

TRANSMUTATION OF COAL FLY ASH TO CONCEIVABLE APPLICATIONS

Anjani RK Gollakota ^{1*}

ABSTRACT

High global energy demands have led to increasing use of coal, which generates Coal Fly Ash (CFA) as a byproduct. CFA is a complex anthropogenic residue and serious threat to the local environment. Improper handling and storage conditions worsen the effects on the environment and nearby species. Current research concerning improved uses for CFA is lacking and developing industrial or commercial uses would improve handling and management of CFA. Thus far, CFA has shown potential as a soil ameliorating agent and catalyst. This paper provides a short review concerning the current research gaps in CFA under-utilization. CFA possesses unique physio-chemical properties that can be directly blended into soil to improve plant growth capacity. Further, all major chemical reactions or product syntheses involve catalysts with silica and alumina as common supports. The Si, Al rich composition in CFA would be a highly suitable alternative if the research were undertaken in depth. Active promotion of CFA's properties by policy makers toward new product streams, alongside conventional utilization, would maximize ecological, industrial and commercial outcomes.

Keywords: Coal fly ash, Soil amelioration, catalysts, environment, policy makers.

1. INTRODUCTION

During the industrial revolution many energy sources were discovered, among which coal became the most utilized. Despite the ubiquity of coal and its versatility, growth in consumption is depleting reserves and causing ecological damage. Presently, around 40% of global energy needs are met by coal. This high degree of utilization to generate energy results in massive quantities of coal fly ash (CFA) production, unfortunately this also poses ecological risks. The recent review of Gollakota *et al.*

(2019) revealed that 750 million tons of CFA were generated in 2015 and this trend is increasing; however, utilization prospects for CFA are less than 25% (*i.e.* < 100 million tons), the remainder being deposited on open land or in lagoons. This improper disposal, handling and exposure to the environment contaminates air and water bodies causing distress to local species. The chemical composition of CFA is quite complex, and the level of threat to the ecosystem increases based on the classification of the coal grade *i.e.*, either class C or class F (see Fig. 1).

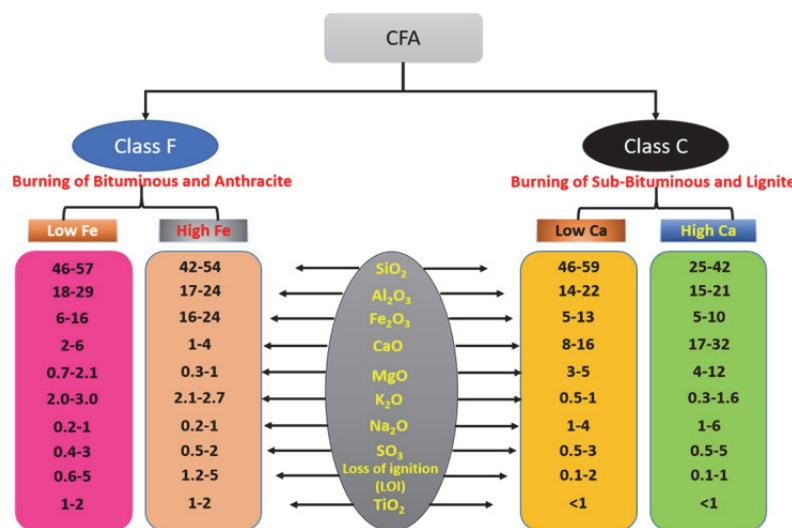


Fig. 1 Schematic of the CFA grading based on the coal origin

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¹ Project Assistant Professor, (corresponding author), Dept. of Safety, health and Environmental Engineering, Douliu, Yunlin County, Taiwan 64002, R.O.C. (e-mail: gollakota.iitg@gmail.com).

There is a present and growing need to find functional applications for CFA. The most common use is the construction industry, followed by production of zeolites for water treatment. Other fields of use are still in relatively premature stages of development. A detailed summary of CFA on the node of pollution control is shown in Table 1.

Despite the negative effects of CFA there are several valuable trace metals that can be found, but extraction remains a challenge (Fig. 2). Over decades of technological progression, and many scientific advancements in CFA utilization, there are some tough challenges such as isolation of radioactive isotopes, especially from Class C CFA. Ignoring these complex possibilities, due to technical limitations, most current research is concentrated on simpler applications of CFA in the construction industry and for zeolite synthesis.

It is well known that CFA plays a vital role in agriculture, for decades it has already been used to treat soil and nutrient deficiencies that would otherwise limit productivity. Also, CFA functions in a majority of the biomass to bio-crude conversion technologies, where complex chemical reactions involve catalysts where Al_2O_3 and SiO_2 are prominently used as support materials. The chemical composition of CFA is rich with alumina and silica, hence this could be a substantial alternative to existing commercial sources. These two possibilities of CFA use, in agriculture and catalysis, could be of significant interest in preserving resources and reducing industry costs. For this reason, this paper gives an overview of the utilization prospects of CFA as a heterogeneous catalyst, and as an inhibitor to soil samples. Further, this manuscript will act as a reference point to future researchers developing new product streams based on CFA.

Table 1 Summary of CFA pollution control

Authors	Purpose	Conditions	Performance indicators
Bhargava <i>et al.</i> (1974)	Removal of detergent from wastewater	2 h contact time, FA concentration (mass%) of 1000 mg/L solution of approximately 23%	-
Singh 2009	Removal of herbicides, such as metribuzin, metalchlor, and atrazine from aqueous solutions	2 h equilibrium time	Removal efficiency of more than 80%, adsorption capacity of FA of 0.56 ~ 3.3 mg/g
Dizgie <i>et al.</i> 2008	Adsorption of reactive dyes remazol brilliant blue and rifacion yellow HED by FA	pH 6.0 Temp: 20°C	Adsorption capacity of FA of 30 mg/g
Dincer <i>et al.</i> 2007	Removal of vertigo blue 49 and orange DNA 13 from wastewater using coal bottom ash	pH up to 7.0 Temp: 25°C	Adsorption capacity of FA of 4.5 ~ 13.4 mg/g
Ahmaruzzaman 2008	Removal of phenolic compounds such as 2,4-dichlorophenol and ortho-chlorophenol	NA	Adsorption capacity of FA of 0.8 ~ 1.7 mg/g
Pengthamkeerati <i>et al.</i> (2008)	Removal of phosphate ions from aqueous solutions	pH 9.0, 25°C	Adsorption capacity of FA of 32 ~ 83 mg $\text{PO}_4^{3-}/\text{g}$
Chaturvedi <i>et al.</i> (1990)	Removal of fluoride from water solutions	pH (2.0 ~ 6.5), high temperature 30 ~ 50°C	Removal efficiency of 94%
Hollis <i>et al.</i> (1986)	Removal of boron	pH 7.0 ~ 12.0	-
Ayala <i>et al.</i> (1998)	Removal of heavy metals (C and Cu) by FA	pH 5.0, high metal retention at higher pH values, grain size < 90 μm	Metal removal of 60% ~ 90%
Cho <i>et al.</i> (2005)	Zn, Pb, Cd, and Cu removal	pH 3 ~ 10, 25°C	Adsorption range of FA of 0.01 ~ 10 mg heavy metal/g
Munoz and Aller (2012)	Lead removal from aqueous solution	pH 5.0, 20°C and 0.5 h retention time	-
Daci <i>et al.</i> (2011)	Removal of heavy metals (Fe, Cu, Mn, Zn, Cd, and Pb)	pH 4.5 ~ 11.0, 20 ~ 30 min of contact time	Sorption rate as high as 99%

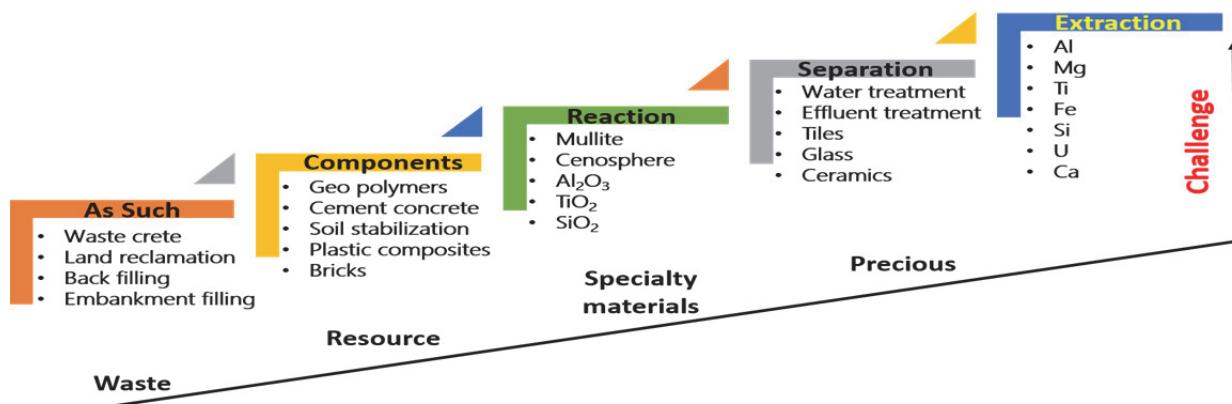


Fig. 2 Challenges remaining the utilization of CFA

2. SOIL AMELIORATION

Although there are existing additives to ameliorate soil such as lime and dolomite, they are not economical, take longer to improve soil health and most importantly they are not ecofriendly. However, the physico-chemical properties of CFA such as the slit and clay sized particles, low bulk density, high water holding capacity, pH and presence of essential plant growing nutrients indicates that CFA could be a strong contender for soil amelioration (Yao *et al.* 2015). The physical properties of the soil such as texture, bulk density and water holding capacity can be substantially raised through use of CFA; however, the extent of change is reliant on the type of soil and the quantity of CFA applied. For instance, Adriano and Weber (2001) observed that the application of CFA did not vary the bulk density, but did improve water holding capacity. Conversely, Pandey and Singh (2010) reported a declining pattern of soil bulk density and the water holding capacity. The reason attributed to the fluctuations in bulk density is the micro porosity and shape of the CFA particles. Furthermore, physical attributes such as hydraulic conductivity vary along the soil type and the quantity of CFA used (Adriano *et al.* 1980).

Chemical properties of soil also acts as a function of the composition of CFA. Variable chemical characteristics alter soil pH and the degree of weathering. For instance, in the case of CFA, excess Sulphur which is acidic in nature, decreases the pH of the soil, and a contrary behavior can be seen in alkaline dominated CFA.

Electrical conductivity is another important property of soil agriculture that is directly related to plant growth. CFA is rich in soluble salts and can thus significantly alter the electrical conductivity of the soil positively, which is undesirable for plant growth (Kim *et al.* 1994). However, not every soil raises its electrical conductivity upon application of CFA, some soils exhibit a declining conductivity due to precipitation of the soluble compounds in CFA that cause the pH to increase (Shaheen *et al.* 2014).

Finally, the microbial properties which are a prime factor in plant growth undergo changes when CFA is applied. CFA acts on microbial properties through several vectors, via pH, salinization, toxicity of boron, and the physical conditions of the soil (Carlson

and Adriano 1993). Pandey and Singh (2010) performed laboratory tests on the application of CFA to test microbial activity and revealed that application of the CFA on sandy soil improved microbial respiration, enzyme activity and N mineralization, contrary to a study by Wong and Wong (1986). The contradiction in data regarding decreasing microbial respiration is ascribed to the potential release of CO₂ and presence of trace elements in the CFA used. A number of salient studies on the application and uses of CFA on soil amelioration are shown in Table 2.

CFA has a substantial effect on physical, chemical and biological properties of CFA. However, the data acquired thus far appears to depend heavily on the nature of the soil and type of CFA used. Much research effort is devoted to delineating physical and chemical property variations, but little is done regarding biological aspects of CFA inhibition on soil.

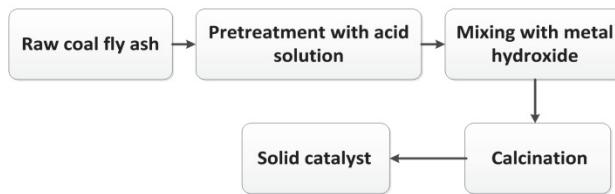
3. CATALYSIS

In terms of real time application, there are no homogeneous catalysts. Heterogeneous catalysts, however, are quite common and vital to industrial and agricultural productivity. Such catalytic activity is due to interaction between the support and active components. Metal oxides such as Al₂O₃, SiO₂, TiO₂, and MgO are the most widely encountered catalytic supports (Yao *et al.* 2015). Research has already shown that CFA is rich in SiO₂ and Al₂O₃, and that it has good thermal stability. This makes CFA a prime contender for use as a catalyst support.

The process of CFA catalyst preparation begins with purification (also referred to as pretreatment) which involves the use of an acidic treatment to improve the surface area and pore volume. Pretreatment is followed by cleaning of the mixture and acid mixing the alkaline agents such as NaOH or KOH to improve the metal oxide content of the catalyst (Gollakota *et al.* 2019). After this, the CFA mixture and metal oxide are subjected to hydrothermal treatment at 700°C for 1 h, which produces a catalyst that can be used for biodiesel synthesis (Go and Yeom 2018). A flowchart of this process is given in Fig. 3.

Table 2 Summary of the literature pertaining the usage of coal fly ash as an inhibitor to soil amelioration

Description	Conditions	Mechanism	Result	Reference
The use of coal combustion fly ash as a soil amendment in agricultural lands (with comments on its potential to improve food security and sequester carbon)			The review presents comprehensive insights about the possibility of using coal fly ash to improve soil fertility and the benefits of preventing carbon sequestration.	Ukwattage <i>et al.</i> (2013)
Phytoextraction of rhenium by lucerne (<i>Medicago sativa</i>) and erect milkvetch (<i>Astragalus adsurgens</i>) from alkaline soils amended with coal fly ash	-	Phytoextraction	Through phytoextraction and using Re as a medium, tested plants were able to from soil to a maximum extent of 40%.	He <i>et al.</i> (2018)
Opportunities and challenges in the use of coal fly ash for soil improvements – A review			This study presents the overall status of coal fly ash disposal practices and problems, focusing particularly on its usage as a soil amelioration medium	Shaheen <i>et al.</i> (2014)
Characteristics of fly ash in relation to soil amendment	-	Homogeneous mixing	Combining fly ash to the soil improves the productivity of agricultural crops due to the presence of K, Na, Zn, Ca, Mg etc	Kumar <i>et al.</i> (2017)
Assessments of Class F fly ash for amelioration of soil acidity and their influence on growth and uptake of Mo and Se by canola	-	Homogeneous blending	Class F fly ash acts as a limiting material in agricultural soils due to its higher alkalinity	Manoharan <i>et al.</i> (2010)
Fly ash for soil amelioration: A review on the influence of ash blending with inorganic and organic amendments			The review elucidates the benefits of coal fly ash in soil amelioration. Upon addition of coal fly ash with organic and inorganic amendments like lime, gypsum, red mud, animal manure will improve the soil quality and higher biomass production	Ram and Masto 2014
Effect of rice husk biochar and coal fly ash on some physical properties of expansive clayey soil (Vertisol)	-	Homogeneous blending	The coal fly ash is able to improve the physical quality and swelling –shrinkage of the soil	Lu <i>et al.</i> (2014)

**Fig. 3 Flowchart showing the CFA to catalyst process**

Other key factors governing the use of CFA as a catalyst are reaction temperature, reaction time, molar ratio, and catalyst loading. The optimal reaction time reveals not only the adsorption efficiency, but helps to refine the processes operational aspects such as cost and reactor design. The second important parameter is the reaction temperature, which controls the chemical reactions. A proper understanding of the reaction temperature not only helps to determine maximum conversion yield, but also assists in controlling the chemical reactions. Catalyst loading and weight/molar ratio are also considered crucial aspects to control when optimizing a catalytic reaction. These factors further help with evaluating the maximum catalytic activity possible under specific conditions. Finally, the kinetic and isotherm studies are required to provide the best insight into the morphological aspects of the CFA catalyst, they also reveal the adsorption capacity of the synthesized catalyst.

4. CONCLUSIONS

This paper has provided an overview of two potential industrial use aspects of CFA, soil amelioration and catalysis.

CFA possesses peculiar characteristics that promote improvement in the physical, chemical and biological qualities of soil. Upon application of CFA, with some organic and inorganic amendments like lime, gypsum, or red mud, plant productive output can be significantly enhanced. CFA primarily influences plant growth through its effect on the water holding capacity and pH of soil. While several studies suggest the potentiality of CFA in soil amendment, a greater understanding of its many and varied facets is still required. For example, determining the alternative materials and blends to optimize amelioration capacity, or scale-up composting technology would strongly improve the current underuse of CFA. Such increased use would also lead to development in the field of N fixation isolation, and potentially some form of policy intervention endorsing CFA usage in agriculture.

Noteworthy progress has also been observed in CFA utilization for heterogeneous catalysis. Owing to its high stability, reusability, low preparation cost, and environmental friendliness, CFA catalysts are efficient and effective when used as a substrate. Again, further study is required to develop this practice and produce novel catalysts that can compete with existing composites. Studies have already been done that assert CAF catalysts could facilitate higher conversion yields and produce unusual and novel precious composites relative to the raw species. That said, the present capacity of CFA utilization does not match the global generation rate. Further technological transformation and industry promotion is required, in conjunction with ecological awareness, to generate a more sustainable approach.

Table 3 Summary of the literature pertaining the usage of coal fly ash as a catalyst support

Description	Conditions	Mechanism	Result	Reference
Fly ash supported calcium oxide as recyclable solid base catalyst for Knoevenagel condensation reaction	900°C, 3 h	Mixing	High conversion rate of 87% achieved with the loading of CaO and coal fly ash based catalyst	Jain <i>et al.</i> (2010)
Transesterification of soybean oil catalysed by fly ash and egg shell derived solid catalysts	1000°C, 2 h	Wet impregnation	Higher conversion yields of about 96.5%, RSM studies were performed to scale up the process	Chakraborty <i>et al.</i> (2010)
Synthesis and characterization of fly ash supported sulfated zirconia catalyst for benzylation reactions	110 ~ 550°C, 2 h	Two-step sol gel technique	Higher conversion of benzene (87%) and toluene (93%) was achieved	Khatri <i>et al.</i> (2010)
Heterogeneous Fenton-like catalytic removal of p-nitrophenol in water using acid-activated fly ash	105°C, 4 h	Mixing	Effective p-NP removal of 98.8% was achieved within 14 h of reaction time	Zhang <i>et al.</i> (2012)
Selective catalytic reduction of NO by ammonia with fly ash catalyst	70°C, 1 h	Molding followed by steam activation	Higher conversion of NO compound of 95% was possible under mild temperatures of 350°C	Xuan <i>et al.</i> (2003)
Application of solid ash based catalysts in heterogeneous catalysis	The author presented a comprehensive review of the utilization of coal fly ash potential as a catalyst in H ₂ production, hydrocarbon oxidation and hydrocracking			Wang 2008
Modified coal fly ash waste as an efficient heterogeneous catalyst for dehydration of xylose to furfural in biphasic medium	400°C, 4 h	Hydrothermal treatment	The catalyst yield is 62% furfural at 170°C in 210 min	Chatterjee <i>et al.</i> (2019)
Biofuel preparation from waste chicken fat using coal fly ash as a catalyst: Optimization and kinetics study in a batch reactor	600°C, 1 h	Hydrothermal treatment	The catalyst obtained the maximum yield of 76.2 wt% biofuel and 24 wt% gasoline	Suchamalawong <i>et al.</i> (2019)

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