The 2nd International Conference on Multidisciplinary Engineering and Applied Sciences (ICMEAS-2023)

A Review on Extraction of Rare Earth Elements (REEs) From Coal Using Acid Leaching

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2023 2nd International Conference on Multidisciplinary Engineering and Applied Science (ICMEAS) | 979-8-3503-5883-4/23/$31.00 ©2023 IEEE | DOI: 10.1109/ICMEAS58693.2023.10429850

***Abstract*- Coal has become a feasible source of rare earth elements (REEs; the 14 stable lanthanides, scandium, and yttrium). It is believed to contain significant amounts of rare earth elements, making it a primary source of REEs which serves as basic raw materials in the production of renewable energy. This review established the feasibility of recovering REEs from coal using acid leaching method. It discusses; the sourcing of REEs from coal, the applications of REEs and acid leaching as an effective hydrometallurgical method for extracting REEs from coal. It also examined the efficiency of methods used by other researchers in extracting REEs from coal. However, the potential of acid leaching as a solution to issues such as: difficult leaching conditions, low recovery and the use of expensive chemicals has not been fully investigated. For a better choice in the extraction of REEs from coal, more study and review are necessary.**

A. Keywords—acid leaching, coal, extraction, leaching, rare earth elements.

I. INTRODUCTION

Vanadium (V) and silver (Ag) were extracted from Poland coal debris in the late nineteenth hundred years, starting the historical backdrop of metals recuperation from coal (otherwise called coal fly debris). The ignition side-effects of coal (for the most part debris) have gotten a ton of interest in the 20th century as potential wellsprings of valuable metals like germanium (Ge), gold (Au), uranium (U) and gallium (Ga). U and Ge were effectively economically recuperated from coal debris by the middle of the 20th century in Japan, the Soviet Association and the UK [1].

Because of its overflow and absence of necessity for complex foundation and innovation, coal is broadly involved and is the most ideal choice for meeting the energy needs of low-pay and developing countries. Coal will remain in huge demand for a while to come since it is also required for the production of concrete and steel (as coke). In any case, gangue minerals such as quartz and pyrite that are normally present in coal can deliver side-effects like debris and various poisons including (Sulfur oxides (SO_X), carbon dioxide (CO_2) and nitrogen oxides (NO_X) [2].

Rare and precious metals are thought to be present in high concentrations in coal, making it a possible source of REEs. Numerous coals have been found to contain significant amounts of rare earth elements, including the Kentucky Fire Clay coal in the Eastern United States (14,700MT), the coal from Southwest China (94,500MT), and the coal from the Far East Russian (1,690MT) [3].

II. COAL: A SOURCE OF REEs

A vital raw material utilized in the metallurgical industries as a source of energy and reductant is coal. High grade coking coals are desirable for use in metallurgy, particularly in blast furnace operations, although they are not widely available. This has prompted the development of new ways that can use coal as a fuel and a reducing agent to remove iron from its ore. Elective strategies incorporate COREX iron creation, direct reduced iron through Rotary kilns and Tunnel. Enormous particles are unavoidably delivered during irregularity lean grade coal extraction, transportation and taking care of. A great many lots of fines (around 3 mm) are purportedly delivered during mining tasks [4].

Coal distribution, stratigraphy and climatic conditions have all been studied in researches. Regional and periodic changes in REE are linked to the original rock's characteristics and the tectonic history of the point of origin area. It has been demonstrated that rare earth elements (REE) in coal and clastic deposits can help us comprehend the sedimentary setting of coal-containing successions. Because of the consistency of their activity across multiple geochemical mechanisms and the regularity of their fragmentation patterns, they can provide information into the history following coal deposition. This is because of the way that a few topographical and physical-synthetic qualities are reflected in their development and overseeing factors. Various examinations on the geochemistry of REE in coal, metal minerals, mudstone and dark shale volcanic rock have explained their starting point, structural setting, paleoenvironment and different conditions encompassing their decomposition [4].

III. REEs RECOVERY FROM COAL

The recuperation of REEs out of coal fly ash and coal is presently ongoing and is supposed to be an interesting and hot topic in the foreseeable future. The speciation, genetic origin of REEs, concentrations and techniques for recuperating REEs out of coal and the byproduct of coal [5].

Rare earth elements often only make up traces of coal (less than 0.1 weight percent). To meet the requirements for industrial extraction, it is important to locate REE-rich coal or enrich REE in coal ash [6]. A rare earth element classification system has been developed for studies pertaining to coal by splitting the REE into; HREE (Eu to Lu) and light REE (La to Sm). REE abundance in natural samples follows the Oddo-Harkins rule, which asserts that elements with even atomic numbers are more prevalent than elements with odd atomic numbers. Coal and coal fly ash have also been shown to have negative consequences [7].

The term "rare earth elements" (REE) refers to a collection of 17 elements that the International Union of Pure and Applied Chemistry (IUPAC) classifies as having different but related physical and chemical properties. They consist of the transition metals Sc and Y as well as the lanthanide series elements (Dy, Sm, Tm, La, Pm, Nd, Tb, Ce, Lu, Er, Sm, Pr, Gd, Er, and Ho) [8].

The 15 lanthanide group elements (from La to Lu) along with chemically related Y and incidentally Sc, make up the REEs. The light rare earth components (LREEs), which incorporate La through Eu and the weighty interesting earth elements (HREEs), which likewise contain the other lanthanide components and yttrium, are isolated into two sub-groups. Scandium's considerably more modest ionic sweep keeps it from squeezing into both of these groupings [9].

Although rare earth components are generally more predominant on the planet's surface than any other regularly utilized elements but their lack in concentration makes their exploitation difficult. This is because their ionic radii are comparable, making them interchangeable in many minerals and difficult to separate [9]. Since REEs have unique luminescent, attractive, electrical and optical abilities because of the consecutive filling of the f-orbitals, they are viewed as fundamental components [10].

IV. APPLICATION OF RARE EARTH ELEMENTS

Most of REEs applications include catalysis (oil refineries, catalytic converters), chemical processes (water treatment plants), permanent magnets (disk drive motors, disk drives, motors, clean energy), polishing (refractories capacitors and sensors, UV resistant glass), metal alloys (aircraft making, steel alloy industrial manufacturing) glass, ceramic, phosphors (fluorescent light, scintillation sensors, cathode ray tubes), magnetic resonance imaging (MRI). The usage of REE has also been concentrated on climate-related challenges. Among other things, REE are used in wind turbine generators, specific alloys that enable automobiles to be lighter (contributing to a drop in fuel consumption) and UV-protected glass (contributing to a decrease in energy use in buildings). To achieve a carbon-free and sustainable global energy source, rare earth elements are essential [11].

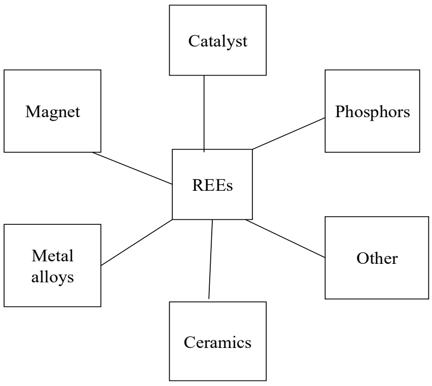

Fig. 1: Application of REEs

The primary resources, which are; monazite, xenotime and bastnasite and the secondary resources are the two main mineral sources of REEs. The subject of the current study, monazite, is made up of REE phosphates and contains thorium and uranium in varying amounts. To remove REEs from monazite, it is now baked with sulfuric acid or digested with alkali. Monazite concentrations are treated via alkaline digestion, whereas lower grade monazites are often baked in sulfuric acid [12]. Many nations now view industrial waste and electronic waste as secondary resources for these elements because there are no operational or economically viable primary reserves for them. As they pose a risk to the environment if improperly managed, this technique may help to lessen the environmental issues related to abstraction and mining of REEs [13].

The properties of REE vary as a result of the variations in electronic configurations, such as the number of electrons present in 4f orbitals and each has unique uses in high-tech industries [14]. To comprehend their distinctive behavior in many processes, including; mineral processing, the history of rock geochemical evolution, environmental mobility etc. [15] [16], various classification approaches have been devised and employed [17] [18]. The percentage of LREE to HREE (L/H) was used to examine the allocation of REE in coal and coal fly ash. A three-part industrial separation of REE, including crucial (Er, Tb, Nd, Y, Eu and Dy), uncritical (Pr, Sm, Gd and La) and excessive (Lu, Yb, Tm, Ce and Lo) groups is proposed [7] in order to further assess coal as a REE raw material. Be aware that the division of rare earth elements into crucial, uncritical and excessive groups is partly arbitrary and ephemeral, depending on the needs of the worldwide market and the available supplies [7]. There have been substantial price variations for some REEs during the last 10 years [19].

V. HYDROMETALLURGICAL PROCESSES

Processes involving hydrometallurgy are very profitable since they operate at low cost, may be able to recover leachates and produce less air pollution. In the hydrometallurgical processes leaching and disintegration step is where rare earth elements are recuperated from a powder blend produced using waste assets. Decomposition and leaching refer to a method for transforming a mixture of powders containing different components into a state that may be decomposed in water or acid to create a form suitable for separation and refining [20]. Additionally, the separated and purified rare earth solution is eventually converted into oxides for use in industries. Lately, hydrometallurgy was examined as a method to recover rare earth elements, primarily; yttrium and europium [21].

Similarly, Innocenzi and his exploration group had the option to recuperate yttrium from utilized cathode ray tubes and fluorescent lights through a step-by-step method that included filtering with H_2SO_4, cleansing and ultimately precipitation with oxalic corrosive to deliver yttrium oxalates as the outcome. The quantity of Y recovered in this strategy was around 55% and the yttrium oxide that was delivered was 99% pure [22] [23].

VI. ACID LEACHING

The typical technique for obtaining rare earth elements (REEs) from parent rock and other sources is acid leaching. The REEs dissolve in an acid solution as part of the procedure, after which the solid residue is removed from the REEs. By maximizing the leaching conditions, such as acid concentration, time and temperature, REEs can be removed more effectively [33].

The process of treating phosphogypsum fertilizer (PGF) to extract REEs is an illustration of acid leaching. PGF, a phosphoric acid industrial byproduct, has trace amounts of REEs. To create calcium carbonate precipitated with REEs, a metathesis of PGF with sodium carbonate is required. Then, nitric acid or citric acid leaches off the REE-containing calcium carbonate. However, acid leaching can be utilized to remove REEs from coal and it very well may be additionally compelling in recuperating metals from minerals and concentrates [33].

In acid leaching, monazite is extracted at high temperatures while the rare earth elements are dissolved under pressure in a sulfuric acid solution. Impurities such as Th and Fe are leached simultaneously in this non-selective process. There will be a greater leaching of contaminants at low working pH values. Processing specimens with large gangue mineral structures in the REE concentrates may necessitate a higher acid/example (w/w) concentration, particularly if iron is present. At the point when temperature and filtering span are expanded, the acid/example (w/w) proportion should likewise go up. Following balance, the fluid is either accelerated as sodium twofold sulfate to isolate the REEs from the thorium or the REEs are isolated by precipitation [24].

The pummeled coal sample and acid solutions are added to the beaker at the portion fundamental for the response. Leaching experiments are much of the time directed in a measuring glass, drenched in a temperature-controlled water shower and furnished with an attractive fomenter and temperature sensor. The temperature is ordinarily the main variable in draining. The start and end of the leaching reactions are regularly controlled utilizing an attractive instigator at a suitable blending rate. The slurry is expected to be vacuum separated after each draining test. One powerful technique for removing REEs from coal is filtering [25].

Acid leaching transfers a portion of the rare earth elements from the solid state to the solution, where they can then be further recovered and purified using different techniques such physical beneficiation. Leaching performance is dependent on the type of the materials being examined; more acid is often consumed; comparatively higher recovery; Leaching is thought to have several potential pitfalls, including low recovery, expensive chemicals and difficult leaching conditions [25].

Similarly, Riya Banerjee and her research team examined five organic carboxylic acids i.e. monocarboxylic (lactic acid), dicarboxylic (tartaric, malonic and succinic acids) and tricarboxylic acid (citric acid) for their efficiency in successfully leaching rare earth elements out of coal ash. Without using mineral acids, it has been shown that a relatively low concentration of organic acids (5%), when present, can significantly increase the efficiency of leaching (to 62%) of REEs from coal ash. Because carboxylic acids extracted REEs from coal ash without affecting its minerology, it was possible to use the cleaned coal ash for other purposes, such as in the brick and cement industries. Because organic acids are thought to be far kinder and more environmentally benign than mineral acids, the work's uniqueness resides in the efficient and focused leaching of lanthanides from coal ash utilizing solely these acids. [30].

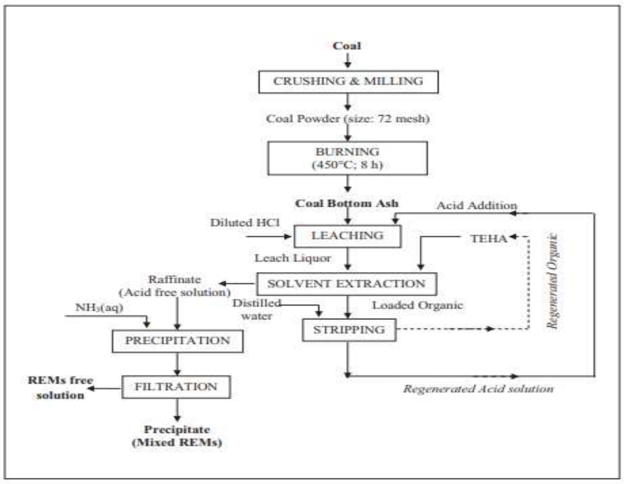

Fig. 2: Proposed stream sheets for the recuperation of REMs from CBA [26].

TABLE I. Summary of the efficiency of methods used in the extraction of REEs from coal by other researchers

EFFICIENCY OF METHODS USED IN THE EXTRACTION OF REEs FROM COAL			
SAMPLE SOURCE	**METHOD**	**CONCLUSION**	**REFERENCE**
Coal fly ash from a chinese power generation plant in using coal as enegy source	Behzad Vaziri Hassas, Mohammad Rezaee, Zhiping Wen, Changchun Zhou, Jinhe Pan, and Tiancheng Nie in 2019, used coupled physical separation operations with REY of CFA boosted from 782 µg·g−1to 1025µg·g−1, and then acid leaching with 80% HCl (optimized for different variables using the Taguchi three-level experimental design)	The appropriate conditions were met for the separation of the physical product after optimization, yielding a leaching efficiency of 79.85%.	[27]
Coal combustion ash Coals from three major U.S coal basin sources	In 2018, Heileen Hsu-Kim, Ross K. Taggart, Ryan C. Smith, James C. Hower, and Jack F. King, employed alkaline leaching, with varied leaching parameters such as leachate-to-ash ratio, extractant type (HCl, NaOH), extractant concentration and CaO addition throughout the leaching process: At 85°C, the sample was treated with 6.25 mol/L NaOH, then subjected to acid leaching with 20% HCl for 80 minutes.	The Appalachian and Illinois low calcium fly ashes from Basin coals exhibited much poorer leaching efficiency (only 40%), whereas the Powder River Basin coals had the highest REE recoveries (approximately 100% of total REE concentrations). Overall, the findings indicate that compared to the other ashes under study, REEs in coal fly ashes from the Powder River Basin leach into acidic solution more fast.	[28]
Coal fly ash Preparation plants treating West Kentucky coal, Illinois coal, and fly ash and surface ash specimens from two FBC power stations, as well as Fire Clay coal mine sources.	R. Q. Honaker, R. Q. Honaker W. Zhang, and J. Werner in 2019, extracted REEs from coal by roasting the sample with Na_2CO_3 (1:1 by weight) at 600°C, then undergoing 2 hours of successive leaching using water and 1.2 mol/L HCl at 750°C.	Pretreatment of coal-based mineral matter in an oxidizing atmosphere at high temperatures (600^0C, 750^0C, and 900^0C) greatly improves the ability of rare earth elements (REEs) to leach. The findings indicated that coal preprocessing at 600°C for 2 hours rose the recovery of REEs from 20-40% to 80% for all coal sources.	[29]
Coal fly ash Coal of Indian origin	Riya Banerjee, Ashok Mohanty, Sanchita Chakravarty, Saswati Chakladar and Paromita Biswa in 2021, used one step process to leach out of REEs from CFA; a coal sample was treated with 5% tartaric acid with a natural solution pH of 1.8 at 90°C for 60 minutes.	Best leaching efficiency was attained (65% for LREE, 19% for HREE, and 62% for total REE). Tartaric acid had the highest efficiency for leaching among the various carboxylic acids.	[30]
Coal fly ash Blue gem coal form Knox County, Kentucky	In 2018, Hower JC, Berti D, Hochella MF, Rimmer SM, and Taulbee DN, used acid digestion and electrowinning processes where REE oxides and REE carbonates were dissolved at pH 5.5 4; apatite at pH 3.5 2; REE phosphates and hematite at pH 1.5; and zircon and glass phase at pH 1.5	Class C fly ash comprises 50-60% REEs in the form of REE oxides and carbonates, 20-30% in the form of phosphates and hematite, and 20% in the form of apatite. Class F fly ash contains 30-70% REEs linked to the glass and zircon phases, 10–40% to apatite, 10% to carbonates and oxides, and 10% to phosphates and hematite.	[31]
Inner Mongolian coal fly ash from the Jungar power station in China	In 2010, Wang X, Zhang Y, Peng S, Chou CL, Zhao L, and Dai S, employed a magnetic separation approach to separate the fly ash into three phases: a ferromagnetic-rich phase, a glass phase, and a mullite-corundum phase.	Glassy phases contain a lot of REE but in limited quantities in crystalline materials.	[32]

VII. CONCLUSION

The research investigated the abstraction of rare earth elements from coal using acid leaching and discovered that high purity rare earth elements may be generated successfully from coal. Charcoal is a dependable source of energy generation and a feedstock for the manufacturing of chemicals, fuels and steel in both industrialized and developing nations. Additionally, it is the least expensive method of producing power worldwide.

REEs are critical components for the advancement of modern industry, as well as the discovery and development of high-tech things that we use on a daily basis. Filtering (leaching) is one of the most successful and reliable methods for removing rare earth elements from coal. The acid leaching method yields a high return of REEs. Significant yields of REEs are right now separated utilizing acid leaching. The most crucial factor in leaching is typically the temperature. This study examines the origins, applications and utilization of rare earth elements and moreover, the extraction of those elements out of coal via acid leaching. A part of rare earth elements is transferred from the solid state into the solution by acid leaching, where they can then be further recovered and purified using different techniques like physical beneficiation. The efficiency of acid leaching is influenced by the type of examined samples, leaching process pitfalls include expensive chemicals, difficult leaching conditions and low recovery.

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