

Pyrolysis Of Coal Dust For The Production Of Tar And Useful Products

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Abstract— Coal mining for electricity production at Morupule coal mine in Palapye generates a considerable amount of fines or dust. The fines are generated through coal disintegration during various processes such as drilling, blasting, crushing and grinding. These coal dust fines have built-up over the years and have now become a nuisance and a health and environmental hazard to the community. If this is not dealt with accordingly, it can pose as a serious health threat as mineral dust can possibly cause diseases such as asbestosis and anthracosis. This work addresses the potential hazards by assessing the feasibility of utilizing the coal through the process of pyrolysis for the production of useful products such as tar/bitumen. Pyrolysis is a viable process as it is a widely used in waste utilization practices including refinery sludge, plastic waste, biomass, rubber from scrap tyres, as well as in metallurgical processes for coke formation. The amount of gaseous emissions (NO_x and SO_x) and heavy metals are less than those released through other processes. The tar will be produced mainly for the construction of roads and pavements. The benefits of such a project, include the reduction of the health and environment hazard, additionally, there will be socio-economic benefits such as job creation and better roads in the village of Palapye.

Keywords—coal, pyrolysis, tar

I. INTRODUCTION

With a record of 212 billion tonnes of coal resources in Botswana, the country relies solely on this resource for electricity, in addition to imported electricity from neighboring South Africa and Namibia. Coal mining for electricity production from the Morupule coal mine in Palapye generates a considerable amount of fines. The fines are generated through the disintegration on the coal during different processes such as drilling, blasting, crushing and grinding [1]. These coal dust fines have built-up over the years and have now become a nuisance and a health and environmental hazard to the community. If this is not deal with accordingly, it can pose as a serious health threat as mineral dust such as this can possibly cause diseases such as asbestosis and anthracosis [1]. This work addresses these issues by looking into utilizing the coal dust build-up through the process of pyrolysis for the production of useful products such as tar/bitumen. The goal is to improve the infrastructure (roads) in the village of Palapye. Such a project will solve the health and environmental concern which is caused by the coal mining activity. Furthermore, jobs

will be creating at the tar production plant as well as during road construction. The primary products of coal pyrolysis include char, tar and inorganic gases [2]. All these products can be utilized in a variety of applications in the chemical metallurgical industries. These range from activated carbon or carbon nanotubes from the solid residue to products of refined coal tar/oil which are chemicals such as creosote oil, naphthalene, phenol and benzene [3].

II. BACKGROUND

A. Health Hazards caused by coal mining

The pyrolysis process in this work is employed to convert coal dust to tar. This has been motivated by the build-up of coal dust at the Morupule Coal mine in Palapye. Coal dust is respirable, and exposure to it by mine workers and inhabitants in the locality of the mine are at risk of lung diseases due to prolonged exposure.

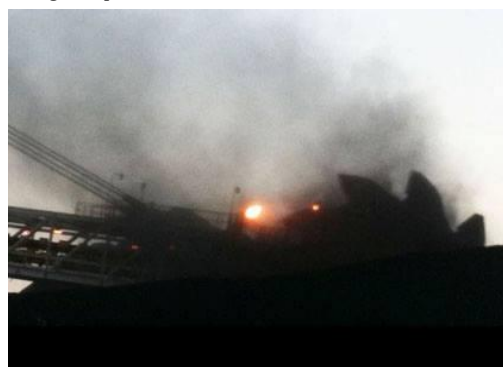


Figure 1: Coal dust from mining environment

B. Benefits of the project

The process was selected as a coal utilization process to eliminate this problem. Pyrolysis is a viable process for this as it is a widely used in waste utilization practices including refinery sludge, plastic waste, biomass, rubber from scrap tyres as well in metallurgical processes for coke formation [4]; [5]. The amount of gaseous emissions (Nitrogen oxides and

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Sulphur oxides) and heavy metals are less than those released by other processes [6]. The tar will be produced mainly for the construction of roads and pavements. The benefits of such a project, include the elimination of the health and environment hazard, additionally, there will be socio-economic benefits such as job creation and better roads in the village of Palapye.

III. PYROLYSIS PROCESS

The main products from pyrolysis include vapors, tar, mixture of gases and charcoal [7]. These pyrolysis vapors can be condensed into light and heavy oils. They can also be converted into syngas via steam reforming, and further processing for synthetic liquid fuels. More applications of the vapors include conversion to non-energy products [8]. This is an indication of the versatility of the products of coal pyrolysis.

Coal → Gas (CH_4 , CO, H_2 , etc.) + Liquid (tar) + Solid (coke)

The yields and nature of the products are influenced by a number of factors including; the coal type/ characterization, coal particle size, carrier gas, pressure, temperature, residence time and heating rate [2]. These parameters also determine the reaction pathway that occurs during the process [9].

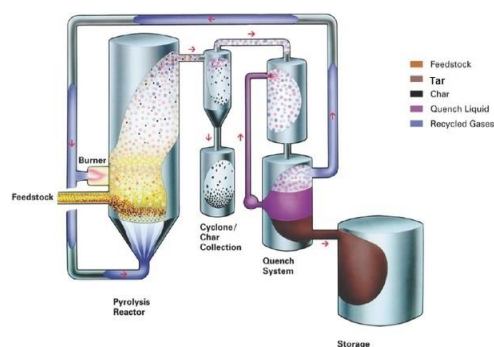


Figure 2: Coal pyrolysis process

A. Pyrolysis process stages

Heydari et al. [10] describe the process to occur in two main stages. The primary stage which occurs at 600°C and below includes the softening of coal, coke formation, release of vapors and development of plasticity. The aromatic carbon retained at this stage causes the coke formation while the hydro-aromatic carbon forms tar. Cracking that occurs in the secondary stage results in gas formation, low mass molecules and poly-condensation products [9]. This is consistent with Serio et al. [11] explanation which is summarized on the figure below.

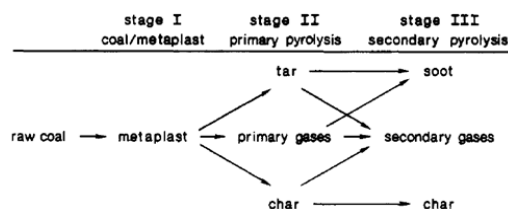


Figure 3: Coal pyrolysis model by Chemin and van Krevelen

Temperature is the most influential factor in pyrolysis, which according to Song et al. [12] enables compartmentalizing when it rises. Thermal degradation reactions start occurring at temperatures $\sim 400^\circ\text{C}$ through the cracking of bridge carbons possessing the lowest bonding energies [9]. Song et al. [11] described the five stages of pyrolysis according to the elevation in temperature as follows; drying stage, slow pyrolysis stage, fast pyrolysis stage, fast poly-condensation stage and slow poly-condensation stage. This information is important in considering coal utilization technology and product of interest.

B. Product evolution

Knowledge on the evolution of species as the process progresses provides understanding of the mechanisms and its relation to coal structure. This can also be used as a guide for selecting suitable parameters leading to high yields of product of interest, which is tar. The previously mentioned parameters influence species evolution and product distributions by altering the reactions occurring in the different stages pyrolysis [2].

The formation of tar results when larger ring fragments become hydrogen-saturated and distil as tar of medium molecular weight [9]. In a number of coals, this is primarily the first species released in pyrolysis reaction therefore it occurs at relatively low temperatures [13]. The tar however is prone to experience secondary vapour-phase reactions forming hydrocarbon carbons, soot and modified tar. As temperature rises, the composition of tar diminishes [6]. Ladner [5] found that at temperature above 1000°C negligible tar is produced because the volatiles would have cracked into gaseous components. Furthermore, rapid heating supports high tar yield as opposed to slow heating. However, in this work, the product of interest is a thick form of the tar which is produced as slow heating rates. High pressure opposes tar production but supports the methane and other hydrocarbon gas production. Table 1 shows the composition of pyrolysis products obtained with slow and rapid heating which were findings by Peters and Betling [7].

C. Tar formation

The formation of tar occurs in a series of stages which includes, a) depolymerisation by rupture of weaker bridges in the coal macromolecule to release smaller fragments, b) re-polymerization of metaplast molecules, c) transport of lighter molecules away from the surface of the coal particles by combined vaporization and gas phase diffusion d) internal transport of lighter molecules to the surface of the coal particles and finally, the liquid phase in softening coal [14]. Figure 4 illustrates the reaction processes that occur in coal pyrolysis [13].

Tar formation occurs as molecular fragments broken down from the particle are released and transported out of the char particle without experiencing cross-linking reactions in the char particle. However in this work, the process should be designed

in a way that cross-linking is promoted in order to produce a thicker form of the tar [15].

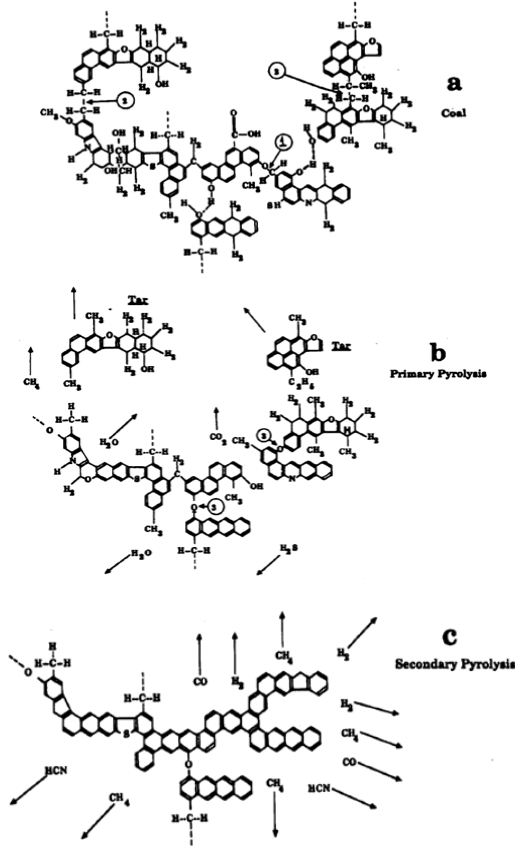


Figure 4: Hypothetical coal molecule during stages of pyrolysis [14]:

IV. REACTION KINETICS

The study of reaction rates in relation to time, concentration and temperature is carried out through the determining the kinetics of a reaction from experimental data [16]. Kinetic evaluation of the pyrolysis varies with different models. Pyrolysis reactions fall under the solid-state reactions, kinetic data from this type of reactions are conventionally obtained using discrete isothermal methods analysis [17]. However, more vigorous methods have been adopted, which are operated under non-isothermal conditions. They have gained much popularity due to their superior ability in terms of speed and efficiency. The single order reaction model is most commonly applied for coal weight-loss and species evolution [14]. In this model, the rate of total volatile species evolved dW_i/dt is

proportional to the instantaneous value of the source material [18].

$$\frac{dW_i}{dt} = k_i(W_{oi} - W_i) \quad (1)$$

Where W_{oi} is the ultimate yield and W_i is the amount already evolved, k_i , the proportionality constant, is the rate constant obtained from the Arrhenius expression:

$$k_i = k_{oi} e^{\left[\frac{E_i}{RT}\right]} \quad (2)$$

This model is applicable to homogeneous systems where the decomposition of the source is caused by a single process with single activation energy E_i . More advanced models have been applied [18]. The kinetic parameters are determined through a set of thermogravimetric experimental data.

The thermal decomposition of coal predicted on a single reaction under isothermal conditions is predicted by the following equation ([16]; [12]):

$$\frac{dx}{dt} = k(T)f(x) \quad (3)$$

From the Arrhenius law:

$$k = A \exp\left(-\frac{E}{RT}\right) \quad (4)$$

Where t denotes time, x is the degree of conversion, dx/dt is the rate of the isothermal process, $f(x)$ is the conversion function which represents the reaction model applied, it depends on the controlling mechanism:

$$X = \frac{w_0 - w_t}{w_\infty - w_{\infty}} \quad (5)$$

Where w_0 and w_∞ are the initial sample weight and final weight respectively, w_t is the remaining sample at time t ([16]; [11])

In the case of non-isothermal conditions, the rate expressions which are defined as a function of temperature at a linear heating rate v can be expressed through a superficial transformation of equation (3) to give ([15]; [17]; [19]).

$$\frac{dx}{dT} = \frac{A}{v} \exp\left(-\frac{E}{RT}\right) f(x) \quad (6)$$

and;

$$v = \frac{dT}{dt} \quad (7)$$

The non-isothermal method employs a heating rate, v which is linear, to raise the temperature. The linear heating follows the equation:

$$T = T_0 + vt \quad (8)$$

Where T_0 is the starting temperature, v is the linear heating rate (K/min) and T is the temperature at time, t .

Now, the following relationship is defined for non-isothermal experiments:

$$\frac{dx}{dT} = \frac{dx}{dt} \cdot \frac{dt}{dT} \quad (9)$$

Substituting (9) in (3) gives (6) [10].

According to the Freeman-Carroll hypothesis:

$$f(x) = (1 - x)^n \quad (10)$$

$$\frac{dx}{dT} = \frac{A}{v} \exp\left(-\frac{E}{RT}\right) (1 - x)^n \quad (11)$$

Where E_a is the activation energy, R is the gas rate constant 8.314, n is the reaction order, T is the temperature and t is the time ([16]; [20]). This relation (11) represents the differential form of the non-isothermal rate law.

Integrating (11), eq. (12) is obtained:

$$\int_0^x \frac{dx}{(1-x)^n} = \frac{A}{v} \int_0^T \exp\left(-\frac{E}{RT}\right) dT = -\frac{AR}{vE} \left(1 - \frac{2RT}{E}\right) \exp\left(-\frac{E}{RT}\right) \quad (12)$$

Natural logarithm on (12) after integration the (13) or (14) is obtained;

$$\ln\left[-\frac{\ln(1-x)}{T^2}\right] = \ln\left[\frac{AR}{vE} \left(1 - \frac{2RT}{E}\right)\right] - \frac{E}{RT} \quad (n = 1) \quad (13)$$

$$\ln\left[-\frac{\ln(1-x)^{1-n}}{T^2(1-n)}\right] = \ln\left[\frac{AR}{vE} \left(1 - \frac{2RT}{E}\right)\right] - \frac{E}{RT} \quad (n \neq 1) \quad (14)$$

The value of E is usually very large, which makes the term $2RT/E$ very small (~ 0). Therefore in the case for a first order reaction, $X = (1/T)$, $a = \ln[(AR/vE)(1-(2RT/E))]$, $b = -(E/RT)$, $Y = \ln[\ln(1-x)/T^2]$. This leads to a linear equation of $Y = a + bx$, which enable the activation energy and pre-exponential factor A to be calculated by slope and line intercept respectively [15]; [11].

The single order model has been applied by a number of researchers, including Song et al. [11] who used the model to study the kinetics of low rank coal from different locations in China. Kok [21] applied this method when determining the

kinetic parameters in the pyrolysis of alsphalite from Silopi, Turkey. Gao et al. [15] also used this model in their work in which they were determining the properties and reaction kinetics of low rank coal from Western China. Dai et al. [20] in their study, which was aimed at determining the kinetics of blended from China in a fluidized bed reactor, used the model.

V. EFFECTS OF PROCESS PARAMETERS ON REACTION KINETICS

A. Effect of heating rate and final temperature on pyrolysis process

The A number of researchers have found that the heating rate has an effect on the oil yield in coal pyrolysis. Seebauer et al. [22] found that high heating rate promotes high yield of volatile matter. However, there was no significant difference in the ultimate yield of the oils, which they attributed to the small range of heating rate used in their work. The high heating rates enhance the tar production process in that, it allows less time for tar cracking which culminated to more tar and less methane [23]. Gao et al. [15] explain that an increase in heating rate, influences the kinetic parameter in that is causes an increase in the activation energy and the pre-exponential factors. The heating rate affects the fracture speed of the coal molecule [16]. Altun et al. [24] studied the effect of heating rate on asphaltite from Silopi. They report that at lower heating rates, heat transfer was more effective in the particles, which was caused by the slow rate of heating. Effective heat transfer results in better cracking in the molecules which led to the release of more volatiles.

B. Effect of particle size

In their work, Gao et al. [15] stated that the kinetic parameters seemed to not show an obvious change with the increase of particle size. They explained that with regards to particle size, pyrolysis is governed by the mass and heat transfer, as well as the physical properties of the raw material. These findings were consistent with Altun et al. [23] who find no significant change in the kinetic parameters with the size of particle. The theory of secondary reactions in coal pyrolysis suggests that as the tar is cracked as it transports in the pores of the coal molecule [22].

C. Effect of pressure

A high pressure in the system promotes gas production more than liquid production [21]. This is due to the fact that pressure influences reactions in the metaplast which produces methane and carbon dioxide [21].

VI. CROSSLINKING IN COAL PYROLYSIS

The break-up of the macro-molecular network in coal structure is controlled by rates of bond breaking, mass

TABLE 1: EXPRESSIONS FOR THE MOST COMMON REACTION MECHANISMS IN SOLID STATE REACTIONS [25]

Reaction model	$f(x)=(1/k)(dx/dt)$	$g(x)=kt$
Reaction order		
Zero order	$(1-x)^n$	X
First order	$(1-x)^n$	$-\ln(1-x)$
nth order	$(1-x)^n$	$(n-1)^{-1}(1-x)^{(1-n)}$
Nucleation		
Power law	$n(x)^{(1-1/n)}$; $n=2/3, 1, 2, 3, 4$	x^n ; $n=3/2, 1, 1/2, 1/3, 1/4$
Exponential law	$\ln x$	X
Avrami-Erofeev(AE)	$n(1-x)[- \ln(1-x)]^{(1-1/n)}$; $n=1, 2, 3, 4$	$[- \ln(1-x)]^{1/n}$; $n=1, 2, 3, 4$
Prout-Tompkins(PT)	$x(1-x)$	$\ln[x(1-x)^{-1}] + C^a$
Diffusional		
1-D	$1/2x$	x^2
2-D	$[- \ln(1-x)]^{-1}$	$(1-x)\ln(1-x) + x$
3-D(Jander)	$3/2(1-x)^{2/3}[1-(1-x)^{1/3}]^{-1}$	$[1-(1-x)^{1/3}]^2$
3-D	$3/2[(1-x)^{-1/3}-1]^{-1}$	$1-2/3x-(1-x)^{2/3}$
Contracting geometry		
Contracting area	$(1-x)^{(1-1/n)}$; $n=2$	$(1-x)^{(1-1/n)}$; $n=2$
Contracting volume	$(1-x)^{(1-1/n)}$; $n=3$	$(1-x)^{(1-1/n)}$; $n=3$

^a Integration constant

transport and crosslinking. Cross linking reactions determine the ultimate tar yield as well as the molecular distribution of the tar product [15]. This means that the greater the extent of these reactions, the higher the molecular weight of the tar, which would mean a more viscous product. Crosslinking can be associated with the decomposition of carboxyl groups in the coal molecule to form carbon dioxide [26]. In order to promote cross-linking, the heating rate should be slow as rapid heating has proven to substantially reduce the occurrence of these reactions [27]. Additionally, they can be promoted by oxidation [25].

A method of monitoring crosslinking reactions in coals works by measurement of solvent swelling ratio as described by Green [27]. It is used to determine the crosslinking changes during pyrolysis. The extent of crosslinking reactions is dependent on the coal rank. Bituminous coal is said to crosslink at higher temperatures than lignites. Coal from Morupule coal mine is sub-bituminous to bituminous [28]; therefore it is expected to crosslink at relatively high temperatures based on the latter statement.

VII. COAL CHARACTERIZATION

In order to apply the most efficient clean coal utilization technology, the coal must undergo analysis to determine its physiochemical properties. The results are crucial in identifying the most suitable utilization technology [29]. These tests include the proximate analysis, ultimate analysis and miscellaneous analysis [3].

The proximate analysis involves determining the moisture, ash content, volatile matter and fixed carbon content. The ultimate analysis provides information on the elements present

in the coal sample which include carbon, nitrogen, hydrogen, sulphur, oxygen as well as the ash content [27]; [28]. This can be used to evaluate combustion characteristics and approximate sulphur and nitrogen oxides [30]. Miscellaneous analysis is commonly carried out for coal buyers, at their request. It comprises of determining the calorific value, swelling index, grindability, ash fusibility and plastic properties of coal.

TABLE 2 adopted from Gupta [29] shows a summary of important bulk properties of coal which influence conversion as well as the analysis required for the estimation of the properties.

TABLE 2: PHYSICAL PROPERTIES OF COAL AND RESPECTIVE ANALYSIS

Properties	Analysis
Chemical properties	Proximate analysis, ultimate analysis and ash analysis
Physical properties	Density, specific gravity, pore structure, surface area, reflectivity
Mechanical properties	Hardness/abrasiveness friability, grindability, dustiness index
Thermal properties	Calorific value, heat capacity, thermal conductivity, plastic, agglomerating index, free swelling index
Electrical properties	Electrical resistivity, dielectric constant, magnetic susceptibility

VIII. CONCLUSION

Coal pyrolysis products carry the potential to be used in a wide range of applications; these include tar for road construction and synthesis of useful chemicals such as benzene. Improving this process to an efficient one which yields quality tar suitable for road construction and other chemicals will broaden the usefulness of the process. This in turn promises viability, which would validate the purpose of this work.

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