Comparative analysis of milling time on the particle sizes of coal fly ash and wood fly ash using Otsu Method for thresholding

O. M. Ikumapayi1, E. T. Akinlabi1,2, P. A. Adedeji1, S. A. Akinlabi2,3

1Department of Mechanical Engineering Science, University of Johannesburg, Johannesburg, 2006, South Africa
2Department of Mechanical Engineering, Covenant University, Nigeria
3Department of Mechanical and Industrial Engineering, University of Johannesburg, Johannesburg, 2006, South Africa
Corresponding Author: oikumapayi@uj.ac.za

Abstract-
South Africa remains one of the countries with an abundance of coal fly ash (CFA) emerging from her abundant coal deposit. Despite the hazardous and environmental unfriendliness of fly ash, so many benefits can be derived therein. With new policies of waste-to-wealth, the country seeks ways by which this resource could be of value addition. However, CFA in its raw form often requires further milling operation to enhance suitability for the intended use. This study investigated the effect of milling time on the particle size of CFA and wood fly ash (WFA) using image segmentation. Both CFA and WFA received at micro-sized particles were washed with distilled water to remove impurities and dried in the oven at 80°C for 48 hours then sieved using 75µm size. Microstructural images of both CFA and WFA milled at varying times ($t = 0, 20, 40, 60$ minutes) were morphologically and physiochemically analysed using scanning electron microscope (SEM), X-ray diffraction (XRD), Energy Dispersive X-ray (EDX), and X-Ray Fluorescence (XRF). SEM images of CFA and WFA were segmented using Otsu thresholding technique and average particle sizes were estimated. CFA contains a higher composition of Al$_2$O$_3$ (30.93%) and SiO$_2$ (51.43%) compared to WFA, which has 10.70% and 46.31% respectively. However, WFA contains more of Fe$_2$O$_3$ (17.28%) than CFA (2.29%). The number of particle size increased with increases in milling time while the particle area decreased with an increase in milling time. At a 95% confidence interval, there exists a significant difference between results obtained at different milling time. Also, a significant difference exists between the mean particle diameters of the two ash sources.

Keywords: Coal fly ash; image segmentation; particle size distribution; Otsu thresholding; wood fly ash

1. Introduction

Material reinforcements from green technology have widely gained acceptance over the years. Fly ash (FA), which is obtained from coal-fired plants [1] is an example less explored. About 184.14 million tons of FA was generated between 2014 to 2015 with only 44.31% unutilized [1]. Within the same period, South Africa produced about 34.4 million tons of ash (fly ash, bottom ash, fresh and weathered ash) with only 7% sold from 6 of its 13 coal-fired power stations [2]. Its abundance in coal-fired plants has raised global interest in exploring more of its economic use. Recent studies have shown that FA is good fillers in polymers and rubbers for...
minimal production cost and enhancement of certain mechanical properties [3]. The synthesis of geopolymers from FA has also been established to have a high prospect of acting as an alternative cementitious material for concrete mix [4]. Some other areas of application where FA has been deployed include its use in acid mine drainage treatment [5], hydrotalcite synthesis [6], zeolites synthesis [7]–[10], Following the wake-up call for the global material efficiency policies, the use of alternative materials like fly ash for reinforcement is on the top list, most especially in the reduction in clinker-to-cement ratio for manufacturing [11].

South Africa generates about 77% of its electricity from 13 coal-fired power plants. With the present utilization rate, the country has about 200 years of coal supply left [12]. Despite the increase in FA generation on an annual basis, it is less utilized in the country as shown in Figure 1.

Figure 1. Estimate in 2016 of annual FA production in Megatons per coal-fired power plant in South Africa [2]

However the FA produced is known for its fine spherical particulate nature, almost zero carbon content with high pozzolanic activity and unusual high consistency [13]. The annual generation of FA from these power plants has triggered new policies on the more economic use of the product to reduce its environmental impact and increase revenue from its market. Despite the fineness of the product, there is a need for milling the ash collected to the desired size. This is because the FA is not separated from the bottom ash, which is associated with large particle size during disposal. The performance, efficiency, and effectiveness of fly ash in any application depend wholly on its particulate nature: macro, micro or nano scale. Milling to smaller particle size aids fast absorption and reaction of FA in any application. The effect of milling time is essential in determining the final microstructure for effectiveness in the desired reinforcement process. For example, particle size distribution controls the effect of water demand and workability of cement paste, which in turn affects its cementitious activity [14]. Milling changes both the particulate nature as well as the microstructure of substances, which affects the coagulative characteristic.

A significant number of studies have been performed on South Africa CFA. Some of these include characterization of its physical and surface properties modified by Sodium Lauryl Sulphate (SLS) [3]. A proposed application of this is seen in the construction of PVC
composites. In the study, a surface modification at multiple conditions was performed at different conditions using the surfactant, SLS. The study concludes on the feasibility of CFA as an alternative to CaCO$_3$ in PVC filling under low filler loadings. Similar to this study is the investigation of the transformation of the CFA into useful compounds to be used in other fields for the economic purpose [8], [9], [15]. Of keen interest to study is the investigation of the effect of grinding time on gasification ash, and Portland Cement clinker [14]. The gasification ash possesses similar which has a similar chemical composition as the South African coal fly ash. Separate grinding on the two samples was performed as well an interground of both samples at the microscale.

Optical granulometry has advanced in several forms over the years. In the past, visual inspection has been in material science for image inference across varying parameters. However, visual inspection is limited to macro-level judgment and physical properties of size and intensity. Several techniques have been developed in recent times to study microstructural images. One of the notable empirical functions, which estimates the particle size distribution is the Rosin-Rammler (RR) distribution function [14], [16]. The function describes the particle size distribution of powders with various sizes and types. It has been tested to be effective in representing powders made by crushing, grinding, and milling operations [17]. The RR model is generally represented by:

$$F(d) = 1 - \exp\left[-\left(\frac{d}{l}\right)^m\right]$$

(1)

where:

- $F(d)$ = distribution function
- $d$ = particle size
- $l$ = mean particle size
- $m$ = measure of spread of particle sizes

the parameters $l$ and $m$ are adjustable parameters peculiar to the distribution. Equation (1) can then be re-written as:

$$\ln\{-\ln[1 - F(d)]\} = m \times \ln(d) - m \times \ln(l)$$

(2)

such that a plot of $\ln\{-\ln[1 - F(d)]\}$ against natural $\ln (d)$ results in a straight line with slope $m$, if the material behaviour fits the RR model. RR model was applied to determine the particle distribution of gasification ash and Portland cement clinker at varying grinding time by [14]. The study established a significant difference between the particle size distribution of gasification ash and Portland cement clinker at same the time interval. The study demonstrates that cement clinker is harder than the gasification ash. On the contrary, in our study, the milling process was investigated on CFA and WFA rather than the grinding operation.

Image segmentation involves breaking-up a digital image into diverse sets of pixels [18]. This helps to better analyse intricate features digitally represented in the image. Image thresholding forms an integral and important process in image segmentation [19]. Image thresholding essentially separates the foreground of an image from its background [20], [21]. This tool becomes effective when significant gray levels exist between background images and objects from which separation is to occur [22]. With the emergence of several classical image segmentation algorithms like histogram shape-based techniques, clustering-based techniques, mutual information techniques, local adaptive techniques, Otsu technique has gained more popularity[19]. In the past decades, there have been improvements on the technique to address
bias between the image background and the object in a case when the object is brighter and the variance is larger [23], [24]. The application of Otsu’s technique requires knowledge about the intensity difference and the object size for successful thresholding. To complement other studies, this study uses a sequence of an algorithm written in MATLAB to estimate the particle size at varying milling time. This is seen as a means of automating the process, such that particle size can be determined in real time during milling by using optimal imagery of samples. Thresholding becomes a potent tool for image segmentation when the variance of the object differs significantly from that of the background. This is often the case in SEM images and change monitoring applications [25]. The ideal threshold for segmentation is the intersection point of the image and its background. This is such that the intersection is far from the class with large variance [19].

Despite the presence of heavy metals in fly ash, which have raised environmental concerns and classified it at a hazardous material, a regulation has been handed to the Department of Environmental Affairs (DEA) in South Africa for an exclusion from the hazardous list. This will foster its use in cement and brick making, soil amelioration, mine backfilling, and road construction in the country [2]. This study is therefore aimed at (i) to investigate the morphological characteristics of CFA and WFA (ii) to determine the effect of varying milling time on the average particle size of CFA and compares it with that obtained from WFA source using image segmentation approach. (iii) to compare the image statistics of both ash sources at different milling time for statistical significance. The novelty of this study is in the determination of the particle size of CFA and WFA using image segmentation approach. The rest of the paper is structured as follows: the materials and methodology used for milling, image acquisition and segmentation of coal and wood fly ash are presented in Section 2. Section 3 presents the results of microstructural and statistical analysis of both images at different milling time. The discussion of findings is presented in section 4 of the work while section 5 concludes the paper.

2. Materials and Methods

2.1 Material collection and processing

The CFA used in this study was collected from the ash dump of Majuba Power Station; a coal-fired power station located in Mpumalanga Province of South Africa. In the plant, as the pulverized coal undergoes burning in the boiler, part of the ashes falls to the bottom of the system (bottom ash), however, others escape with the flue gas but are trapped using filter bags. The two categories of ashes are channeled to the ash dump.

WFA used in this study was obtained after a bunch of firewood was subjected to complete combustion until ash was formed. WFA formed were washed with distilled water to remove unwanted particles. After discharging and draining, the WFA was dried inside an electric oven at a temperature of 80°C for 48 hours. The resulting ash was sieved using a sieve size of 75µm on a KingTest Sieve Shaker (VB 200/300) operated at 220V/50Hz and 5A. The proportions that passed through the mesh size was then taken for milling. Similar processing was performed on CFA collected in its raw state from the coal power station.

2.1.1 Vibratory Disc Milling Machine (VDMM)

Milling operation was performed using a digital vibratory disc grinding mill Lab Pulveriser (Model 2MZ-200). The machine has specification as presented in Table 1.
Table 1. Vibratory disc milling machine specification

| Property             | Specification       |
|----------------------|---------------------|
| Dimension            | 740 x 740 x 950 mm  |
| Number of bowls      | 2                   |
| Capacity per bowl    | 200g                |
| Feed Size            | < 15mm              |
| Motor                | 380V/50Hz, 1.5KW    |
| Motor speed          | 940rpm              |

Dry mechanical milling at varying milling time of 20, 40 and 60 minutes was performed on the two ash samples. The machine was thoroughly washed, dried and cleaned with acetone before and after use to remove any contaminants that may be present. For WFA and CFA, 40g of each were charged into each bowl of the VDMM simultaneously and then set for milling. The machine was interrupted every 10 minutes of operation in order to avoid a rise in temperature and to limit adherence of the powder within the container walls. A cooling interval of 30 minutes was ensured before the next running. Samples of CFA and WFA were milled at 20 minutes interval up to 60 minutes.

2.2 Microstructural characterization
The milled and un-milled CFA and WFA were analyzed through various microstructural characterization processes. For this study, the Scanning Electron Microscopy (SEM), Energy Dispersive X-ray (EDX), X-Ray Diffraction (XRD), and X-Ray Fluorescence (XRF) were used. The processes involved are discussed as follows.

2.2.1 Energy Dispersive X-Ray Spectroscopy (EDXS)
The elemental composition of un-milled samples of CFA and WFA was analysed using EDXS. The variation in the trend of the composition of each element present across the un-milled samples was identified.

2.2.2 X-Ray Fluorescence (XRF) and X-Ray Diffraction (XRD)
In this study, the chemical composition of both CFA and WFA were analyzed at 75 µm using X-Ray Fluorescence (XRF) spectroscopy (model PHILIP PW1404 XRF) Wavelength Disperse Spectrometer. Principal chemical compositions of both CFA and WFA obtained were compared with selected similar studies. Similarly, the XRD test was performed using the PHILIPS X’Pert (model number 12NC: 943003040601) operated at PW: 3040/60, 240V, 8.5KVA, and 50Hz and with the specifications in Table 2. XRD test was carried out on the ash samples to confirm the crystal structure and mineralogical compositions of the samples used. The XRD was acquired using copper Kα radiation (λ = 1.5406 Å) and kβ radiation (λ = 1.39225 Å) and an automatic divergence slit; i.e., an irradiated sample length that is independent of the Bragg angles ((2θ) in degree). The diffraction patterns and crystal phases at different milling times were obtained.

Table 2. Diffractometer machine specification

| Property     | Specification |
|--------------|---------------|

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2.2.3 Scanning Electron Microscope (SEM)
A TESCAN model, type VEGA 3 LMH was the type of SEM machine that was employed for the study. In order to have the sample more conductive and to have better resolution, the samples were sputter-coated with a thin layer of carbon just before the SEM analyses coupled with Energy Dispersive Spectrometer (EDS) analyses. The beam intensity used in the analysis was 12 and the accelerating voltage used was 20KV, all micrographs were taken at SEM magnification of 1000 (1.0 kx). The particle size and surface of both CFA-NPs and WFA-NPs were analysed at different milling times 20, 40 and 60 mins and also the un-milled (0 min) were also analysed by SEM.

2.3 Image thresholding and segmentation
Otsu’s method [26] was used for image thresholding. The technique is a nonparametric as well as unsupervised thresholding technique, which selects optimum thresholds from a maximization of the intraclass variance of binary images. A significant assumption in the algorithm is the bimodal class of the image: the foreground and background pixels which the SEM images in this study satisfy. The method is apparently a 1-dimensional. The global optimal threshold value was obtained for the SEM images.

Given the local thresholds \( t_1, t_2, t_3, \ldots, t_n \) to be selected from, the thresholds subdivide the images into \( n+1 \) classes of \( C_1, C_2, C_3, \ldots, C_n \) by maximizing the objective function given by the sum of all threshold variances [24];

\[
J_1(t_1, t_2, t_3, \ldots, t_n) = \sigma_0^2 + \sigma_1^2 + \sigma_2^2 + \sigma_3^2 + \cdots \sigma_n^2 \tag{1}
\]

where

\[
\sigma_0^2 = \omega_0 (\mu_0 - \mu_T)^2, \omega_0 = \sum_{i=0}^{t_1} p_i, \mu_0 = \sum_{i=0}^{t_1} \frac{ip_i}{\omega_0}
\]

\[
\sigma_1^2 = \omega_1 (\mu_1 - \mu_T)^2, \omega_1 = \sum_{i=t_1}^{t_2} p_i, \mu_1 = \sum_{i=t_1}^{t_2} \frac{ip_i}{\omega_1}
\]

\[
\sigma_2^2 = \omega_2 (\mu_2 - \mu_T)^2, \omega_2 = \sum_{i=t_2}^{t_3} p_i, \mu_2 = \sum_{i=t_2}^{t_3} \frac{ip_i}{\omega_2}
\]

\[
\sigma_3^2 = \omega_3 (\mu_3 - \mu_T)^2, \omega_3 = \sum_{i=t_3}^{t_4} p_i, \mu_3 = \sum_{i=t_3}^{t_4} \frac{ip_i}{\omega_3}
\]

\[
\sigma_n^2 = \omega_n (\mu_n - \mu_T)^2, \omega_n = \sum_{i=t_n}^{t_{n+1}} p_i, \mu_n = \sum_{i=t_n}^{t_{n+1}} \frac{ip_i}{\omega_n}
\]

and \( \sigma_0^2, \sigma_1^2, \sigma_2^2, \sigma_3^2, \ldots, \sigma_n^2 \) are the class variances, \( \omega_0, \omega_1, \omega_2, \omega_3, \ldots, \omega_n \) represents the class probabilities, \( \mu_0, \mu_1, \mu_2, \mu_3, \ldots, \mu_n \) represents the mean values of the segmented classes. The intensity of the whole image is \( \mu_T \leq \mu_T = \omega_0 \mu_0 + \omega_1 \mu_1 + \omega_2 \mu_2 + \omega_3 \mu_3 + \cdots + \omega_n \mu_n \) and \( \omega_0 + \omega_1 + \omega_2 + \omega_3 + \omega_n = 1 \).
Segmentation of the SEM images was performed at different milling time using a MATLAB script implemented in MATLAB R2015a installed on a desktop computer workstation with configuration 64 bits, 32GB RAM Intel (R) Core (TM) i7 5960X. Shown in Figure 2 is the graphical flow chart of the process.

Threshold Segmentation is one of the easiest methods of Image processing to detect morphological maps[27] and image colour intensity and one of the commonest parallel techniques of segmentation. The method uses a segmentation algorithm which sectionalised image grayscale processed information based on the targets of various gray value[28]. Grayscale Images are converted into binary Images in this method and the binary images consist of the whole important data in respect of the shapes and the location of the objects. This conversion into a binary image is of importance since it reduces data complexity. One of the merits of threshold segmentation method has been that the speed of operation is fast and the calculation is simple[29]. Image segmentation is an essential ingredient to distinguish foreground pixel from background pixel as well as enhances better visual perception of the Image[30]. It helps in recognising the patterns of the Images by so doing gives high quality and resolutions of the final result of the analysis[31]. One must not shy away from demerits of threshold segmentation which was the inability to get accurate results where there is no adequate variation in grayscale or enough overlap of the grayscale values in the Image[32], [33]. It is worth to mention that it is sensitive to noise and uneven grayscale as a result of the whole consideration of gray information without spatial information of the image.
3.0 Results and Discussions

3.1 Chemical composition

Presented in Table 3 is the chemical composition of both CFA and WFA and its elemental composition is presented in Figure 3. The major compounds identified in order of increasing composition in CFA include $\text{Al}_2\text{O}_3$, $\text{SiO}_2$, $\text{CaO}$ and $\text{Fe}_2\text{O}_3$ whose percentage by composition outwit others. CFA possesses a high percentage of $\text{SiO}_2$ (51.43 wt%), $\text{Al}_2\text{O}_3$ (30.93 wt%), which confirms the bituminous nature of the CFA [34], [35]. This constituent also confirms the chemical properties of South African coal [9], [36]. The high percentage of $\text{SiO}_2$ and $\text{Al}_2\text{O}_3$ makes CFA a good component for stable insulators. CFA also demonstrates the presence of rare earth elements like Cr and Ti found in the lanthanide series. These elements have gained a wide range of application in automotive, electronics, optics, and defence [37]–[40].

Table 3: The chemical composition analysis of CFA and WFA using XRF

| Chemical Formula | CFA (wt%) | WFA (wt%) |
|------------------|-----------|-----------|
| $\text{Al}_2\text{O}_3$ | 30.93 | 10.7 |
| $\text{SiO}_2$ | 51.43 | 46.31 |
| $\text{MgO}$ | 1.95 | 0.36 |

Figure 2. Image segmentation flow chart
| Chemical Formula | CFA (wt%) (this study) | WFA (wt%) (this study) | CFA-Badarpur [41] | Rice Husk Ash (wt%) [42] | Type II Portland Cement (wt%) [43], [44] |
|------------------|-----------------------|-----------------------|--------------------|--------------------------|-------------------------------------|
| Al₂O₃            | 30.93                 | 10.70                 | 27.69              | 0.01                     | 6.67                                |
| SiO₂             | 51.43                 | 46.31                 | 59.52              | 90.0                     | 21.56                               |
| MgO              | 1.95                  | 0.36                  | 0.50               | 0.12                     | 4.51                                |
| Fe₂O₃            | 2.29                  | 17.28                 | 4.85               | 0.03                     | 6.17                                |
| CaO              | 6.75                  | 3.002                 | 0.68               | 0.60                     | 49.88                               |
| K₂O              | 0.77                  | 2.53                  | 1.61               | 2.30                     | 0.76                                |

On the other hand, the major compounds present in WFA in order of increasing composition are SiO₂, Fe₂O₃, Al₂O₃, CaO, and SO₃. The percentage of SiO₂ is high in both ash types, giving both a tendency of being suitable as binders, though CFA offering more suitability than WFA. Heavy metals in both ash types have a propensity for leaching, which is harmful to the environment [41] if left unused.

Comparing the chemical compositions of major compounds obtained from CFA and WFA in this study with other materials in the literature, we have the results presented in Table 4.

### Table 4: The chemical composition analysis of CFA and WFA from XRF

Comparing the major chemical composition of CFA and WFA in this study with those obtained in selected literature as presented in Table 4, CFAs possess a high concentration of Al₂O₃ and SiO₂ than fly ashes obtained from biomass. Table 4 also presents the Portland cement Type II principal composition following the ASTM C150 standard. The two ash categories contain calcium oxide (CaO) about 86% lower Type II Portland Cement, however, they both have the concentration of SiO₂ higher than that of the Type II Portland Cement. CFA-Badarpur analysed by [41] contains nearly similar compositions of Al₂O₃ and SiO₂ but differs significantly in other chemical compounds.
Figure 3: CFA (a) and WFA (b) elemental compositions at un-milled state as analysed by EDX.

Figure 4 shows the XRD diffractograms of CFA and WFA micro-particles at un-milled state. The crystal phases detected by the XRD in WFA are rhombohedral, hexagonal, cubic and tetragonal while that of CFA are hexagonal, orthorhombic, rhombohedral and anorthic. From XRD analysis, the dominant crystalline structures in WFA were calcite (CaCO3), quartz (SiO2), Sylvite (KCl), Lime (CaO), maghemite-Q (Ɣ-Fe2O3), nitratetine (Na(NO3)) and magnetite (Fe3O4) phases. However, in CFA, the dominant structures were Quartz (SiO2), Mullite (Al2.32Si6.68O12.84), Sillimanite (Al2(SiO4)O, Calcite high (CaCO3), Hematite (Fe2O3), Microcline (KAlSi3O8).

From Figure 4, the intensity decreased with milling time for the WFA, which remained almost constant for CFA. Also, comparing the peak intensity of CFA with WFA, a higher value of count was observed at every stage of milling CFA. At almost 30° (2Θ) CFA and 30° (2Θ) WFA, the peaks were observed to higher. When considering, CFA, it was higher in 0 minutes and at 20, 40 and 60 minutes, the peak maintained almost same peak, which dictates that there is the stability of quality in CFA while in WFA, 0 minutes proved to have the highest peak and
the was gradual reduction as the milling time increases. This reduction could be as a result of contaminations in WFA from the source of preparation or the nature of the timber or wood-fired to generate ash. Hence, the integrity of the CFA remained evenly and subjective of the fact that it is a suitable material (powder reinforcement) and with relative to its elemental composition; show in high percentage proportion of Al₂O₃ and less of Iron Oxides. The opposite is the case of WFA.

3.2 Image segmentation

The SEM images of both types (CFA and FWA) were originally obtained in 768 by 828 pixels dimension. The images were cropped into 750 by 745 pixels. SEM images obtained were binary images with the ash being of white intensity and the sputtering giving black shades. Hence, an image complementing process is essential. The resulting cropped image was complemented, such that black and white are revered in the binary images. This makes the ash the regions of interest other than the black spots signifying the sputtering.

The micrographs of different milling time are shown in figures part (a) of Figures 5-12 for CFA and WFA. It is observed that both CFA and WFA contains large particle size before milling. From SEM images before segmentation, particle aggregation occurs, with the smaller particles sticking to larger. It was also established from the micrographs that further milling affect the size of the particles and there still agglomeration of the particles.

Shown in Figures 5 – 12 are the SEM images before and after segmentation of both ashes. White spots represent the ash content and the black spots the sputtering effect. By physical inspection, a comparison between the segmented images of un-milled CFA (Figure 5b) and WFA (Figure 9b) samples, and the milled samples show smaller diameters of particles identified in the milled samples. The algorithm developed identifies prospective ash spots within the domain of the image size and magnification. Theoretically, it is established that the object position in an image does not influence the thresholding efficiency unless the position of the current object possesses varying levels of intensity [45]. In this case, the object differs distinctively from the background in terms of intensity. From the original images of prior to segmentation, the intensity difference= 100% (i.e. a white object with a black background).

During segmentation, object elements are morphologically structured such that structuring element members consists of pixels with centres not greater than the minimum pixel of each object from the origin of the identified object.

Original and Segmented SEM Images of Coal FA
Figure 5a-b. Original SEM and segmented image of un-milled coal FA.

Figure 6a-b. Original SEM and segmented image of coal FA at 20 minutes milling time.
Figure 7a-b. Original SEM and segmented image of coal FA at 40 minutes milling time.

(8a) 

(8b) 

Figure 8a-b. Original SEM and segmented image of coal FA at 60 minutes milling time.

Original and segmented SEM images of wood FA

(9a) 

(9b) 

Figure 10a-b. Original SEM and segmented image of un-milled (0 min) WFA
Figure 11a-b. Original SEM and segmented image of wood FA at 20 minutes milling time.

Figure 11a-b. Original SEM and segmented image of wood FA at 40 minutes milling time.
For each milling case, the mean particle size within each image was calculated (Table 5). The microstructure decreases across the milling times compared to the initial diameter of 75 µm obtained from the sieve. The milling process is expected to decrease particle size with time. However, the particle size decreases until a stationary point is reached (Figure 13), where further milling does not result in a decrease in particle size. A similar scenario was recorded by [14, 46 - 47] where results obtained at grinding time of 1.5hrs and 2hrs were similar. The final particle size of fly ash depends on the area of application. However, consideration should be given to the milling time to preserve the microstructure as well as the particle size of the ash.

Table 5. Mean particle size of CFA and WFA at varying milling time

| Milling Time (mins) | Mean (µm)- CFA | Mean (µm)- WFA |
|---------------------|----------------|----------------|
| 0                   | 3.66           | 3.69           |
| 20                  | 3.64           | 3.43           |
| 40                  | 2.74           | 3.22           |
| 60                  | 2.44           | 3.20           |
Figure 13: Graph indicating mean particle size (µm) at different milling time

Table 6 presents an analysis of variance performed on the two ash sources and the varying milling times.

| Source of Variation | SS    | df  | MS    | F      | P-value | F crit |
|---------------------|-------|-----|-------|--------|---------|--------|
| Milling Time        | 0.31571 | 2     | 0.157855 | 1.926955 | 0.341652 | 19     |
| Ash Sources         | 0.000122 | 1     | 0.000122 | 0.001483 | 0.972778 | 18.51282 |
| Error               | 0.163839 | 2     | 0.08192 |        |         |        |
| Total               | 0.479671 | 5     |         |        |         |        |

Conclusion
The particle size distribution is highly essential in determining the effectiveness of fly ash as reinforcements in material composites. The distribution is associated with bulk material which affects the processability and functionality as excipients in reinforcements. New policies by the South African government on making economic use of CFA is under implementation. However, CFA needs further milling for easy absorption for the intended purpose. This study considered estimating the particle size of CFA from images obtained from the scanning electron microscope. The result compared with WFA shows no significant difference between the particle sizes across varying milling time. However, the chemical composition of WFA differs significantly from CFA. This, unfortunately, does not make WFA a perfect substitute for CFA in certain areas of application. Increasing the milling time does not necessarily decrease the particle size of ash samples, which validates the literature. In industrial applications, automation of the process can be established such that the microstructural analysis of the samples can be taken in real time while the milling process is on-going. For further research, other segmentation techniques can be explored and the results compared with that obtained in this study for in terms of effectiveness and efficiency.

References
[1] Kumar, A. and Mandal, J. N. (2017). Effect of Reinforcement on Multi-tiered Fly Ash Wall, *Procedia Eng.*, vol. 189, no. May, pp. 446–453.
[2] Rynolds-Clausen, K and Singh, N (2016). *Eskom’s revised coal ash strategy and implementation progress*, pp. 1–11.
[3] Van der Merwe, E. M., Mathebula, C. L. and Prinsloo, L. C. (2014). Characterization of the surface and physical properties of South African coal fly ash modified by sodium lauryl sulphate (SLS) for applications in PVC composites, *Powder Technol.*, vol. 266, pp. 70–78.
[4] Xie, J. and Kayali, O.(2016). Effect of superplasticiser on workability enhancement of Class F and Class C fly ash-based geopolymers, *Constr. Build. Mater.*, vol. 122, pp. 36–42.
[5] Petrik, L. F., White, R., Link, M. J., Somerset, V. S., Burgers, C. L. and Fey, M. V (2013). Utilization of South African fly ash to treat acid coal mine drainage, and
production of high quality zeolites from the residual solids,” 2003 Int. Fly Ash Symp., vol. 2, pp 1-8.

[6] Murithi, G. N., Petrik, L. F., Gitari, W. M. and Doucet, F. J (2017). Synthesis and characterization of hydrotalcite from South African Coal fly ash, Powder Technol., vol. 312, pp. 299–309.

[7] Koshy, N. and Singh, D. N. (2016). Fly ash zeolites for water treatment applications,” J. Environ. Chem. Eng., vol. 4, no. 2, pp. 1460–1472.

[8] Missengue, R. N. M. et al., (2017). Transformation of South African coal fly ash into ZSM-5 zeolite and its application as an MTO catalyst,” Comptes Rendus Chim., vol. 20, no. 1, pp. 78–86.

[9] Musyoka, N. M., Petrik, L. F. Fatoba, O. O. and Hums, E. (2013). Synthesis of zeolites from coal fly ash using mine waters,” Miner. Eng., vol. 53, pp. 9–15.

[10] Chareonpanich, M., Namto, T., Kongkachuichay, P. and Limtrakul, J. (2004). Synthesis of ZSM-5 zeolite from lignite fly ash and rice husk ash, Fuel Process. Technol., vol. 85, no. 15, pp. 1623–1634.

[11] IEA, (2015). World Energy Outlook.

[12] Eskom, (2016). Coal Power,

[13] Eskom, (2017) “Ash management in eskom,”

[14] Du Plessis, H., Kearsley, E. P. and Matjie, H. (2007). Effect of grinding time on the particle size distribution of gasification ash and Portland cement clinker,” J. South African Inst. Civ. Eng., vol. 49, no. 4, pp. 28–34.

[15] Matjie, R. H. Bunt, J. R. and van Heerden, J. H. P (2005). Extraction of alumina from coal fly ash generated from a selected low rank bituminous south African coal, Miner. Eng., vol. 18, pp. 299–310.

[16] Rosin, P. and Rammler, E. (1933). The laws governing the fineness of powdered coal,” Journl Inst. Fuel, vol. 7, pp. 29–33.

[17] Vítěz, T. and Travniček, P. (2011). Particle Size Distribution of a Waste Sand From a Waste Water Treatment Plant With Use of Rosin–Rammler and Gates–Gaudin, Acta Univ. Agric. ..., vol. 59, no. 3, pp. 197–202.

[18] Patricia, F. and Joao Manuel, R. (2012). Computational Vision and Medical Image Processing. Taylor & Francis Group, London.

[19] Xu, X., Xu, S., Jin, L. and Song, E. (2011). Characteristic analysis of Otsu threshold and its applications, Pattern Recognit. Lett., vol. 32, no. 7, pp. 956–961.

[20] Abutaleb, A. S. (1989). Automatic thresholding of gray-level pictures using two dimensional entropy, Comput. Vis. Graph. Image Process., vol. 47, pp. 22–32.

[21] Liang, K. H. and Mao, J. J. W. (1995). Image thresholding by minimizing the measures of fuzziness, Pattern Recognit., vol. 28, no. 1, pp. 41–51.

[22] Sezgin, M. and Sankur, B. (2004). Survey over image thresholding techniques and quantitative performance evaluation,” J. Electron. Imaging, vol. 13, pp. 146–168.

[23] Elbayoumi Harb, S. M.N. Isa, A. M. and Salamah, S. A. (2015). Improved image magnification algorithm based on Otsu thresholding, Comput. Electr. Eng., vol. 46, pp. 338–355.

[24] Khairuzzaman, A. K. M. and Chaudhury, S. (2017). Multilevel thresholding using grey wolf optimizer for image segmentation,” Expert Syst. Appl., vol. 86, pp. 64–76.

[25] Hu, Q. M., Hou, Z. J. and Wieslaw, L. N. (2006). Supervised range-constrained thresholding, IEEE Trans. Image Process, vol. 15, pp. 228–240.

[26] Otsu, N. (1979). A Threshold Selection Method from Gray-Level Histograms, IEEE
Trans. Syst. Man. Cybern., vol. 9, no. 1, pp. 62–66.

[27] Rohilla, L., Garg, V., Mallick, S. S. and Setia, G. (2018). An experimental investigation on the effect of particle size into the flowability of fly ash,” Powder Technol., vol. 330, pp. 164–173.

[28] Kuruvilla, J., Sukumaran, D., Sankar, A. and Joy, S. P. (2016). A review on image processing and image segmentation, 2016 Int. Conf. Data Min. Adv. Comput., pp. 198–203.

[29] Zaitoun, N. M. and Aqel, M. J. (2015). Survey on Image Segmentation Techniques,” Procedia Comput. Sci., vol. 65, no. lccmit, pp. 797–806.

[30] Zhu, S., Xia, X., Zhang, Q. and Belloulata, K. (2007). An Image Segmentation Algorithm in Image Processing Based on Threshold Segmentation,” 2007 Third Int. IEEE Conf. Signal-Image Technol. Internet-Based Syst., pp. 673–678.

[31] Aly, A., Deris, S., and Zaki, N. (2011). Research review for digital image Segmentation techniques, Int. J. Comput. Sci. ..., vol. 3, no. 5, pp. 99–106, 2011.

[32] Kaur, D. and Kaur, Y. (2014). International Journal of Computer Science and Mobile Computing Various Image Segmentation Techniques: A Review,” Int. J. Comput. Sci. Mob. Comput., vol. 3, no. 5, pp. 809–814.

[33] Kaur, A. (2012). A Review Paper on Image Segmentation and its Various Techniques in Image Processing, Int. J. Sci. Res. ISSN (Online Impact Factor, vol. 3, no. 12, pp. 2319–7064.

[34] Belviso, C. (2018). State-of-the-art applications of fly ash from coal and biomass: A focus on zeolite synthesis processes and issues, Prog. Energy Combust. Sci., vol. 65, pp. 109–135.

[35] Vamvuka, D. and Kakaras, E. (2011). Ash properties and environmental impact of various biomass and coal fuels and their blends, Fuel Process. Technol., vol. 92, no. 3, pp. 570–581.

[36] Makgato, S. S. and Chirwa, E. M. N. (2017). Waterberg coal characteristics and SO2minimum emissions standards in South African power plants, J. Environ. Manage., vol. 201, no. 2, pp. 294–302.

[37] Taggart, R. K., Hower, J. C. and Hsu-Kim, H. (2018). Effects of roasting additives and leaching parameters on the extraction of rare earth elements from coal fly ash,” Int. J. Coal Geol., vol. 196, no. May, pp. 106–114.

[38] Long, K. R., Van Gosen, B. S., Foley, N. K. and Cordier, D. (2010). The principal rare earth elements deposits of the United States - a summary of domestic deposits and a global perspective.

[39] Mayfield, D. B. and Lewis, A. S. (2013). Environmental review of coal ash as a resource for rare earth and strategic elements.,” in Proceedings of the 2013 World of Coal Ash (WOCA) Conference, Lexington, KY, USA, 2013, pp. 22–25.

[40] Seredin, V. V. (1996). Rare earth element-bearing coals from the Russian far east deposits, Int. J. Coal. Geol., vol. 30, pp. 101–129.

[41] Gupta, N., Gedam, V. V., Moghe, C. and Labhasetwar, P. (2017). Investigation of characteristics and leaching behavior of coal fly ash, coal fly ash bricks and clay bricks, Environ. Technol. Innov., vol. 7, pp. 152–159.

[42] Darsanasiri, A. G. N. D., Matalkah, F., Ramli, S., Al-Jalode, K., Balachandra, A. and Soroushian, P. (2018). Ternary alkali aluminosilicate cement based on rice husk ash, slag and coal fly ash, J. Build. Eng., vol. 19, no. November 2017, pp. 36–41.

[43] Mohseni, E., Hosseiny, S. S., Ranjbar, M. M., Roshandel, E. and Yazdi, M. A.
(2015). The effects of silicon dioxide, iron (III) oxide and copper oxide nanomaterials on the properties of self-compacting mortar containing fly ash,” *Mag. Concr. Res.*, vol. 67, no. 20, pp. 1112–1124.

[44] ASTM, *ASTM C150: Standard specification for Portland cement*. Philadelphia, PA, USA. 11: Annual Book of ASTM Standards., 2011.

[45] Goh, T. Y., Basah, S. N., Yazid, H., Aziz Safar, M. J. and Ahmad Saad, F. S. (2018). Performance analysis of image thresholding: Otsu technique, *Meas. J. Int. Meas. Confed.*, vol. 114, pp. 298–307.

[46] Ikumapayi, O. M. and Akinlabi, E. T. (2018). Characterizations of the evolving properties during the milling time of wood fly ash Nanoparticles (WFA-NPs). Proceedings of the International conference on Industrial Engineering and Operational Management, Pretoria/Johannesburg, South Africa, October 29 - November 1, pp 2040 – 2049.

[47] Ikumapayi, O. M. and Akinlabi, E. T. (2019). Effects of Vibratory Disc Milling Time on the Physiochemical and Morphological Properties of Coal Fly Ash Nanoparticle, *Key Engineering Materials.*, vol. 796, pp. 38–45  
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