (Dy), holmium (Ho), erbium (Er), thulium (Tm), ytterbium (Yb), lutetium (Lu), along with scandium (Sc) and yttrium (Y). [6] examined the levels of rare earths in South African coals and coal ash. Samples were collected from eight different coal-fired power stations across the country. Analysis conducted on these samples includes petrography, X-ray diffraction (XRD), Malvern particle size analysis, inductively coupled plasma-mass spectrometry (ICP-MS), and instrumental neutron activation analysis (INAA). The results indicate that Total REY concentrations ranged from 95 to 149ppm in the coal samples, and total REY concentrations ranged from 402 to 599ppm in the ash samples, far higher than

the average upper crustal values, and higher than global average hard coal ash values, but lower than REY-rich coal ashes recorded for various deposits globally [6].

A similar study was conducted in Nigeria by [7] where eight coal fly ash samples generated from coals from Nigeria's major coal belts were collected. The Silica and alumina accounted for the bulk of the major elements in the samples, with respective ranges of 38.1–44.5% and 14–15.98%. Total REE contents in the samples ranged from 874 ppm to 1127 ppm, while the cerium, yttrium, neodymium and lanthanum-dominated rare oxide totals were found to be in the range of 941–2145 ppm across the samples. In Tanzania, a study to examine the viability of REEs in coal and fly ash samples using X-ray fluorescence (XRF) spectrometry and laser ablation inductively coupled plasma mass spectrometry (LA-ICP-MS) reveals that the total concentration of REEs in the coal samples ranged from 89.48 parts per million (ppm) to 196.30 ppm, while in CFA samples, ranged from 362.55 ppm to 475.77 ppm. These findings on the viability of coal products across the African continent as a source of REEs have important implications for governments and other relevant authorities, as they provide valuable insights into the feasibility of investing in coal and coal ash as promising sources of REEs [7], [8], [6].

Factors affecting REE concentration

The findings of studies on the feasibility of recovering REEs from coal ash across several locations in Africa suggest that certain coal types may be more favourable for REE recovery than others. As an illustration, the study by [6] investigating Main Karoo Basin coal ash samples from South Africa indicates that none of the REY elements were particularly enriched, as the REY content is below 1000ppm. However, samples from UNICRANE power plants around Kogi State, Nigeria, analysed in [7] with a REE content ranging from 941 to 2145 ppm across the samples, suggesting the promising availability of the elements in these samples. Thus, coal type and location of the coal samples play crucial roles in their viability.

Another important factor that determines the concentration of REEs in coal ash is the combustion process. Specifically, the temperature, combustion efficiency, and mineralogical transformations during coal combustion all influence how REEs are retained or volatilized. Some REEs, such as La, Ce, Nd and Sm have low-boiling-points. These elements are partially volatilized at high temperatures (> 1200 °C) and may condense in fly ash particles. As an illustration, [9] found that REEs in South African coal ash samples were enriched in fine fly ash particles due to condensation effects at high combustion temperatures. Similarly, high-boiling-point elements such as Y, Dy, Ho and Er tend to remain in the bottom ash or slag, leading to variations in REE concentration between fly ash and bottom ash [10]. The mineral composition of coal also influences REE behaviour during combustion. [11] provides an in-depth understanding of REE behaviours during coal combustion, including potential mineral transformations of REE-organic complexes and REE carbonate minerals, as well as the retention of REE phosphate and silicate minerals.

Since several factors, such as mineral composition, coal types, and combustion temperatures, influence the

concentration and the viability of recovery of REEs from coal ash, it is therefore important to explore the existing recovery attempts for REEs from coal ash and the methods involved.

III. REES RECOVERY METHODS

Analysis and Characterisation

To recover rare earth elements (REEs) from coal ash, different analytical methods are usually adopted to determine the concentration, mineral associations, and spatial distribution of REEs in the ash. Some common techniques used in literature are discussed.

A. Inductively Coupled Plasma Mass Spectrometry (ICP-MS)

Reference [12] analysed a fly ash sample from an eastern U.S. coal power plant to determine and quantify the mineralogical association of rare earth elements (REE). These analyses were completed using laser ablation inductively coupled plasma mass spectrometry (LA-ICP-MS) and a scanning electron microscope, equipped with an energy-dispersive X-ray spectrometer (SEM-EDS). For particle size determination, a Malvern Mastersizer 3000 was used with the fly ash dispersed in water. The particle size analysis result is shown in Figure 2. The figure shows a bimodal distribution in volume density with a smaller particle size maximum at 0.872 μm and a larger maximum at 24.1 μm . The particle size in number density shows that the $D_n(90)$ value is 1.31 μm . This implies that on a number density basis, nearly all fly ash particles are smaller than 1.31 μm in diameter.

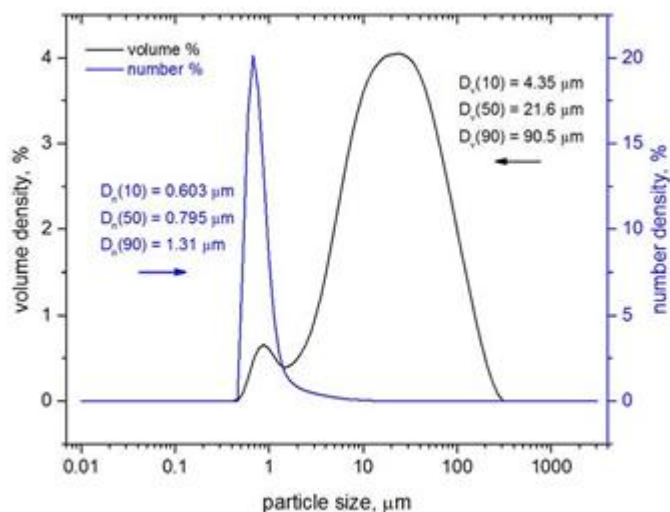


Fig.2. Particle size analysis using ICP-MS [12]

B. Neutron Activation Analysis (NAA)

Reference [13] combined instrumental neutron activation analysis (INAA) with inductively coupled plasma mass spectrometry (ICP-MS) to study the elemental content of coal combustion residues from Polish, Colombian, and Kazakh mines. The neutron activation analysis was performed in a MARIA nuclear reactor (Swierk, Poland) with a thermal neutron flux of 10^{14} $\text{n}\cdot\text{cm}^{-2}\cdot\text{s}^{-1}$ for 30–50 min. The cooling time was 1–7 days. The process of the use of INAA includes (1) preparation of the samples and standards; (2) irradiation in the

neutron flux in the nuclear reactor; and (3) ray spectrometric measurements [13].

Some other techniques for analysis and characterisation of REEs in coal ash discussed in literature include Energy-Dispersive X-ray Spectroscopy (EDS or EDX), X-ray diffractometer (XRD) techniques, Mineral Liberation Analysis (MLA), among others [1], [11], [14].

Recovery of REEs

Typically, a sequential process is used for the recovery of REEs from coal ash. This sequence usually involves a combination of physical and chemical or hydrometallurgical processes. Some of these physical techniques include sieving, magnetic separation, and flotation, among others. The chemical or hydrometallurgical processes include acid leaching, alkali fusion, among others. Some of the common techniques involved in REEs recovery from coal ash, as discussed in recent literature, are as follows.

C. Acid Leaching

This recovery method involves treating coal ash with strong acids, such as hydrochloric (HCL) or sulfuric acid (H_2SO_4), to dissolve REEs into a solution. Subsequent processes, like solvent extraction or precipitation, are used to recover the REEs from the leachate. [15] used a sequential chemical roasting, water leaching, and acid leaching process for the recovery of REEs from CFA. The study found that NaOH and Na_2CO_3 were the most effective additives to break the glassy phases of sodium silicate and sodium aluminosilicate, which can then be readily dissolved in water or acid. The study further found that Water leaching was very effective for the dissolution of the sodium silicate product, removal of most of the silica, and for turning the glassy phases into a porous structure. Consequently, mass transfer limitations were reduced, and acid could easily diffuse into the particles, dissolve the majority of remaining, and extract REEs along with other elements such as Al. The study found that this process enhanced the REE recovery to 79% and 89% using NaOH and Na_2CO_3 roasting, respectively [15]. A study by [16] developed a green approach for REE recovery from coal fly ash, generating multiple products and upcycling waste residues. The process involves three modules including consists of three modules: REE leaching using citrate, REE separation and concentration using oxalate, and zeolite synthesis using secondary wastes [16].

D. Ionic Liquids (IL)

Ionic liquids are liquids composed of cations and anions. Ionic liquids (IL) and deep eutectic solvents (DES) have emerged as a new class of green solvents for REE extraction and separation processes and are often called “designer solvents” due to their ability to offer selective recovery of REEs; since literature suggests that oxides of Si and Al may be less soluble in these solvents than other metal oxides [17]. [18] studied the use of some ionic liquids for the purification of REEs from the leaching solution of coal combustion products (CCPs). In the study, the CCP samples from the power plant were obtained by calcining the coal samples and coal fly ash samples at 1020°C for 6h, so that the samples were completely combusted. Then, the ionic liquid type extractants were studied

for the purification of REEs from the leaching solutions of CCPs. Using the purification and recovery procedures, the purification multiples of Al, Fe, and Ca arrived at 94, 40,588 and 8.7 times, and the recovery rate of REE was 37.4% [18].

E. Bioleaching methods (microbial-assisted extraction)

Bioleaching involves the use of microorganisms to oxidise metal sulphides in ores and wastes, making them soluble and recoverable. A study by [19] tested three microbial strains, namely *Candida bombicola*, *Phanerochaete chrysosporium* and *Cryptococcus curvatus* on their performance of leaching trace elements and REEs from fly ash. The study found that *C. bombicola* was identified to be the best, leading to the highest mineral loss and extracting efficiencies of trace elements and REEs among the three strains. Similarly, [20] adopts the use of bioleaching and precipitation for the recovery of REEs from coal-based resources. In the study, an acidified bioleaching solution was made from the oxidation of the pyrite with *Acidithiobacillus ferrooxidans*. This generated acidified solution was used to extract REEs from coal waste, with 13–14% yields for most REE elements (~72 h of leaching) (MIAO et al., 2008) [20].

There are other advanced techniques discussed in the literature which are used for REEs recovery from coal ash such as ion-exchange, solvent extraction, membrane separation, etc. [17].

IV. CHALLENGES WITH VIABLE EXTRACTABILITY OF RARE EARTHS FROM COAL ASH

Even though an extensive level of research has been carried out on the extractability of REE from coal ash, researchers are still confronted with some challenges, especially as regards commercial and economic recovery of the elements. Firstly, unlike high-grade REE ores, coal ash contains REEs at relatively low concentrations (typically <1000 ppm) [6]. Thus, it becomes necessary that large volumes of material are processed, increasing operational costs. Another challenge is with the mineralogy of coal ash. REEs in coal ash are often dispersed in various mineral phases, including phosphates, silicates, and oxides. Some phases are resistant to leaching, requiring aggressive reagents or high temperatures, which raises costs. For example, the aluminosilicate glassy phase is quite resistant to leaching since a significant portion of REEs is encapsulated, making them difficult to extract through acid leaching [1]. These concerns revolve around and are not limited to feasibility vs efficiency, sustainability vs selectivity and the ability to incorporate a continuous process with minimum downtime [17]. These research gaps are continually being explored in recent studies, with some of the questions surrounding sustainability being addressed with benign and novel solvents, such as with the help of deep eutectic solvents and organic solvents, to efficiently extract REEs from waste streams and coal residue [17].

V. CONCLUSION

The recovery of rare earth elements (REEs) from coal ash presents a promising opportunity to secure a sustainable and alternative supply of these critical materials. This review

highlights various recovery techniques. It begins with an overview of rare earths in coal ash. It then examines some recovery techniques beginning with physical separation methods such as sieving, flotation, and magnetic separation, which serve as pre-concentration steps to enhance REE content. Analytical techniques like ICP-MS, SEM-EDS, and XRD are essential for characterisation, ensuring an efficient extraction process. Recovery methods, including acid leaching, bioleaching, and ionic liquid extraction, have demonstrated significant potential in maximising REE yield while minimising environmental impact.

Despite the progress in REE recovery from coal ash, challenges remain in scaling up these processes, improving selectivity, and optimising cost-effectiveness. Future research should focus on integrating multiple recovery techniques, developing environmentally friendly reagents, and enhancing the economic viability of these approaches. With continued advancements, coal ash could emerge as a valuable secondary source of REEs, contributing to global supply chain stability and resource sustainability. Integrating these recovery technologies within South Africa's coal-based energy system presents a circular economy opportunity to minimise waste, recover critical minerals, and contribute to the green industrial transition.

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