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IIT (ISM): Indian Institute of Technology

Research Article

Keywords: Pulverized coal, bulk density, particle size, specific surface area, spontaneous combustion, explosion

Posted Date: September 27th, 2021

DOI: <https://doi.org/10.21203/rs.3.rs-828214/v1>

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Version of Record: A version of this preprint was published at Environmental Science and Pollution Research on November 26th, 2021. See the published version at <https://doi.org/10.1007/s11356-021-17626-9>.

Physico-chemical characteristics of pulverized coals and their interrelations- A spontaneous combustion and explosion perspective

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Abstract

Characteristics of pulverized coals have significant influence on the spontaneous combustion and explosion processes. This paper presents an experimental and theoretical framework on physico-chemical characteristics of coal and analyzes their interrelations from spontaneous combustion and explosion perspectives. The chemical properties, morphology, bulk density, particle size and specific surface area of pulverized coals from nine different coal subsidiaries of India are vividly investigated in five distinct sizes. Moreover, the effects of particle size on bulk density, specific surface area and N₂ adsorption capacity of pulverized coals are critically analyzed. The micrographs revealed that the coal particles are mostly irregular in shape with angular outlines and sharp edges. With decrease in particle size, the bulk density of pulverized coals decreased and the specific surface area and N₂ adsorption capacity increased. The relationships of bulk density and specific surface area of pulverized coals with particle size are established. Moreover, the specific surface areas determined by both the particle sizing and BET methods are compared and correlation factors between them are determined. This study led to the generation of insightful coal characteristic data which can be used as reference material for furthering researches on spontaneous combustion and explosion involving pulverized coals.

Keywords: Pulverized coal; bulk density; particle size; specific surface area; spontaneous combustion; explosion.

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1. Introduction

Coal still maintains its legacy as the largest source of energy across the globe. Pulverized coal finds applications as raw material in several process industries, wherein its physical properties, viz. bulk density, shape, size, surface texture, moisture content, etc. play a major role. Bulk density is a useful parameter in the characterization and handling of coal. Under a given processing condition, the bulk density of coal is significantly affected by the particle size distribution and moisture content among many other factors (Yu et al., 1995; Braga et al., 2019). In contrast, coal dust explosion and spontaneous combustion of coal leading to fire are the two major threats to the safety of coal mines, process industries and utilities sector (Amyotte et al., 2003; Yuan et al., 2015; Yong et al., 2019). Apart from imposing direct threat, they pollute the workplace and surrounding environment and adversely affect the human health by releasing harmful toxic gases, combustion residues and significant amounts of particulates (Li et al., 2020). Coal dust produced during the mining process constitutes one of the major causes of explosion (Cashdollar, 1996; Mishra and Azam, 2018) and health hazards in coal mines (Laney and Weissman, 2014). Yuan et al. (2015) reported that coal dust contributes to 35% of the dust explosions in China and one of the main causes of the explosions is high dependence on coal for energy consumption.

The particle size and surface area of pulverized coal are often correlated and greatly influence the spontaneous combustion susceptibility and explosion severity of coal dust (Cashdollar, 1996; Mishra and Azam, 2018; Azam and Mishra, 2019; Li et al., 2020; Pan et al., 2020). The explosibility and spontaneous combustion susceptibility or oxidation rate of coal increase with increase in the fineness and exposed surface area of coal. The finer the pulverized coal particle size, the greater the exposed specific surface area available for oxygen adsorption, the stronger the coal oxidation and coal-oxygen recombination abilities, and the greater the susceptibility of coal to spontaneous combustion (Pan et al., 2020). Hence, characterization of particle size and surface area of pulverized coal is important from the spontaneous combustion susceptibility and explosion severity assessment perspectives.

In recent years, the effects of particle size and surface area on ignition sensitivity (Amyotte et al., 1993; Mishra and Azam, 2018; Azam and Mishra, 2019), explosibility (Cashdollar, 2000; Gao et al., 2010; Harris et al., 2015; Li et al., 2016; Cao et al., 2012; Liu et al., 2018) and spontaneous combustion susceptibility (Rifella et al., 2019; Li et al., 2020) of pulverized coals are being extensively investigated by the researchers worldwide. These studies used pulverized coals in various size ranges for experimentation. Mishra and Azam Mishra and Azam (2018)

studied the effects of particle size on minimum ignition temperature (MIT) and combustion process of coal dust in five different sizes ranges. Hosseinzadeh et al. (2019) studied the minimum ignition energy (MIE) to assess the explosion risk of six different dusts, including Indonesian coal dust cloud, based on their particle size distribution, density and flammability. Li et al. (2016) studied the effects of particle size (D_{50}) and size dispersity (σ_D) on explosion severity of coal dust and observed that the presence of finer particles greatly increases the total effective specific surface area and speed up the devolatilization rate, which would accelerate the dust explosion process. Li et al. (2020) investigated the influence of fuel concentration and coal particle size on multiple explosion parameters for methane–coal particle mixtures. They considered coal dust particles in three sizes, such as 38, 48 and 75 μm for the experiments. Li et al. (2020) studied the effects of particle size on self-ignition behaviour of coal dust layer on a hot plate in three different size ranges, viz. 100-150, 150-200 and 200-300 μm . Xu et al. (2020) investigated the microstructure and oxidation reactivity of crushed coal of different ranks in five particle size ranges, namely, 0–74, 74–125, 125–180, 180–224 and 224–280 μm . Saleh and Nugroho (2013) studied the effect of particle size on spontaneous combustion of coal in the size groups of -599+299, -299+249, -249-150, -150+76 and -76 μm and observed that the propensity of coal to spontaneous combustion increases with decreasing particle size. In view of this, understanding of the characteristics, especially the particle size distribution and surface area, finds an important place in the application and implications of pulverized coal in mining and allied industries.

Coal is a complex and highly porous material with ‘sponge-like’ structure. It contains numerous interconnecting pores of different shapes and sizes. The specific surface area (i.e., surface area per unit mass, m^2/g) of coal particles depends upon the method of its measurement, such as particle sizing, photoextinction, methylene blue dye adsorption and gas adsorption (both noble gas and CO_2). Among all the methods, particle size-based measurement is most readily visualized as being the ‘easily’ accessible coal surface, since the specific surface area is measured based on the average sieve size of the particles in the coal powder (Linge, 1989). The laser scattering instruments are widely used for this purpose. In particle sizing by laser diffraction technique, the particle size and specific surface area (S) are related as (ISO 13320-1: 2009)

$$S = \frac{6 \sum \frac{V_i}{d_i}}{\rho \sum V_i} = \frac{6}{\rho D_s} \quad (1)$$

where, V_i is the relative volume by particle size class d_i , ρ is the material density and D_s is the mean diameter based on surface area, also known as Sauter mean diameter. The above equation relates the specific surface area of a collection of smooth spherical particles with the average diameter of an equivalent spherical particle (D_s) of density (ρ).

Gas adsorption is the preferred technique for specific surface area determination, as it takes into account the surface roughness and crevices of particle exterior and porous interior of the particles. Among several methods developed, Brunauer-Emmett-Teller (BET) adsorption method (Brunauer et al., 1938) is commonly used to measure the specific surface area of particulate matter by physisorption of N_2 gas molecules at the boiling point temperature of liquid nitrogen of about 77 K (-196°C) (Clarkson and Bustin, 1999; Cheng et al., 2015; Zhao et al., 2016).

The volume of gas adsorbed on a monolayer over the particles surface is determined as per the BET isotherm equation (Brunauer et al., 1938)

$$\frac{p}{v(p_0 - p)} = \frac{1}{v_m c} + \frac{c-1}{v_m c} \left(\frac{p}{p_0} \right) \quad (2)$$

where, p and p_0 are the equilibrium and saturation pressure of adsorbate gas, $\frac{p}{p_0}$ is the relative pressure, v is the

total volume of absorbed gas, v_m is the volume of gas required to form a complete unimolecular adsorbed layer, and

c is the BET constant. In Eq. (2), the plot of $\frac{p}{v(p_0 - p)}$ against $\frac{p}{p_0}$ is a straight line, whose intercept is $\frac{1}{v_m c}$ and

slope is $\frac{c-1}{v_m c}$. The constants v_m and c can be evaluated from the slope and intercept. In BET method, the specific

surface area (S) of solid particles is determined by dividing the total surface area (S_{total}) by mass of the solid sample or adsorbent (w). It is given by equation (Thommes et al., 2015)

$$S = \frac{S_{total}}{w} \quad (3)$$

$$S_{total} = \frac{(v_m N A_{cs})}{v} \quad (4)$$

where v is the molar volume of the adsorbate gas, v_m is the monolayer volume of the adsorbed gas, N is Avogadro's number ($6.023 \times 10^{23} \text{ mol}^{-1}$), A_{cs} is the cross-sectional area of the adsorbate ($16.2 \text{ \AA}^2$ for nitrogen).

BET method assumes that the particles usually are neither spherical nor smooth and it takes into account the surface area of internal pore spaces. Conversely, particle sizing method assumes the particles as smooth, spherical and nonporous. Therefore, the specific surface area measured by gas adsorption (BET method) is greater than that measured by the laser diffraction instruments (Zlochowet al., 2018).

Several researchers have measured the particle size and specific surface area of different types of pulverized coals using various methods (Table 1). From the table, it may be observed that N₂ gas adsorption is the most commonly used method among the others. Linge (1989) compared the surface area of coal particles measured by different methods, including, particle sizing, photoextinction, methylene blue dye adsorption and gas adsorption (both N₂ and CO₂). For the coal particles in size range of 15-9 µm, he reported the specific surface area by particle sizing and N₂ gas adsorption methods in the ranges of 0.16-0.30 and 1-7 m²g⁻¹, respectively. Cheng et al. (2015) studied the effect of different experimental conditions on the specific surface area calculation and reported that BET theory more accurately calculates the specific surface area of coal. Dubois et al. (2011) studied the dependency of BET surface area on particle size for some granitic minerals of different particle sizes. They observed a linear relationship between the BET surface area and inverse of the particle size, up to a certain particle size.

Table 1

This paper aims at investigating the (1) important physico-chemical properties of pulverized coals, such as proximate and ultimate analyses, morphology, bulk density, particle size and specific surface area, (2) effect of particle size on bulk density, specific surface area and N₂ adsorption capacity of pulverized coals and (3) establishing possible correlations among these parameters. Nine different Indian pulverized coals in five different size ranges were considered in this study. The outcome of this study can have applications in the safety and risk engineering of coal mines, process industries and utilities sector dealing with pulverized coals, and serve as reference material for further researches on combustion and explosion involving coal dusts.

2. Materials and Methods

2.1 Collection and preparation of coal samples

In this study, nine coal samples (A, B, C, D, E, F, G, H, I) collected from different coal mining companies situated in different parts of India were used. The coal samples were pulverized in a dry ball mill and screened into five

different particle size ranges, viz. <38, 38-74, 74-212, 212-425 and 425-850 μm , using a series of sieves for experimentation (Fig. 1). The morphology of pulverized coals was assessed using FE-SEM Supra 55 (Carl Zeiss, Germany).

Fig. 1

2.2 Proximate and ultimate analyses of coal

The proximate analysis of collected coal samples was done with a Thermogravimetric Analyzer (TGA-2000A, Navas Instruments, USA) according to ASTM D7582 for determining the moisture (M), volatile matter (VM), ash (A) and fixed carbon (FC) contents of coal samples. The ultimate analysis of coal samples was done using 'Flash 2000' Organic Elemental Analyzer (Thermo Scientific) as per ASTM D3176-15 for determining the elemental composition of coal, such as carbon (C), hydrogen (H) nitrogen (N) and oxygen (O) contents.

2.3 Measurement of bulk density of pulverized coals

The bulk density of coal is influenced by its physical characteristics, such as relative density, shape, particle size distribution, surface properties and moisture content, and on the dimensions of the measuring container. The bulk density of pulverized coals was determined using a fixed volume stainless steel container as per ISO 23499:2013, to study the effect of particle size on bulk density of coal. Sufficient amount of coal powder was filled in the container and compacted. Excess coal powder was scraped carefully from top of the vessel with the sharp edge of a spatula and the weight of the coal powder in the container was taken. The bulk density of the sample was determined by dividing the sample weight by container volume. The bulk density of each sample was determined thrice and the average value was considered as the bulk density of the sample.

2.4 Measurement of particle size and specific surface area of pulverized coals

The particle size of pulverized coals of different sizes was determined with a particle size analyzer. However, the specific surface area was determined using both particle size analyzer and surface characterization analyzer utilizing multipoint Brunauer–Emmett–Teller (BET) method.

2.4.1 Measurement by particle sizing method

The particle size distribution and specific surface area of pulverized coal samples were determined using Microtrac S3500 Particle Size Analyzer system (Microtrac, USA) on dry basis in air entrainment mode operation. The analyzer works on the principle of light scattering and utilizes three precisely placed red laser diodes to accurately characterize particles as a volume equivalent sphere diameter in size range of 0.02 to 2800 μm . The device measures the light scattered from the particles in a laser beam of wavelength 780 nm. It analyses the intensity and angle of light scattered from the particles measured by optical detector arrays to determine the particle size distribution based on algorithms that utilize Mie compensation and Modified Mie calculations for non-spherical particles. In this study, the refractive index of coal was taken as 1.6 for particle size analysis (Mengüç et al., 1994).

2.4.2 Measurement by BET method

In this study, the BET surface area of pulverized coals was determined using N_2 adsorption at 77 K with a 3Flex 3500 high resolution surface characterization analyzer (Micromeritics, USA). This method utilizes the fact that the extent of N_2 adsorption is directly related to the available surface area. Prior to the adsorption experiments, degassing of the coal samples was done at 130° for almost 12 hours using sample degas system (Micromeritics Vac Prep 061) to remove the volatile substances from the coal. Then the samples were kept in the liquid N_2 canister for almost 2 hours for saturated adsorption of N_2 on available coal surfaces. The N_2 adsorption isotherms were captured at a relative pressure (p/p_0) range of 0.05 to 0.3.

3. Results and Discussion

3.1 Results of proximate and ultimate analyses

The results of proximate and ultimate analyses of coal samples are presented in Table 2. The proximate analysis results show that the moisture, volatile matter, ash and fixed carbon contents of the coals varied in the ranges of 1.12-8.06, 19.27-37.2, 14.23-36.61 and 22.97-58.54%, respectively. The results of ultimate analysis show that the carbon, hydrogen, nitrogen and oxygen contents of the coal samples varied in the ranges of 36.01-63.34, 3.06-4.51, 1.29-2.71 and 30.41-59.32%, respectively. Moisture is an important parameter affecting the handling, storage and

transport of coal. The VM content greatly influences the thermochemical reactivity and rank of coal (Speight, 2015). The coal samples A, B, G and I possess lower VM content of 19.27, 21.1, 23.86 and 26.11%, respectively, which indicates their better storage potential and lower spontaneous combustion propensity (Nyakuma et al., 2017).

Table 2

The fuel ratios (ratio between the FC and VM) of coal samples were calculated to determine the rank of coals. Based on the fuel ratio (FR), Frazer (1877) classified the coals as: Anthracite (FR: 12-100), Semi-anthracite (FR: 8-12), Semi-bituminous (FR: 5-8) and bituminous (FR: 0-5). Since the fuel ratios of the coal samples determined in the range of 0.62-2.8, the coals are confirmed to be of bituminous rank.

3.2 Morphology of pulverized coals

The micrographs of pulverized coals of different sizes, viz. <38, 38-74, 74-212, 212-425 and 425-850 μm , examined with FE-SEM Supra 55 (Carl Zeiss, Germany) are presented in Fig. 2. The micrographs revealed that the coal particles are mostly irregular in shape with angular outlines and sharp edges. Particle agglomeration and sticking of fine particles with the coarser ones were also observed in the micrographs.

Fig. 2

3.3 Effect of particle size on bulk density of pulverized coals

The variations of bulk density of different pulverized coal samples with particle size are shown in Fig. 3. The results show that the bulk density of pulverized coals increases with increase in the particle size. Among all the samples, the highest and lowest bulk density values were determined for samples H and D, respectively. While the bulk density of pulverized coals of <38 μm size varied in the range of 0.39-1.05 g/cm^3 , for coals of 425-850 μm size, it varied in the range of 0.76-1.35 g/cm^3 . The variations in bulk density of different pulverized coals of same particle size may be due to the difference in coal properties. The mean bulk density of nine pulverized coals of sizes <38, 38-74, 74-212, 212-425 and 425-850 μm were determined 0.63 ± 0.24 , 0.71 ± 0.23 , 0.84 ± 0.21 , 0.92 ± 0.21 and 0.97 ± 0.22 g/cm^3 , respectively.

Fig. 3

The variation of average bulk density of pulverized coals with particle size shown in Fig. 4 depicts an increasing trend, which best fitted the polynomial trend with $R^2 = 0.989$. The polynomial regression equation obtained between the bulk density and particle size ($y = -0.007x^2 + 0.136x + 0.492$) in Fig. 4 can be used to predict the bulk density of pulverized coals of known sizes. The average bulk density of pulverized coals increased from 0.63 to 0.97 g/cm³, or increased by 1.54 times, with increase in the particle size from <38 to 425-850 μm . The reason behind decrease in bulk density with decrease in the particle size is attributed to the fact that with decrease in particle size, the total surface area and voids between the particles are increased, which results in decrease of bulk density due to increase in the volume of coal particles (Braga et al., 2019). Elliott (1981) and Braga et al. (2019) also demonstrated that for the same moisture content, bigger coal particles have higher bulk density. Moreover, in the coal particles of higher size range, the inert petrographic materials having higher hardness and resistance to crushing are concentrated and thus, the bulk density of pulverized coals increases with increase in the particle size (Silva et al., 2011). Sadovnikov and Gusev (2018) also observed decrease in the pycnometric density and increase in the specific surface area with decrease in the average particle size of silver sulfide powders.

Fig. 4

3.4 Effect of particle size on specific surface area of pulverized coals

The median particle diameter (D_{50}) and specific surface area of pulverized coals of different sizes determined by particle sizing method are presented in Table 3. The particle size distribution curves of a coal sample for different sizes are shown in Fig. 5. The D_{50} values of coal particles of <38, 38-74, 74-212, 212-425 and 425-850 μm were determined in the ranges of 20.37-36.9, 39.07-57.57, 117.2-170.4, 343.3-386.0 and 763.3-837.3 μm , with mean values of 29.58 ± 4.99 , 50.56 ± 6.74 , 153.2 ± 18.78 , 359.36 ± 13.15 and 802.28 ± 28.58 μm , respectively. The specific surface area of coal particles of <38, 38-74, 74-212, 212-425 and 425-850 μm were determined in the ranges of 0.239-0.423, 0.163-0.338, 0.043-0.112, 0.017-0.035 and 0.007-0.01 m²/g, with mean values of 0.332 ± 0.052 , 0.234 ± 0.051 , 0.066 ± 0.027 , 0.021 ± 0.006 and 0.008 ± 0.001 m²/g, respectively.

Table 3

Fig. 5

The variations of median particle diameter (D_{50}) and specific surface area of pulverized coals with particle size are shown in Fig. 6. Similarly, the variations of average D_{50} and average specific surface area of pulverized

coals with particle size are depicted in Fig. 7. From these figures, it is evident that D_{50} increases with increase in the coal particle size. Generally, the specific surface area of pulverized coals increased with decrease in the particle size. With decrease in particle size from 425-850 to $<38 \mu\text{m}$, the mean value of D_{50} decreased by 27 times and the mean specific surface area increased by 41.5 times. The finer coal particles with greater exposed surface area interact more with oxygen resulting in higher oxygen adsorption. Moreover, size reduction of coal due to crushing generates more free radicals and accelerates the coal oxidation reactivity (Xu et al., 2020). Therefore, the finer coal particles are more prone to spontaneous combustion. Moreover, they require lesser ignition energy and consequently, increase the severity of coal dust explosion in mines (Mishra and Azam, 2018).

Fig. 6

Fig. 7

In order to establish relationship between the specific surface area and particle size of pulverized coals, a graph between the average median particle size (D_{50}) and average specific surface area was plotted as shown in Fig. 8. It may be observed that the specific surface area of pulverized coals decreased with increase in the particle size, exhibiting an asymptotic trend for particles beyond $350 \mu\text{m}$. Sadovnikov and Gusev (2018) and Gómez-Tena et al. (2014) also observed a similar trend. A good correlation ($R^2 = 0.994$) was observed between the specific surface area (S) and median particle size (D_{50}) of pulverized coals, and the relationship obtained is given by

$$S = 18.39 (D_{50})^{-1.14} \quad (4)$$

This equation can be used for predicting the specific surface area of pulverized coals of known median size.

Fig. 8

3.5 Effect of particle size on BET surface area and N_2 adsorption capacity of pulverized coals

The BET surface areas (S_{BET}) of pulverized coals (A, H and I) were measured in order to compare the specific surface areas determined by both the particle sizing and BET methods, and the results are presented in Table 4. The BET method includes the pore and external areas to compute the total specific surface area and provides critical information regarding the effects of particle size and porosity on N_2 adsorption and specific surface area.

Table 4

The N₂ adsorption isotherms of pulverized coal samples of A, H and I for different particle sizes are presented in Fig. 9. From the figure it is evident that N₂ adsorption capacity of coals increases with decrease in the particle size. It means, the finer coal particles adsorb greater quantity of N₂ molecules than the coarser ones. Similar phenomenon was also observed by other researchers (Dudzińska et al., 2017; Hou et al., 2017).

Fig. 9

The variations of BET surface area and monolayer N₂ adsorption capacity with particle size of different pulverized coals of samples A, H and I are shown in Fig. 10. With decrease in particle size from 425-850 to <38 µm, the N₂ adsorption of coal samples A, H and I increased from 0.0513 to 0.6438, 0.7218 to 0.9063 and 0.0341 to 0.4439 cm³/g, or in other words, it increased by 12.55, 1.26 and 13.02 times, respectively. This may be attributed to the fact that as the particle size of coal increases, the mesopores get confined by more constricted pore openings and thereby, reducing the accessibility of mesopores to the N₂ molecules. These constrictions are removed with decrease in particle size by comminution, and consequently, increasing the pore accessibility to N₂ molecules (Cui et al., 2004; Chen et al., 2015; Hou et al., 2017). Thus, with size reduction of coal during mining process, the propensity of coals to spontaneous combustion enhances due to sorption of greater amount of oxygen on the coal surface. Xiumin et al. (2002) reported that when the coal particle size reduced from 83.77 to 19.30 µm, the BET surface area increased by 20.81 times. They explained, the main reasons for increase in the specific surface area with decrease in coal particle size are due to the decrease in average pore diameter and exposure of more and more small pores within the particles.

Among the three samples, N₂ sorption was found to be high in sample H, medium in samples A and low in sample I. This suggests that sample H is of high porous structure and easily accessible to N₂ molecules as compared to the samples A and I.

Fig. 10

Figure 10 also depicts that the variation of BET surface area with coal particle size is consistent with the N₂ adsorption. BET surface area increased with decrease in the particle size of coal. With decrease in particle size from 425-850 to <38 µm, the BET surface area for coal samples A, H and I increased from 0.22 to 2.80, 3.14 to 3.94 and 0.15 to 1.93 m²/g, or in other words, it increased by 12.73, 1.25 and 12.87 times, respectively. A similar trend was also reported by other researchers. Hou et al. (2017) determined the BET specific surface area of original coal and

tectonized coal in five particle size fractions, i.e., 500–1000, 250–500, 125–250, 63–125 and 32–63 μm , and observed an increase in surface area from 0.22 to 3.06 and 1.00 to 2.07 m^2/g in original coal and tectonized coal, respectively with decrease in particle size. The BET surface area decreased in the order of coal samples $\text{H} > \text{A} > \text{I}$, irrespective of the particle size. The high specific surface area of sample H may be attributed to its high porosity (Dudzińska et al., 2017).

Hou et al. (2017) reported that the decrease in particle size makes some inaccessible mesopores accessible to N_2 molecules and increases the mesopore specific surface area and volume. In contrast to mesopore characteristics, the micropore characteristics are independent of particle size. Liu et al. (2010) also observed that the BET specific surface area increases with decreases in the coal particle size, while the mean pore size shows an approximately opposite trend. The reason they cited for this is that, when the particle diameter is reduced, more and more small pores inside the matrix are exposed, resulting in the increase of specific surface area.

3.6 Comparison between particle sizing and BET surface areas

A comparative analysis of surface areas of coal particles determined by both the particle sizing and BET methods was done as shown in Fig. 11. It may be observed that the specific surface areas of coal samples determined by both the methods decrease in the order $\text{H} > \text{A} > \text{I}$. Among the three samples, the specific surface area of sample H was found to be highest in all the particle sizes. This indicates that sample H is highly porous in nature among the three samples.

Fig. 11

An attempt was also made to find the relationships and determine correlation factors between the specific surface areas determined by both the particle sizing and BET methods (Fig. 12). Good linear correlation between the specific surface area values determined by both the methods was observed. In case of samples A and I, the correlation factor was determined about 7 and 5, respectively, which means the BET specific surface area is approximately 7 and 5 times, respectively greater than the particle sizing surface area. Gómez-Tena et al. (2014) determined correlation factor of 6 for ceramic materials. In contrast, in case of sample H, though a good correlation was observed between the surface area values measured by both the methods, the correlation factor was found to be approximately 2 with a greater y-intercept of 3.17. The deviation signifies the highly porous nature of the sample. It

is worth mentioning here that the correlation between the specific surface areas determined by both the methods depends on the characteristics of the coal, and especially on the coal particle size and porosity. The correlation factors determined in this study are specific to the coals analyzed and can be used as a reference for comparison purpose by other studies.

Fig. 12

4. Conclusions

Characterization of coal finds an important place in researches concerning spontaneous combustion of coal and coal dust explosion aiming at safety of coal mines, process industries and utilities sector. This study investigated the physico-chemical properties, such as proximate and ultimate analyses, particle morphology, bulk density, particle size, specific surface area and N₂ adsorption capacity of pulverized coals at five distinct size ranges. The moisture, volatile matter, ash and fixed carbon contents of coals varied in the ranges of 1.12-8.06, 19.27-37.2, 14.23-36.61 and 22.97-58.54%, respectively, and the carbon, hydrogen, nitrogen and oxygen contents of coals varied in the ranges of 36.01-63.34, 3.06-4.51, 1.29-2.71 and 30.41-59.32%, respectively. The coal particles are mostly found to be irregular in shape with angular outlines and sharp edges. Interesting results with respect to the variations of bulk density, specific surface area and N₂ adsorption capacity of pulverized coals with particle size were obtained. The study revealed that the bulk density, specific surface area and N₂ adsorption capacity of pulverized coals greatly influenced by the particle size. The bulk density of pulverized coals increased with increase in the particle size. The bulk density of the smallest (<38 µm) and coarsest size (425-850 µm) coals varied in the ranges of 0.39-1.05 g/cm³ and 0.76-1.35 g/cm³, respectively. With increase in the particle size from <38 to 425-850 µm, the average bulk density of pulverized coals increased from 0.63 to 0.97 g/cm³, or it increased by 1.54 times. The polynomial regression equation obtained between the bulk density and particle size ($y = -0.007x^2 + 0.136x + 0.492$) can be used to predict the bulk density of pulverized coals of known sizes.

The specific surface area of pulverized coals increased with decrease in the particle size. With decrease in particle size from 425-850 to <38 µm, the mean D₅₀ value decreased by 27 times and the mean specific surface area increased by 41.5 times. The relationship obtained between the specific surface area (S) and median particle size (D₅₀) of pulverized coals [$S = 18.39 (D_{50})^{-1.14}$] can be used to predict the specific surface area of pulverized coals of known median size. The monolayer adsorption of N₂ and BET surface area increased with decrease in the particle

size of pulverized coals. With decrease in particle size from 425-850 to <38 μm , the BET surface area for coal samples of A, H and I increased from 0.22 to 2.80, 3.14 to 3.94 and 0.15 to 1.93 m^2/g , or it increased by 12.73, 1.25 and 12.87 times, respectively. This signifies that the finer coal particles are more prone to spontaneous combustion and explosion due to exposure of greater surface area for oxygen sorption. Linear correlation was obtained between the particle sizing and BET specific surface areas of pulverized coals. In case of samples A, H and I, the correlation factors between the particle sizing and BET specific surface areas were determined 7, 2 and 5, respectively.

Acknowledgment

The financial support received for this study from the Science and Engineering Research Board (SERB), Govt. of India [No. EMR/2016/004210] is greatly acknowledged.

Declarations

Ethical approval:

Not applicable. This research does not involve the use of any animal or human data or tissue.

Consent to Participate:

Not applicable.

Consent to Publish:

Not applicable.

Author Contributions:

D. P. Mishra: Conceptualization, Methodology, Data curation, Formal analysis and investigation, Writing - original draft preparation, Writing - review and editing, Funding acquisition, Project administration, Resources.

Funding:

This research was funded by Science and Engineering Research Board (SERB), Govt. of India [Grant No. EMR/2016/004210].

Competing interests:

The author declares no competing interests.

Availability of data and materials:

All relevant data generated during the study are included in the article.

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487 Table 1. Particle size and specific surface area of pulverized coals measured by particle sizing and gas adsorption
488 methods

Type of coal	Coal particle size	Specific surface area (m ² g ⁻¹)	Method of measurement	Reference
Coals of various ranks	<10-100 μm	0.16-0.30	Particle sizing	Linge (1989)
		0.55-1.20	Photoextinction	
		0.4-2.5	Methylene blue dye adsorption	
		1-7	N ₂ adsorption	
		165-311	CO ₂ adsorption	
Medium-volatile bituminous coal	-250 μm	0.7-5.0	N ₂ adsorption	Clarkson and Bustin (1999)
Pulverized coal	14.71-44.26	7.2-6.29	N ₂ adsorption	Liu et al. (2010)
	12.56-52.78	8.56-5.46		
	6.9-33.68	10.88-9.07		
Coals of different ranks	60–80 mesh	0.04-0.93	N ₂ adsorption	Zhao et al. (2016)
		77.4–198.4	CO ₂ adsorption	
Bituminous coal	63-700 μm	0.36-18.95	N ₂ adsorption	Dudzińska et al. (2017)
		90.44-170.12	CO ₂ adsorption (Dubinin-Raduszkiewicz model)	
Original coal	500–1000, 250–500,	0.22-3.06	N ₂ adsorption	Hou et al. (2017)
Tectonized coal	125–250, 63–125 and 32–63 μm	1.00-2.07		

Mean particle diameter				
Coal dust	(D ₅₀):	2.76-0.04	Laser particle size analyser	Liu et al. (2018)
	5.42-190.46 μm			

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Table 2: Results of proximate and ultimate analysis of coal samples

Coal sample	Proximate analysis (w/w %)				Fuel ratio (FC/VM)	Ultimate analysis (w/w %)			
	M	VM	A	FC		C	H	N	O
A	1.24	19.27	25.44	54.05	2.80	60.79	3.58	1.96	33.66
B	1.63	21.1	36.61	40.66	1.93	36.01	3.06	1.61	59.32
C	8.05	33.21	23.26	35.48	1.07	54.44	4.51	1.88	39.17
D	4.33	36.82	35.88	22.97	0.62	44.28	3.57	1.65	50.51
E	7.71	36.84	22.81	32.64	0.89	58.96	4.22	1.97	34.85
F	5.88	30.82	20.88	42.42	1.38	58.00	3.54	2.71	32.47
G	8.06	23.86	31.77	36.31	1.52	46.24	3.30	1.29	45.67
H	6.63	37.2	14.32	41.85	1.13	63.34	4.13	2.12	30.41
I	1.12	26.11	14.23	58.54	2.24	61.81	3.84	1.87	32.47

492 M: moisture; VM: volatile matter; A: ash; FC: fixed carbon

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494 Table 3: Median particle diameter and specific surface area of different sizes of pulverized coals

Particle size (μm)	Median particle diameter (D ₅₀), μm		Specific surface area, m ² /g	
	Range	Mean±SD	Range	Mean±SD
<38	20.37-36.9	29.58 ±4.99	0.239-0.423	0.332±0.052
38-74	39.07-57.57	50.56±6.74	0.163-0.338	0.234±0.051
74-212	117.2-170.4	153.2±18.78	0.043-0.112	0.066±0.027

212-425	343.3-386.0	359.36±13.15	0.017-0.035	0.021±0.006
425-850	763.3-837.3	802.28±28.58	0.007-0.01	0.008±0.001

SD: Standard deviation

Table 4: BET specific surface area and monolayer adsorbed N₂ (Q_m) of pulverized coals of various sizes

Particle size (μm)	Sample A		Sample H		Sample I	
	S _{BET}	Q _m	S _{BET}	Q _m	S _{BET}	Q _m
	m ² /g	cm ³ /g	m ² /g	cm ³ /g	m ² /g	cm ³ /g
<38	2.80	0.6438	3.94	0.9063	1.93	0.4439
38-74	1.33	0.3046	3.54	0.8143	0.82	0.1881
74-212	0.47	0.1075	3.39	0.7783	0.75	0.1724
212-425	0.24	0.0542	3.17	0.7293	0.60	0.1377
425-850	0.22	0.0513	3.14	0.7218	0.15	0.0341

Figure captions

Fig. 1. Pulverized coal samples of various sizes

Fig. 2. SEM pictographs of pulverized coals of different sizes

Fig. 3. Variations of bulk density of different pulverized coal samples with particle size

Fig. 4. Variation of average bulk density of pulverized coals with particle size

Fig. 5. Particle size distributions of pulverized coals of different sizes: (a) < 38 μm, (b) 38-74 μm, (c) 74-212 μm, (d) 212-425 μm and (e) 425-850 μm

Fig. 6. Variations of D₅₀ and specific surface area of pulverized coals with particle size

Fig. 7. Variations of average D₅₀ and specific surface area of pulverized coals with particle size

Fig. 8. Relationship between the median particle size (D_{50}) and specific surface area of pulverized coals

Fig. 9. N_2 gas adsorption isotherms of coal samples at different particle sizes: (a) Sample A, (b) Sample H, (c) Sample I

Fig. 10. Variations of BET specific surface area and monolayer adsorbed N_2 with particle size of different pulverized coal samples

Fig. 11. Comparison between the particle sizing and BET specific surface areas of pulverized coal samples of different sizes

Fig. 12. Relationship between the particle sizing and BET specific surface areas of pulverized coals

Fig. 1.

(a) $< 38 \mu\text{m}$ (b) $38\text{-}74 \mu\text{m}$ (c) $74\text{-}212 \mu\text{m}$

(d) 212-425 μm (e) 425-850 μm

Fig. 2.

Fig. 3.

Fig. 4.

Fig. 5.

Fig. 6.

Fig. 7.

Fig. 8.

Fig. 9.

Fig. 10.

Fig. 11.

Fig. 12.