

Extraction of alumina from fly ash by pyro-hydro metallurgical routes: A review

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Abstract

Fly ash is a resulting by-product formed due to the burning of pulverized coal on a steam-generating plant. Fly ash mainly contains alumina and silica, hence it can be an excellent secondary source for alumina. Today only 30% of global fly ash can be reused commercially, the remaining was dumped. This causes waste of resources and environmental pollution too. The alumina in the fly ash was in the form mullite phase which is a stable phase so decomposition of this phase is difficult. The mullite phase can be disilicated by calcination, roasting, etc which were pyro-metallurgical routes. Alumina can be extracted as aluminum hydroxide and other unstable forms from fly ash by hydrometallurgical routes. This process requires a huge amount of energy for reaction which is a major drawback of this process. Till now industrial extraction of alumina from fly ash cannot be feasible. Different researchers and scholars did various works to overcome this difficulty. This paper presents a review for extraction of alumina via these routes. It starts with different concepts used in extraction of alumina from fly ash. The middle portion presents the survey of work done by different scholars. The work finally concludes with future possibilities.

Keywords: Fly ash; Mullite; Kaolite; Zeolite; Extraction; Alumina

1. Introduction

Most countries in the world depend on pulverized coal for electricity production. Grounding of coal into fine pieces or powder is known as pulverized coal. The fly-ash is a resulting by-product formed due to pulverized coal combustion in electric and steam-generating plants. According to the chemical composition, in the cement industry, fly ash can be classified into two categories, namely, class C and class F [1]. Class C ash has a low silicate and high calcium content, whereas vice versa for class F ash [1,2]. They were produced by burning of ignite and anthracite, respectively.

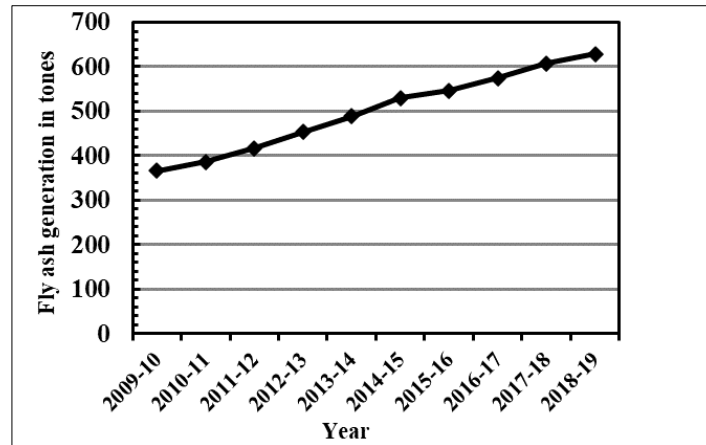
Table 1 Chemical composition of fly ash in %

Compounds	Class F Fly Ash	Class C fly Ash	Portland cement
SiO ₂	56	40	23
Al ₂ O ₃	26	17	4
Fe ₂ O ₃	7	6	2
CaO	9	24	64
MgO	2	5	2
SO ₃	1	3	2

(Source: US Department of Transportation: Federal Highway Administration)

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Silica and alumina were the major components of fly ash, as listed in Table -1. Million tons of fly ash are produced globally, of which only 20%-30% reutilized [3]. The remaining fly ash was dumped, which resulted in the wastage of secondary resource and environmental pollution. In India, 70% of the total power is produced through coal, which produces a huge amount of fly ash [4]. The output of fly ash in India is fractionally expanding, as shown in Figure 1.



(Source: INDIA'S THERMAL POWER ASH MISMANAGEMENT)

Figure 1 Annual generation of fly ash in India

Alumina offers a wide range of applications in various fields. It has high mechanical strength, wear resistance, corrosion resistance, the capability to withstand high temperatures and thermal stresses, high electric insulation, and improved dielectric properties [5]. It can be used in the electronics industry, wear-resistant parts, corrosion-resistant parts, chemical processing, translucent enclosures for highly corrosive sodium vapor (high-pressure sodium vapor lamps), synthetic gems, oil and gas processing, refractories [5]. Advancements in different sectors, such as automobiles, defense, construction, industrial results in an increasing demand for alumina.

Currently, bauxite is the major source for alumina production via Bayer's process. The depository of bauxite in the natural world is finite. The extraction of alumina from secondary sources is necessary to meet future demand. Fly ash can be an excellent secondary source of aluminum. The alumina content in the fly ash is configured as mullite, which is an exceptionally stable phase. To extract aluminum from fly ash, the mullite phase must be transformed into less stable phases. The soda lime sintering method, the acid method, and some salt methods are useful in extracting alumina from fly ash, but they have some disadvantages. The soda lime method has the efficiency of transforming the mullite phase into the kaoline phase. This makes it an efficient method for aluminum extraction. High energy consumption is a major disadvantage of this process. Researchers and scholars have used various methods for the removal of aluminum and its forms from fly ash. Methods such as sintering, roasting and calcination involve changing mullite phase, which belongs to the pyrometallurgical techniques. Techniques such as acid leaching belong to the hydrometallurgical category. Pyrometallurgical routes can be used to change the mullite phase to a less stable state. A hydrometallurgical route was used to dissolve the less stable form of alumina. Hydrometallurgical routes have advantages such as high treatment capacity, low-temperature operation, and high removal capacity of different impurities, whereas pyrometallurgical routes have environmental friendliness and low chemical consumption [7].

In this study, we provide an overview of the different methods used to extract aluminum from fly ash in different forms. It involves only pyrometallurgy and hydrometallurgy techniques. A schematic flow chart of the complete work is shown in Figure 2.

2. Important Terms Related to Fly Ash in Aluminum Extraction

2.1. Pozzolanic Material

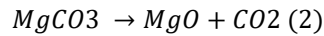
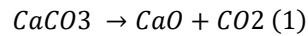
Pozzolanic materials are the class of materials that possess the properties of pozzolana. There are two categories of pozzolana, 1st category is pyroclastic rocks, and the second is siliceous and aluminous materials when mixed with calcium hydroxide and water have cementing properties [8].

2.2. Mullite

Mullite is a solid-phase solution of alumina and silica with superior temperature stability as compared to corundum [9]. Mullite exists in two stoichiometry, namely 3:2 mullite and 2:1 mullite [9]. They have lower thermal expansion coefficients and high creep resistance. A lower thermal expansion coefficient makes the phase change difficult. In fly ash, alumina and silica were present in mullite, which makes it difficult to extract.

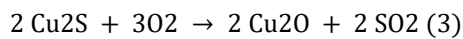
2.3. Calcination

Calcination is used to remove volatile substances from carbonate ore via heating. This helps to convert carbonate ore into oxides [10]. The reaction mechanism of calcination is stated in Equations 1 and 2.



2.4. Roasting

Roasting is a type of heating process where sulfide ore is converted to its oxide ore in the existence of surplus air. The mechanism for the conversion of copper sulfide to copper oxide is presented in Equation- 3.



2.5. Leaching

Leaching is the process of extractive metallurgy where valuable metals are converted to soluble salts whereas impurities are converted to insoluble salts.

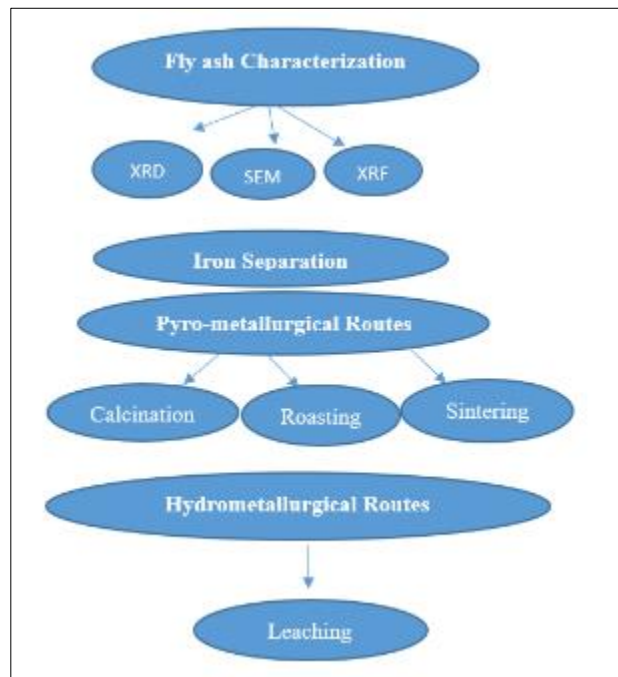


Figure 2 Schematic Flow Chart on the Routes used in Aluminium Extraction

2.6. Iron Separation:

Magnetic separation is an efficient method to separate iron from fly ash. Fly ash contains both di- and paramagnetic materials. High-intensity magnets attract paramagnetic materials (Fe_2O_3) whereas they repel di-magnetic materials. They can efficiently divide the fly ash into 20% and 80% fractions, respectively [13]. The magnetic separator's current, voltage, and magnetic field should be set at 0.2 A, 50 V, and 0.14- 0.16 Tesla, respectively [31]. S. Prakash et al. reported that loading a small amount of sodium oleate in fly ash can effectively change iron recovery via magnetic separation [49].



(Source- Strzałkowska, E. (2021). Morphology, chemical and mineralogical composition of magnetic fraction of coal fly ash. International Journal of Coal Geology, 240. doi.org/10.1016/j.coal.2021.103746) @ 2024 copyright clearance.

Figure 3 Schematic Diagram for Recovery of Iron from Fly-ash By Magnetic Separation

2.7. Pyro-metallurgical Routes:

The word pyro has been derived from the Greek word ‘pura’ means fire. The use of fire helped ancient people extract gold from their ores by melting and casting small particles found in rocks. When the removal of metals from sulfide ore became difficult by smelting, the roasting process was discovered. It can change sulfides into oxides, efficiently recover metals from their ores. In fly ash, aluminium was covered with silica as shown in the figure below. Silica acts as a passivation layer for aluminum extraction. The extraction of aluminum, fly ash should be desilicated at the initial step. It could be done by applying pyro-metallurgical techniques such as calcination, roasting, and sintering. The different techniques used for desilication have been listed in the Table-2.

Table 2 Summary of Desilication of fly ash by Pyrometallurgical Techniques

Desilication Technique	Chemical Used	Desirable Temperature & Time	% of Silica Separated	Reference
Sintering	CaO	1000-1250°C 30 Min	NA	[16]
Calcination	Ammonium Sulfate	600°C 2 hours	NA	[19]
Lime Soda Sintering	Sodium Hydroxide	1200°C	37.21%	[22]
Sintering	Coal + CaO	1100°C 3 hours	NA	[20]
Sintering	Na ₂ CO ₃ + CaCO ₃	1300°C 1 Hour	NA	[13]
Calcination	CaCO ₃ + Soda Ash	1050°C 2 Hours	45.66 %	[26]
Roasting	Ammonium Hydrogen Sulfate	400°C 45 Minutes	90%	[32]
Sintering	Na ₂ CO ₃ + CaCO ₃	1050°C 2 Hours	36.44%	[34]

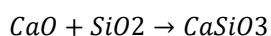
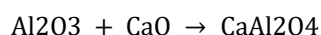
Sintering Leaching	after Coal + CaCO ₃	1150°C 3 Hours	49.24%	[35]
Sintering	Ammonium Sulfate	400°C 3 Hours	NA	[52]
Calcination	Leachant after H ₂ SO ₄ - Leaching	500-1000°C 5 Hours	NA	[39]
Roasting	Charcoal + Fe ₂ O ₃	1250°C 2 Hours	NA	[40]
Roasting	Ammonium Sulfate	400°C 1 Hour	NA	[53]
Calcination	CaCl ₂	900°C 1 Hour	NA	[38]

2.8. Role of Pyro-metallurgy techniques in Aluminium Extraction

The aluminum content of fly ash is in the form of mullite, corundum, and amorphous alumina-silicate glass [34]. The stability of this phase makes it difficult to extract aluminum. Pyrometallurgy techniques can efficiently convert its stable phase into a less stable phase. In most cases, it can be done by heat treatment with sodium or calcium-based compounds. Sodium can convert silica and alumina contents into sodium silicate and sodium aluminate, respectively. Calcium can convert silica and alumina content into calcium silicate and calcium aluminate. Then, it can be dissolved into a requisite chemical to separate aluminum and silicon in different forms. The reaction mechanism is stated below. A summary of the desilication of fly ash is stated in the Table-2.

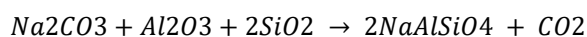
2.9. Lime Sintering

To overcome the issue caused by the stability of aluminosilicate, it can be sintered with CaO. It can efficiently convert alumina in the mullite phase into calcium aluminate and silica into calcium silicate. Calcium aluminate can easily dissolve in different solutions, whereas calcium silicate cannot. This is the key factor for treating it with CaO. The mechanism can be better understood using the equation shown below.



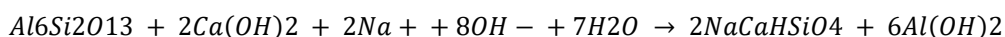
2.10. Soda Sintering

Soda sintering can efficiently convert the alumina-silica glass phase of fly ash into a less stable sodium aluminum silicate phase. After hydrometallurgical processing, alumina and silica can be separated. The mechanism for this process can be better understood using the equation below.



2.11. Calcium Hydroxide Sintering

Fly ash was sintered with CaOH to obtain calcium sodium hydrosilicate. This can be further processed to obtain kaolite and sodalite [50]. These were less stable phases of alumina which can easily be separated by hydrometallurgical routes. Replacing calcium hydroxide with sodium hydroxide can efficiently convert the mullite phase of fly ash into zeolite phase [16].



2.12. Hydrometallurgical Routes

Hydrometallurgy was first invented due to the conversion of some metal ions to gold. The modern hydrometallurgy was born due to the recovery of gold from cyanide treatment and Bayer's process for aluminum production in 1887 [11]. In the twentieth century development in leaching was started. The First World War resulted in the development of the

zinc electrolytic process for brass production and in the course of Second World War development of different leaching processes were established [11]. During the 1960's leaching through bacteria was developed which was known as bioleaching.

Leaching plays an important role in the recovery of aluminum from fly ash. Sulphuric acid leaching was majorly used for the extraction of alumina from fly ash. Direct sulphuric acid leaching was not very efficient in recovering aluminum from fly ash. It can extract only 24% of aluminum even after 6 hours [20]. This is high acid consumption & time consumption process. This may be due to the stable phase of alumina in fly ash. Before going to the hydrometallurgical process its mullite phase must be converted to a less stable phase. The leaching efficiency of fly ash depends upon various factors i.e. solid-liquid ratio, acid concentration, temperature, and time [21]. These are listed in the Table-3.

Table 3 Parameters Used for Leaching

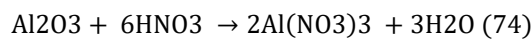
Leaching Type	Pyrometallurgical Technique Used Before Leaching	S/L ratio	Concentration	Temperature	Time	Extraction Efficiency	Reference
Acid Leaching (HCL)	-	0.17	8N	105°C	3 Hours	80%	[16]
Acid Leaching (H ₂ SO ₄)	Sintering with CaO	0.15	2N		15-30 Minutes	96-98%	[16]
Alkali Leaching (Sodium Carbonate + Sodium Hydroxide)	Sintering	0.25	10gpl + 60.78 gpl	85°C	10 Minutes	NA	[22]
Acid Leaching (H ₂ SO ₄)	-	0.5	26N	Room Temperature	24 Hours	Low	[21]
Acid Leaching (H ₂ SO ₄)	Sintering by Pelletizing it with CaO and Coal	0.25	3.06 mol/dm ³	80°C	8 Hours	76%	[20]
Acid Leaching (H ₂ SO ₄)	Sintering by Pelletizing it with CaO and Coal	0.33	6.12 mol/dm ³	80°C	12 Hours	78%	[20]
leaching (NaOH + NaF)	-	0.1	Pure	90°C	2-3 Hours	91%	[41]
Allkali Leaching (NaOH)	-	13.7 Molar ratio	8M in first step & 20M in 2 nd step	85°C & 260°C	150 Minutes & 1 Hour	89%	[23]
Na ₂ CO ₃ Leaching	Sintering	0.33	8%	60°C	40 minutes	60-70%	[28]
Alkali Leaching (Na ₂ CO ₃ + NaOH)	Lime Sintering	0.33	15 gpl	75°C	15 Minutes	96%	[54]
Sodium Hydrogen Sulfate +	-	0.4	15Mol/Lit	220°C	2-5 Hours	87-91%	[33]

H ₂ SO ₄ Leaching (1:1)							
HCL Leaching	-	0.2	345 gpl	200°C	1 Hour	95%	[55]
H ₂ SO ₄ Leaching	Calcination of leachate obtained from 1 st stage leaching	0.2	6M	60°C	4 Hour	94.3%	[39]

2.13. Future Prospective

2.13.1. Nitric acid

Silica can be dissolved by the use of nitric acid [48]. Alumina cannot be attacked by nitric acid. Hence, based on this fact a novel method can be made for the extraction of alumina from fly ash by desilicating it through nitric acid. The aluminum and silicon can dissolve in nitric acid. The major advantage of forming aluminum nitrate is it can dissolve in water.



2.13.2. Potassium Hydrosulfide

Chunbin et al. effectively extracted activating fly ash with potassium bisulfate and potassium pyrosulfate. Potassium hydrosulfate shows similar properties to it, an attempt has never been made to extract aluminum from potassium hydrosulfate.

2.13.3. Gypsum

Gypsum is the natural source of calcium oxide. This is cost efficient as compared to calcium oxide. Dandan Liu et al. reported 90% aluminum extraction from fly ash by using CaO. Using same procedure and replacing calcium oxide with gypsum a trial can be made to extract aluminum.

2.13.4. Carbon

R.H. Matjie et al. reported that the mixing of fly ash with carbon and coal at a specific ratio can enhance its efficiency [20]. Coal is pure form of carbon. Hence, an attempt can be made by mixing graphite or carbon black from different sources instead of coal to check its extraction efficiency

2.13.5. Bioleaching

There are very less no of attempts made to extract aluminum via bioleaching. Bioleaching may be a low energy consuming, cost efficient and environmental friendly way to extract aluminum from fly ash.

3. Conclusion

The following points concluded from this work has been presented below:

- The major issue on the extraction of alumina was the transformation of mullite phase.
- There are large number of processes like soda lime sintering, acid leaching, calcination which were efficient to convert mullite phase to other unstable phases. Large amount of energy requirement makes it cost inefficient. This reason makes it industrially non-profitable.
- The alumina extract of fly ash can be dissolved only up-to a certain limit. After certain limit extraction of alumina from fly ash becomes difficult because alumina content of fly ash was covered with silica particles. Hence, more efficient disilication makes more efficient aluminum extraction.

Credit of Work

Rahul Mandal has written introduction part, constructed the tables and written the abstract of the work. Manaranjan Mohanta, IGNTU, India, withdraw conclusion, and guided the work.

Compliance with ethical standards

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Disclosure of conflict of interest

The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be constructed as a potential conflict of interest

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