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Techniques for Rare Earth Elements Recovery from Coal Fly Ash: A Comparative Analysis

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ABSTRACT

Rare Earth Elements (REE) are essential for high-tech and renewable energy applications; however, extracting these elements from primary sources presents significant environmental and economic challenges. Coal fly ash (CFA), a byproduct of coal combustion, offers a viable alternative as a secondary source for REE recovery due to its abundance and REE content. This approach also supports coal downstreaming efforts. This study evaluates physical, chemical, and biological techniques for recovering REE from CFA, focusing on efficiency, cost, and environmental sustainability. Physical methods such as particle sizing and magnetic separation effectively concentrate REE as a preparatory step, enhancing the efficiency of subsequent chemical processes. Chemical techniques, particularly acid and alkaline leaching, achieve recovery rates exceeding 90%, though they require careful waste management to mitigate environmental impacts. Meanwhile, biological methods like bioleaching and biosorption provide a more sustainable alternative with minimal waste, albeit with lower recovery efficiencies. An integrated approach combining these techniques demonstrates significant potential to optimize REE recovery while reducing costs and environmental risks, aligning with circular economy principles.

INTRODUCTION

Rare Earth Elements (REE) consist of 17 metallic elements, including the lanthanides, yttrium, and scandium, which are essential to a wide range of contemporary industries. These elements are essential in manufacturing high-tech devices such as smartphones, wind turbines, electric vehicles, and defense products, making them integral components of a global sustainable technology-based economy (Park and Liang, 2019). The growing importance of REE has led to a surge in global demand over the past few decades. However, the production of REE is highly concentrated in a few countries, particularly China,

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which controls over 80% of the global supply. This dependency and supply uncertainty in international markets have driven other countries to explore sustainable alternative sources to meet the increasing demand (Wang et al., 2019).

One promising alternative source of REE is coal fly ash (CFA), a byproduct of coal combustion. In Indonesia, coal downstreaming strategies have become a national priority to enhance the added value of domestic coal resources, supported by a coal production total of 296 million tons in 2020, even during the COVID-19 pandemic (Haq et al., 2021). These efforts extend beyond converting coal into energy products, exploring its potential as a raw material for high-value products like REE. Utilizing CFA as a source of REE could reduce import dependence, strengthen national supply independence, and integrate industrial waste management into the circular economy. Furthermore, developing REE recovery technologies through coal downstreaming offers opportunities for job creation, advancements in extraction technologies, and reduced environmental impacts from coal mining and combustion (Haq et al., 2022). However, challenges persist due to the variability of CFA's chemical composition and the complex distribution of REE within its mineral structures, necessitating innovative and environmentally friendly extraction methods (Sreenivas et al., 2021).

REE recovery techniques from CFA can generally be divided into three main categories: physical, chemical, and biological (Das et al., 2018). Physical approaches, such as magnetic separation, particle size reduction, and roasting, aim to concentrate REE in CFA fractions to facilitate subsequent extraction steps (Kumari et al., 2019). Chemical methods, including alkali fusion and acid leaching, are more effective at releasing REE from CFA's mineral matrix through chemical reactions, but these methods are typically more expensive and have a greater environmental impact due to high energy and chemical consumption (Zhang et al., 2020). On the other hand, biological technologies using microorganisms or other biological materials to dissolve or adsorb REE from CFA offer a more environmentally friendly approach, although they often require longer processing times and yield lower efficiencies compared to chemical methods (Pan et al., 2020; Park et al., 2020).

This study aims to analyze these three techniques comparatively to determine the most efficient and environmentally friendly approach for recovering REE from CFA. Each method will be evaluated based on recovery effectiveness, operational costs, and environmental impact. The research intends to provide a comprehensive guide for industries in selecting the most suitable method for REE recovery from CFA, thereby delivering optimal economic and environmental benefits. By understanding the advantages and limitations of each extraction method, the study seeks to identify techniques that are commercially viable with minimal environmental impact.

METHODOLOGY

This review article begins by analyzing a collection of studies discussing methods for recovering REE from CFA using physical, chemical, and biological approaches. The review process starts with data collection from relevant articles, focusing on key aspects such as REE recovery methods, recovery efficiency, and novelty of the research. Keywords used for the search include: "REE from CFA", "REE recovery from CFA", "REE leaching", "REE extraction from CFA", "chemical REE extraction", "alkaline REE extraction", and "biological REE extraction from coal fly ash". The selection of articles was carried out by evaluating their titles and abstracts to confirm their relevance to the focus of the review.

Subsequently, evaluation and data extraction from the selected articles were conducted using MS Excel to analyze five key aspects: extraction techniques, REE recovery outcomes, research objectives, publication year (limited to the last ten years), and contextual relevance. These findings are systematically presented in **Table 2**. Each technique—whether physical, chemical, or biological—was evaluated based on recovery efficiency, cost analysis, and environmental impact, with the aim of identifying promising combinations of techniques for further development.

The purpose of this review is to provide a comprehensive understanding of the topic and to identify the most effective and promising REE recovery methods from CFA. This approach is expected to establish a solid foundation for further analysis of the effectiveness and potential application of the reviewed methods.

RESULT

Recovery REE Techniques

Physical Techniques

Physical techniques are utilized to enrich the concentration of REE in CFA before chemical extraction processes are applied. Typically, three methods are employed: particle size classification, magnetic separation, and roasting treatment (Manurung et al., 2020; King et al., 2018; Cao, 2018; Taggart et al., 2018; Pan et al., 2020; Sreenivas et al., 2021; Kumari et al., 2019).

Particle Size Classification: Particle size separation is an effective physical technique to enhance the REE content in specific fractions of coal fly ash (CFA). According to Pan et al. (2019), the concentration of REEs in smaller-sized fractions can reach up to 1200 ppm, nearly double that found in coarser particles. This size-based separation allows for the enrichment of REE content in finer fractions, which can then be optimized in subsequent chemical leaching processes (Pan et al., 2019). These findings align with Tang et al. (2019), who reported that fractions smaller than 45 micrometers contained up to 1.5 times more REEs than coarser fractions. This separation technique, as part of the pre-treatment process, reduces chemical consumption and operational costs during leaching by targeting REE-rich fractions (Taggart et al., 2018). Additionally, studies have shown that particle size separation decreases the total mass that needs to be processed in subsequent extraction stages, thereby improving overall process efficiency (Park and Liang, 2019). By isolating REE-rich fine fractions, the chemical extraction steps can be more focused, increasing efficiency by up to 50% compared to direct leaching without pre-treatment (Peiravi et al., 2017).

Magnetic Separator: Magnetic separation is utilized for CFA containing ferromagnetic minerals or particles with high Fe content. According to Manurung et al. (2020), CFA with high Fe content can employ magnetic separation as a pre-treatment step. This technique enables the separation of approximately 30% of magnetic material associated with REEs in the tested fly ash. These ferromagnetic minerals often contain trace amounts of REEs linked to specific magnetic minerals. The findings indicate that magnetic separation is not only effective in isolating iron-rich fractions but also enhances the REE concentration in the fractions selected for subsequent extraction stages (Manurung et al., 2020). Furthermore, previous studies by King et al. (2018) and Cao et al. (2018) reinforce the effectiveness of magnetic separation, which can separate 10–30% of the total CFA mass. These fractions can then be processed using chemical methods for higher REE recovery (King et al., 2018; Cao, 2018).

Roasting Treatment: The roasting method applied to CFA involves the use of additives such as CaO, NaOH, Na_2CO_3, $(NH_4)_2SO_4$, and $CaSO_4$ prior to leaching, which has been proven effective in enhancing REE recovery rates (Taggart et al., 2018; Liu et al., 2019). In a study conducted by Liu et al. (2019), the extraction process began with an alkali roasting stage using Na_2CO_3 at a temperature of 1133 K. Following this step, H_2O leaching was performed to extract Al and Ga released during roasting. The next stage involved HCl acid leaching to recover REEs, followed by the use of ion adsorption resin to separate Al and Ga from the solution. This study reported an REE extraction efficiency of approximately 80% (Liu et al., 2019). In another study by Taggart et al. (2018), the combination of roasting treatment and HNO_3 acid leaching for REE recovery from CFA achieved even higher extraction rates, exceeding 90%.

By combining various physical techniques, including particle size classification, magnetic separation, and roasting, the efficiency of REE recovery in subsequent stages can be significantly enhanced. The detailed flowchart illustrating this process is presented in **Figure 1**.

Chemical Techniques

Acid leaching: Acid leaching is the most commonly used chemical method for recovering REEs from CFA. This process involves the use of strong acids such as H_2SO_4, HCl, and HNO_3 to dissolve REEs bound within the mineral matrix of CFA. In a study conducted by Taggart et al. (2018), HNO_3 was used as the leaching agent, achieving REE recovery rates of over 90%. This effectiveness is attributed to the ability of acid ions to break the bonds between REEs and the aluminosilicate structure within CFA (Taggart et al., 2018). Another study by Wang et al. (2019) demonstrated that using 2 M HCl with a solid-to-liquid ratio of 1:20 at a temperature of 80°C achieved recovery efficiencies of up to 75% for elements such as La, Nd, and Y. However, one major drawback of this method is the risk of acid waste contamination and the need for complex chemical residue management. Despite these challenges, acid leaching remains a reliable option, particularly for CFA with high REE content.

Alkaline leaching: Alkaline leaching utilizes alkaline solutions such as NaOH or Na_2CO_3 to dissolve silicate or aluminosilicate minerals in CFA, facilitating the release of REEs. According to Wen et al. (2020), alkaline leaching with NaOH at 120°C for 2 hours successfully dissolved 41.1% of the silica in CFA, thereby enhancing REE availability for subsequent separation steps. Furthermore, Pan et al. (2019) observed that this method is particularly effective for CFA with high silica content, achieving REE recovery rates of up to 60%. While alkaline leaching is less effective for direct REE recovery compared to acid leaching, it is often employed as a preparatory step to condition CFA for further leaching processes. This pre-treatment ensures that REEs are more accessible, improving the overall efficiency of subsequent extraction methods.

Alkaline leaching-acid leaching: A highly effective method for recovering REEs from CFA involves combining alkaline leaching with subsequent acid leaching. This process begins with alkaline leaching to dissolve silicate fractions and other obstructive minerals, followed by acid leaching to extract the remaining REEs, as illustrated in **Figure 1**. Research by Ma et al. (2019) demonstrated that using 5 M NaOH at 80°C for 2 hours, followed by 2 M HCl, achieved REE recovery rates of up to 88.15%, particularly for elements such as Ce, Nd, and La. Additionally, Taggart et al. (2018) highlighted that this combined process reduces acid consumption, making it both more environmentally friendly and cost-effective. Tang et al. (2019) further noted that this combination could improve recovery efficiency to 90% for CFA containing REEs in complex mineral forms. This approach is particularly effective for CFA with REEs bound in challenging aluminosilicate matrices, ensuring higher recovery rates while optimizing resource utilization.

Biological Techniques

Bioleaching: Bioleaching is a biological method that utilizes microorganisms to dissolve REEs from CFA. Microorganisms such as *Acidithiobacillus thiooxidans* produce organic or inorganic acids, such as H_2SO_4, which can leach REEs from the mineral matrix of CFA. Research by Su et al. (2020b) demonstrated that bioleaching with *Acidithiobacillus thiooxidans* achieved recovery rates of up to 76.37% for Ce and 64.77% for La at 30°C over a 7-day period. These results indicate a reasonable efficiency, albeit with a longer processing time compared to chemical methods. Additionally, the bioleaching process generates acidic waste with lower toxicity, simplifying waste management and making it more environmentally friendly than chemical leaching methods (Su et al., 2020b). The advantages of bioleaching lie not only in the microorganisms' ability to selectively dissolve REEs but also in their capacity to operate under relatively moderate environmental conditions without requiring high temperatures or pressures. However, the primary challenges of this technique include its relatively lengthy extraction time and the need to maintain a stable microbial culture environment (Sreenivas et al., 2021).

Biosorption: Biosorption is a biological technique that utilizes microorganisms or biomaterials to adsorb REEs from CFA leachate solutions. According to Park et al. (2020), Escherichia coli modified with lanthanide-binding peptides exhibits high selectivity toward heavy REEs (HREEs), such as La and Nd. This technique is capable of recovering up to 85% of REEs from leachate derived from Powder River Basin (PRB) fly ash. The main advantage of biosorption lies in its high selectivity for specific elements, enabling the separation of REEs with higher purity compared to other techniques. Additionally, biosorption uses renewable biological materials, making it a more sustainable solution. However, this technique requires pre-processing steps to produce REE-rich leachates, such as prior acid or alkaline leaching, to ensure effective biosorption (Park et al., 2020). A comparison of bioleaching and biosorption methods is presented in **Table 1**.

Table 1. Advantages and disadvantages of biological techniques

Techniques	Advantages	Disadvantages
Bioleaching	Environmentally friendly; acid waste is easier to manage.	Slow processing (up to 7 days); moderate efficiency.
Biosorption	High selectivity towards HREE; increasing REE purity.	Limited to leaching solutions; requires microbial modification.

Table 2. Summary study of REE Recovery Techniques from CFA

Physical Techniques	Chemical Techniques	Biological Techniques	Persen Recovery REE	Ref, [Code Ref]
Filtration, magnetic separator, hydrocyclone	Leaching water, leaching ammonium acetate, leaching ammonium acetate + acetic acid, acid leaching (HCl), leaching HNO_3 + H_2O_2, leaching (HF-HNO_3), Fusion (Na_2O_2)	x	98% LREE, 81% HREE	Sreenivas et al. (2021)
x	Alkali pretreatment (NaOH), acid leaching (HCl)	x	100% La, 90% Ce, 93% Nd	Wen et al. (2020)
x	Super-critical CO_2, tributylphosphate (TBP)	x	>99% Y, >99% Eu, <7% La, Ce, Tb	Das et al. (2018)
x	Acid leaching (HNO_3), solvent extraction	x	90.9% LREE, 94.2% HREE, 90.5% TREE*	Peiravi et al. (2017)
Alkaline roasting	Acid leaching (HNO_3)	x	>90% TREE	Taggart et al. (2018)
x	Alkali pretreatment (NaOH), acid leaching (HCl)	x	88.15% REE*	Wang et al. (2019)
x	Alkaline leaching (NaOH), acid leaching (HCl)	x	Alkali: 85% REE Acid: 40% REE	King et al. (2018)
x	Acid leaching (HCl)	x	80% REE	Honaker et al. (2019)
Precipitation	Acid leaching (HCl), solvent extraction	x	78% Ce, 80% La, 88% Nd, and 35% Y	Kumari et al. (2019)
Physical separation	Acid leaching (HCl)	x	80% TREE	Pan et al. (2020)
x	Hydrothermal alkali treatment (NaOH)	Bioleaching (*Acidithiobacillus thiooxidans*)	100% Sr, 90.8% V, 79.3% Y, 76.37% (Ce), 64.77% (La)	Su et al. (2020b)
x	Acid leaching (HCl)	Biosorption (*E. Coli*, *A. nicotianae*)	85% (La, Nd), 80% TREE	Park et al. (2020)

x	Alkali pretreatment (NaOH), Acid leaching (citric acid)	x	77.6% TREE	Rosita et al. (2023)
x	Liquid emulsion membrane (LEM), supported liquid membrane (SLM)	x	LEM: >70% TREE* SLM: 75% HREE, 50% LREE	Smith et al. (2019)
x	Alkali fusion (Na_2CO_3), Acid leaching (HCl)	x	72.78% TREE	Tang et al. (2019)
x	Acid leaching (HCl)	x	71.9% La, 66.0% Ce, 61.9% Nd	Cao et al. (2018)
x	Acid leaching (HNO3)	x	69.90% TREE	Zhang et al. (2020)
x	x	Indirect bioleaching (*C. Bombicola*)	63.0% Sc, 67.7% Yb, 64.6% Er	Park and Liang (2019)
x	Combined acid (HCl), alkali extraction (NaOH)	x	55% TREE*	Ma et al. (2019)
x	Acid leaching (HCl)	x	62.1% Y, 55.5% Nd, 65.2% Dy	Tuan et al. (2019)
x	Acid leaching (H_2SO_4, HNO_3, HCl)	x	H2SO4: 60% TREE HNO3: 29% TREE HCl: 2% TREE	Rao et al. (2020)
Physical separation (Particle separator, magnetic separator)	Alkali leaching (NaOH), Acid leaching (Acetic acid)	x	20.58% Ce, 43.53% Dy, 17.38% La, 40.96% Nd, 18.45% Y, 32.74% Yb	Manurung et al. (2020)

Economic Evaluation

The recovery of REE from CFA offers significant economic opportunities, particularly by transforming industrial waste into high-value products. In terms of processing costs, physical techniques such as magnetic separation and particle size classification require relatively low initial investment. According to Manurung et al. (2020), these methods can reduce material volume by up to 30% before further processing, lowering chemical leaching costs by approximately 20%. Meanwhile, roasting treatment, although highly effective with recovery rates exceeding 90% (Taggart et al., 2018), demands substantial thermal energy, making it more expensive than other physical techniques.

Chemical methods, such as acid leaching with HNO_3, achieve recovery efficiencies of over 90% (Taggart et al., 2018). However, the cost of chemicals and acid waste management can account for up to 40% of the total process cost (Wang et al., 2019). Alternatively, combining alkaline and acid leaching is more economical, as NaOH is used to break silicate matrices before acid leaching, reducing chemical consumption by up to 25%. Ma et al. (2019) reported that this combination achieved recovery rates of 88.15% at a cost of $500–700 per ton of ash, making it one of the most cost-efficient approaches for large-scale applications.

Biological techniques, such as bioleaching, provide a more environmentally friendly approach with lower operational costs compared to chemical leaching. Su et al. (2020) demonstrated that bioleaching with *Acidithiobacillus thiooxidans* recovered up to 76.37% Ce and 64.77% La, while generating waste that is easier to manage. This method reduces waste management costs by 30% compared to chemical leaching. However, the longer processing time (up to 7 days) and lower efficiency make it more suitable for small-scale projects or areas with strict environmental regulations. Biosorption, as highlighted by Park et al. (2020), offers high efficiency, recovering up to 85% of heavy REEs (HREEs), but requires microbial modifications, which add to operational costs.

From a value-added perspective, REE recovery from CFA holds attractive economic potential. REE content in CFA can reach 2100 mg/kg (Rao et al., 2020), with an economic value of approximately $3.5–5.0 per kg, depending on global markets. Additionally, high scandium (Sc) content further enhances economic value by up to 15%, with scandium priced at $2000 per kg in global markets (Das et al., 2018). Utilizing CFA also reduces the cost of coal ash waste management by $10–30 per ton (Sreenivas et al., 2021).

Overall, the combination of roasting, alkaline, and acid leaching techniques offers the best balance between recovery efficiency (>90%) and operational costs, making it the preferred choice for industrial-scale applications. Meanwhile, biological methods are better suited for small-scale applications due to their lower costs and minimal environmental impact, despite longer processing times. Integrating physical, chemical, and biological approaches can optimize REE recovery from CFA, providing competitive costs and controlled environmental impacts.

Environmental and Sustainability Review

Physical techniques such as magnetic separation, particle size classification, and roasting treatment generally have minimal environmental impact, as they do not produce direct chemical waste. According to Manurung et al. (2020), particle size classification utilizes the natural properties of materials without adding chemicals, resulting in waste limited to unused particle fractions. However, roasting treatment has a higher environmental footprint due to significant thermal energy consumption, which can increase carbon dioxide emissions (Taggart et al., 2018). Consequently, this method is better suited for facilities powered by sustainable energy sources.

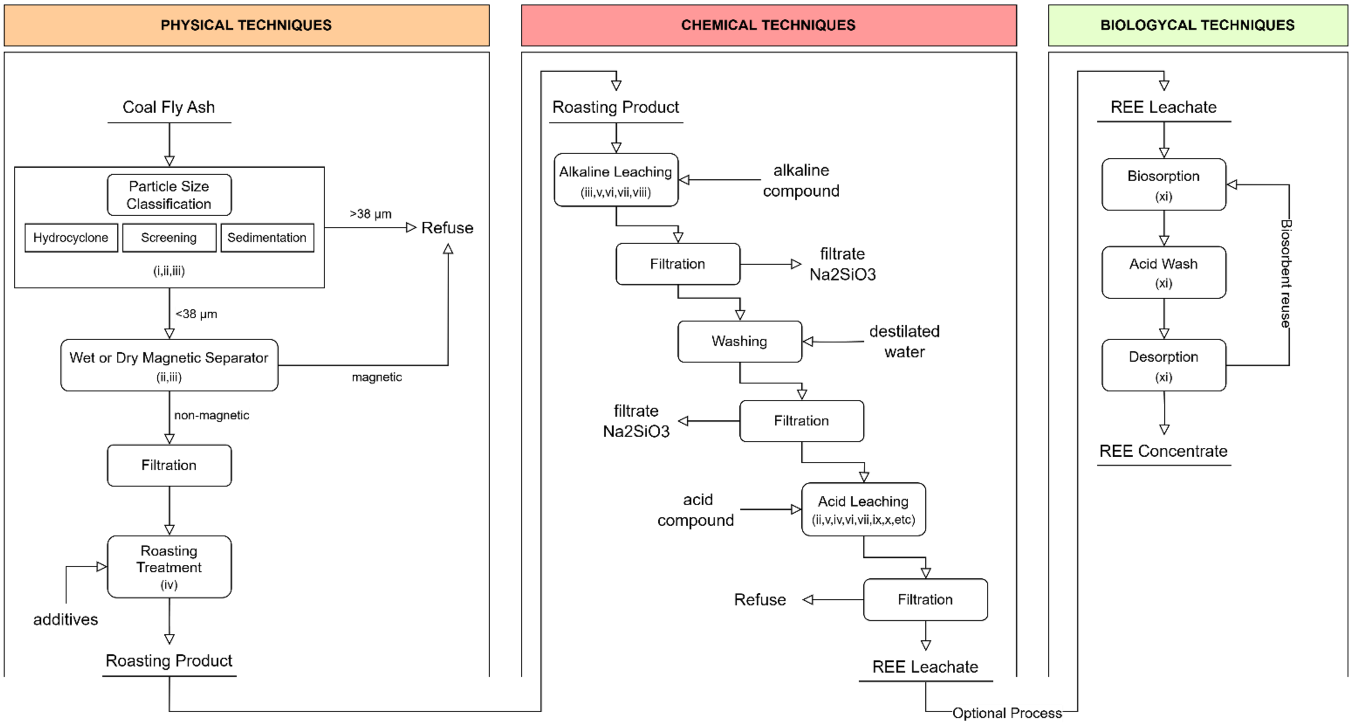
Chemical techniques like acid leaching and alkaline leaching, while highly efficient, generate liquid waste containing hazardous chemicals. Leaching with HCl or HNO_3, as reported by Wang et al. (2019), produces acidic waste that poses risks of groundwater and soil contamination if not managed properly. However, the combination of alkaline-acid leaching reduces acid consumption by up to 25% (Ma et al., 2019), thereby decreasing the volume of liquid waste generated. Chemical waste management remains a critical challenge for these techniques. Heavy metals such as arsenic and mercury often mix with the leachate, increasing toxicity risks (King et al., 2019). As such, stringent waste management systems are essential to prevent environmental contamination.

Unlike chemical techniques, biological approaches such as bioleaching and biosorption offer more environmentally friendly alternatives. In Su et al. (2020), bioleaching with *Acidithiobacillus thiooxidans* produced less liquid waste compared to chemical leaching. Moreover, the near-neutral pH of the solution made the waste easier to manage. This method also relies on natural microorganisms, reducing dependence on hazardous chemicals. Biosorption, as described by Park et al. (2020), employs modified microbes to adsorb REEs from leachate. This technique generates negligible liquid waste and requires only relatively safe buffer solutions. Such approaches are highly environmentally friendly and can be applied in the final stages of extraction to enhance selectivity without adding to the waste burden. By integrating physical, chemical, and biological techniques, REE recovery can be optimized while maintaining better control over environmental impacts. This combination ensures a more sustainable recovery process with reduced ecological risks.

Potential Technique for Recovery REE from CFA

Based on **Figure 1**, the evaluation of REE recovery techniques using a combination of physical and chemical methods demonstrates significant potential for selectively extracting REEs from coal fly ash. This approach includes physical steps such as particle sizing, wet magnetic separation, and roasting treatment to enhance REE concentration. Subsequently, chemical or hydrometallurgical processes are applied, involving alkaline solutions and acidic reagents for digestion and acid leaching. This combined method enables higher and more efficient REE recovery, especially when tailored to the characteristics of the processed ash (Sreenivas et al., 2021).

Alternatively, biological methods can be employed to reduce reliance on costly and environmentally impactful alkaline and acidic reagents. Bioleaching, utilizing microorganisms such as *Acidithiobacillus* or modified *E. coli*, offers a more environmentally friendly approach. However, to achieve optimal REE recovery rates, microorganisms need to be engineered for greater selectivity, particularly for heavy REEs such as lanthanides (Park et al., 2020). Integrating biological methods with physical and chemical approaches holds promise for enhancing process sustainability without compromising extraction efficiency.



Sources index: (i) Pan et al. (2020); (ii) Sreenivas et al. (2021); (iii) Manurung et al. (2020); (iv) Taggart et al. (2018); (v) Wen et al. (2020); (vi) Wang et al. (2019); (vii) King et al. (2018); (viii) Rosita et al. (2023); (ix) Peiravi et al. (2017); (x) Honaker et al. (2019); (xi) Park et al. (2020)

Figure 1. Potential flowchart techniques for recovering REE

CONCLUSION

This study highlights various techniques for recovering REEs from CFA, encompassing physical, chemical, and biological approaches, each with its own advantages and challenges. Physical techniques, such as particle separation, wet magnetic separation, and roasting treatment, are effective in enhancing REE concentration as a pre-treatment step for subsequent chemical processes. Chemical methods, particularly acid leaching and the alkaline-acid combination, offer high recovery efficiencies of up to 90%, although they require careful waste management to minimize environmental impacts. Meanwhile, biological techniques, such as bioleaching and biosorption, provide a more environmentally friendly alternative, albeit with lower efficiency compared to chemical methods. An integrative approach combining these three techniques shows significant potential for improving recovery efficiency, reducing costs, and minimizing environmental impacts, thereby supporting the principles of a circular economy and sustainability in the REE industry.

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