Pilot Scale Tar Production From Morupule Coal Dust Through Flash Pyrolysis

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ABSTRACT

The fourth industrial revolution promotes green and cleaner processes, pyrolysis being one of the strategies for waste utilization. In Palapye, Botswana, dust produced in the process of mining, beneficiation and utilization of coal at Morupule Mine and Power Plants causes serious health hazards to the employees, residents of Palapye and surrounding communities. Pyrolysis of coal dust is one of the wastes to energy technique that can be adopted to utilize waste coal dust. Botswana relies on imported tar to upgrade dusty roads while it has sufficient resources to produce its own tar from coal. One of the main products for pyrolysis of coal dust is tar which can be used to construct roads and pavements. This study utilizes coal dust from Morupule coal mine for tar production using flash pyrolysis in a batch reactor at pilot scale. Morupule coal dust was characterized before pyrolysis and the highest recorded yield is 22.9% kg at 600 °C.

II.

Keywords; Coal dust, flash pyrolysis, proximate analysis, tar, thermal cracking, ultimate analysis

I. INTRODUCTION

There is an estimated 212 billion tons of coal reserves in Botswana making coal one of the abundant natural resources in the country. Only about 2.4 metric tons a year is being mined at Morupule colliery mine which is utilized at the adjacent Morupule coal fired power plant for the production of 132 MW of power. Morupule colliery located 4.5km west of residential area of the urban village of Palapye produces and processes the highest quantities of coal in Botswana. Coal processing includes crushing and grinding which produce dust. Coal dust poses serious health hazard especially lung diseases to the employees and the surrounding communities. According to (Parry, 2016), coal dust causes diseases such as asbestosis and anthracnose. Coal pyrolysis technology can be employed in the utilization of coal dust, thus reducing dust stockpiles which occupies large landspace and negatively affect the environment and human life.

Pyrolysis is a thermal cracking process which involves the breaking down of coal in an oxygen free environment yielding tar, gas and char (Saydut *et al.*, 2010). The process parameters such as residence time, temperature, heating rate, particle size, coal rank reactor type and pressure influence the yields of rapid de-volatilization (Saydut *et al.*, 2010); (G Speight, 2019) Flash pyrolysis, of late has gained researchers' interest due to its potential to produce tar in greater yields than

other pyrolysis procedures which is suitable for upgrading to valuable liquid fuels (Tyler, 1979). Flash pyrolysis occurs at high heating rates above 500 °C in a short residence time. This process yields gas, tar and char. (Saydut *et al.*, 2010) carried out an experiments on the pyrolysis of coal in a fixed bed reactor and concluded that the utilization of coal in the flash pyrolysis process is more economical than the other processes of coal liquefaction due to the fact that the primary pyrolysis and the second reactions occur outside or inside the coal particles, the aliphatic structures can be replaced by the aromatic structures with the increasing pyrolysis temperature.

METHODOLOGY

A proximate analysis of Morupule coal dust using thermogravimetric analysis (TGA) was performed to predict the pyrolytic conditions of the coal dust. Coal characterization helps in the establishment of any possible relationships between coal dust feedstock and the resulting products. X-ray diffraction (XRD) was used for ultimate analysis at standard conditions (Speight, 2015) to dertermine minerals available in the sample, their sturcture and morphology.

A. TGA 701

The mass of the crucible was weighed and recorded then a sample of 1g was added into each of the crucibles. For moisture content and volatile matter, the temperature was increased to 107°C and to 950°C respectively to measure the weight loss until it was constant. The software measured and record three values in order to establish that the weight loss is constant and stops the process. The nitrogen flow valve on the line connected to the furnace was opened to 10L/min to provide an inert environment. After 2 hours, oxygen flowing at 3.5L/min was introduced and the sample was heated from 600 °C-750 °C for 10 minutes for complete combustion to occur producing ash residue.

B. XRD analyser

Three samples of 0.1g coal dust grinded into fine powder in fluid form of size less than $10\mu m$ were placed onto the sample surface by smearing uniformly onto a glass slide to produce a flat surface. For unit cell determinations, a small amount of a standard with known peak positions was first added and used to correct peak positions. The intensity of diffracted X-rays was continuously recorded as the sample and detector rotated through their respective angles (Dutrow Barbara L, 2019).

C. FTIR

Fourier transform infrared spectroscopy was used to characterize the produced tar and is one of the multifaceted analysis techniques available for analysis of coals and coal tar (Zhu, 2010) It identifies the molecular vibrations due to absorption of infrared radiation. If a sample is continuously exposed to changing wavelength of infrared diffraction, it absorbs light and the incident radiation corresponds to the energy of a particular molecular vibrations have wavelength ranges of 1200cm⁻¹ -4000 cm⁻¹ and 500 cm⁻¹-1200 cm⁻¹ respectively (Zhu, 2010). At these wavelengths the functional groups have unchanging absorption peaks and the infrared spectrum is useful in detecting functional groups (Gupta, 2007).

The sample was finally pyrolyzed in a batch reactor, varying the temperature of flash pyrolysis, feeding rate and heating rate. The reactor was first heated to the set temperature before the coal dust sample was fed into the reactor at 0.053g/s feed rate. Inside the reactor, heat liberates volatiles which diffuse through the product line through to the condensation train. Three condensers at 150°C, 75°C and 25°C were used to cool volatiles into oils of varying densities. The non-condensable gases were collected into the gasometer. At each chosen temperature, four experimental runs were performed.

III.

A. Thermogravimetry analysis

The thermal cracking reactions of coal are studied using TGA which measures the weight loss of a sample as a function of time and temperature at a given heating rate. The peaks in the graph produced are used to identify temperatures for primary cracking and secondary cracking. Moisture and volatiles were liberated in temperature ranges of 50°C - 150°C and 107°C - 900°C respectively. The sample was dried to remove the free moisture content before pyrolysis. This study has shown that 23.33% of Morupule coal dust can be converted into tar during pyrolysis. With reference to the study made by (Bowen & Irwin, 2008) which outlines the classification values for different grades of coal, Morupule coal dust falls mostly in the ranges of bituminous coal. The range according to (Bowen & Irwin, 2008) is 5-10% moisture and 20-40% volatiles. The TGA results are summarized in table I.

TABLE I. PROXIMATYE ANALYSIS OF MORUPULE COAL DUST USING TGA

Parameter	Amount Recorded (%)
Moisture	5.06
Volatiles	23.33
Fixed Carbon	50.41
Ash Content	21.2



B. X-ray difraction (XRD)

Figure 1: X-ray diffraction curve for coal dust

XRD was used to identify the morphology and mineral phases present in Morupule coal dust, which was found to have an amorphous structure. Morupule coal dust as depicted from peaks figure 1 contains minerals such as kaolinite, dolomite, quarts, pyrite, calcite and siderite in addition to the graphitic carbon. The minerals available in raw coal dust determine the ashing behavior of coal when it is burnt at high temperatures. For instance, this sample is highly concentrated with kaolinite and quartz which contains high amounts of ash.



Figure 2: Influence of process temperature on oil yield

Thermal energy is required in pyrolysis to break the bonds in coal matrix. Tar yield increased from 11.2%, 12.6%, and 22.9% at 500 °C, 550 °C and 600 °C respectively. The coal dust conversion yield increases with increasing temperature. This is further explained by the collision theory. This theory stipulates that molecules need sufficient energy for a reaction to occur. When the temperature increases, the energy levels within a particle rises causing rapid collision. As a result, the reaction proceeds faster, producing a much higher yield. Above 600 °C, the oil production decreases to 21.5% at a temperature of 650 °C. Beyond 600 °C, the energy of reaction is so high causing secondary cracking which enhance the production of gas than oils. The maximum yield of 22.9% was achieved at 600°C which is 98.2% of the value of volatiles determined in the proximate (TGA) analysis. Production of tar of Morupule coal dust reaches peak (optimum) production at 600°C, figure 2.



Figure 3: Influence of feed rate on the oil yield

D. Effects of Feed Rate on the Yield

The effect of feed rate on production and distribution of products i.e., gases and oils depend on the reactor configuration. For this study, a fixed bed reactor was used. Several feed rates were computed manually through measuring the feeding time intervals and the mass fed into the reactor through a lock valve. According to literature, lower feed rates promotes faster heat transfer thus speeding the rate of devolatilization of a sample leading to increased yield (Guedes, et al., 2018). Lower feed rates allows for longer residence times for the volatiles to be liberated from the coal dust. Feed rates 0.054kg/s, 0.0622kg/s and 0.0505kg/s were used and produced yields of 57mL, 49mL and 39mL respectively when the temperature was held at 650°C. Although a feed rate of 0.0505kg/s is the lowest feed rate, it yielded the lowest tar / oil, figure 3. This can be attributed to the fact that during the experiments the lock valve frequently blocked thus interfering with the flow of the material into the reactor. Hence, less mass was deposited into the reactor resulting in lower conversion of coal dust into condensable oils.



Figure 4: FTIR spectra for coal tar

E. FTIR analysis

Fourier transform infrared spectroscopy was used to identify the functional groups present in oil. Wavelengths were found at 750cm^{-1} peak, 1250cm^{-1} , 1400cm^{-1} , 1650cm^{-1} and $2700-2850 \text{cm}^{-1}$. The molecular mass increases with the increase in peak and so is the oxidative stability. The triglycerides and ethers can be located at 1250cm^{-1} and 1650cm^{-1} may contain the C=C, C=O or C=N bonds which are carboxylic compounds (aldehydes, acids etc.), figure 4. The $2700-2850 \text{cm}^{-1}$ peak can either show the presence of C₂₂ or glycerin.

Bitumen is a thermo-plastic derivative of crude oil that possess waterproofing and adhesive properties used in roads paving. Bitumen can also naturally occur in rock asphalt. According to (Saliani *et al.*, 2019), the FTIR spectra of bitumen show high intensity on the carbonyl group on wavelengths 1700cm⁻¹ and 2900cm⁻¹ and aliphatic group (triglycerides and ethers) on a wavelength of 1460cm⁻¹.

The tar from coal dust pyrolysis shows high concentration of the carboxylic acids, triglycerides and ethers at 2850cm⁻¹ and 1400cm⁻¹ which are good for their binding in road construction (Ibarra, et al., 1991). Hence this study has shown that, tar produced from Morupule coal dust through pyrolysis can be used for road construction.

F. TGA Analysis of Coal Dust Chars

After pyrolysis thermogravimetric analysis was performed on the coal dust char and the results are presented in table II. The indicated the amounts 12.84%, 12.40%, 11.97% and 15% volatiles in chars after c pyrolysis at 500 °C, 550 °C, 600 °C and 650 °C respectively. These chars can potentially be further pyrolyzed or utilized in other processes such as gasification and cracking. The fixed carbon content also increased from 52.59% to 61.35%, thus making it suitable for heating purposes and syngas production for synthetic fuels (Ho¨o¨k & Aleklett, 2010). The production of charcoal for heating and cooking contributes to the reduction of energy poverty especially for the poor, unemployed and rural communities.

TABLE II. THERMOGRAVIMETRY ANALYSIS FOR COAL CUST CAHARS AT DIFFERENT TEMPERATURES

Parameter	Temperatures (°C)				
	500	550	600	650	
Moisture (%)	2.40	1.78	1.62	2.01	
Volatiles (%)	12.84	12.40	11.97	15.0	
Fixed Carbon (%)	60.25	61.20	61.35	59.06	
Ash content (%)	26.32	25.91	26.22	25.31	

IV. CONCLUSION

V.

Proximate and ultimate analysis were carried out on Morupule coal dust. Coal dust contains 23.3% volatiles and these condensable volatiles were collected during flash pyrolysis resulting in an optimum yield of 22.95 at 600°C. The oil is to be elementary analyzed to classifications. The FTIR spectra of the oils was found to be similar to that of coal tar and hence the collected oil can be regarded as coal dust tar. The highest intensity found at 1250cm⁻¹ peak indicated the presence of triglycerides and ethers. The highest tar yield achieved in this setup was 1.42bbl./ton. This ratio falls within the range of 1-1.4barrels/ton of coal achieved by Sasol in the production of synthetic fuels. Thus, the production of tar from coal dust is feasible with the potential to contribute towards socio – economic development considering and abundance of coal dust at Morupule Coal Mine and Power Plant.

RECCOMENDATIONS

For future studies, flash pyrolysis will be carried out in an auger reactor because accumulation of char in a fixed reactor reduces the heating surface area. Feasibility are required to test the viability of this project on a commercial scale. Tar used in roads construction must have certain physical properties and these tests will form also part of the future studies.

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- [2] Cui, L. J., Lin, W. G. & Yao, J. Z., 2006. Influences of temperature and coal particle size on the flash pyrolysis of coal in a fast-entrained bed. *Chemical Research in Chinese Universities*, 22(1), pp. 103-110.
- [3] Gattering, B. & Karl, J., 2014. The influence of particle size, fluidization velocity, and fuel type on ash-induced agglomeration in biomass combustion. *Frontiers in Energy Research*.
- [4] Guedes, R. E., Luna, A. S. & Torres, A. R. T., 2018. Operating parameters for bio-oil production in biomass pyrolysis: A review. *Journal of Analytical and Applied Pyrolysis*, Issue 129, pp. 134-149.
- [5] G Speight, J. (2019) Handbook of Alternate Fuel Technology, Handbook of Alternate Fuel Technology. doi: 10.1142/11366.
- [6] Hautagalung, M., 2008. Understanding Coal Analysis. *Majari Magazine*, 2 June, pp. 1-4.
- [7] Ho¨o¨k, M. & Aleklett, K., 2010. A review on coal-to-liquid fuels and its coal consumption. *INTERNATIONAL JOURNAL OF ENERGY RESE*, p. 848–864.
- [8] HUCHLER, L. A., 2005. Audit your deaerator performance. HYDROCARBON PROCESSING, august.p. 102.
- [9] Ibarra, V., J. & Moliner, R., 1991. Coal characterization using pyrolysis-FTIR. *Journal of Analytical and Applied Pyrolysis*, Volume 20, pp. 171-184.
- [10] James, S. G. (2015) Handbook of coal analysis. 2nd editio. Edited by M. f Vitha. John Wiley & Sons.
- [11] Li, S. et al. (2005) 'Characterization of Coal by Thermal Analysis Methods'. Available at: <u>https://core.ac.uk/download/pdf/61908918.pdf</u>.
- [12] Parry, D. C., 2016. Morupule B Phase II Units 5 & 6: To Design, Finance, Construct., Gaborone: s.n.
- [13] Radenović, A. (2006) 'Pyrolysis of coal', Journal of Chemists and Chemical Engineers, 55(7–8), pp. 311–319.
- [14] Saydut, A. et al. (2010) 'The characterization of liquid product via flash pyrolysis of coal (Hazro, SE Anatolia, Turkey)', Energy Sources, Part A: Recovery, Utilization and Environmental Effects, 32(19), pp. 1821– 1828. doi: 10.1080/15567030902882992.
- [15] Speight, J. G., 2015. Handbook of Coal Analysis. 2 ed. New Jersey: John Wiley & Sons
- [16] Tyler, R. J. (1979) 'Flash pyrolysis of coals. 1. Devolatilization of a Victorian brown coal in a small fluidized-bed reactor', Fuel, 58(9), pp. 680–686. doi: 10.1016/0016-2361(79)90223-0.
- [17] Zahid, A. & Robert, W. Z., 2007. The Influence of Water System Impurities on the Performance of Deposit Control.

REFERENCES

 Cliff, D. I. et al. (1984) 'Products from rapid heating of a brown coal in the temperature range 400-2300 °C', Fuel, 63(3), pp. 394–400. doi: 10.1016/0016-2361(84)90018-8.