Research Journal of Environmental Sciences

ISSN 1819-3412

Academic Journals Inc.

www.academicjournals.com

OPEN ACCESS
Research Journal of Environmental Sciences
ISSN 1819-3412
DOI: 10.3923/rjes.2018.98.105

Research Article
Chemical, Mineralogical and Morphological Investigation of Coal Fly Ash Obtained from Mpumalanga Province, South Africa

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Abstract

Background and Objective: Coal fly ash generated from the combustion of coal by thermal power plant stations are enormous and the disposal there of is a big problem. In the present study, the chemical, mineralogical and morphological characterization of coal fly ash samples (CFAs) obtained from Mpumalanga province, South Africa were investigated. **Materials and Methods:** The CFAs were characterised by X-ray diffraction (XRD), X-ray fluorescence (XRF), scanning electron microscopy-energy dispersive spectroscopy (SEM-EDS), high resolution transmission electron spectroscopy (HRTEM) and the selected area electron diffraction (SAED) analytical technique. **Result:** Experimental results showed that the CFAs contain hematite, magnetite, calcite, lime, mullite and quartz. The CFAs are polycrystalline, could be categorized as class F fly ash and the particles are spherical in shape. **Conclusion:** The phase characterization suggested that CFA could be used for the synthesis of nanoparticles, as well as extraction of valuable metals, more so, the elemental composition of the CFAs suggested that the storage and disposal of CFAs could lead to the release of salts and toxic elements into the environment, thereby, contaminating surface and ground waters.

Key words: Coal fly ash, physicochemical properties, thermal power plant, mineralogical characterization, morphological characterization

Citation: John Alegbe, Olushola Sunday Ayanda, Patrick Ndungu, Nechaev Alexander, Ojo Olanrewaju Fatoba and Leslie Felicia Petrik, 2018. Chemical, mineralogical and morphological investigation of coal fly ash obtained from Mpumalanga province, South Africa. Res. J. Environ. Sci., 12: 98-105.

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Competing Interest: The authors have declared that no competing interest exists.

Data Availability: All relevant data are within the paper and its supporting information files.

INTRODUCTION

Coal fly ash (CFA) is a solid waste formed from the combustion of coal in thermal power stations to generate electricity. The CFA is produced at a temperature between 1200-1700°C from feed coal containing organic and inorganic materials. During the combustion of coal at very high temperature, the minerals in the coal melt to become fluid and on cooling form amorphous spherical CFA particles[1]. Consequently, the quality of CFA generated depends on the type of coal, chemical composition, boiler operating conditions and process undergone before combustion[2]. About 40 million tons of CFA are produced annually in South Africa by power stations[3] and Syngas processes in Sasol plants[4]. The volumes of CFA generated from the combustion of coal by thermal power plant stations are enormous and the disposal thereof is a big problem. The CFA production has led to the encroachment of agricultural land for food production. The exposure of CFA in the open for a long time can cause surface and underground water system interaction by leaching to contaminate the water aquifer depending on the degree of the weathering process[5]. Serious environmental problems may occur from the dispersal of silica minerals with elevated concentrations of specific trace elements in a quartz-mullite matrix[6]. Trace elements in CFA are slightly mobile in very high alkaline medium and it can either be considered as an ecological nuisance due to the environmental effect and high economic costs during disposal or can be regarded as a valuable raw material when utilized in an economical and beneficial way[6]. The CFA is highly contaminated because it captures potentially toxic elements which condense during the cooling of combustion gases[7].

Although, CFA generated from the combustion of coal in thermal power stations has found its usefulness in cement and concrete industry, construction industry, agriculture, low-cost adsorbent, erosion control, geopolymers, magnetic sphere recovery, catalyst, carbon recovery, zeolites etc. The CFA is still under utilized for beneficial purposes due to the huge amounts produce in different countries. The World Bank reported that India will require 1000 km^2 of land to dispose CFA generated until 2015[8]. The disposal problem has prompted the need to seek for new and innovative ideas to alleviate the effect of CFA on the environment. Therefore, several investigations worldwide are focusing on the characterization of CFA in order to evaluate its environmental impact and its significance as a material resource for industrial applications[9]. One way of combating the problems caused by CFA is by optimizing its use so that it could become a valuable raw material.

In this study, authors present and discuss the results of the characterization of fresh fly ash samples (CFAs) obtained from various sampling sites (Hendrina, Kriel, Matla, Secunda and Tutuka) in Mpumalanga province, South Africa.

MATERIALS AND METHODS

Sampling: Fresh CFAs were collected in 2014 from 5 different power plant stations namely, Hendrina, Kriel, Matla, Secunda and Tutuka. The power stations were all situated in Mpumalanga province, South Africa (Fig. 1).

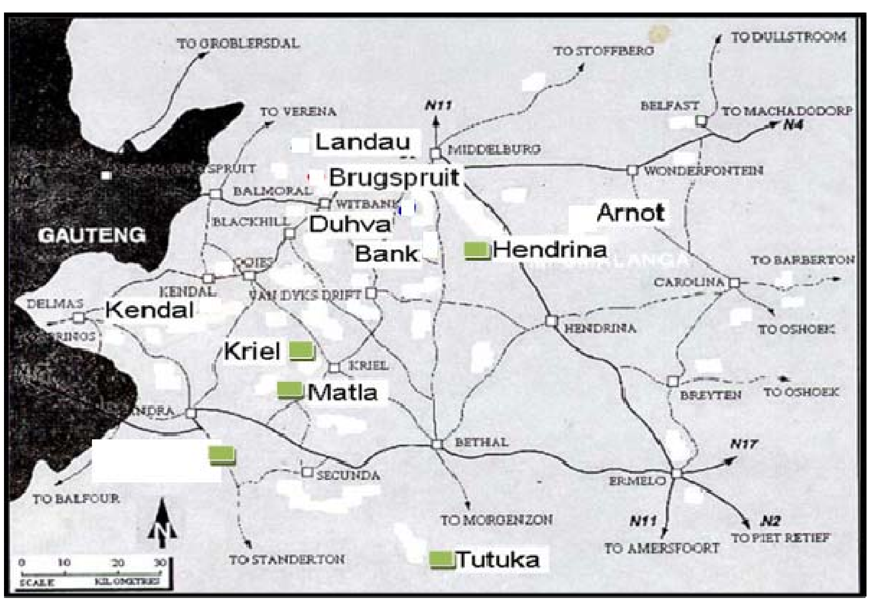

Fig. 1: Locations of pulverized coal-fired thermal power plant stations in the Republic of South Africa

The CFAs were collected directly from the ash handling conveyor belt of the different power stations. The samples were sealed in plastic bags tightly closed to prevent ingress and absorption of air to minimize the reaction of CaO with CO_2 to form calcite ($CaCO_3$) which could reduce the CaO content.

Instrumentation: The mineralogical characterization of the CFAs was carried out by X-ray diffraction (XRD). Morphological characterization was conducted by scanning electron microscopy (SEM), high resolution transmission electron spectroscopy (HRTEM) and selected area electron diffraction (SAED). The chemical investigation was carried out by X-ray fluorescence (XRF) and energy dispersive spectroscopy (EDS), an attachment to SEM.

Statistical analysis: The data obtained on the elemental composition of CFAs were subjected to descriptive statistical analysis such as mean and standard deviation using statistical packages for social sciences (SPSS) software (version 17.0 for Windows; SPSS Inc., Chicago, IL).

RESULTS AND DISCUSSION

Mineralogical composition of coal fly ash: The XRD results of the CFAs are presented in Fig. 2. The results revealed that the CFAs contained magnetic iron fractions such as hematite, magnetite and non-magnetic fractions such as calcite, lime, mullite and quartz. In all the analysed CFAs, mullite ($3Al_2O_3.2SiO_2$) and quartz (SiO_2) were the major non-magnetic fractions, while magnetite and hematite were the major magnetic fractions. This agrees with the reports by Hower *et al.*[10] and Shoumkova[11].

The mineralogical analysis showed that the CFAs contained different mineral phases, an indication that CFA could be used as a source of raw material for the synthesis of nanoparticles, as well as extraction of valuable metals from CFA.

Morphological studies of coal fly ash: The surface and internal morphological analyses of CFAs were conducted by means of SEM and HRTEM-SAED analytical techniques. The results of the morphological analysis by SEM are presented in Fig. 3.

Figure 3 shows that the particles of the CFAs are spherical in shape as well as irregular non-spherical shapes with sizes ranging from 0.2-25 µm. Some of the CFAs comprise of broken and unbroken microspheres. The images revealed the aggregation of the microspheres with small particle size, attached to surfaces which formed large aggregates of microspheres with different sizes[12]. The morphology of broken microspheres in the CFAs contained cenospheres and plerospheres as observed from the SEM micrograph. Cenosphere microspheres are hollow light weight microspheres with cavities containing only gas inside them. Plerospheres are hollow light weight microspheres cavities filled with small mineral particles, spongy, foam or other porous frame work inside it[13,14]. Plerosphere microspheres are noticeable in all the CFAs, this is similar to the results obtained in the literature[9].

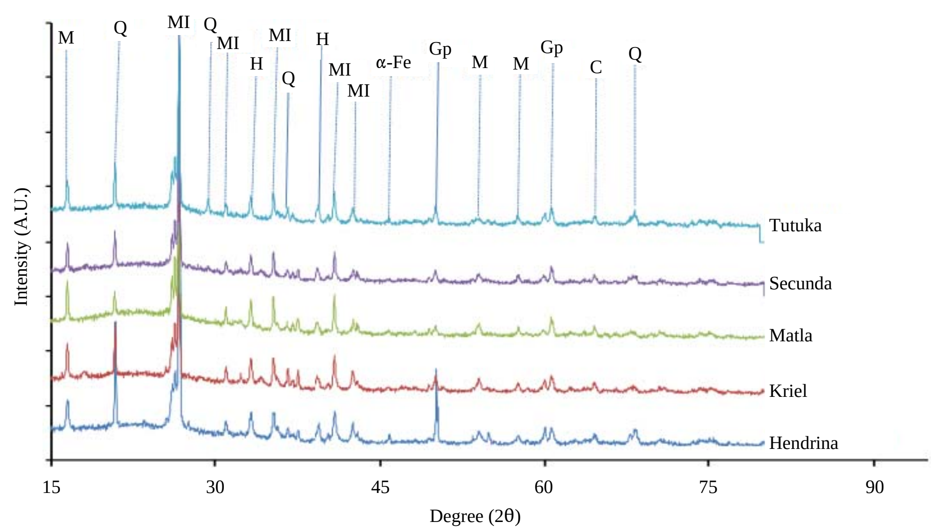

Fig. 2: XRD diffractograms of CFA collected from Hendrina, Kriel, Matla, Secunda and Tutuka power plant stations (Ml: Mullite, C: Calcite, H: Hematite, M: Magnetite and Q: Quartz, GP: Gypsum, α-Fe: Syn Fe)

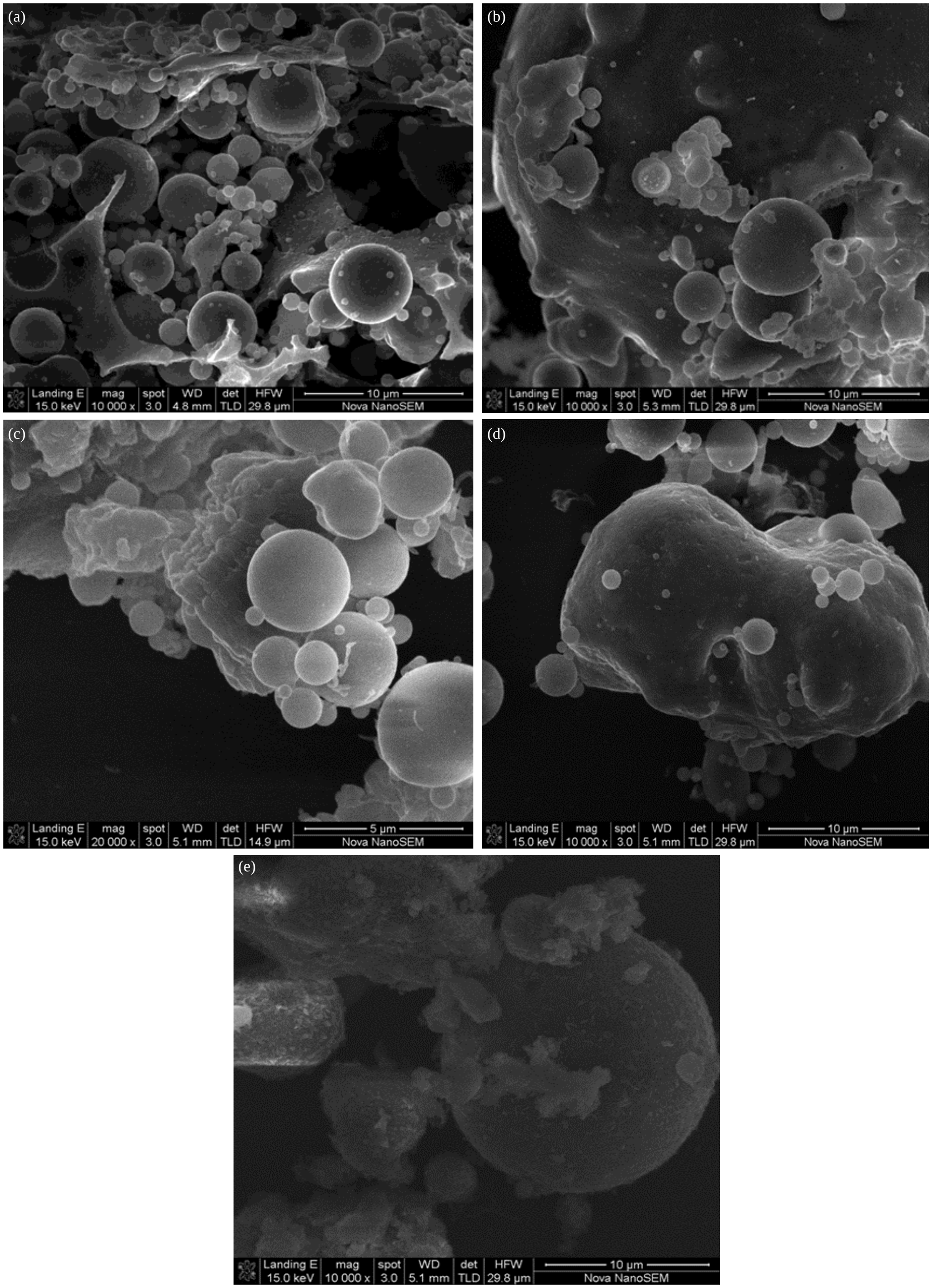

Fig. 3(a-e): SEM micrograph of CFA from (a) Hendrina, (b) Kriel, (c) Matla, (d) Secunda and (e) Tutuka power plant stations

The HRTEM was considered in order to investigate the structure and crystallinity of the CFA particles. The HRTEM and SAED results of the CFAs are presented in Fig. 4.

The morphology of the CFAs showed that the particles are spherical in shape with some irregular shaped structures in agglomerated form. The high resolution images obtained for

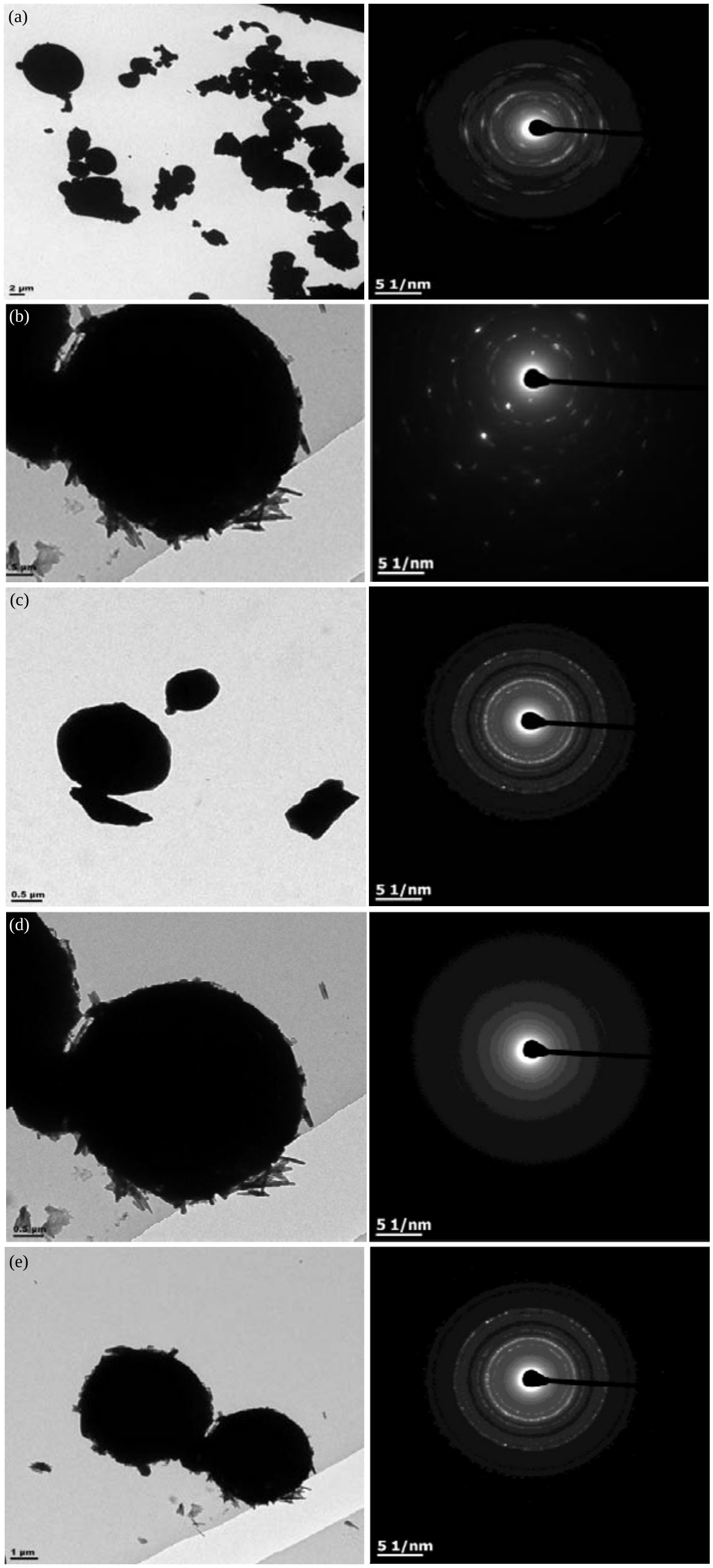

Fig. 4(a-e): HRTEM image (left) and SAED (right) of CFA from (a) Hendrina, (b) Kriel, (c) Matla, (d) Secunda and (e) Tutuka

Table 1: EDS elemental composition of CFAs (n = 3)

Elements	Hendrina CFA	Kriel CFA	Matla CFA	Secunda CFA	Tutuka CFA
Al	10.52±8	15.05±1.95	17.21±2.01	10.5±4.97	11.69±5.82
Ca	5.55±6.98	3.04±1.21	3.31±2.29	17.78±13.73	4.18±2.24
Cl	0.44±0.77	ND	ND	ND	ND
Fe	1.07±1.33	1.04±0.34	0.97±0.85	1.15±0.12	2.26±2.35
K	ND	0.73±0.22	0.59±0.3	0.46±0.28	0.7±0.37
Mg	1.81±3.13	ND	ND	1.31±0.63	1.28±0.15
Na	0.59±1.03	ND	ND	ND	ND
O	63.93±6.27	56.04±1.66	56.12±1.52	53.51±3.39	54.37±3.04
S	0.52±0.63	ND	ND	ND	ND
Si	15.56±9.69	22.25±2.05	20.74±1.18	14.52±6.29	24.13±3.57
Ti	ND	ND	1.06±0.67	0.77±0.41	1.4±0.8

ND: Not detected, Mean±SD

Table 2: Elemental composition of major elements (as oxides in % w/w) in CFAs (n=3)

Metal Oxide	Hendrina	Kriel	Matla	Secunda	Tutuka
SiO_2	53.83±1.18	52.43±0.67	48.35±0.29	49.75±0.17	51.80±0.30
Al_2O_3	26.06±0.10	30.81±0.55	30.89±0.16	26.55±0.56	24.53±0.15
CaO	4.80±0.03	7.66±0.29	6.79±0.12	9.47±0.31	5.91±0.20
Fe_2O_3	4.52±0.06	2.58±0.32	2.91±0.09	2.69±0.29	5.40±0.00
MgO	1.87±0.12	2.06±0.04	2.11±0.04	2.25±0.02	1.41±0.01
TiO_2	1.39±0.07	1.18±0.12	1.26±0.02	1.58±0.02	1.51±0.01
P_2O_5	0.56±0.02	0.45±0.03	0.90±0.03	0.89±0.03	0.50±0.00
K_2O	0.79±0.02	0.55±0.03	0.84±0.01	0.81±0.04	0.80±0.01
Na_2O	0.03±0.01	0.66±0.04	0.55±0.03	0.61±0.01	0.30±0.00
SO_3	4.78±3.47	0.05±0.01	0.20±0.04	0.05±0.00	0.10±0.00
MnO	ND	0.03±0.0	0.02±0.00	0.02±0.00	ND
Cr_2O_3	0.04	ND	ND	ND	ND
NiO	0.01	ND	ND	ND	ND
LOI	ND	1.54±0.37	5.12±0.46	4.61±0.27	7.81
Sum	98.68	100.33	99.93	99.28	100.07
Si/Al	2.07±0.05	1.70±0.04	1.57±0.00	1.87±0.03	2.11±0.00

ND: Not detected, Mean±SD

the CFAs revealed that they are opaque spheres. The irregular non-spherical shaped images could be regarded as the amorphous opaque structures. The SAED revealed that the particles are polycrystalline as shown by the tiny spots scattered round in a ring presented in the Fig. 4a-e.

Elemental composition of coal fly ash: The EDS analysis of CFAs is a qualitative analysis, it was used for elemental identification. The EDS qualitative analysis of CFAs are shown in Table 1. Table 2 presented the elemental composition of major elements (as oxides in % w/w) in the CFAs by XRF.

There are variations in the elemental composition of the CFAs and this can be attributed to the difference in the geological composition of the mined coal sources. The SiO_2 and Al_2O_3 are dominant in all the samples. All the CFA contained over 50% of SiO_2 except for Matla and Tutuka samples whose Si content was between 48-49%. The composition of Al_2O_3 in the CFAs was in the order Matla>Kriel>Secunda>Hendrina>Tutuka. The Kriel and Matla CFAs contained over 30% Al_2O_3 while Hendrina, Secunda and Tutuka contained between 24-26%. The content of lime (CaO) in the CFAs shows that the degree of alkalinity of CFAs was in this order Secunda>Kriel>Matla>Tutuka>Hendrina. The degree of iron composition in the CFAs was in this order Tutuka>Hendrina>Matla>Secunda>Kriel. The composition of MgO in the CFAs was in this order Secunda>Matla>Kriel>Hendrina>Tutuka, whereas, the composition of TiO_2 in the samples was in this order Secunda>Kriel>Matla>Tutuka>Hendrina. The elemental characterization shows that all the CFAs showed similar trends in the composition of their major elements.

According to the American society for testing and materials (ASTM) classification method of CFA[15], all the CFAs could be categorized as class F fly ash. This is because the summation of the percent composition of the major mineral phases: SiO_2, Al_2O_3, CaO, MgO, Na_2O and Fe_2O_3 present in the CFAs was above 70%. Many of the results obtained for the elemental analysis of these CFAs agree with that of previous studies[11,16,17]. The loss of mass on ignition (LOI) was used to measure the unburnt carbon content present in the CFAs. The unburnt carbon could be used to measure the combustion rate of coal feed during electricity generation from thermal power stations. The LOI of the CFAs are in this order Tutuka>Matla>Secunda>Kriel>Hendrina.

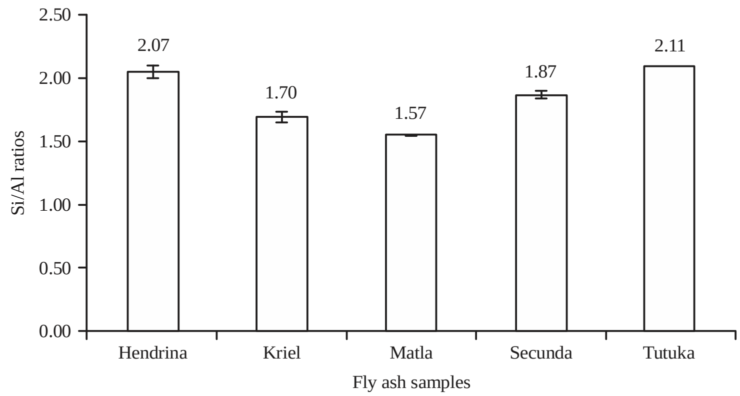

Fig. 5: SiO_2/Al_2O_3 ratios of CFAs (n = 3)

The results of the SiO_2/Al_2O_3 ratio of the CFAs are presented in Fig. 5. The SiO_2/Al_2O_3 ratio for Hendrina and Tutuka fly ash are above 2, whereas, those of Kriel, Matla and Secunda CFAs are below 2. Figure 5 shows that the Si/Al ratio of the CFAs was in the order Tutuka>Hendrina>Secunda>Kriel and Matla.

The results obtained indicated that CFAs are mostly aluminosilicate materials that also contained Ca, Fe, Na, Mg, K and Ti in their oxide forms, as well as unburned carbon and other elements in trace amount, some of which could accumulate with time and be toxic[17]. Thus, the storage and disposal of CFA could lead to the release of leached toxic metals and oxides into the environment[18].

CONCLUSION

The CFAs from Hendrina, Kriel, Matla, Secunda and Tutuka power stations were collected and analysed for their chemical, mineralogical and morphological properties. The morphological studies showed that the particles of the CFAs are spherical with some irregular shaped structures in agglomerated form. The mineralogical compositions of the CFAs are hematite, magnetite, mullite, quartz, gypsum and calcite. The composition of SiO_2, Al_2O_3 and Fe_2O_3 in the CFAs is above 70% which is an indication that all the CFAs belong to class F fly ash. The CFAs from the different power plant stations are relatively consistent in their major oxide contents but the sulphate in Hendrina CFA could be due to the electrostatic precipitator dosing with sulphuric acid. In conclusion, the CFAs contained significant amount of useful metals and oxides, therefore, it is recommended that industrial application be devised for CFA rather than disposal, the disposal of CFA could lead to the release of various salts and toxic elements into surface and ground waters.

SIGNIFICANCE STATEMENT

This study discovers valuable metals and oxides in fly ash that can be beneficial for industrial applications. It also established the potential health hazards associated with fly ash storage and disposal. The study will help researchers to uncover the critical areas of the beneficiation of fly ash that many researchers were not able to explore.

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