Coal Fly Ash

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Keywords: coal fly ash ; carbon nanominerals (CNMs) ; carbonaceous particle

1. Introduction

Coal fly ash (CFA) is a complex material produced from the combustion of pulverized coal in thermal power plants during the production of electricity [1]. Due to its complexity, until now, nearly 316 minerals were found individually while 188 minerals were present in groups identified in various coal fly ash samples from different parts of the world [2][3]. CFA is a fine glass-like, spherical shaped powder, heterogeneous in nature and has sizes varying from 0.01 to 100 microns [1][4]. It is generally light-coloured and consists mostly of silt and clay-sized glass spherical particles which provide a consistency somewhat like that of talcum powder. It is one of the most familiar and widely used pozzolanic materials [5][6], with its two most important factors, minerals and composition, depending on the various factors applied during their handling. Therefore, there is a possibility that one sample of CFA may vary with respect to the next one depending on the source of coal used; on the various environmental conditions during burning and cleaning with pulverization; on the design, types, and operations in the power plant boiler unit; on the degree of coal manufacture, storage, and handling of the fly ash; on the additives used for facilitating burning of coal or improving precipitation performance; on the productivity of emission control devices; and on the prevalent climatic conditions [1][7]. The typical compositions of higher-grade coal-derived fly ashes have SiO_2, Al_2O_3, and iron and calcium oxides with varied weights of carbon, as analyzed by the loss on ignition (LOI) [8]. Due to the presence of value-added minerals in CFA, it is widely applied in ceramics [9], construction, civil engineering, and geopolymers [4]. However, with the progress of technology every year, there is a gradual and progressive increase in CFA utilization in India as per the data provided by the central energy authority of India (CEA 2020). Table 1 shows the detailed fly ash production and utilization in India from 2010–2011 to 2019–2020.

Table 1. Fly ash production and utilization in India during the last decade (2010–2011 to 2019–2020 *).

Descriptions	2010–2011	2011–2012	2012–2013	2013–2014	2014–2015	2015–2016	2016–2017	2017–2018	2018–2019	2019–2020
Fly ash production	131.09	145.42	163.56	172.87	184.14	176.74	169.25	196.44	217.04	226.13
Fly ash utilization	73.13	85.05	100.37	99.62	102.54	107.77	107.10	131.87	168.40	187.81
% Utilization	55.79	58.48	61.37	57.37	55.69	60.97	63.28	67.13	77.59	83.05

* Source: CEA (Central Electricity Authority) 2010 to 2020.

A major portion of CFA is used for manufacturing bricks, tiles, cement, panels, metallurgy, landfills, etc. It is also used for river embankments, civil engineering, geopolymers, zeolites, and other lightweight materials [1][10]. In the current year, 2019–2020, the total CFA production was 226.13 million ton (MTs), out of which 187.81 MTs used, i.e., 83.05%, while 17% were left unused, especially near fly ash ponds. Of the total fly ash used (83.05%), in 2019–2020, about 4.69%, i.e., 10.62 MTs, was used for land and mine-filling purposes while 9.46%, i.e., 21.39 MTs, was used for manufacturing bricks and tiles and about 19.08%, i.e., 43.14 MTs, was used for civil engineering purposes like for fly ash overs, roads, embankments, and dykes. Besides these, about 35.06 MTs, i.e., 15.50% CFA, were used for reclamation of low-lying areas; about 25.60%, i.e., 57.88 MTs, CFA was used for the cement industry; and about 0.06%, i.e., 0.14 MTs, CFA was used for agricultural purposes, especially in the form of fertilizers [4][11]. As far as developed countries are concerned, European countries like France have successfully achieved about 100 per cent utilization of CFAs [12]. Among the South-Asian countries, China has nearly reached 100% CFA utilization in the last few years.

Besides basic minerals, i.e., ferrous, silica, and alumina, CFAs are rich sources of carbon and polyaromatic hydrocarbons (PAHs) [13] which finds applications in many industries [14]. Currently, the carbon-based industries rely on several other industries to meet their demand for raw material. Therefore, CFAs could act as the best substitutes for the recovery of both forms of carbon materials, i.e., organic and inorganic [15]. Both of these carbon sources are coal, present in the organic form. After combustion in a thermal power plant furnace, due to the reduced efficiency of a furnace, they are not completely burnt. Therefore, CFA can act as a potential raw material for carbon with several advantages including being economical, being eco-friendly, and minimizing solid waste arising due to CFA dumping in fly ash ponds [16].

2. Importance of Carbon Nanomaterial and PAHs

Nanoscience and nanotechnology is an interdisciplinary area of investigation that offers several opportunities for scientists in diverse areas to explore the possibilities of novel research [17][18]. Several nanostructures have been formed and planned for use in altered systems and devices. Amongst them, these carbon nanostructures have received vast attention and have been extensively manufactured and inspected [19]. More recently, the science of carbon has gained great visibility with the discovery of fullerenes in 1985 along with the first high-resolution transmission scanning electron microscopy (HRTEM) observations of carbon nanotubes (CNTs) in 1991. The carbon nanomaterials (CNMs) have controlled porosity and surface chemistry, and superior thermal and mechanical features with acceptable stability [20]. The arrival of nanotechnology, fullerene discovery in 1985, and the identification of carbon nanotubes (CNTs) in 1991 have boosted carbon chemistry. There is a continuous increase in the importance of CNMs in science and technology, energy, environment, water, or biomedicine. They have excellent mechanical properties that can attract much attention for use in a variety of applications [21][22]. Moreover, their minute size, high surface-to-volume ratio (SVR), and electron-rich and lightweight properties make them a suitable material for nanotechnology-based applications [23]. Carbon nanostructured materials show different allotropes on the 0-, 1-, 2-, and 3-dimensional nanoscales such as CNTs, nanocarbon coating, fullerene, graphene, and diamond or porous carbonaceous particles [24]. Moreover, carbon nanomaterials can have new applications for their physical, electrical, and chemical properties by clubbing with unlike functional nanomaterials, such as CNT nanocomposites with functional NPs, metal or oxide nanomaterials and carbon nanocoatings with functional metallic particles, or graphene modified with carbonaceous particles [25]. Carbon-rich solid waste, like high carbon CFA; agricultural waste [14]; and other industrial waste have been rarely utilized as a source for nanomaterial production.

3. Polycyclic Aromatic Hydrocarbons (PAHs) Presence in CFA

PAHs are a huge group of ubiquitous chemicals possessing higher than 100 organic compounds comprised of two or more fused C-rings derived from benzene [26][27]. The formation takes place at the time of incomplete combustion of coal in coal-fired TPPs. Moreover, PAHs can also be generated from oil, garbage, gas, and organic substances like tobacco or charbroiled meat [26]. PAHs can be synthesized by both natural and anthropogenic methods. The natural synthesis method includes volcanic content and forest fires, while the anthropogenic method includes industries, internal combustion, and incomplete or partially burned fossil fuel and, through exhausts, diesel engine, aviation, and cigarette smoke. Fossil fuels like coal combustion are one of the major sources of PAHs [28]; apart from this, it is also present in the soil, water surface and groundwater, air, and sediment.

4. CFA as a Natural Source of Carbon Nanomaterials (CNMs) and PAHs

4.1. CFA as a Natural Source of CNMs

The carbonaceous elements of CFA are usually termed unburnt carbon particles or chars, which are made up of porous char particles and aggregated submicron particles [29]. These carbon nanoparticles are basically carbon that is unable to burn during the burning of pulverized coal in TPPs. Some of the unburnt carbon (UC) residues are collected with the CFA in the precipitators [30], which can be precisely and appropriately identified with the use of HRTEM. The parent source of CFA is coal, which is a rich source of carbon [31]. Generally, coals of higher grades (like anthracite and bituminous) have higher percentages of carbon. The CFA, which has a higher amount of UC, is generally known as high carbon fly ash (HCFA) [32][33][34]. Based on the carbon content, CFA can be further classified into three classes, i.e., low or ultralow, moderately high, and very high carbon ash [35]. The percentage of UC in CFA mainly varies from 2–12%, but it could be more than 20% and, in exceptional cases, could reach up to 57% [29]. Therefore, considering an average of 1–12% carbon content in CFA, there are around 8–96 MTs of carbon residues annually around the world (IEA, 2017) [36]. However, there are several other factors that affect the composition of carbon in the CFA. The factors affecting UC value in the CFA could be categorized within two main groups—the effect of coal characteristics (e.g., mineral matter, type of coal, their particle size, presence of moisture, maceral composition, calorific value (CV), and volatile matter) and the consequence of the design of a combustion system and their operating conditions (e.g., residence time available for burning in a furnace,

furnace heating loading, oxygen feed, temperature and pressure of the boiler, air/coal ratio, the flow rate of heat, and pattern of flame) [37][38]. Unlike sub-bituminous coal, bituminous-coal-derived CFA includes both amorphous as well as crystalline carbons that bind together with other CFA particles [31][39].

The coal undergoes pyrolysis, during which the carbon sources are burned and there is the formation of new minerals at the desired temperature [40]. The ash is left with the unburned carbons due to the incomplete burning of the coal. This unburnt carbon in CFA is considered an undesired material for civil engineering [41]. For use in cement, tiles, bricks, roads, etc., the carbon must be eliminated and should be brought to the desired range [1], while the removed carbon can be utilized either directly or after purification in place of carbon materials as an adsorbent, as fuel, or as a precursor for the synthesis of carbon-based nanomaterial [37][42]. Such applications of carbon particles from waste CFA will reduce the problem not only related to CFA but also of an alternative source of carbon and carbon-based nanomaterials. The UC in the CFA produced from low-NOx pulverized coal combustion revealed that these are the mixtures of soot and coal char. The UC or carbon particles of CFA are always accompanied by traces of elements like Si, Al, Hg, Se, Fe, and others [1][43][44]. Such carbon particles require HRTEM for suitable identification and material information.

4.2. Coal Fly Ash as a Source Material of PAHs

The 3-D network of coal is comprised of condensed aromatic and hydroaromatic compounds that are allied by short alkyl bridges and linkages of ether and thioether. Moreover, such 3-D structures of coal also possess PAHs as an integral part [45][46]. The organic content of coal includes complementary structures, where the major constituent is a macromolecular, non-soluble, 3-D network with condensed aromatic and hydroaromatic units which in turn are joined by ether and thioether linkages (like methylenes) [47], while the other fraction is a molecular phase of compounds in which the molecular mass is in the ranger of low to medium. Such compounds can be typically solubilized in the organic solvents with varying dispersals of aliphatic and several types of aromatic hydrocarbons like hydroxylated polycyclic aromatic compounds, polycyclic aromatic, and hydroaromatic hydrocarbons. [48]. In comparison to the bottom fly ash or CFA, their parent molecule, i.e., coal, has a higher content of total PAHs. The PAH concentration in coal may fall in the range of up to 100–1000 mg/kg based on the coal ranking and its higher values especially for hard coals [49][50].

There are two ways for PAHs to be emitted into the surrounding from a combustion source [26]. PAHs are bound to release into the atmosphere mainly in the vapour phases directly arising from the combustion facility. PAHs could also be released along with the solid phases (CFA or bed ash), from where they could be either evaporated or dissipated into the surrounding environment [51]. The PAH percentage in CFA mainly depends on the type of coal or fuel used, the technology applied for the burning of coal, and the residence time of ash inside the combustion facility [52].

5. Estimation of Carbon Content in CFA

The estimation of UC in any compound like CFA can be easily carried out by an LOI at a higher temperature [53]. This LOI acts as an indicator for the UC content of CFA. In order to utilize the class F or class C coal fly ash for concrete purposes, it has to be nearly 6% LOI [54]. However, it is very difficult to report whether the LOI under high temperature is either due to the UC or due to the breakdown of various chemical bonds in the different mineral phases present in CFA. In addition to these two factors, sometimes the LOI is also caused by the moisture adsorbed physically on the surface of the molecule [55][56]. In addition to this LOI-based conventional technique for the UC estimation, there are a few other techniques, i.e., thermogravimetric analysis (TGA) [57] and elemental analysis techniques [58]. Both of these techniques are more precise than the LOI-based conventional method for UC estimation in the CFA. The only problem associated with the TGA and elemental analysis techniques is that, along with UC, some inorganic carbon in the form of carbonates are frequently encountered in UC estimation [15][59]. Valeev et al., 2019, reported an accurate and advanced method for the estimation of carbon contents in the CFA. In this technique, carbon content was analyzed by a fractional gas analyzer CS-600 (LECO Corporation, USA). The CFA samples of about 1 g were kept in ceramic crucibles and then placed into an induction furnace. Finally, the C concentrations were analyzed by infrared absorption of carbon dioxide present in the gas phase during the burning of the sample in the surplus oxygen atmosphere [60][61]. There are also some new emerging techniques like carbon, hydrogen, nitrogen, and sulfur (CHNS) element analyzer; Electron diffraction spectroscopy (EDS); Total organic carbon (TOC) analyzer; x-ray photoelectron spectroscopy (XPS); and electron scattering chemical analysis (ESCA), which could be more promising for the estimation of UC from the CFA [62].

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