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The multiple value characteristics of fly ash from Indian coal thermal power plants: a review

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Abstract Coal-powered thermal plants are the primary source of energy production around the globe. More than half (56.89%) of the Indian power plants use coal for power production. Coal burning in power plants results in coal combustion residuals, which contain coal fly ash (CFA) that is recognized as principle by-product. CFA is difficult to characterize due to its broad compositional variation. Hence, the present article summarizes the various physical, chemical, mineralogical, and petrological characterizations of CFA to its use in different applications. Indian coal thermal power plants are found to release two types of CFA: F (fine) and C (coarse). CFA particles are identified as unburned carbon particles with a large fraction of silica oxides, alumina oxides, and iron oxides with a small fraction of calcium oxide (CaO). Morphologically, CFA particles are spherical, with large carbon molecules and a smooth texture surface. In terms of mineralogy; quartz, mullite, magnetite, and hematite are the dominant mineral phases of CFA and tend to be non-plastic, with permeability levels ranging from 8×10^{-6} to 1.87×10^{-4} cms^{-1}. Petrographically, CFA is enriched in inertinite and liptinites as well as collotelinite, collodetrinite, and vitrodetrinite particles. Moreover, CFA is found to be composed of various organic and inorganic particles. By virtue of multiple characterizations, it has been utilized in several applications for decades, which is still quite limited. Therefore, current study aim to provide helpful insights into the potential use of CFA-derived products in different ways to increase sustainability.

Highlights
- Types of minerals like quartz, mullite, magnetite, and hematite are identified as significant crystalline minerals.
- CFA particles were found enriched in inertinite and liptinites as well as collotelinite, collodetrinite, and vitrodetrinite.
- Based on silicon dioxide (SiO2) and aluminum trioxide (Al2O3), CFA is majorly classified into two classes, namely class F and class C.

Keywords Coal power plants · Coal combustion products · Types of CFA · Characterization · Utilization

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Introduction

Why does coal fly ash (CFA) generation emerge as a significant waste problem for India? This is a question that needs a long time to be answered. A thorough evaluation is still required for a correct assessment of the CFA production from thermal power plants in India. Several pieces of research have been focused on the influence of coal and CFA characterization. This review focuses on the physical, chemical, geotechnical, and petrological characterization of CFA in order to its use in different applications. The multiple characteristics of CFA have been the subject of considerably fewer studies. But before entering into the details of CFA characterization, this becomes important to have an idea regarding the current scenario of coal power production unit, CFA generation, and corresponding utilization.

Coal is the fifth-largest reserve of fossil fuels on the planet and is a prime source of power production. With 54.42% of the overall electricity generated through coal-fired power plants, worldwide India ranks third after China and the USA (Mittal, 2010). The country's total coal power demand has increased from 154,664 to 374,199 MW from 31.03.2007 to 30.11.2020. Based on proximate and ultimate properties and the fuel ratio, coal is characterized as lignite, bituminous, sub-bituminous, and anthracite. Indian thermal power industries currently uses bituminous and sub-bituminous coal, which are extracted mainly from the major coalfields of Korba, Hasdeos-Arrand, Mand-Raigarh, Jharia, Bokaro (West and East), Talcher, Godavari Valley, Raniganj, Wardha Valley, Kamptee from Madhya Pradesh, Bihar, Orissa, Andhra Pradesh, West Bengal, and Maharashtra states in India (Pandey et al., 2011).

Ash particles (fly ash and bottom ash), boiler slag, and flue gas desulfurization materials are coal combustion by-products (CCBs) produced in power plants. The physical and chemical qualities of these CCBs are highly varied since they are determined by the coal supply, moisture, coal burning process, particle size, handling and storage procedures used (Ahmaruzzaman, 2010; Soco & Kalembkiewicz, 2009). Minerals endure thermal degradation, fusion, agglomeration, and disintegration during coal combustion. Many volatile organic fractions and elements exist in the vaporized form. Approximately half of the elements in the boiler turn into bottom ash or slag, while the remaining inorganic materials are released as flue gas, vapors, or CFA. A dominant part of the total CFA is typically obtained by electrostatic precipitation or mechanical filtration, particularly in pulverous-fuel combustion systems.

CFA production is substantial, particularly in India, where it relies heavily on thermal power plants to supply its energy needs. Approximately 200 million tonnes of CFA are produced yearly by the country's coal-fired power plants. It is anticipated that by 2022, it will be around 300 million tonnes. India's Ministry of Power expects to use 1800 million tonnes of coal per year and produce 600 million tonnes of CFA by 2031–2032 (Energy statistics India, 2021; Lal et al., 2015). In addition, the poor quality of coal utilized in Indian coal power plants is a cause of high ash yield, around 35–45% (Mathur et al., 2003). Landfill and ash ponds (dykes) are the two methods followed to dispose of CFA. For landfill disposal, the large land requirement is a big challenging issue for the country. In India, CFA occupies an area of 65,000 acres of land for its disposal, which is expected to increase in the near future and is a major cause of concern (Haldive & Kambekar, 2013). In many areas of the country, the unscientific disposal of CFA has been noticed. This results in adverse effects on local ecosystems through erosion and the production of leachate, and mobilization of dangerous metals in soil and groundwater, posing public health risks (Pandey & Singh, 2010). Numeral studies have reported the groundwater, surface water, and soil pollution around the coal thermal power plant due to fly ash (Mor et al., 2022; Vig et al., 2022a, b; Singh et al., 2020). Keeping this in view, the Ministry of Environment, Forest, and Climate Change has issued various notifications aimed at reducing the environmental impact of fly ash by its scientific disposal and 100% utilization to reduce the land requirements and many other environmental issues (MOEF, 1999).

Figure 1 shows the data for country, viz. distribution of CFA generation and utilization. Denmark, Italy, and Netherlands are the countries involved in 100% utilization of coal fly ash, followed by Japan 96.3%, France 85%, and Canada 75%. India is showing only 38% utilization. According to recent data of Central Electricity Authority of India, CFA's utilization rate has increased from 38 to 78% and the remaining is still disposed of in ash ponds and landfills (CEA, 2020) (Fig. 2).

Hence, it is crucial to determine various strategies for maximizing CFA utilization in order to lessen the load on the environment. In terms of environmental pressure, the reuse techniques for CFA can only reduce the pressure of excessive production (Wang

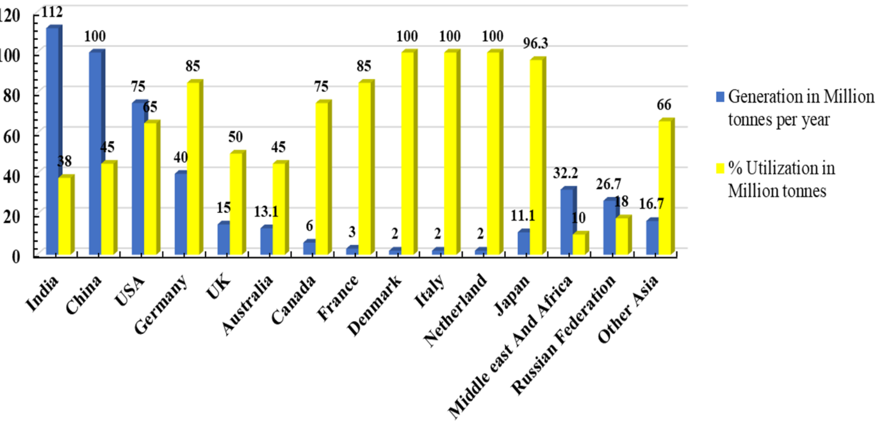

Fig. 1 Presenting the coal fly ash generation (million tonnes) and % utilization (million tonnes) of different countries

et al., 2020a, b). Due to the complexity of CFA's properties, research on the reuse of CFA has lagged behind significantly. Although CFA has been used for decades in the construction industry and various other sectors, its current utilization percentage is still fairly low. CFA was formerly seen as annoying waste that needed safe disposal.

However, its utilization in building and construction material have offered safe disposal options. Because of this, this study is crucial while emphasizing 100% utilization of CFA to reduce the burden of land and disposal to highlight various ways of employing CFA with its full potential in the production of ceramics, cement, and wastewater treatment (Panday et al., 1985; Diamadopoulos et al., 1993; CEA, 2021). The study discussed how Indian CFA is classified, and its properties and different applications using the literature survey approach so that it can be applied in more domains to reduce environmental burden.

CFA classification

CFA is classified according to its composition and multiple characteristics. Based on silica, CFA is categorized into 2 classes, class F (fine), which has low lime (<8%) content, and class C (coarse), which has an extensive lime content (>8%). SiO_2 is the most abundant mineral in this type, and Na_2O is the least abundant mineral present in class F fly ash.

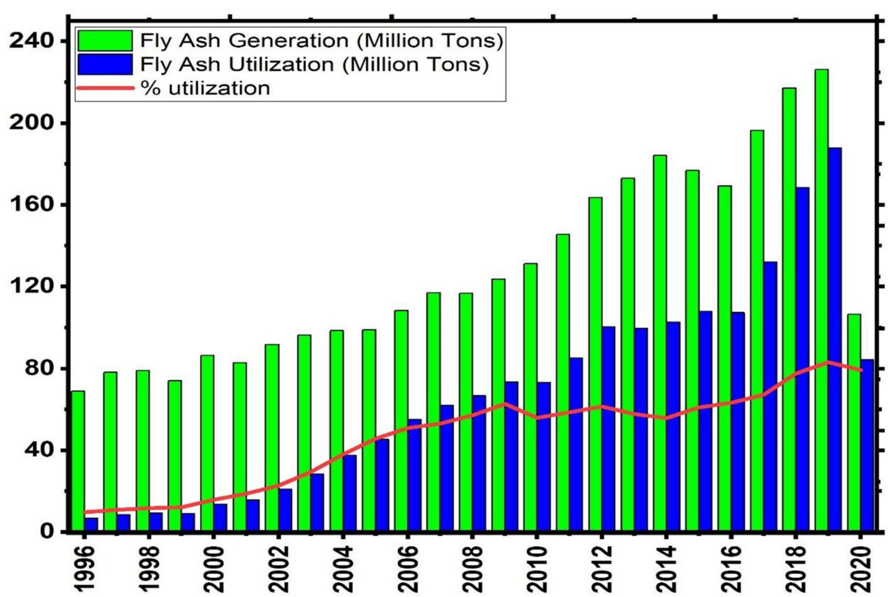

Fig. 2 Growth in the fly ash production and utilization from Indian coal thermal power plants across the country

Additionally, class F is a pozzolanic substance composed of siliceous and aluminous products and has no natural cement-containing properties. On the other hand, class C fly ash contains iron (Fe), SiO_2, and Al_2O_3. Class F fly ash is generated by the power stations that ignite bituminous or anthracite types of coal, whereas class C fly ash is produced by the plants that ignite sub-bituminous types of coal (Wang et al., 2020a, b). In this type, a fraction is not more than 50% after sub-bituminous and lignite coal combustion as per the American Society for Testing Materials Standard (ASTM C618, 2003). In most Indian power plants, F type of fly ash is emitted. In addition to this, there is another class of fly ash, according to ASTMC618; it is class N, which depicts the mineral admixtures that are natural pozzolans like diatomaceous earth, shales and opaline cherts, pumicites, uncalcined, calcined, and several other minerals (Bhatt et al., 2019). Table 1 shows the types of CFA emitted from Indian coal power plants.

CFA characterization

Physico-chemical properties

The knowledge of the general aspects of CFA particles is important as it is a fundamental nature of fly ash. It is categorized by its physicochemical composition, the type of coal utilized in thermal power plants, the technique for coal combustion used, and combustion temperature regulation at the time of power production (Mukherjee et al., 2008). In several studies, CFA has been reported to be fine particles with sizes varying from 1 to 100 µm and an average of < 20 µm. The CFA's bulk density and surface area are found to vary from 0.54 to 0.86 g/cm^3 and 200 to 700m^2/kg, respectively (Kosmatka et al., 2002). Indian CFA has a pH value ranging between 1.2 and 12.5 (Li et al., 2018). In accordance with the pH, ash is classified into 3 groups, i.e., acidic, mildly alkaline, and alkaline, with pH values of 1–7, 8–10, and 11–15, respectively. Moreover, pH is found to vary with the ratio of Ca/S (Kolbe et al., 2011).

The specific gravity of CFA relies on the level of coal pulverization, the shape of its particles, and the type of coal. Iron and calcium contents of CFA are essential in determining its specific gravity. Prior studies have reported that the specific gravity of Indian thermal power plants tends to range between 1.86 and 2.83 (Pandian & Krishna, 2002; Reddy et al., 2018). The variation in the specific gravity of CFA is affected by particle gradation, shape, and chemical composition (Pandian et al., 1998). According to studies, there is an inverse relationship between CFA surface area and total adsorption capacity and particle size is also inversely related to surface area. Generally, the CFA particles have a large surface area due to their non-plastic nature and low cation exchange capacity, which depicts lesser water absorption (Ram & Masto, 2014; Yousuf et al., 2020). In some studies, the surface area of CFA derived from the power plants has been analyzed; for example, the specific surface areas of Neyveli and Muddanur thermal power plants are 9.6 and 8.2m^2/g, respectively (Schure et al., 1985). Singrauli as well as Vindhyachal thermal power plants are revealed to be 4450 cm^2/g and 4590 cm^2/g. Moreover, the CFA has an extremely low moisture content (0.11%) and an extremely high yield of ash (> 90%) (Saikia et al., 2021).

In general, CFA consists of fine-grained materials and falls into the silt category of 1 to 100 µm and typically < 75 µm (Cao et al., 2008; Liu, 2009; Ahmaruzzaman, 2010). ASTM B822-0 with laser diffraction is a valuable method to determine particle size. Few studies reported the silt and clay size fraction of Indian coal ash with particle sizes between

Table 1 List of Indian coal thermal power plants with the type of fly ash generated

Plant, city, or state	Fly ash class
Neyveli	C
Badarpur	F
Korba	F
Ramagundam	F
Vijayawada	F
Dadri, New Delhi	F
Rajghat, New Delhi	F
Orissa	F
Captive Power Plant, Rourkela	F
Kolaghat Thermal Power Station West Bengal	F
Gulbarga Karnataka	F
Neyveli, Tamilnadu	F
Vijayawada, AP	F
Assam	F
Orissa	F

Source: Bhatt et al. (2019)

1 and 100 μm (Moreno et al., 2005, Vassilev & Vassileva, 2007; Hower et al., 1996; Kim & Prezzi, 2008; Diaz et al., 2010; Liu et al., 2004; Qi & Yuan, 2011; Mishra & Das, 2010; Dutta et al., 2009; Reddy et al., 2018; Pandian, 2004). Compared to cement particles, CFA particles are larger in size. Rawat and Yadav (2020) identified CFA particles in the range of 3150 to 4580 nm by dynamic light scattering analysis. Understanding the grain size fraction can provide estimates of engineering properties of the materials like strength, permeability, and expansivity which can be applied to cement industries or beyond (Das et al., 2006; Duncan & Thompson, 1992; Sarkar et al., 2006). Coefficient of uniformity (C_u) and curvature (C_c) and mathematical (D10; D50; D90) expression can be used for estimation of the CFA size. The greater the C_u value, the larger the size fraction. If the values are noted as $C_u \geq 4$ and $C_c < 3$, then CFA is categorized as sand and gravels with "well" grade. If C_u is ≥ 6, and $C_c \leq 3$, CFA is categorized as soil particles with "poor" grade. Numerous Indian thermal power plants classified CFA under poor and well categories (Pandian, 2004; Kaniraj & Gayathri, 2003; Mishra & Dash, 2010; Mohanty, 2012; Ghosh & Subbarao, 1998; Prashanth et al., 2001; Goswami & Mahanta, 2007; Singh et al., 2008; Ram & Masto, 2014).

CFA with a well-consolidated structure has a hydraulic conductivity between 10^{-4} and 10^{-6} cms^{-1}, which is essentially identical to the soil permeability of silty sand to silty clay (Kumar et al., 2019; Reddy et al., 2018). The CFA's permeability is determined by grain size, compaction level, and pozzolanic activity (Pandian, 2004; Prashanth et al., 2001) and reported between 8×10^{-6} to 1.87×10^{-4} cms^{-1} for Badarpur thermal power station (Sahu et al., 2009; Sahu & Gaythri, 2014).

Geotechnical properties

Table 2 presents the geotechnical properties of CFA. The geotechnical properties of CFA are affected by the presence of loss of ignition (LOI), iron content (Fe_2O_3), lime content (CaO), mineralogy and morphology of CFA. LOI value is considered a preliminary primary factor that gives an idea regarding carbonaceous material existing in CFA. Scanning and transmission electron microscope provides the morphological details, energy-dispersive X-rays provide elemental composition, and X-ray dispersion gives the presence of mineralogical nature of CFA particles.

Morphologically, coal combustion leads to spherical, cenospherical, irregular, glassy agglomerates, smooth texture surface, and empty sphere shape of particles (Rawat & Yadav, 2020; Scaccia et al., 2019; Reddy et al., 2018; Zhao et al., 2018). According to researchers, particles of CFA are classified into 11 morphological types (Fisher et al., 1978). Char is considered to be one of the carbonaceous particles present in the CFA, which can be identified as irregular, hollow, and solid spherical particles. These are formed after the last combustion and remain as micrometric to super-micrometric particles of CFA and likewise bottom ash. Almost all coal-fired power stations produce CFA that contains some unburnt char. These are also considered high carbon contents in CFA (Bailey et al., 1990; Valentim et al., 2006). Cenospheres, ferrospheres, and plerospheres are the most important and value-added particles (Torrey et al., 1978; Tripathy & Juengst, 1997; Fomenko et al., 1998; Roy et al., 1984; Anshits et al., 2000; Sokol et al., 2002; Blanco et al., 2000). Morphological variations in the structure of CFA are noted due to the coal operation at different temperatures which creates different compositions (Bruno et al., 2009). It is also interesting to note that CFA particles are generally unburnt carbon content which can be distinguished by their color, which varies from orange to dark red, white to yellow, and brown (Fisher et al., 1978; Ahmaruzzaman, 2010). Moreover, the disposal methods may also contribute to the morphology variations. The above-described CFA's properties make it suitable for multiple utilizations such as construction, landfilling, roadway, railway embankments, road bases, sub-bases, liners, and covers (Pal & Gosh, 2010).

Mineralogical properties

Figure 3 presented the identified minerals from some Indian coal thermal power stations. X-Ray Diffraction (XRD) is the technique to determine the mineralogical properties of CFA. The XRD peaks are identified at different angles. During XRD analysis, the variations in the peaks can be noticed due to the various types of coal utilized, the process of coal combustion and the storage of either precipitator or ash ponds. Factors mentioned above may result in the weakening or removal of peak intensity, changes in the width of the peak, and the formation of new crystals, which has been described in the previous studies (Ibanez et al., 2013a, b; Li et al., 2016; Tennakoon et al., 2015). Some previous studies reported that CFA

Table 2 Geotechnical characterization of fly ash of Indian thermal power plant

Thermal power plant	Silt fraction (%)	Clay fraction (%)	Coefficient of uniformity (Cu)	Coefficient of curvature (Cc)	Permeability tests (cm/s)	Liquid limit (%)	Plastic limit	Plasticity index (%)	Specific gravity	Surface area (m^2/g)
Badarpur, 2004	–	–	5.5	2.47	–	–	–	–	–	–
Captive & Rourkela, 2012	–	–	5.88	1.55	–	51.5	–	–	–	–
Dadri, New Delhi, 2003	–	–	5.65	0.9	–	–	–	–	–	–
Gulbarga, 2001	–	–	2.14	0.95	–	62	–	–	–	–
Kakatiya, 2018	71.86	0	3.6	1.87	$1.01E^{-4}$	–	–		1.86	–
Kolaghat, 1998	–	–	5.44	3.12	–	–	–	–	–	–
Korba, 2004	–	–	6	1.14	–	–	–	–	–	–
Muddanur, 2013	94.5	5.5	–	–	–	29	Non-plastic	Non-plastic	2.17	8.2
Neyvelli, 2013	87	13	–	–	–	39	Non-plastic	Non-plastic	2.83	9.6
Orissa, 2010	–	–	4.02	0.94	–	–	–	–	–	–
Orissa, 2010	–	–	3.96	0.93	–	–	–	–	–	–
Orissa, 2010	–	–	4	0.91	–	–	–	–	–	–
Panipat, 2007	–	–	–	–	–	43	–	–	–	–
Rajghat, 2003	–	–	4.82	1.01	–	–	–	–	–	–
Ramagundam, 2004	–	–	1.59	1.09	–	–	–	–	–	–
Singrauli, 2019	75.41	2.92	4.75	1.85	10^{-4} to 10^{-6}	31.47	Non-plastic	Non-plastic	2.13	0.445
Vijayawada, 2001	–	–	5.7	0.61	–	–	–	–	–	–
Vijayawada, 2001	–	–	3.67	0.76	–	49	–	–	–	–
Vindhyachal, 2019	73.98	2.37	4.37	1.78	10^{-4} to 10^{-6}	36.5	Non-plastic	Non-plastic	2.19	0.459

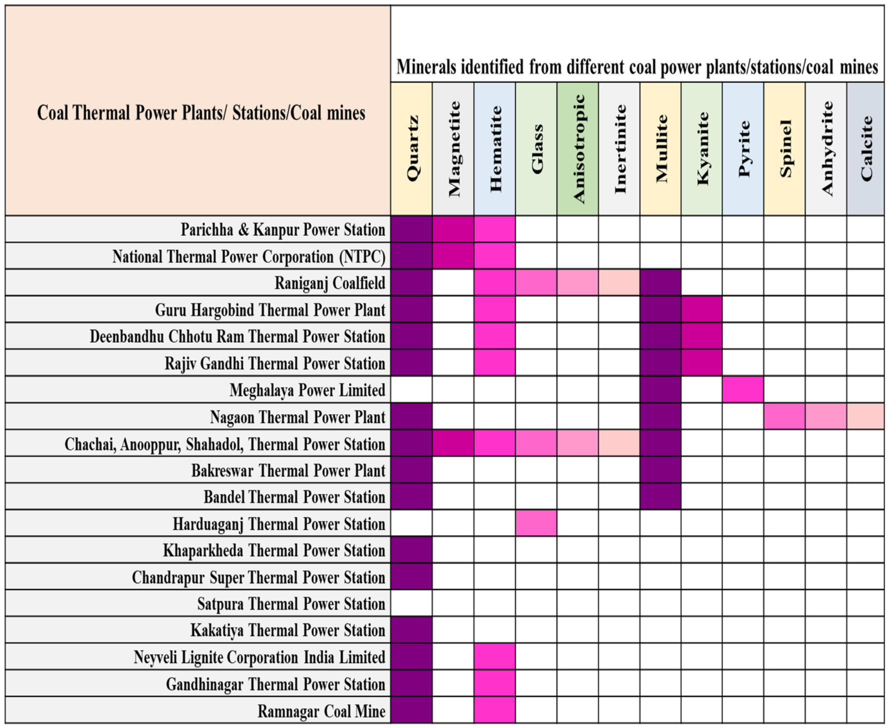

Fig. 3 Mineralogical assessment of Indian coal thermal power plants

particles with a size > 75 m are enriched in high calcium content, whereas CFA particles with a size < 45 m are enriched in quartz, hematite, and magnetite (Das, 2006). For example, lignite CFA is dominated by quartz, hematite, anorthite, gehlenite, and hematite minerals (Ilic et al., 2003; Bayat, 1998; Sakorafa et al., 1996). The number of minerals identified in different CFAs ranges from 316 to 188 (Vassilev & Vassileva, 2005). Studies also revealed that glass, mullite ($3Al_2O_3{\cdot}2SiO_2$), cristobalite, quartz (SiO_2), magnetite (Fe_3O_4), kyanite, pyrite, spinal, mullite, and hematite (Fe_2O_3) are the predominant crystalline phases in CFA. On the other hand, anhydrite-gypsum ($CaSO_4$–$CaSO_4{\cdot}2H_2O$), calcite-ankerite, lime-portlandite (CaO–Ca $(OH)_2$, feldspar ($KAlSi_3O_8$–$NaAlSi_3O_8$–$CaAl_2Si_2O_8$), corundum, and several Ca–Mg silicates are the less predominant minerals of CFA (Vassilev & Vassileva, 2007). The mineralogical composition of CFA is determined by geological characteristics such as coal deposition types, and the inorganic composition of the feed coal, in addition to combustion technology and power plant operational mechanisms (Gupta et al., 1998; Tomeczek & Palugnoik, 2002).

Similarly, the presence of minerals also differed from the level of calcium substance of the CFA. Low calcium content in CFA indicates the presence of quartz and mullite, while high calcium content indicates quartzite and a substantial amount of calcium such as C_3A C_4A_3S, Cs, CaO, and C_3AS (Sarkar et al., 2006). Several studies have worked on the mineralogical assessment of CFA throughout the country (Ibanez et al., 2013a, b; Li et al., 2016; Tennakoon et al., 2015). Quartz, magnetite, and hematite minerals have been reported in Parichha and Panki thermal power plants, Kanpur and National Thermal Power Corporation (NTPC), Kaniha Orissa (Sivalingam & Sen, 2018). Quartz, hematite, glass, anisotrop, inertinite, and mullite have been reported in the Raniganj coalfield (Sivalingam & Sen, 2018). Quartz, hematite, mullite and kyanite have been reported in the Guru Hargobind TPP, Lehra Mohabbat, Punjab; Deenbandhu Chhotu Ram TPP, Haryana; and Rajiv Gandhi TPP, Yamunanagar, Hisar, Haryana (Kumar et al., 2016). Mullite and pyrite minerals have been reported in Meghalaya Power Limited (Oliveira et al., 2014). Quartz, mullite, spinal, anhydrite, and calcite have been reported in Nagaon thermal power plants (Saikia et al., 2015; Ward et al., 2015). Quartz, magnetite, hematite, glass, anisotrop, inertinite, and mullite have been reported in thermal power stations of Chachai Anooppur, Shahasol, Madhya Pradesh (Singh & Pragya, 2015). Quartz and mullite have been reported in Bakreswar thermal power plant, Bandel power plant, and Kakatiya thermal power (Reddy et al., 2018). Only glass minerals were identified in the Harduaganj thermal power plant (Dwivedi et al., 2012). Quartz was identified in the Khaperkheda, Chandrapur, and Satpura power plants and Kakatiya thermal power plant (Gedam et al., 2013; Reddy et al., 2018). Quartz and hematite minerals have been identified in Neyveli lignite corporation, Tamilnadu. Angular particles of CFA samples have been noticed with the highest fraction of quartz, mullite, and hematite derived from the Gandhinagar coal thermal power plant (Rawat & Yadav, 2020). Quartz and hematite are the predominant minerals identified in the Ramnagar coal mine, West Bengal, India (Saikia et al., 2021).

Petrological properties

The International Committee for Coal and Organic Petrology uses protocols to analyze the petrography of coal and CFA. According to studied literature, India has a much lower number of studies on the petrological examination of CCBs. For petrological investigation, the coal pellets are prepared from epoxy-bound particulates to a thin coating of 0.05-lm-alumina. Leitz Orthoplan microscope with a 50×reflected-light oil immersion is used for the observation (Hower et al., 2017). Previous studies on petrological analysis revealed glass, vitrified rock fragments, and quartz anisotropic coal minerals at high temperatures. Isotropic, inertinite, and anisotropic coke particles have been noticed during the incomplete combustion of bituminous coals (Saikia et al., 2015, 2021). Photomicrographs of coal fragments inclusive of the mesosphere, fusinoid char and partly fused inertinite, iso-inertoid char with tunnel-type pores, and primary and secondary vesicles have been reported by Verma et al. (2014). Glassy rims, iron mineral isotropic, and quartz, anisotropic, inertinitic, and total char particles have also been observed by Shreya et al. (2015) and Valentim et al. (2016) in CFA obtained from, Jharia, Jharkhand power plants, Bokaro thermal power station, Jharkhand, and Damodar Valley Corporation respectively.

Furthermore, CFA particles have been noticed as glass matrix with spinel, sulfide, glass mullite, quartz, spinel, isotropic, sulfate rock fragments, inertinite, anisotropic, and pet coke particles (Saikia et al., 2015). Delineate, total telovitrinite, collotelinite, collodetrinite, vitrodetrinite, total detrovitrinite, gelinite, corpogelinite, total vitrinite, total gelovitrinite, fusinite semifusinite, macrinite micrinite, and silicate sulfide carbonates have all been reported by Saikia et al. (2015). Inertinite, liptinites collotelinite, collodetrinite, and vitrodetrinite have been found to be abundant in coal and specify the types of fly-ash carbons. These are the principle vitrinite macerals, whereas fusinite and semi-fusinite constitute the major proportion of inertinites that can be observed in samples of Sattupalli coal mines of Andhra Pradesh (Singh et al., 2012). CFA samples have also got reported the presence of vitrinite, liptinite, and inertinite derived from Bokaro and Chandrapura power plants (Valentim et al., 2016). The carbon particles of CFA are exhibited as anisotropic, isotropic, and inertinite coke particles (Saikia et al., 2021).

Atterberg's limits

The plastic limit (PL), the liquid limit (LL), and the distinction among them (plasticity index (PI)) can be applied to investigate volumetric variations in CFA potential. ASTM D4318 is a widely used Atterberg test in favor of evaluating the PL, LL, and PI of CFA. These characteristics are important in classifying and identifying materials, as well as predicting technical properties, including strength, hydraulic conductivity, and compressibility. The liquid limit values of Indian thermal power plants have been observed between 29 and 62% (Moghal, 2013). No plastic limit % for different power plants has been found for the majority of the ashes, which is due to the non-plastic nature of CFA.

Composition of CFA

Minerals and oxides

According to the chemical characterization of CFA, worldwide, CFA is identified as a mixture of amorphous and ferro-aluminosilicate minerals. India has the highest mass % of SiO_2 in the form of amorphous and crystalline (50.2–59.7), followed by Australia (48.8–66.0), the USA (37.8–58.5), China (35.6–57.2), and Europe (28.5–59.7). The maximum range of aluminum was reported in the USA (19.1–28.6) and the minimum in Europe (12.5–35.6) (Diaz et al., 2010; Dutta et al., 2009; Hower et al., 1996; Kim & Prezzi, 2008; Liu et al., 2004; Mishra & Das, 2010; Moreno et al., 2005; Qi & Yuan, 2011; Vassilev & Vassileva, 2007). As per previously conducted studies, Indian CFA is also found to be composed of various oxides of aluminum (Al), calcium (Ca), iron (Fe), magnesium (Mg), potassium (K), and phosphorous (P) (Rawat & Yadav, 2020; Sivalingam & Sen, 2018; Hower et al., 2015; Tiwari et al., 2014; Patil & Anandhan, 2012). The order of oxides in CFA has been reported as $SiO_2 > Al_2O_3 > Fe_2O_3 > CaO > MgO > K_2O > Na_2O > TiO_2$ (Blisset & Rowson, 2012) (Fig. 4).

Organics and inorganics

Additionally, CFA samples have been reported with various organic and inorganic elements. In organics, CFA is composed of low carbon and hydrogen content (<7%, <0.2%) (Saikia et al., 2021). Organic CFA is also composed of PAHs such as naphthalene, acenapthalene, fluorene, phenanthrene, pyrene, anthracene, fluoranthene, benzo(b)fluoranthene, benzo(a)anthracene, perylene, benzo(k)fluoranthene, benzo(a)pyrene, benzo(ghi)perylene, dibenzo(ah) anthracene, and polychlorinated biphenyl congeners including PCB (18, 28, 44, 52, 77, 101, 126, 138, 153, 169, 180, 194) (Sahu et al., 2009).

CFA is also found to be composed of some inorganic components inclusive of various trace and heavy elements such as arsenic (As), boron (B), beryllium (Be), cobalt (Co), cadmium (Cd), chromium (Cr), lithium (Li), copper (Cu), molybdenum (Mo), lead (Pb), manganese (Mn), antimony (Sb), nickel (Ni), titanium (Ti), and zinc (Zn) (Rani & Jain, 2015; Sarkar et al., 2006; Wang et al., 2020a, b). It consists of the highest fraction of Si, Al, Fe, Ca, Na, K, P, and S (Depoi et al., 2008). Most of the elemental concentration has been observed at the surface of the CFA (Markowski & Filby, 1985). The elemental concentration of CFA depends on the elements that exist in coal and is divided into three general groups: (i) major elements with a high concentration of more than 1000 parts per million (ppm) of carbon (C), hydrogen (H), nitrogen (N), oxygen (O), and sulfur (S); (ii) minor elements, primarily coal-mineral materials such as aluminum (Al), iron (Fe), silicon (Si), magnesium (Mg), calcium (Ca), potassium (K), sodium (Na), manganese (Mn), and titanium (Ti) with concentrations more than 100 and less than 1000 ppm; and (iii) trace elements, with concentration less than 100 ppm (Vejahati et al., 2010). Moreover, the elemental composition is also influenced by the major mines from where the coal is imported, like Raniganj (West Bengal), Giridhih, (Jharkhand), East Bokaro and West Bokaro (Jharkhand), Singrauli (Madhya Pradesh and Utter Pradesh), Korba (Chhatisgarh), Wardha Valley (Maharashtra), and Talchar (Orrisa). On the other hand, the high sulfur content in coal also affects the composition of CFA (Goodarzi & Sanei, 2009). The corresponding major elemental concentrations in the coal of different mines are presented in Table 3.

This has also been noted that CFA particles have been found with numerous valuable constituents like Al, Si, Fe, B, Ca, Cu, Mg, Mn, P, S, and Zn, besides the suitable level of hazardous elements like As, Ba, Cr, Hg, Pb, Ni, and V (Mashau et al., 2018). The most dangerous heavy metals in fly ash can be identified as Hg, As, Cr, Cd, and Pb and they appear as flue gas constituents (Zhao et al., 2018; Liu et al., 2020; Singh et al., 2012). These elements originate from improper disposal of CFA (Chaudhary & Banerjee, 2007). Many studies have determined the concentration of trace and heavy metals in CFA using the energy-dispersive X-rays, inductively coupled plasma-optical emission spectrometry, and inductively coupled plasma mass spectrometry and atomic absorption spectroscopy. One of the interesting studies conducted by Bhangare et al. (2011) reported different elements in CFA samples in the order Fe > Mn > Zn > Cu > Cr > Ni > Mg > Pb > Li > Co > Hg > Cd > As. Singh et al. (2020) identified As, Ba, Cr, Cd, Hg, Pb, and Mn in NTPC thermal power plants located in Utter Pradesh and Madhya Pradesh. As, Ba, Cr, Cd, Hg, Mn, and Pb in the CFA samples have been reported in the range of 5.15–25.74 mg/kg, 777.05–970.70 mg/kg, 43.25–64.61 mg/kg, 0.56–0.56 mg/kg, 0.17–1.26 mg/kg, 163.83–831.47 mg/kg, and 28.94–119.57 mg/kg individually.

Radionuclides

The coal used in CFPP has trace levels of the uranium (U), thorium (Th), and actinium (Ac) series elements; when the coal is burned, some of these radionuclides are released into the atmosphere. Few important radionuclides in CFA are 226 Ra, 228Ac, 40 K, 238U, 226Ra, 228Ra, 230Th, 210Pb, 210Po, 238Th,

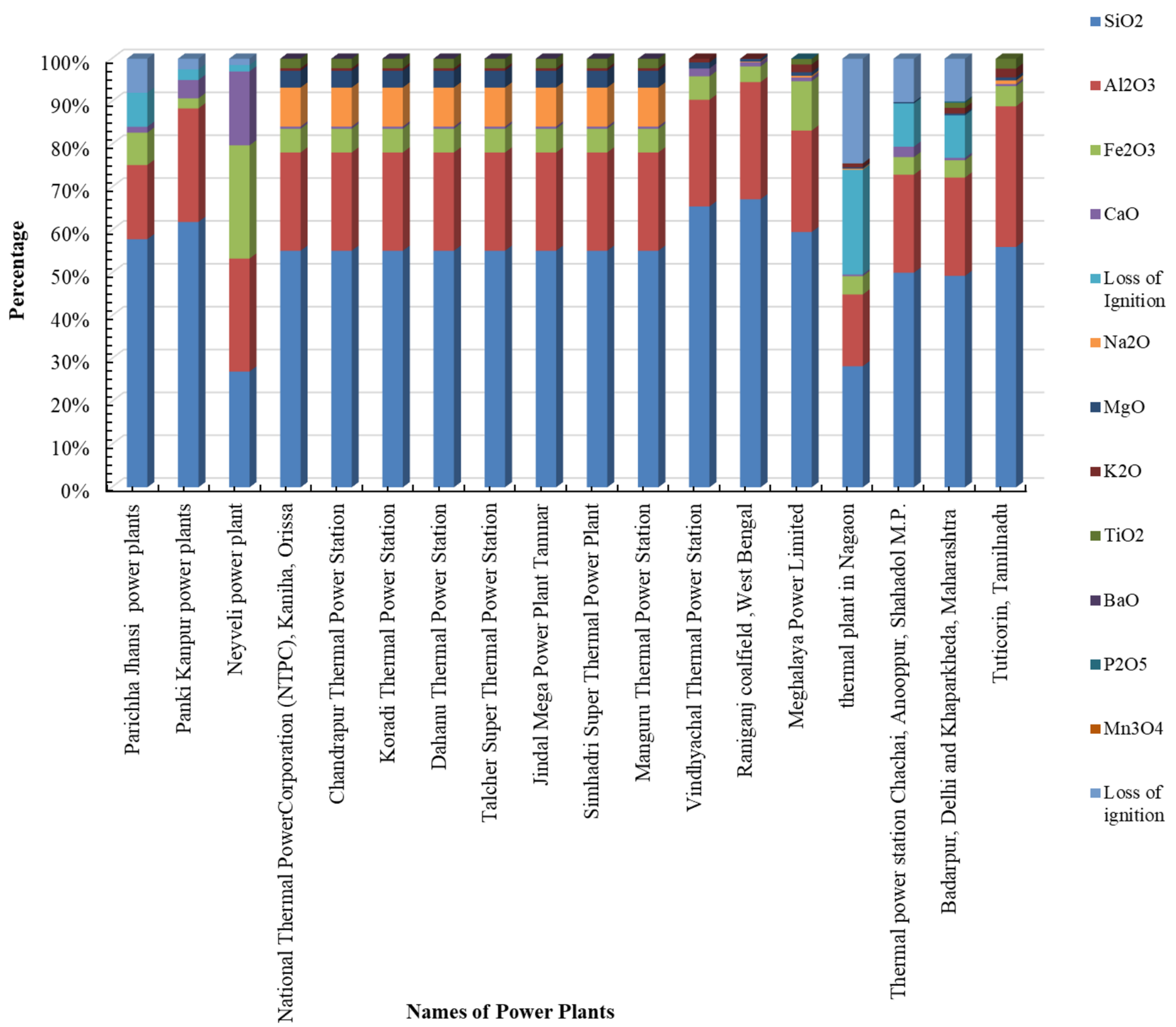

Fig. 4 Components identified in coal fly ash of different Indian coal thermal power plants from previous literature

Table 3 Presenting the quality of Indian coal by weight percentage (%)

Coal field	Carbon	Hydrogen	Sulfur	Nitrogen	Oxygen	Ash fraction	Moisture content	CV (kcal/kg)
Raniganj West Bengal	60.2	4.2	0.3	1.8	7.1	22.9	3.5	4280
Giridhih, Jharkhand	50.8	3.8	0.5	1.3	5.9	34	3.7	4058
East Bokaro, Jharkhand	61	4.1	0.49	1.53	7.2	21	4.68	4300
West Bokaro, Jharkhand	52.3	3.3	0.41	1.23	4.9	36	1.86	4098
Singrauli, MP and UP	39.27	2.8	0.55	0.92	9.18	39	8.28	3850
Korba, Chhatisgarh	42.93	2.8	0.4	1.03	8.5	38	6.3	3997
Wardha Valley, Maharashtra	46.4	.9	0.41	1.16	9.3	32	7.83	4020
Talchar, Orrisa	40.56	2.76	0.38	0.93	9	40	6.37	3910

Source: Chandra and Chandra (2004)

232Th, and Cs137. Various radionuclides released in the atmosphere is determined by the radionuclide concentration in the coal type and the radionuclide partitioning into gaseous, sub- and super-micron fragments and aerosol (Flagan & Friedlander, 1978; Smith et al., 1979; Biermann & Ondov, 1980; Damle et al., 1981; Senior & Flagan, 1982). According to UNSCEAR, 2020, the mean mass concentrations of radionuclide in emitted CFA (Bq/kg) are 240 for 226Ra, 930 for 210Pb, 265 for 40 K, 70 for 232Th, 200 for 238U, 1700 for 210Po, 110 for 228Th, and 130 for 226Ra. For instance, it is estimated that in 1974, coal-fired electric generating units in the USA alone discharged roughly 1400 tonnes of uranium into the atmosphere (Tadmor, 1986). Many investigators have reported the radioactive elements in CFA around coal thermal power plants enlisted in Table 4 (Dai et al., 2012; Dhadse et al., 2008; Asokan et al., 2005; Finkelman, 2006). It is noted that China and India are the leading countries in case of radioactive elemental pollution. Worldwide, the high concentration of 40 K can be seen in the different coal power plants of Poland, Iraq, Turkey, and Nigeria. According to the literature survey, fewer studies have been conducted on 228Ra, 230Th, 210Pb, 210Po, and Cs137. The highest level of 232Th has been reported from a power plant in China (118.7 Bq/kg) (Ademola & Onyema, 2014; Ahmed et al., 2021; Amin et al., 2013; Baba, 2002; Bem et al., 2002; Cevik et al., 2008; Habib et al., 2019; Lu et al., 2006; Ozden et al., 2018; Weng & Chu, 1992).

The major radioactive elements reported in Indian coal power plants are 226 Ra, 228Ac, and 40 K. The highest average concentration of 226Ra was reported in Kolaghat power stations (West Bengal), followed by Choudwar thermal power plant (Odisha). In the case of 228Ac, Choudwar thermal power plant (Odisha) was reported with the highest values (140.7 Bq/kg), followed by Allahabad thermal power plant, Uttar Pradesh (110.3 Bq/kg). On the other hand, 40 K was reported to be with the highest value among other radionuclides. Allahabad thermal power plant, Uttar Pradesh, reported the highest concentration, 422.9 Bq/kg, followed by Raichur thermal power plant, Karnataka (363.8 Bq/kg) (Asokan et al., 2005; Dhadse et al., 2008). This has been reported that radionuclide discharge from the heap of the power plants in the form of particulates can reach up to the level of 750 MBq year^{-1} (BARC report, 2021). The mean concentrations from different thermal power plants of 210Po, 40 K, 232Th, 226Ra, and 238U (Bq/kg) were 25, 54, 19, 60, and 85 from Tamnar Chhattisgarh; 70, 200, 125, 105, and 101 from Talcher power plant, Orrisa; 34, 43, 40, 78, and 86 from Simhasri, Andhra Pradesh; 59, 81, 34, 93, and 67 from plant 4; and 52, 72, 85, 62, and 116 from Manguru, Andhra Pradesh, as reported in a previous study (BARC, 2021).

CFA waste control strategies

On 14 September 1999, the Ministry of Environment Forest and climate change implemented a law for 100% utilization of coal fly ash within 15 years (Dwivedi & Jain, 2014). Global CFA utilization rate is estimated to be nearly 25%, whereas current utilization rates for the USA, EU, and India have been estimated as 39%, 47% (CCPs Europe and the USA), and 78%, respectively. Central Electricity Authority of India (CEA, 2018) has reported that usage of CFA which has increased from 1 to 131.87 MT from 1994 to 2018 due to the attempt of various government and private organizations. Different technologies have been adopted by the Ministry of Science and Technology for the safe and productive utilization of fly ash since 1996, which has increased fly ash utilization. Numerous investigations have been conducted for the establishment of recycling CFA. However, the 100% usage of CFA in different sectors is still lagging, which is a cause of major concern. Therefore, thermal power industrialists need to be strictly regulated to avoid environmental pollution (Zhang et al., 2014). There is great potential in recovering non-hazardous wastes (or by-products) generated in coal combustion thermal power plants. Numerous life cycle assessment and waste management, including ways to minimize, reuse, recovery, and disposal strategies, has been discussed by various researchers (Choudhry & Hadley, 1992; Senapati, 2011). The best available technique has an important contribution in the waste management. Somehow waste prevention and minimization are not possible. Below are some new technologies that can be used to recover and dispose of waste.

For instance, alumina, the principal component of CFA, has been used as a substitute for bauxite. It acts as a suitable adsorbent for various gases and shows effective wastewater treatment processes. CFA has found its growing utilization in agriculture, manufacturing

Table 4 Average natural radionuclides (Bq/kg) in fly ash from coal thermal power plants worldwide

Thermal Region power plant	226 Ra	228Ac	40 K	238U	226Ra	228Ra	230Th	210Pb	210Po	238Th	232Th	Cs137
Allahabad, India	94.4	110.3	422.9	–	–	–	–	–	–	–	–	–
Angul, India	82.2	97.3	315.4	–	–	–	–	–	–	–	–	–
Badarpur, India	71.3	102.7	294.8	–	–	–	–	–	–	–	–	–
Bakreswar, India	83.7	96.9	329.7	–	–	–	–	–	–	–	–	–
Choudwar, India	100.1	140.7	375.9	–	–	–	–	–	–	–	–	–
Dhamanjodi, India	82.5	95.5	309.3	–	–	–	–	–	–	–	–	–
Farakka, India	88.9	101.2	309.6	–	–	–	–	–	–	–	–	–
Kolaghat, India	694.6	103.3	341.9	–	–	–	–	–	–	–	–	–
Manguru, Andhra Pradesh, India	62	–	72	–	–	–	–	–	–	–	–	–
Neyveli, India	44.9	61.2	284.9	–	–	–	–	–	–	–	–	–
Raichur, India	99.1	108.5	363.8	–	–	–	–	–	–	–	–	–
Rihandnagar, India	80.8	94.7	284.5	–	–	–	–	–	–	–	–	–
Satpura TPP (Sarni, Central India)	67.20	77.27	314.0	–	–	–	–	–	–	–	–	–
Simhasri, Andhra Pradesh	78	-	43	–	–	–	–	–	–	–	–	–
Talchar (Orissa)	83.8	102.7	333.1	–	–	–	–	–	–	–	–	–
NLC (Tamil Nadu, Southern India)	48.8	69.2	291.9	–	–	–	–	–	–	–	–	–
Tamnar Power Plant, Chhattisgarh	85	–	54	–	–	–	–	–	–	–	–	–
Talcher Power Plant, Orrisa	105	–	200	–	–	–	–	–	–	–	–	–
Vindhyachal, Madhya Pradesh (2250 MW)	93	–	81	–	–	–	–	–	–	–	–	–

Table 4 (continued)

Thermal Region power plant	226 Ra	228Ac	40 K	238U	226Ra	228Ra	230Th	210Pb	210Po	238Th	232Th	Cs137
Yenikoy Coal Power Plant, Turkey	–	–	134 ± 11–382 ± 17 Bq kg^{-1}	129 ± 12–600 ± 23	358 ± 14–689 ± 26	22 ± 2–42 ± 3	123 ± 62–649 ± 100	186 ± 14–867 ± 44	104 ± 12–543 ± 32	–	–	–
Lin-Kou Thermal Power Plant, Taiwan	–	–	572	216	143	101		359	273	68	–	–
Orgi River Thermal power station Nigeria	–	–	287–285		18.1–38.8	–	–	–	–	–	31.6–44.7	–
Afsin-Elbistan Coal-Fired Thermal Power Plant, Turkey			404 Bq kg^{-1}		167						44	–
Baoji Thermal Power Plant, China			261.5–520.8		76.1–1665.7						118.7–38.5	–
Lodz Thermal Power Plant, Poland			448.5–758.0		54.2–119.3						47.5–91.5	
Thermal Power Plants, Iraq			395.71 (site I), 475.54 (site II)	64.14 (site I), 46.18 (site II)							42.71 (site I), 39.90 (site II)	59.14 (site I), 35.81 (site II)
Barapukuria Coal-Based Powerplant, Bangladesh			277.8	329.5	175.4						263.7	
2420 MW Thermal Power Plant, Malaysia			291.2		138.7						108.0	
Yatagan Thermal Power Plants, Turkey				25.03–36.40 ppm							20.10–30.13 ppm	

zeolites, glass and ceramics, mesoporous material formation, and geopolymer synthesis (Blissett & Rowson, 2012). CFA has a tremendous potential to be utilized as a valuable catalyst for the degradation of different environmental pollutants (Mostafa et al., 2018).

The recycling by-product of CFA, bottom ash, ash of rice husk, and sugarcane bagasse ash in self-consolidated concrete production has been reported by Sua-Iam and Makul (2015). CFA-based adsorbents are reported to be used as a substitute for the removal of dissolved pollutants, such as alloys of petrol, dyestuff, heavy and trace metals, radioactive elements, COD, SS, and various gaseous pollutants like CO_2, SO_2, H_2S, H_2SO_4, Hg, HNO_3, benzene, toluene, and xylene. Unburnt carbon in CFA can be applied to form adsorbents for organic contaminants such as phenol, dyes, herbicide, and polychlorinated biphenyls: 2,3,4-trichlorobiphenyl and 2,2′,3,3′,4,5,6-heptachlorobiphenyl (Nollet et al., 2003). Morphologically, CFA particles resemble clay-sized particles, having low density, greater water retention capability, and desirable pH, and are therefore used for soil amendments. It is also reported as the richest source of essential plant nutrients like phosphorus, sulfur, potassium, calcium, magnesium, copper, manganese, and zinc required for plant metabolism (Ram et al., 2007). Based on geotechnical properties, CFA can be used in multiple applications, namely cementing, asphalt plaster, sub-grade equilibrium, soil amendment, general engineering fill, and structural fill and infill (González et al., 2009). Furthermore, CFA's use in soil systems has also been noted: this enhances the soil aeration and water retention property. It also acts as a bactericidal effect and can decrease the availability and mobility of different metals in the soil (Pandey & Singh, 2010).

Based on the properties mentioned above, CFA can also be applied for copper (II) adsorption (Panday et al., 1985). In a few studies, coal ash by batch adsorption experiment has been used to synthesize zeolites, which proved a good technique for extracting Cd(II) (Javadian et al., 2015). CFA's function as a geopolymer for Pb(II) extraction from wastewater has also been reported (Al-Zboon et al., 2011). The maximum adsorption capacity of dyes from CFA in wastewater has been achieved at pH 7.5–8.5 and 5–6 (Sun et al., 2010). Bench-scale batch experiments have been performed for acid modification of F class CFA palygorskite (MPal) (Li et al., 2016). CFA can be used to eliminate 2,4-dichlorophenol, phenol from wastewater, a typical effluent from plastics and oil refineries, rubber proofing, steel, and pharmaceuticals, etc. (Haribabu et al., 1993). However, several industries have used CFA in construction for decades.

For successful waste management, countries have established different techniques to regulate disposal and lessen their environmental impact. The infrastructure development in India should focus on recovering resources like materials, energy, and nutrients. Using current technology and India's long history of recycling, resources can be retrieved from CFA. In the most popular WtE method, combustion is utilized to generate both heat and power from residual waste (World Energy Council, 2015). CFA is the main by-product of the coal power industry. Utilizing fly ash in building materials helps to reduce the creation of dangerous pollutants. The two primary uses of fly ash in India are the production of biomass and the creation of building materials. Agriculture, forestry, and floriculture are included in the biomass production process, and the production of cement, bricks, and road embankments is a part of the construction industry. Additionally, the other important technologies for fly ash utilization in India include phytoremediation and phyto-management (Pandey et al., 2009; Verma et al., 2014). In addition to these numerous additional solutions, backfilling technology is employed to reduce power plant CO_2 emissions (Arenillas et al., 2005; Fang et al., 1999). Because coal fly ash contains a significant amount of alkali earth metal, carbon capture and storage technology can be utilized to sequester CO_2. Since fly ash has a high concentration of alkali components, the primary approach relies on mineral carbonation which are essential components for mineral carbonation, even if there are a few other ways to use fly ash to reduce CO_2 emissions (Fauth et al., 2002; Teir et al., 2009; Wee, 2013). The least energy-intensive and secondary-pollution option for the long-term disposal of aluminosilicate fly ash could be to transform them into soil conditioners or ecological restoration materials (Liu et al., 2022).

Use of fly ash as a geopolymer binder is also an intriguing approach. The innovative geopolymer binder is emerging for use in concrete technology by the construction sectors. To fully utilize geopolymer binders, more research is needed, especially in ambient curing and durability issues (Yao et al., 2015; Kosmatka et al., 2002). Fly ash can also be used in wastewater treatment using practical methods such as membrane filtration, adsorption, photocatalysis,

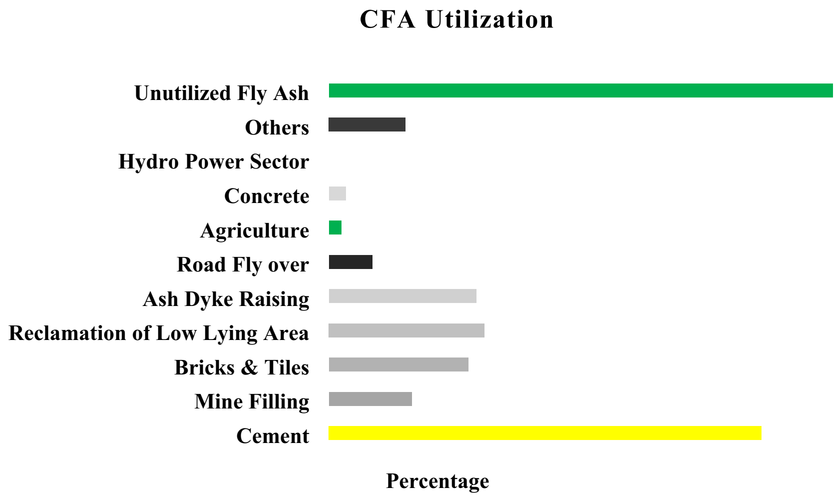

Fig. 5 Percentage (%) utilization of coal fly ash in different sectors

and the Fenton process technologies. Fly ash–based Fenton catalysts are an efficient, affordable, and environmentally friendly technology (Mushtaq et al., 2019).

In addition to this, numeral mathematical application can also be taken into consideration like WGF means waste generation factor which is used for the waste generation estimation is based on the amount of production. This value has an important contribution verifying the amount of waste generation declared by an industry (Demir et al., 2019). Another one is the emission factor calculation. The emission factor is crucial in assessing the pollutant emission from the power plant. The concentration of gaseous pollutants (CO_2, SO_2, NOx, and PM) released by coal-fired power plants over the preceding two decades must be ascertained using this information. Pollutant mass can be determined per unit of time by determining the gaseous concentration emitting form the stacks and flue gas flow rate (Nazari et al., 2010; Zhao et al., 2008). The State Environmental Protection Administration (the predecessor of the Ministry of Environmental Protection) in China issued an emission factor database that only provided SO_2 and total particulate matter emission rates for the power sector (SEPA, 1996). An emission factor is also a crucial tool for creating emission control strategies. This is the conventional way to represent this variable to divide the product's or material's original weight, volume, distance, energy generation, or pollutant dispersion time by the resulting pollutant's intensity. There are many ways to compute emission factors, including material balancing, modeling, and utilizing a flue gas analyzer, which is thought to be the dominant method for computing emission factors (Wang et al., 2020a, b). The use of CFA for various purposes shows that the cement forming units consumed 26.85% of total CFA, out of which 9.66% consumption was documented in the recovery of low-lying areas, 9.15% in ash dykes, and 8.65% in making building materials. On the other hand, 5.15% was used in storage mining areas, 2.70% in roads and embankments, 1.04% in concrete, 0.77% in agriculture, and 4.76% in different areas (Fig. 5).

Summary

Based on the comprehensive examination of various studies, this review suggests broad range of applications of CFA. The characterization includes physicochemical, mineralogical, petrological, and morphological using previous literature surveys. On this basis, the following observations are made:

1. Coal thermal power plants produce CFA as their major by-product. Based on silicon dioxide (SiO_2) and aluminum trioxide (Al_2O_3), CFA is broadly classified in two categories, namely class F and class C, and the majority of the Indian power plants predominately consist F type of fly ash.
2. In terms of mineralogical properties, several minerals, like quartz, mullite, magnetite, and hematite, are identified as significant crystalline minerals.

3. Indian CFA is also composed of heavy and trace elements along with radioactive elements.
4. The no plastic limit % for different power plants has been found for the majority of the ashes, which is due to the non-plastic nature of CFA.
5. It is noted that CFA particles can be categorized into 11 morphological classes in which char is considered to be one of the carbonaceous particles presents which is identified as irregular, solid, and hollow spherical particles. Cenospheres, ferrospheres, and plerospheres are the most important and value-added particles noticed in the CFA.
6. CFA tends to be non-plastic in nature, with a permeability level between 8×10^{-6} and 1.87×10^{-4} cms^{-1}. C_u and C_c of CFA are found in the range between 2.14–5.65 and 0.76–2.47, respectively.
7. Petrographically, CFA particles were found enriched in inertinite and liptinites as well as collotelinite, collodetrinite, and vitrodetrinite.

The retrospective look added considerable knowledge to CFA characterization, suggesting that it is possible to create new industry synergies through the CFA characteristics. Governments and individuals must utilize fly ash to its fullest potential following environment friendly technologies for the purpose to reduce its negative effects on the environment and the economy. This also suggests that the sustainable uses of CFA would have economic advantages and eradicate the disposal problems of CFA with its linked environmental hazards and disadvantages.

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