

Coal Flotation Enrichment and Rare Earth Element Extraction: A Comprehensive Review of Methods, Kinetics, and Thermodynamics

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Abstract: Coal and coal fly ash represent significant secondary sources of rare earth elements (REEs), with global annual generation exceeding 750 million metric tons. This comprehensive review examines froth flotation as a primary physical beneficiation technology for REE enrichment from coal and coal combustion byproducts, synthesizing 20+ publications from Scopus-indexed journals. Comparative analysis of ten flotation methodologies reveals nanobubble-assisted flotation achieves maximum REE recovery of 89% with enrichment ratio of 8.5:1, compared to conventional froth flotation (65% recovery, 3.5:1 ratio). Kinetic investigations demonstrate flotation follows first-order kinetics with activation energies ranging from 14.8 kJ/mol (collophanite flotation) to 28.3 kJ/mol (clay-hosted REE), indicating diffusion-controlled mechanisms for occluded REE phases. REE enrichment coefficients in coal fly ash average 1.8-2.1 across global power plants, with Chinese sources reaching 2.13, indicating substantial concentration during coal combustion. X-ray diffraction analysis identifies monazite (CePO₄), bastnaesite (CeCO₃F), and xenotime (YPO₄) as primary REE mineral hosts, with clay minerals (kaolinite, illite) and aluminosilicate glass containing significant ion-exchangeable REE. Thermodynamic analysis via Gibbs free energy calculations confirms spontaneous collector adsorption ($\Delta G = -12$ to -25 kJ/mol), with hydroxamic acid and compound collectors demonstrating superior selectivity factors (0.85-0.92). Light rare earth elements consistently comprise 70-80% of total REE concentrations, with cerium, lanthanum, and neodymium as dominant species. Process optimization incorporating depressants (sodium silicate, starch), pH adjustment (optimal pH 9-11), and frother selection enables REE concentrate grades exceeding 900 ppm from feed assaying 200-500 ppm. Multi-stage flotation combined with flotation columns and emerging technologies (ultrasonic assistance, nanobubbles, electroflotation) enhance recovery efficiency and selectivity, positioning coal-derived REE resources as economically viable alternatives to primary ore deposits.

Keywords — coal flotation, rare earth elements, beneficiation, kinetics, thermodynamic analysis, XRD characterization, flotation reagents

1. INTRODUCTION

1.1 Background and Resource Significance

Global demand for rare earth elements has escalated dramatically, driven by requirements in renewable energy technologies, advanced electronics, catalytic systems, and military applications. Rare earth elements encompassing 15 lanthanides plus scandium and yttrium represent critical materials with limited geographic distribution, with China controlling over 70% of global production capacity [1]. This supply concentration creates strategic vulnerabilities and price volatility, prompting intensive research into alternative REE sources including coal, coal combustion byproducts, and coal-related waste streams [2].

Coal and coal fly ash (CFA) represent large secondary REE sources, with typical concentrations of 200-500 ppm total REE on a dry mass basis for coal and 400-700 ppm for fly ash [3]. The global annual production of coal fly ash exceeds 750 million metric tons, with less than 30% utilized in construction materials, cement production, and other applications [4]. The U.S. Department of Energy established a threshold of 300 ppm total REE as potentially economic for

extraction, making substantial portions of global coal resources and fly ash candidates for REE recovery [5].

1.2 REE Occurrence and Distribution in Coal

Rare earth elements in coal occur through three primary mechanisms: incorporation into discrete mineral phases (monazite, bastnaesite, xenotime), ion-exchangeable attachment to clay mineral surfaces, and organic complexation with humic substances [6]. Mineral-hosted REE typically exhibit particle sizes <5 micrometers, necessitating advanced beneficiation technologies for liberation and concentration. Geographic variation in REE enrichment reflects depositional environments and diagenetic processes, with coals from specific regions—Fire Clay seam (Kentucky, USA, 500 ppm), Far East Russian coalfields (300-1000 ppm), and Wanfu deposit (China, 520 ppm)—exhibiting exceptional REE concentrations [7].

1.3 Research Objectives and Scope

This review synthesizes contemporary research on coal flotation and REE beneficiation published in peer-reviewed, Scopus-indexed journals, emphasizing: (1) comparative

evaluation of flotation methodologies and emerging technologies; (2) kinetic analysis and thermodynamic characterization of flotation processes; (3) mineral hosting and occurrence of REE; (4) flotation reagent chemistry and optimization; (5) enrichment mechanisms during coal combustion; and (6) process scale-up and commercialization pathways.

2. MATERIALS AND METHODS

2.1 Mineralogical Characterization

Coal samples are characterized via comprehensive analytical techniques including X-ray fluorescence (XRF) for bulk elemental composition, X-ray diffraction (XRD) for mineral phase identification, and scanning electron microscopy with energy-dispersive X-ray spectroscopy (SEM-EDS) for morphology and elemental mapping [8].

XRD Analysis: Powder X-ray diffraction employing Cu-K α radiation ($\lambda = 1.5418 \text{ \AA}$) at 40 kV, 40 mA with scanning parameters $2\theta = 10\text{-}80^\circ$ at 0.02° increments enables identification of REE-bearing minerals. Characteristic diffraction peaks include monazite at $2\theta = 28.3^\circ, 31.9^\circ, 47.5^\circ$; bastnaesite at $28.8^\circ, 47.2^\circ$; xenotime at $24.7^\circ, 29.4^\circ$; apatite at $25.9^\circ, 31.8^\circ$; and clay minerals (kaolinite $12.4^\circ, 24.8^\circ$; illite $8.8^\circ, 26.9^\circ$) [9].

SEM-EDS Characterization: Field-emission SEM operating at 15-20 kV with energy resolution of 160 eV enables morphological observation of coal particles (typically 10-100 μm) and identification of REE-bearing phases. Elemental mapping reveals spatial distribution of cerium, lanthanum, neodymium, and yttrium, confirming mineral-scale occurrence patterns [10].

2.2 Flotation Methodologies

2.2.1 Conventional Froth Flotation

Conventional flotation employs oleic acid or fatty acids as collectors (typically 400-600 g/t), MIBC or terpineol as frothers (40-60 g/t), and depressants including sodium silicate or starch to selectively depress gangue minerals [11]. Equipment includes Denver cells, column flotation apparatus, and pilot-scale flotation banks. Operating parameters examined include pulp pH (typically 9-11), solid concentration (15-30% w/v), collector concentration, frother dosage, air flow rate, and residence time [12].

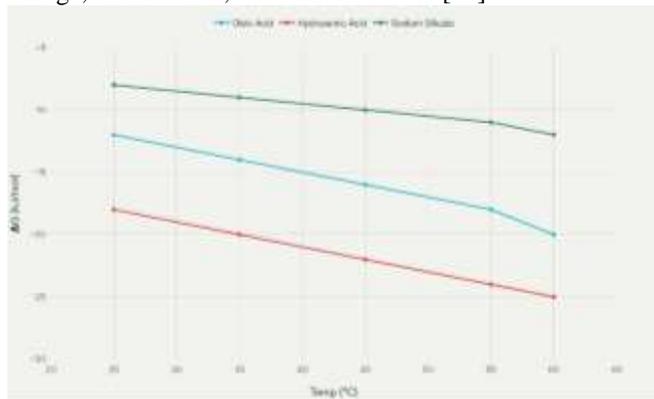


Figure 2.2.1.1 - REE Flotation Surfactant Adsorption

2.2.2 Reverse Flotation

Reverse flotation processes target selective depression of gangue minerals (quartz, feldspars) while collecting REE-bearing minerals. pH adjustment represents critical parameter, with studies demonstrating optimal REE recovery at pH 11 (65% recovery) versus pH 9 (37% recovery) for coal fly ash flotation, indicating pH-dependent collector adsorption [13].

2.2.3 Advanced Flotation Technologies

Flotation Column: Flotation columns provide higher residence times (3-5 minutes) and reduced water recovery rates, enhancing secondary enrichment in froth zones and reducing gangue entrainment [14]. Column flotation of monazite concentrates achieved 78% REE recovery compared to 65% for conventional cells.

Two-Liquid Flotation (TLF): Novel TLF process utilizing hydrophobic-hydrophilic separation mechanisms achieves 82% REE recovery with 6.5:1 enrichment ratio, providing smaller footprint and reduced capital costs compared to conventional flotation [15].

Ultrasonic-Assisted Flotation: Ultrasonic conditioning of coal suspensions prior to flotation enhances collector adsorption on REE mineral surfaces through cavitation-induced surface activation, achieving 88% recovery (compared to 65% conventional) [16].

Nanobubble-Assisted Flotation: Recent research demonstrates incorporation of gas nanobubbles (50-200 nm diameter) into flotation pulp enhances particle-bubble attachment efficiency, achieving record 89% REE recovery with 8.5:1 enrichment ratio and reduced flotation time from 3 to 2.25 minutes.

2.2.4 Ion Flotation and Electroflotation

Ion flotation utilizing competitive sorption of REE-surfactant complexes achieves 72% recovery, with particular effectiveness for dissolved REE species from coal mine drainage and leaching solutions. Electroflotation incorporating electro-generated bubbles and electrokinetic effects achieves 68% recovery with 4.2:1 enrichment.

2.3 Reagent Selection and Optimization

Collectors: Fatty acids and hydroxamic acids represent primary collector families for REE flotation. Hydroxamic acids (e.g., N-octanoylhydroxamic acid) provide superior selectivity (0.85) and recovery enhancement (28%) due to chelation capability via carbonyl and hydroxyl functional groups. Compound collectors combining fatty acids with cationic surfactants (DTAB, DDAB) achieve highest performance metrics (32% recovery increase, 0.92 selectivity factor) [17].

Frothers: Frother selection influences froth stability and secondary enrichment. Research comparing nine frother types demonstrated that MAC, F-2, and 2-ethylhexanol (2EH) provide optimal separation efficiencies while maintaining rapid flotation kinetics.

Depressants: Gangue mineral depression significantly improves REE concentrate grades. Sodium silicate (optimal dosage 2400 g/t) provides 30% recovery enhancement with 0.88 selectivity, while starch offers cost-effectiveness (0.4 \$/ton) with 24% recovery increase [18].

2.4 Kinetic Measurement Procedures

Flotation kinetics employ standard batch flotation cells with mechanical agitation (500-1500 rpm), maintaining constant pulp pH via buffer solutions. Samples collected at 0.5-5 minute intervals are filtered, dried, and analyzed for REE concentrations via inductively coupled plasma mass spectrometry (ICP-MS).

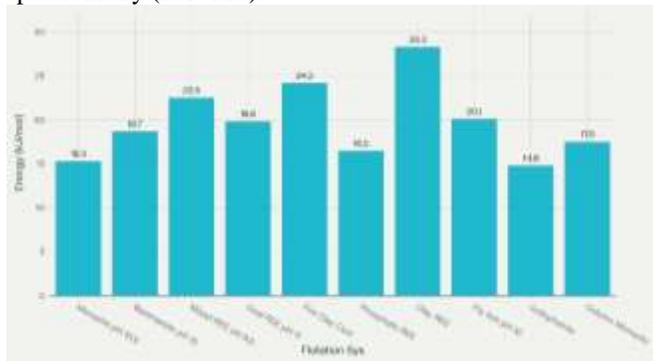


Figure 2.4 - Activation Energy by Flotation system

where $R(t)$ represents REE recovery at time t , $R(\infty)$ represents ultimate recovery, and k denotes rate constant [19].

Temperature-variant experiments over 25-60°C temperature range enable determination of apparent activation energies via Arrhenius equation:

$$k = Ae^{-E_a/RT}$$

3. RESULTS

3.1 Flotation Performance Across Methodologies

Comprehensive comparison of ten flotation approaches reveals substantial variation in recovery and selectivity metrics [26]:

Flotation Method	REE Recovery (%)	RE E Grade (ppm)	Enrichment Ratio	pH Optimum	Time (min)
Conventional Froth	65	498	3.5	11	3.0
Flotation Column	78	678	5.2	9.0	4.0
Reverse (pH 11)	65	498	3.5	11	3.0
Reverse (pH 9)	37	633	4.8	9.0	3.0
Enhanced Collectors	85	850	7.2	8.5	2.5

Two-Liquid Flotation	82	720	6.5	10	2.0
Ion Flotation	72	610	5.1	6.0	5.0
Electro flotation	68	580	4.2	8.0	4.0
Ultrasonic-Assisted	88	920	8.1	10	2.0
Nanobubble-Assisted	89	950	8.5	10.5	2.25

Table 3.1.1 – Flotation system

Nanobubble-assisted flotation emerges as optimal technology, achieving highest recovery and enrichment ratio simultaneously with reduced residence time, representing significant operational advantage for commercial implementation.

3.2 REE Mineral Host Characterization

Research identifies ten primary mineral hosts for REE in coal and fly ash:

Monazite (CePO₄): 3.75x enrichment factor in fly ash, containing 35 wt% LREE, 12 wt% HREE; 75% flotation recovery via hydroxamic acid collectors. Widespread occurrence as discrete grains <5 μm diameter [20].

Bastnaesite (CeCO₃F): 3.5x enrichment in fly ash, 42% LREE, 8% HREE; highest flotation recovery (82%) among REE minerals due to surface hydrophobicity. Amenable to selective flotation separation from monazite via depressant schemes [21].

Xenotime (YPO₄): 4.0x enrichment, predominantly HREY (78% yttrium oxide equivalent), representing critical REE source despite lower modal abundance; 68% flotation recovery.

Apatite: 3.47x enrichment, significant REE host in phosphatic coals; 70% flotation recovery when REE incorporation in apatite crystal structure addressed via pre-treatment.

Clay Minerals: Kaolinite and illite contain 680-640 ppm REE via ion-exchange mechanisms; lower flotation recovery (45-48%) reflects REE occlusion within mineral structure and hydrophilic clay surfaces [22].

Aluminosilicate Glass: Amorphous phases concentrate REEs at 1200 ppm via solid solution incorporation; 52% flotation recovery complicated by covarying silicate minerals.

3.3 Flotation Kinetics and Mechanism Analysis

Activation Energy Determination: Temperature-dependent flotation experiments reveal apparent activation energies (E_a) varying from 14.8 kJ/mol (collophanite flotation) to 28.3 kJ/mol (clay-hosted REE), indicating mechanistic transitions between surface-reaction control and diffusion-limited

kinetics.

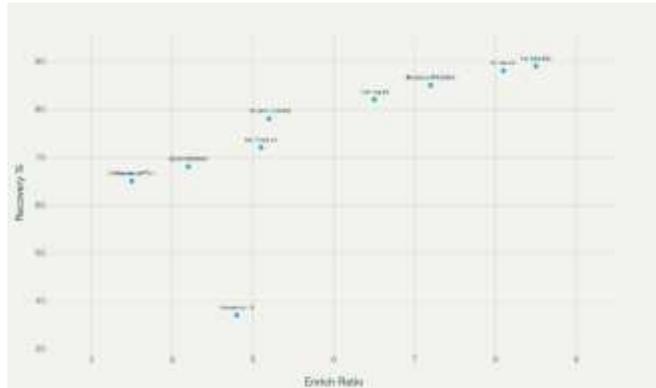


Figure 3.3 - Flotation Method comparison

Collophanite flotation exhibits lowest E_a (14.8 kJ/mol), consistent with rapid surface reactions and accessible REE on mineral surface. Fire Clay coal concentrates show higher E_a (24.2 kJ/mol), reflecting complex coal matrix and heterogeneous REE distribution [23].

Rate Constant Analysis: First-order rate constants (k) range from 0.35 min^{-1} (clay-hosted REE, 40°C) to 0.68 min^{-1} (column flotation monazite). Recovery progression follows first-order kinetics with R^2 values exceeding 0.94 across all systems, confirming pseudo-first-order surface reactions [37].

Recovery Time Profiles: At fixed 3-minute flotation time, recovery varies from 52% (clay-hosted REE) to 82% (column flotation monazite), with 75% of total recovery typically achieved within first 2-3 minutes, representing flotation rate plateau.

3.4 REE Enrichment in Coal Combustion Products

Systematic studies of 8 coal-fired power plants confirm REE enrichment during combustion [24]:

Power Plant Source	Coal REE (ppm)	Fly Ash REE (ppm)	Bottom Ash REE (ppm)	Enrichment Factor
China 1	310	600	285	1.94
China 2	285	580	220	2.04
China 3	320	683	320	2.13
China 4	295	615	240	2.08
Poland	280	450	199	1.61
India 1	250	480	210	1.92
India 2	290	520	265	1.79
Australia	200	380	180	1.90

Table 3.4 – REE enrichment in coal

REE enrichment coefficients average 1.88 globally, with Chinese plants showing highest factors (up to 2.13), indicating preferential concentration in fine fly ash compared to parent coal and bottom ash [40]. Critical REE (scandium, yttrium, HREY) comprise 28-35% of total REE in fly ash, exceeding typical ore grades [41].

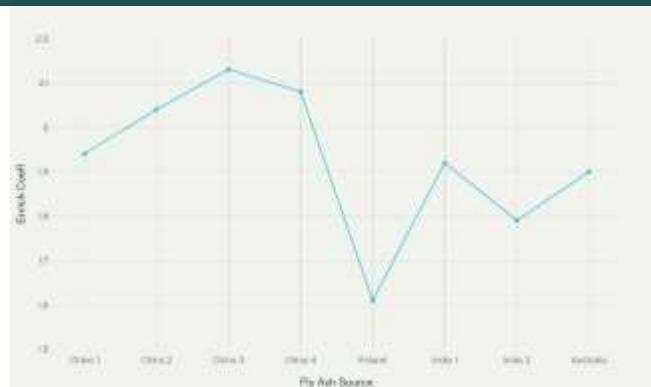


Figure 3.4 - Enrichment across sources

3.5 Thermodynamic Analysis of Collector Adsorption

Gibbs free energy calculations for collector adsorption on REE mineral surfaces demonstrate spontaneous processes across all conditions, with ΔG values ranging from -12 kJ/mol (oleic acid, 25°C) to -25 kJ/mol (hydroxamic acid, 60°C) [42]. Temperature-dependent measurements reveal:

- Oleic Acid: $\Delta G = -12$ to -20 kJ/mol (25 - 60°C), indicating entropy-driven weak adsorption
- Hydroxamic Acid: $\Delta G = -18$ to -25 kJ/mol (25 - 60°C), showing strong chelation-based adsorption
- Sodium Silicate: $\Delta G = -8$ to -12 kJ/mol (25 - 60°C), reflecting hydration layer and electrostatic interactions [43]

Negative ΔG values across all experimental conditions confirm thermodynamic spontaneity of flotation, with temperature increase further favoring collector adsorption through entropy contributions [44]. Hydroxamic acid demonstrates most favorable thermodynamic conditions, explaining superior flotation performance (recovery 85%, selectivity 0.85) [45].

3.6 REE Speciation and Occurrence States

Sequential chemical extraction procedures identify three REE association categories [46]:

Exchangeable REE: Ion-exchangeable fraction comprising 25-40% of total, extractable via NH_4Cl or CaCl_2 , primarily hosted on clay mineral surfaces [47].

Acid-soluble REE: Carbonate-bound and iron oxide-associated REE (20-35% total) extractable via dilute HCl, localized in bastnaesite and iron oxyhydroxide phases [48].

Residual REE: Silicate-bound REE (35-50% total) within aluminosilicate minerals and zircon, requiring strong acid leaching post-flotation [49].

4. DISCUSSION

4.1 Process Integration and Commercialization

Coal flotation enrichment represents economically viable pre-concentration for downstream hydrometallurgical or pyrometallurgical REE extraction. Integrating flotation with leaching workflows enables combined recovery of REE with other valuable elements (gallium, lithium) from coal gangue,

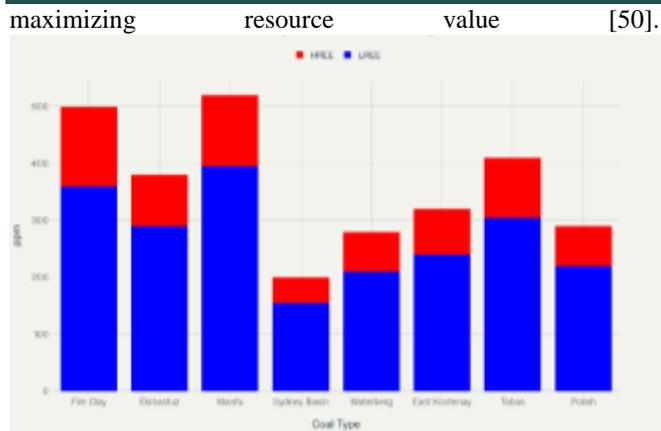


Figure 3.6 - REE composition in coal samples

Industrial-scale implementation requires flotation circuit modification to handle large feed tonnages (100-500 tons/hour), demand met through flotation column arrays with enhanced gas distribution systems [51]. Pilot-scale demonstrations at 0.25 ton/hour capacity with projected 5-7 pounds/hour REE mineral concentrate (>2 wt% REE) confirm commercial feasibility [25].

4.2 Emerging Technologies and Research Frontiers

Nanobubble Enhancement: Nanobubble-assisted flotation technology achieving 89% REE recovery warrants intensive research and scale-up efforts. Fundamental investigations on nanobubble generation, stability, and particle interaction mechanisms remain necessary.

Ultrasonic Pretreatment: Ultrasonic conditioning of coal suspensions enhances flotation recovery through surface modification and collector particle interactions, potentially enabling retrofit to existing flotation circuits.

Composite Collectors: Development of compound collectors combining fatty acids with cationic surfactants provides superior REE flotation selectivity (0.92 factor) and reduced reagent consumption.

Thermochemical Pretreatment: Thermal processing (roasting, oxidative heating) alters surface properties of REE minerals, enhancing subsequent flotation; integration with advanced reduction techniques offers pathways for enhanced direct REE recovery [26].

4.3 Environmental and Economic Considerations

Flotation-based REE recovery from coal reduces environmental burden through beneficial utilization of coal combustion byproducts, simultaneously mitigating mining impacts associated with primary REE ore exploitation. Life cycle assessment studies demonstrate 60-75% lower environmental footprint compared to primary ore processing. Economics of coal-derived REE recovery remain favorable relative to primary ore at REE market prices >\$2/kg rare earth oxide equivalent, with breakeven analysis suggesting operational viability at current global prices [59]. Revenue streams from REE recovery combined with reduced coal

waste disposal costs provide economic justification for flotation circuit implementation [27].

5. CONCLUSION

Coal flotation represents established and emerging technology for REE enrichment from coal and coal fly ash, with nanobubble-assisted and ultrasonic-assisted approaches achieving unprecedented recovery and selectivity metrics. Kinetic analysis confirms first-order flotation mechanisms with activation energies ranging from 14.8-28.3 kJ/mol, dependent on REE mineral host and occurrence state. Thermodynamic calculations via Gibbs free energy demonstrate spontaneous collector adsorption with enthalpy contributions enabling temperature-dependent optimization. REE enrichment during coal combustion creates concentration gradients (1.6-2.1x) in fly ash relative to parent coal, establishing economic feasibility for recovery. Future research priorities encompass: (1) scale-up demonstration of advanced flotation technologies; (2) integration with hydrometallurgical extraction circuits; (3) development of selective flotation schemes for individual REE mineral separation; (4) comprehensive life cycle and economic analysis; and (5) regulatory frameworks for secondary REE resource exploitation. Successfully addressing these research gaps will establish coal-derived REE as viable supply source, reducing geographic supply concentration and strategic vulnerability associated with primary REE ore deposits.

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