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Comparative investigation of spontaneous combustion of biomass, hydrochar, coal and their blends using Wits-Ehac and thermogravimetric analysis

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ABSTRACT

Biomass, hydrochar, coal and hydrochar/coal blends have been proposed as alternative energy sources to coal. Given that the new fuels are derived from biomass, which is highly reactive, it is necessary to investigate their potential for spontaneous combustion (SPONCOM). Through the characteristics of coal discard, biomass, hydrochar, and hydrochar mixed at different ratios with discard coal, this study examined the factors that contribute to SPONCOM. The SPONCOM liability of these fuels was examined using the thermogravimetric analysis (TGA) and the Wits-Ehac index. The physicochemical analysis revealed an increase in the energy characteristic of the hydrochar produced from biomass. All samples showed a transmittance of the C=O stretch, which promotes SPONCOM, according to the results of the Fourier Transform Infrared Spectroscopy (FTIR) analysis. The TGA findings revealed that biomass is highly reactive, while discard coal was found to be non-reactive. A significant correlation was seen between the TG_{spc} index and the physicochemical properties of the samples. The Wits-Ehac results showed that biomass had the lowest SPONCOM liability index, while the 50% hydrochar/50% discard coal blend had the highest SPONCOM liability index. There was no correlation between the TGA and Wits-Ehac results, as the Wits-Ehac results showed some inconsistencies, particularly for samples derived from biomass. This study establishes the mechanism responsible for SPONCOM of hydrochar blended with coal in relation to their properties using the TGA. In addition, contributed to the understanding of techniques for predicting the SPONCOM of likely fuels for this application.

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Introduction

Coal accounts for 37% of world's electricity (WCA 2020), but there is a public outcry to decrease its use due to its greenhouse gas emissions. The conventional production of electricity from coal-fired plants is estimated to produce an estimated 15 billion tonnes of carbon dioxide (CO₂) per year. The World Nuclear Association (2020)

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asserts that this is the cause of global warming. In addition to other pollutants like particulate matter, sulfur dioxide, and nitrous oxide ([IEA 2018]). Natural gas has also been considered as an alternative source for electricity generation due to its relatively reduced greenhouse gas emissions per kWh of electricity produced (Oboirien et al. 2018). However, the fluctuation in the cost of natural gas is a key detrimental factor to the world's acceptance of the approach, as natural gas is scarcely distributed worldwide. Solar and wind energy are both clean and renewable energy sources that could be a potential alternative source for electricity generation. Nonetheless, they are considered intermittent, and the cost of solar energy storage is high (Slabbert 2017; Williams 2013). The increase in both human population and electricity demand necessitates the advancement of clean coal technology to reduce emissions and prevent climate change. In addition, it also implies exploring new potential energy sources for clean electricity generation to reduce coal dominance. Biomass is by far the largest renewable energy source, accounting for 55% of the world renewable energy and over 6% of global energy supply (IEA 2023). With the availability of different and suitable combustion technologies, newly produced hydrochar/biocoal fuel can be blended with coal and co-fired (Setsepu et al. 2021). South Africa has over 2 billion tonnes of discard coal of various grades, and this coal resource can be co-fired with the new hydrochar in existing coal power plants to meet the regulated emission standard. The fuel's (hydrochar coal blend) spontaneous combustion (SPONCOM) liability must therefore be understood as hydrochar is composed of biomass, which is highly reactive. The reliable data on the oxidation of these fuels and the necessary measures to mitigate this tragedy will be provided.

Biomass has been viewed as a potential alternative to coal for power generation in centralized boilers (Schwarzer, Jensen, and Glarborg 2017). However, substituting coal with another fuel is not simple, given that several factors should be considered. The majority of SPONCOM research has focused on coal because it is a prominent phenomenon in the coal mining industry. However, this phenomenon could affect other materials like biomass, forestry residue, coal-biomass mix and other opportunity fuels (Avila 2012; Matsobane et al. 2023). Regardless of the type of material involved, the mechanism that controls SPONCOM's process is the same. Biomass burns more quickly than coal, which means there is a higher possibility of a rapid propagation of any ignition flame for biomass in co-firing plants (Wang et al. 2012). According to Krause (2009), there have been numerous explosions and SPONCOM occurrences observed when processing, transporting, and storing various types of biomass. The degree of the SPONCOM of coal is impacted by variables including moisture, surface area, coal rank, pore structure, maceral content, etc (Gbadamosi et al. 2021; Genc, Onifade, and Cook 2018; Said et al. 2022). The oxygen content, temperature, and moisture content all influence biomass's SPONCOM, just as they do for coal (Manic et al. 2021). Additional important parameters such as bacterial activity, porosity, temperature, activation energy, and others have been reported to have significant impacts on biomass' SPONCOM characteristic (Ashman, Jones, and Williams 2018; Avila 2012; Chansa, Luo, and Yu 2020; Matsobane et al. 2023). Before 2015, approximately 24 biomass combustion incidents were reported worldwide, and most incidents occurred in wood pellet companies (The Linde Group 2015; Mullerova 2014). Another incident occurred at Inferno Wood Pellets Co Rhode Island in 2013 (Mullerova 2014; Pfecke and Warrenville 2010). The SPONCOM of biomass occurs mostly during long-term bulk

storage in silos and during transportation (Ebadat 2019; Larsson 2017; Persson 2013), therefore, SPONCOM liability of biomass or hydrochar must be understood.

The SPONCOM liability of biomass has been predicted using a limited number of techniques (Avila 2012; Rupa-Gadd 2006). Those techniques are limited to the final phase of oxidation, which is close to the ignition point. The significance of the material's biological activity is typically not considered. In such instances, biomass is assessed using standard thermal analysis methods such as Frank-Kamenetskii, heating basket, and modified crossing point temperature (XPT) methods (Chen 1999). In addition, the original XPT method and the isothermal oven test, also called the constant temperature method, have also been utilized (Buggeln and Rynk 2002). The number of prior studies to predict the SPONCOM of biomass by using certain approaches, including that of hydrochar coal blend, is inadequate to support their efficacy. For this reason, Wits-Ehac index and thermogravimetric analysis (TGA), which have been used to determine the SPONCOM liability of coal and coal shale (Gbadamosi et al. 2021; Onifade, Genc, and Bada 2020) were explored in this study. The same Wits-Ehac index was also used by Abdulsalam et al. (2020) to determine the SPONCOM liability of different coals used for producing activated carbon. Based on the proven capability and reliability of the Wits-Ehac index, along with the TGA, hydrochar produced from *Searsia lancea* and blends of hydrochar/discard coal were evaluated for their SPONCOM characteristic. The results of this research provide more knowledge of the most suitable technique to predict the potential of raw *Searsia lancea* biomass and its hydrochar/coal pellets toward SPONCOM.

Material and Methods

Sample Preparation

Twelve-year-old *Searsia lancea* tree samples harvested from the Mispah tailings facility (MTF) of the Vaal River Mine in South Africa were used in this study. The tree samples harvested from sections 11 and 12 (S11 and S12) were sun dried, and then milled to $-212\ \mu\text{m}$ for all characterization and SPONCOM experiments. The *Searsia lancea* tree compartments were combined to create a mixture of 50% S11 and 50% S12 raw biomass after milling (Matsobane et al. 2023). The same blend made from 50% S11 and 50% S12 were subjected to hydrothermal treatment carbonization (HTC) to produce hydrochar. The HTC process was carried out using a 1.75 L Berghof BR-1500 high-pressure reactor at a temperature of 280°C , for 180 minutes (residence time), and solid: liquid ratio of 1:8. This condition was the best test condition attained by Setsepu et al. (2021) to produce hydrochar from the same sample.

Characterisation

The proximate analyses for all pulverized samples were conducted in accordance with ASTM D-5142, with approximately 1 g used to determine the inherent moisture, ash content, and volatile matter content, with fixed carbon calculated by difference. The ultimate analysis and total sulfur for all the samples were carried out in accordance with ASTM D 5373-02 and ASTM D 4239-05 for CHN and sulfur content respectively. The heat content of the 100% raw biomass was determined in accordance with ASTM (D5865-04). The identification of functional groups in each sample was made using a PerkinElmer

Frontier FTIR spectrometer. A sample of 0.5 g was placed on the FTIR spectrometer lens to conduct the analysis. An infrared beam is transmitted through the sample by the apparatus, and the frequency at which the sample absorbs the beam falls between 4000 cm^{-1} and 450 cm^{-1} at a step size of 4 cm^{-1} . An absorbance spectrum is produced by the FTIR spectrometer, and the peaks in the spectrum correspond to various molecular structures and chemical bonds in each sample. A functional group, such as an alkane, an alcohol, an aromatic, an aliphatic, etc., is represented by each peak.

SPONCOM Tests

TGA Tests

A prediction of the biomass's combustion reactivity in an oxidative media was made using the TGA and DTG (differential thermogravimetric) thermographs. At different heating rates (4, 8, 12, 16, 20, and $24^\circ\text{C}/\text{min}$), a 100 mg sample of $-212\text{ }\mu\text{m}$ was thermally heated from 25°C to 850°C for each test run. Each sample is maintained at 850°C until there was no change in mass loss. From the DTG and TGA thermographs for all samples, the derivatives of weight loss ($\%/ \text{min}$) as a function of time and temperature as well as the slope of the derivative plot in the linear region of the curve at low temperatures were calculated. A plot displaying a profile of the heating rates versus the slope for each of the examined samples was produced using the data from each sample (Avila 2012). The thermogravimetric SPONCOM index TGA_{spc} was used to predict the liability of biomass to SPONCOM using the data as given in Equation 1.

$$TG_{spc} = \frac{\Delta \text{weight loss rate}}{\Delta \text{Temperature}} \quad (1)$$

Wits-Ehac Test

The Wits-Ehac setup comprises an air circulator, oil bath, oil circulator, a flowmeter, a heater, an air supply compressor, six-cell assemblies (three cells for the discard coal sample or hydrochar/coal blends and the other three for calcined aluminum, which is the inert material) and a computer (Fig. 1a). The samples (discard coal, raw *Searsia lancea*, hydrochar and three hydrochar/coal blends) were pulverized to $-212\text{ }\mu\text{m}$, and 20 to 25 g of the pulverized samples were fed into three cells of the Wits-Ehac apparatus, while the remaining three cells fed with calcined aluminum. The sample cells and the inert material cells were placed in an oil bath heated to 200°C . As the oil bath was heated, the temperatures of the samples and the inert material were recorded on the system every 30 seconds using a micro-computer. The time taken for each sample test depends on the SPONCOM characteristic of the sample. The tests for samples with high SPONCOM liability take less time than samples with low SPONCOM liability. The Wits-Ehac apparatus incorporates the XPT and the differential thermal analysis (DTA) thermograph to determine the sample's SPONCOM index (Wits-Ehac index). The DTA thermograph provides an insight into the degradation of the sample tested in three stages, as illustrated in Figure 1b.

The first stage denotes the temperature at which the inert material exceeds that of the tested sample. The region where the exothermic reaction takes place is known as Stage II. This stage provides information on the temperature difference between the inert material and the tested sample. Also, at this stage, the tested sample's heating rate could

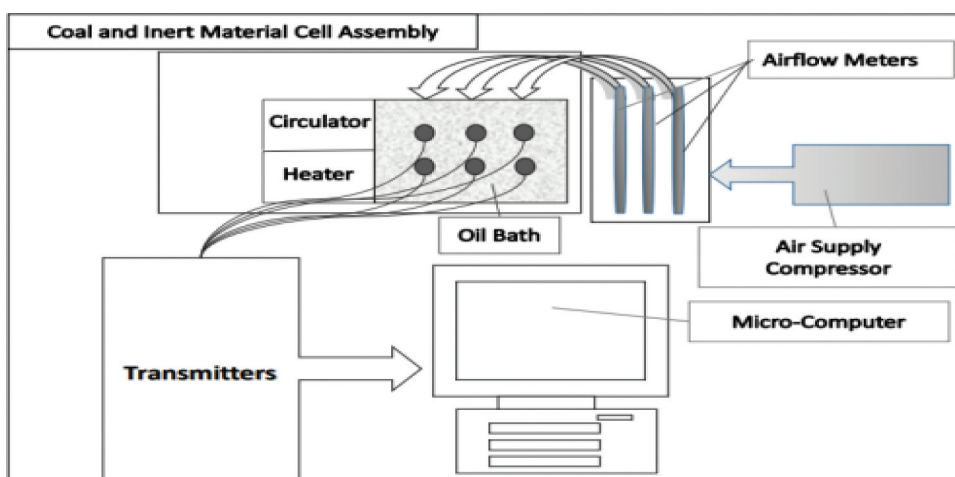


Figure 1a. A schematic diagram of the Wits-Ehac apparatus (Wade, Gouws, and Phillips 1987).

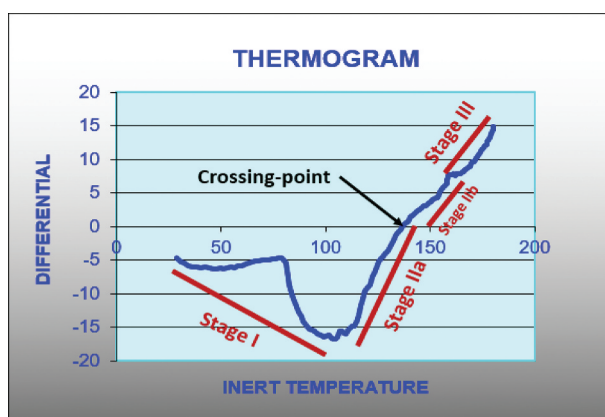


Figure 1b. A typical DTA thermogram profile (Wade, Gouws, and Phillips 1987).

be higher than that of the inert material because it is more likely to self-heat, and hence reach the temperature of the surroundings (oil bath temperature). The point at which the differential is zero in Stage II is the XPT. Stage III is when the tested sample burns out completely. The Wits-Ehac index was calculated according to Equation 2. It is expected that in Stage II, a sample with a high SPONCOM liability will have a very steep slope and a low XPT compared to a sample with low SPONCOM liability. According to Wade et al. (1987), a coal sample with a Wits-Ehac index below three has a low SPONCOM liability. Whereas, between 3 and 5 five, the likelihood of the coal undergoing SPONCOM is medium, and when the index is above 5, the sample SPONCOM liability is high.

$$\text{Wits - Ehac index} = \left(\frac{\text{Stage II slope}}{\text{XPT}} \right) \times 500 \quad (2)$$

Results and Discussion

This section presents the characterization and SPONCOM analysis of biomass, discard coal, hydrochar, and hydrochar/coal blends.

Characterisation of Biomass, Discard Coal, Hydrochar and Hydrochar/Coal Blends

The proximate analysis, ultimate analysis and the heat content of the samples are shown in Table 1. The inherent moisture of biomass, discard coal and hydrochar was 8.01%, 3.10%, and 1.92%, respectively. The biomass's moisture content may make it more vulnerable to SPONCOM. According to the study by Miyawaki et al. (2021), biomass with a high moisture content may result in rapid microbial heat generation that leads to combustion. The moisture content of 25% hydrochar + 75% discard coal, 50% hydrochar + 50% discard coal and 75% hydrochar + 25% discard coal used in this study ranged from 2.04 to 2.76%. According to Hao et al. (2014), carbonaceous materials with high moisture content act as catalysts by accelerating the oxidation reaction, which results in heat release and SPONCOM. Therefore, a hydrochar/coal blend with a higher moisture content could have a relatively high SPONCOM liability index compared to other blends with lower moisture content.

where VM is the volatile matter, A is the ash, M is the inherent moisture, FC is the fixed carbon, TC is the total carbon, N is the nitrogen, O is the oxygen by difference, H is the hydrogen, TS is the total sulfur, CV is the calorific value, H is the hydrochar, DC is the discard coal.

$$\text{Total oxygen} = 100 - (M\% + A\% + TS\% + TC\% + H\% + N\%) \quad (3)$$

Hydrochar, biomass, and discard coal had ash content of 0.36%, 2.92%, and 36.23%, respectively. The ash content for the hydrochar/coal blends used, i.e., 25% hydrochar + 75% discard coal, 50% hydrochar + 50% discard coal and 75% hydrochar + 25% discard coal blends, range from 9.88 to 27.20%. High mineral concentration in ash-containing samples serves as a heat sink and obstructs fuel pores and active sites (Beamish and Arisoy, 2008; Onifade et al., 2020). This is because ash created an oxidative barrier during low temperature oxidation and preventing moisture and high volatile organics from escaping the internal surface of the fuel. Another study reported by Blazak et al. (2001) shows that as ash content increases, spontaneous combustion liability decreases due to the reduction in the amount of inert and organic material acting as a heat sink. According to Onifade (2018), coal with high ash content is less susceptible to SPONCOM. Manic et al. (2021) observed the same trend during their investigation of biomass. Therefore, it is expected that discard

Table 1. Proximate and ultimate analysis of biomass, discard coal, hydrochar and hydrochar/coal blends.

Samples	Proximate Analysis (Air dried)				Ultimate Analysis (Dry basis)					CV
	M%	A%	FC%	VM%	TS%	TC%	H%	N%	O%	MJ/kg
Biomass	8.01	2.92	19.54	60.52	0.07	44.60	6.26	0.47	37.67	17.28
Hydrochar	1.92	0.36	52.12	45.60	0.08	70.90	5.01	0.47	21.26	29.58
Discard coal	3.10	36.23	40.01	20.65	1.64	48.20	2.92	1.11	6.80	18.55
25% H + 75% DC	2.76	27.20	43.24	26.79	1.21	54.30	3.55	0.95	10.03	21.42
50% H + 50% DC	2.45	18.46	46.06	33.03	0.87	60.10	4.03	0.82	13.27	25.23
75% H + 25% DC	2.04	9.88	48.56	39.51	0.45	65.30	0.66	0.66	21.01	26.97

coal and the three blends of hydrochar and coal might have a lower SPONCOM index than biomass and hydrochar.

The fixed carbon content results for the samples correspond to the trend seen in their total carbon content. Samples with a high SPONCOM susceptibility had a high carbon content, according to Onifade et al. (2020). A higher SPONCOM liability for the hydrochar used in this study compared to other fuels could be a result of the hydrothermal carbonization of biomass. It is reported in the literature that coalification of coal increases carbon content leading to the densification and reduction of coal porosity (Ceballos, Hawbolbt, and Helleur 2015). This is the same phenomenon that occurs when raw biomass is carbonized, leading to a hydrochar with high total carbon content. The high carbon content of hydrochar produced in this study may result in a higher SPONCOM liability than discard coal.

The volatile matter content in fuels can be used to predict their liability to SPONCOM. According to Manic et al. (2021), biomass feedstock with high volatile matter is highly susceptible to SPONCOM. Onifade et al. (2020) reported that coal samples with a high average content of volatile matter have a high TG_{spc} index. Given the high proportion of volatile matter in biomass and hydrochar, both samples are likely more reactive and easily combustible than others (Liu et al. 2021; Shah 2022). Both biomass and hydrochar fuels are likely more prone to SPONCOM as they had the highest oxygen content compared to other fuels (Table 1). This is because high oxygen concentration increases the combustion risk and self-ignition property of fuels (Huangfu et al. 2018; Ren et al. 2021).

As reported in Table 1, the HTC of biomass to hydrochar improved the heating content of the biomass from 17.25 MJ/kg to 29.58 MJ/kg. The hydrochar produced in this study can be classified as a grade A fuel, as according to Steyn and Minnit (2010), coal or solid fuel with a calorific value ≥ 27.5 MJ/kg is deemed as a grade A fuel. Hydrochar also has an identical characteristic to medium-rank bituminous coal with fixed carbon ranging from 45% to 86% and a calorific value in the range of 19.3 to 30.2 MJ/kg (Pang 2016). Biomass had a calorific value of 17.28 MJ/kg, followed by discard coal with a calorific value of 18.55 MJ/kg. Furthermore, the calorific value increases as the ash content decreases as seen in Table 1. The blending of the discard coal with hydrochar resulted in improved calorific value among other properties. The 75% hydrochar + 25% discard coal can be classified as a grade B fuel given that it has a calorific value greater than 26.5 MJ/kg (Steyn and Minnit 2010). Based on the physicochemical properties of the fuels, the 100% biomass may be more susceptible to SPONCOM than the other samples.

FTIR Analysis

The active functional groups in the samples have a significant impact on SPONCOM liability, so FTIR was used to determine changes in these functional groups. The spectra obtained for each sample are presented in Figure 2. The makeup of each functional group in carbonaceous material affects the material's propensity to SPONCOM (Geng et al. 2009; Xu 2017). Hydroxyl, carbonyl and carboxyl are the three predominant oxygen functionalities, especially in coal (Kadioglu and Varamaz 2003). A high concentration of the three oxygen functional groups in a sample may result in an increased contact time with air leading to increased self-heating. The absorption peaks at 3691 cm^{-1} was assigned to the hydroxyl group ($-\text{OH}$), which was found in discard coal, 25% hydrochar + 75% discard coal, 50% hydrochar + 50% discard coal and 75% hydrochar + 25% discard coal. The discard coal had

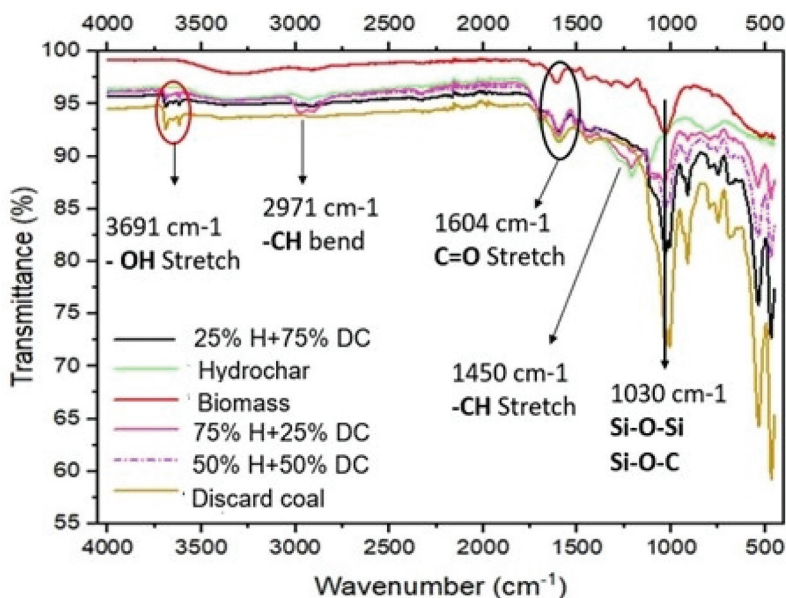


Figure 2. The FTIR spectra of six samples. OH: Hydroxyl; -CH: Aliphatic; C=O: Carbonyl; -CH: Aromatic; Si-O-Si: Silicates.

a well pronounced -OH group on its surface, indicating that this sample is highly oxidized due to weathering conditions. With -OH being dominant in this sample, it is expected to be loosely packed, leading to the likelihood of undergoing SPONCOM at low temperatures (Zhai et al. 2019). Furthermore, the -OH stretch is responsible for the mass loss when SPONCOM takes place (Zhang et al. 2021).

The aliphatic hydrocarbon group is also one of the most reactive groups that form a part of the main reactant leading to SPONCOM, given that it has reactive side chains (Lu and Hu 2007). The absorption peaks at 2971 cm^{-1} are ascribed to the symmetric stretching aliphatic hydrocarbon, which can be seen in the 75% hydrochar + 25% discard coal and hydrochar. This shows the formation of methyl groups in the hydrothermally treated biomass. Aliphatic compounds are reported to release heat energy, which is responsible for self-heating as this group oxidizes readily (Moroeng 2015). Tang (2015) and Zhong et al. (2019) also showed that high levels of aliphatic hydrocarbons increase coal's liability to SPONCOM. All samples also had a carbonyl group, which is known to decompose thermally leading to SPONCOM at temperatures above the accepted low temperatures required for oxidation (Moroeng 2015). The stretch on 1604 cm^{-1} (carbonyl group) is present in all samples as seen in Figure 2 and may exist in various forms depending on the type of sample. From the ultimate analysis, all samples were rich in carbon content (Xu 2017). The aromatic group at 1450 cm^{-1} is more prominent in hydrochar and 75% hydrochar + 25% discard coal as seen in Figure 2. This group is formed from the upgrade made to the low proportion of lignin in raw biomass through pyrolysis, hydrothermal liquefaction and gasification (Carlson et al. 2009; Mahajan et al. 2020). According to Kumar and Anand (2019), an aromatic group in biomass-derived fuels is associated with volatile matter that is composed of aromatic hydrocarbons, sulfur and different chains of

hydrocarbons. It is expected that materials with high volatile matter will be more susceptible to SPONCOM (Manic et al. 2021; Onifade 2018).

The sharp peak at 1030 cm^{-1} in the fingerprint region represents the silicates associated with the inorganic minerals in the samples. This peak is more pronounced in the biomass sample but was completely removed in the hydrochar as seen in Figure 2 because of biomass minerals that are released into the water phase during hydrothermal treatment (Setsepu et al. 2021). The fingerprint section reveals that discard coal had the most mineral content among all fuels. It is expected that coal will be the most mineral-rich with the highest ash content of all samples as seen in Figure 2. According to Onifade (2018), the higher the ash concentration, the lower the sensitivity to SPONCOM. Consequently, biomass and hydrochar are expected to be more susceptible to SPONCOM than other high-ash fuels.

SPONCOM Analysis

The propensity of different fuels to undergo SPONCOM can be predicted using various techniques. Even though some techniques have been well-established in their application, no technique has been chosen as a standard among the various approaches (Onifade and Genc 2019; Said et al. 2022; Wongthonglueang et al. 2022). This is because the dependability of all the procedures has not been validated. The procedure applied has always been based on the idea that if a substance can experience an exothermic reaction, it is also capable of SPONCOM (Onifade 2018). TGA and Wits-Ehac tests were conducted for this study to have a detailed understanding of fuel's self-heating and SPONCOM liability.

TGA Analysis

The objective of the TGA is to quantify the weight loss of a sample as the temperature changes given that the information can provide insight into the propensity of a sample to undergo SPONCOM. For some fuels like coal, the weight change, which is a factor of temperature and time, can be investigated experimentally using TGA. For this study, the self-heating characteristics of the fuels used was evaluated based on the approach used by Avila (2012), Torrent-Garcia et al. (2015) and Manic et al. (2021) on coal, biomass and coal-biomass blends. The derivative weight vs time and temperature, as well as the slope based on the linear portion of the derivative curve at low temperatures, are determined from the combustion profiles of each fuel. Equations 4 and 5 were used to derive these major variables from the TGA thermographs.

$$m = \frac{\partial m}{\partial t} f(T, t) \quad (4)$$

$$m = \frac{\partial m}{\partial T} f(T, t) \quad (5)$$

The discard coal, biomass, hydrochar, and hydrochar plus coal blends at different ratio's derivative weight versus temperature and time profiles were produced at six heating rates (4, 8, 12, 16, 20 and $24^{\circ}\text{C}/\text{min}$). A linear trend line was utilized to simulate the linear segment that occurs at the first heating stage, where the devolatilization reaction starts for each sample exposed to the six heating rates. This linear section is relevant to SPONCOM phenomenon since it shows the oxidation process in its early phases (Avila 2012).

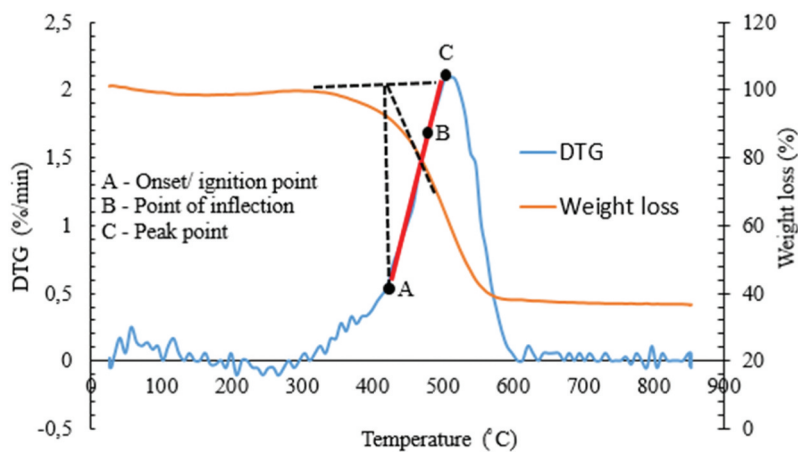


Figure 3. Derivative weight vs time profile used to calculate the gradient.

Since each fuel has a unique linear segment at different heating rates, a graphical evaluation (Fig. 3) was conducted to examine the ignition temperature, point of inflection and peak temperature of the fuels. The point of inflection on the curve was calculated using linear interpolation in Equation 6. The onset point known as ignition temperature was found using the intersection approach (Isaac 2019). The three crucial locations that were utilized to determine the slope of the linear line on the curve are shown in Figure 3.

$$Y = Y1 + (X - X1) \frac{(Y2 - Y1)}{(X2 - X1)} \tag{6}$$

The TGA analysis was first carried out on 100 mg of discard coal, which was the same mass used by Avila (2012) and Onifade et al. (2020). The slopes and derivative profile of the discard coal at six heating rates are shown in Figure 4. Table 2 also lists the three locations

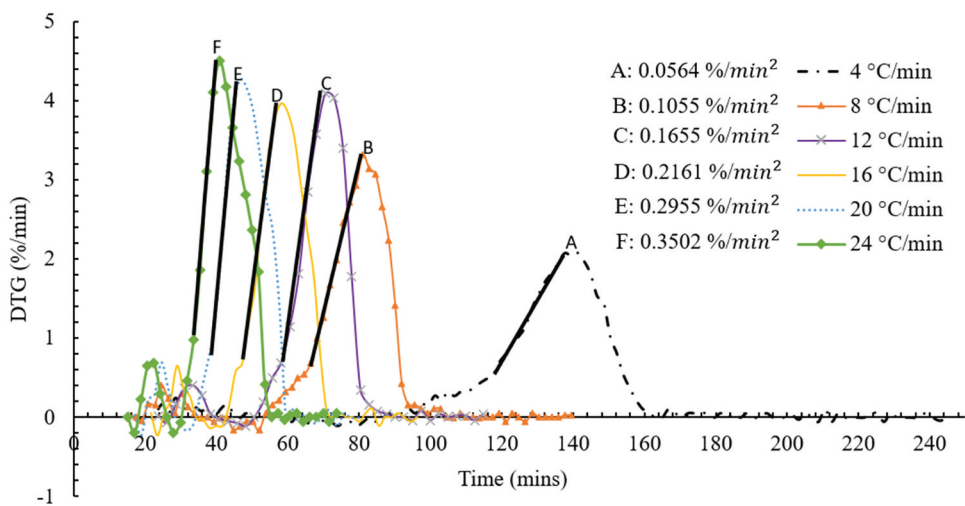


Figure 4. DTG profiles and slopes produced for discard coal at different heating rates.

Table 2. Specific temperatures and differential thermogravimetric values used to calculate the slopes of the derivative profile of discard coal at various heating rates.

Heating rate °C/min	Unit	Ignition Point	Inflection Point	Peak Point	Slope r	R^2
4	Time (min)	117.55	137.65	141.28	0.0564	0.9999
	DTG (%/min)	0.74	1.88	2.08		
8	Time (min)	66.45	79.17	8.95	0.1055	0.9989
	DTG (%/min)	1.79	3.13	3.32		
12	Time (min)	6.55	68.00	7.48	0.1655	0.9937
	DTG (%/min)	2.45	3.68	4.10		
16	Time (min)	49.07	56.40	58.23	0.2161	0.9832
	DTG (%/min)	1.99	3.58	3.97		
20	Time (min)	39.22	44.70	46.53	0.2955	0.9760
	DTG (%/min)	2.11	3.73	4.27		
24	Time (min)	33.50	39.02	4.82	0.3502	0.9824
	DTG (%/min)	1.95	3.88	4.51		

and their regression coefficients that were utilized to determine the slope of the linear segment for each heating rate.

The heating rate and the linear segment on the discard coal derivative profile have a direct proportional relationship, as demonstrated in Figure 4. As the heating rate increases, the slope of the linear segment increases as the sample burns more quickly and becomes more reactive (Avila, Wu, and Lester 2014; Bada et al. 2015). In contrast to those tested at higher heating rates, the discard coal sample that was examined at lower heating rates burned out after 160 minutes. It is also noteworthy that more reactive samples are linked to higher peak values. Figure 4 further shows that the sample becomes extremely reactive as the heating rate rises. There is a significant correlation between the variables under investigation, as seen by the regression coefficients (R^2) for the approximated linear segment at various heating rates becoming closer to 1.

The slope values in Table 2 were used to calculate the SPONCOM index (TG_{spc} index) of discard coal. The TG_{spc} index is the ratio between the weight loss rate and the temperature change caused by an external heating source (Avila 2012; Onifade, Genc, and Bada 2020).

The heating rates were divided into three ascending sets: lower (4, 8, 12, 16°C/min), middle (4, 8, 12, 16, 20°C/min), and higher (4, 8, 12, 16, 20, 24°C/min) to calculate the SPONCOM liability indices for the fuels. The TG_{spc} index of the samples at the lower, middle, and higher heating rates was calculated for each sample by plotting the three sets of heating rates against the slopes of the derivative curves. The regression coefficient was also considered when producing the TG_{spc} index to ensure accurate findings. Figure 5 displays the TG_{spc} index, the regression coefficient for discard coal, and the slopes of the derivative curve profile against heating rate. Appendix A indicate the figures showing similar information for the other samples and their TGA profiles, while Appendix B shows the heating rates used against the slopes of the derivative curve for other samples.

The slope of the approximate linear line that passes through the point of the thermographs, as depicted in Figure 5 was used to calculate the sample's TG_{spc} index. The discard coal's TG_{spc} index was 0.0135%/C.min at lower heating rates (4, 8, 12, and 16°C/min). Given that the conditions at lower heating rates are considerably more like the actual conditions, Onifade et al. (2020) claim that the SPONCOM liability of a sample may be anticipated based on the TG_{spc} index of the sample. For discard coal at the higher and middle heating rates, the TG_{spc} index was 0.0147 and 0.0149%/C.min, respectively. The indices at the lower, middle, and

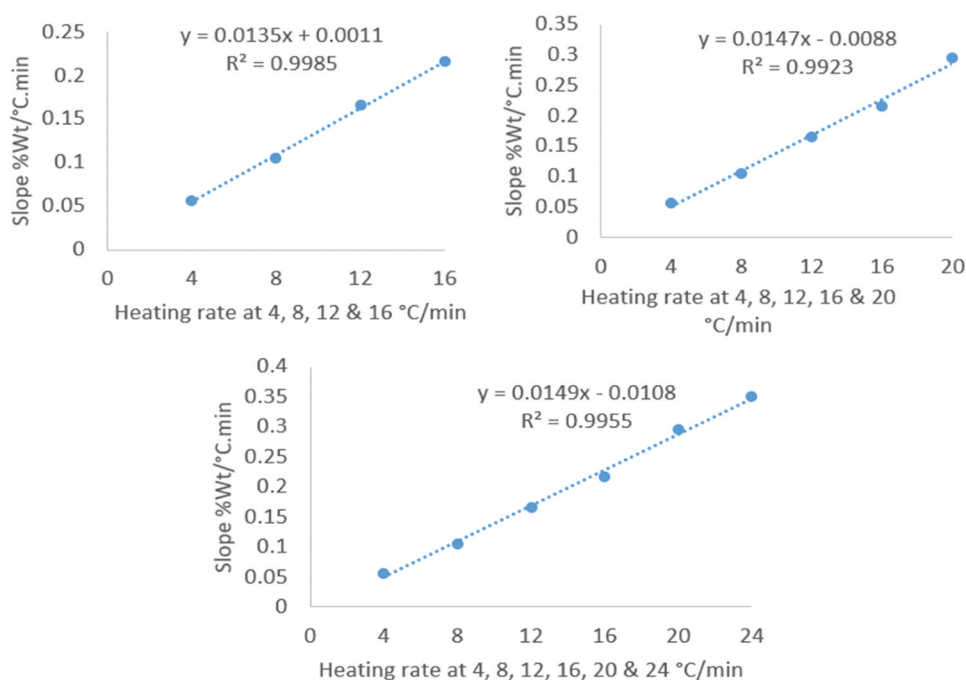


Figure 5. Heating rates for discard coal against the slopes of the derivative curve.

Table 3. Calculated slopes for all samples at different heating rates.

Sample	Heating Ramp Applied (°C/min)					
	4	8	12	16	20	24
Biomass	0.2545	0.8543	1.3922	2.0177	2.6788	3.3056
Hydrochar	0.0599	0.1601	0.2533	0.3320	0.4326	0.5621
25% H + 75% DC	0.0800	0.1584	0.2530	0.3202	0.4133	0.5007
50% H + 50% DC	0.0330	0.0992	0.1859	0.2779	0.3573	0.4499
75% H + 25% DC	0.0586	0.1346	0.2286	0.3093	0.4071	0.4942

Table 4. TG_{spc} and regression of fuels in descending order at the various heating rates.

Order	Samples	4, 8, 12, 16 (°C/min)		4, 8, 12, 16, 20 (°C/min)		4, 8, 12, 16, 20, 24 (°C/min)	
		TG_{spc}	R^2	TG_{spc}	R^2	TG_{spc}	R^2
1	Biomass	0.1457	0.9992	0.1503	0.9987	0.1525	0.9990
2	Hydrochar	0.0227	0.9971	0.0229	0.9985	0.0243	0.9948
3	75% H + 25% DC	0.0212	0.9985	0.0218	0.9984	0.0220	0.9970
4	50% H + 50% DC	0.0205	0.9948	0.0207	0.9974	0.0211	0.9981
5	25% H + 75% DC	0.0204	0.9962	0.0207	0.9979	0.0210	0.9986
6	Discard coal	0.0135	0.9985	0.0147	0.9923	0.0149	0.9955

higher heating rates did not differ significantly from one another. These results also indicate that the SPONCOM liability of discarded coal rises as the heating rates increase because the ignition temperature is reached faster as the heating rate increases (Manic et al. 2021).

The procedure for calculating the TG_{spc} index was applied to the other fuels, biomass, hydrochar, 25% hydrochar + 75% discard coal, 50% hydrochar + 50% discard coal and

Table 5. SPONCOM liability based on TG_{spc} (Manic et al. 2021).

TG_{spc} Index	Class
<0.02	Non-reactive
0.02–0.03	Low reactive
0.03–0.05	Reactive
>0.05	High reactive

75% hydrochar + 25% discard coal. Table 3 presents the calculated slopes for each sample at different heating rates while Table 4 shows the TG_{spc} indices and regression coefficients of the samples at various groups of heating rates in descending order. Lastly, Table 5 presents a general TG_{spc} index classification values for fuel's SPONCOM liability.

From the results presented in Table 4, biomass was highly reactive compared to other fuels since its TG_{spc} index is 0.1457. Any samples with a TG_{spc} index > 0.05 are categorized as highly reactive by Table 5. With biomass containing a high amount of volatile matter and moisture, but a low amount of ash, it is anticipated that it will be more susceptible to SPONCOM than the other five fuels in Table 4. The TG_{spc} index for the samples increased with increasing heating rates as expected because the rate of oxygen absorption becomes rapid as the temperature increase as shown in Table 3 (Mandal et al. 2022).

The discard coal and 25% hydrochar + 75% discard coal, which have a high ash content and low volatile matter (Table 1), have the lowest SPONCOM liability. The findings agree with those made public by Manic et al. (2021) and Torrent- Garcia et al. (2015). The assessment of the self-ignition risks of several types of solid biofuels by thermal analysis by Torrent- Garcia et al. (2015) revealed a similar indirect association between the fixed carbon and the TG_{spc} index. The correlation between the total sulfur content of the samples used in this study and their SPONCOM liability is opposite to that reported by Onifade et al. (2020). For this study, the samples that exhibit high SPONCOM liability had low total sulfur content. The difference might be a result of the difference in the composition of the samples. Onifade et al. (2020) used coal and coal-shale, whereas this study used biomass, hydrochar and hydrochar/coal blends.

Wits-Ehac Analysis

The sample's SPONCOM liability was determined from the DTA thermographs generated for the individual samples. The two important parameters used in determining the Wits-Ehac index are the XPT and Stage II slope of the DTA curve. To generate a DTA thermogram, the difference between the temperature of the sample and that of the inert material (differential temperature) is plotted against the temperature of the inert material. This technique has been used for coal, coal-shale and activated carbon several times, but not to predict the SPONCOM liability of biomass and hydrochar/coal blends. The results in the form of a DTA thermogram of the samples are presented in Figure 6.

Table 6 summarizes the sample's XPT and Stage II slopes in descending order of the Wits-Ehac index. The Wits-Ehac computer system generates the DTA thermogram after analyzing each sample, providing the XPT and Wits-Ehac index values. Equation (2) was used to obtain the Stage II slope values, and Table 7 shows the risk rating classification of the samples based on Wits-Ehac index values.

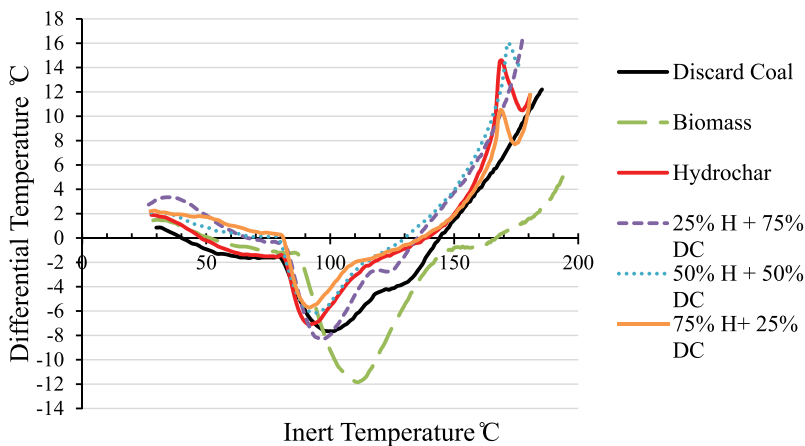


Figure 6. DTA thermogram for the samples.

Table 6. XPT, stage II slope and Wits-Ehac slope for the samples in descending order.

Order	Sample	XPT (°C)	Stage II Slope	Wits-Ehac Index
1	50% H + 50% DC	13.80	1.237	4.73
2	25% H + 75% DC	136.00	1.257	4.62
3	100% Hydrochar	139.40	1.252	4.49
4	75% H + 25% DC	137.00	1.178	4.30
5	Discard coa1	144.20	1.214	4.21
6	Biomass	167.50	1.169	3.49

Table 7. Classification of the SPONCOM liability (Wade, Gouws, and Phillips 1987).

Wits-Ehac Index	Risk
<3	Low risk
3–5	Medium risk
>5	High risk

The XPT for the six samples ranged from 130.8°C to 167.5°C, while their Stage II slope ranged from 1.169 to 1.257, and the Wits-Ehac index range was from 3.47 to 4.73 respectively. The results in Table 6 show that the higher the XPT, the lower the Wits-Ehac index. A low XPT shows that a sample ignites quickly and is, therefore, highly reactive and prone to undergoing SPONCOM easily. The results also show that 50% hydrochar + 50% discard coal is highly liable to SPONCOM, given that it had the highest SPONCOM index, followed by 25% hydrochar + 75% discard coal and hydrochar. Contrary to the TGA SPONCOM test, the Wits-Ehac results further show that biomass had the lowest Wits-Ehac index.

Comparison Between the TGA and Wits-Ehac Results

Given that the two SPONCOM tests investigated in this study were the TGA and the Wits-Ehac index, the results of the SPONCOM liability of the six samples from the two tests are shown in Table 8 for comparison. The results presented for the TGA test are those that were

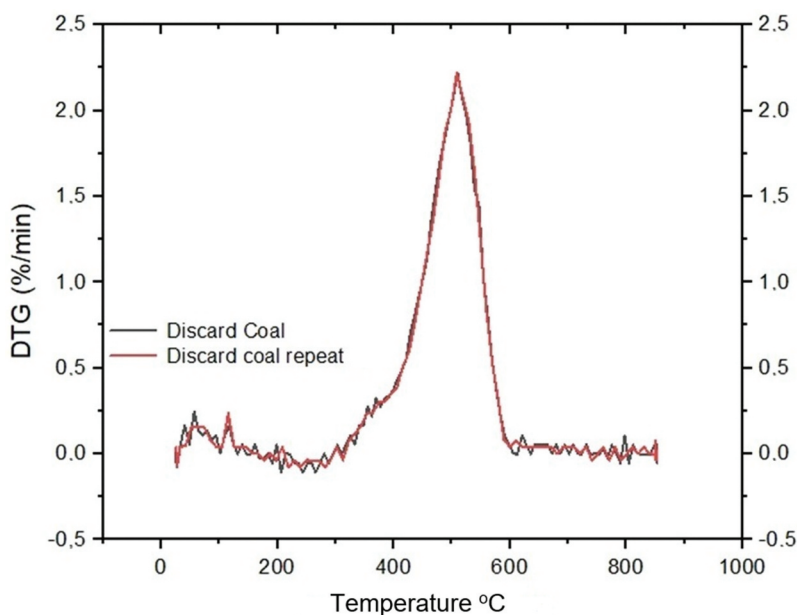
Table 8. Comparison of the TGA and wits-ehac results in descending order.

Order	Samples	TG_{spc}	Order	Samples	Wits-Ehac Index
1	Biomass	0.1457	1	50% H + 50% DC	4.73
2	Hydrochar	0.0227	2	25% H + 75% DC	4.62
3	75% H + 25% DC	0.0212	3	Hydrochar	4.49
4	50% H + 50% DC	0.0205	4	75% H + 25% DC	4.30
5	25% H + 75% DC	0.0204	5	Discard coal1	4.21
6	Discard coal	0.0135	6	Biomass	3.49

found at lower heating rates, given that situations with lower heating rates are the ones that are most similar to the real-world circumstances as reported in Section 3.3.1.

The results presented in Table 8 are arranged in descending order for each test. For the TGA test, biomass was found to have the highest TG_{spc} index at low heating rates. Whereas the same sample had the least SPONCOM liability centered on the Wits-Ehac index. The hydrochar had the second highest TG_{spc} index and the third highest Wits-Ehac index. Based on the TGA test, the discard coal was predicted to have the lowest SPONCOM liability and the second lowest based on the Wits-Ehac index. The two tests show no correlation for any sample. It is important to note that the results obtained for the TGA test can be supported by the physicochemical properties of the samples, as discussed in section 3.2.1. However, the opposite may be said about the Wits-Ehac index results, as there is no direct relationship between the Wits-Ehac data, proximate and ultimate analysis results.

Since biomass had the utmost and minimal SPONCOM potential for the TGA and Wits-Ehac test, respectively, the sample was re-tested to verify the repeatability of the biomass sample using the Wits-Ehac test. The repeatability test for TGA test was carried out using discard coal and the results are presented in Figure 7. The TGA results were reliable, reproducible and associated with the sample characterization results. Figure 8 presents a DTA thermogram for a repeated biomass test.

**Figure 7.** Repeatability test of discard coal at a heating rate of 4°C/min.

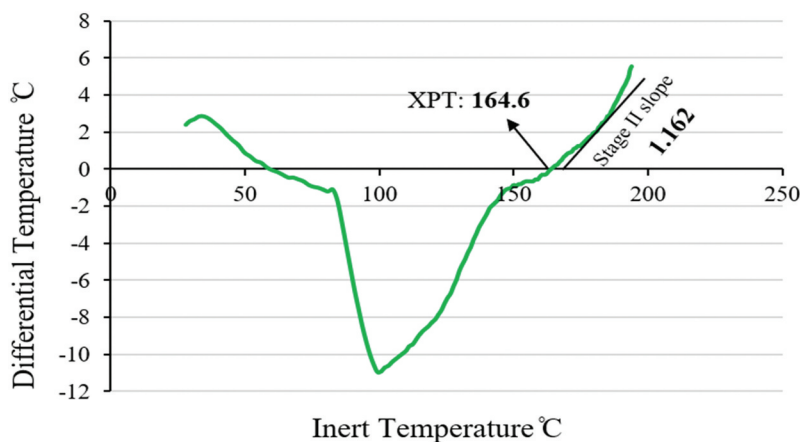


Figure 8. DTA thermogram for a repeated biomass.

The XPT for the first biomass sample as seen in Figure 6 was 167.5 °C, and the repeat test was 164.6 °C. The first and repeat tests had a Wits-Ehac index of 3.49 and 3.53, respectively. This shows that the Wits-Ehac results are repeatable; however, the data obtained from the biomass and hydrochar/coal blends are not supported by the sample's characterization results or any data from previous studies. This method has been proven to be reliable and accurate for the analysis of coal samples (Onifade 2018; Onifade, Genc, and Bada 2020), but this was not the case for biomass used in this study. The difference in the bulk density of the biomass (18 kg/m³), hydrochar and hydrochar/coal blends to coal (700 kg/m³) might be responsible for the discrepancy. In addition, since the sample holding cells of the Wits-Ehac apparatus are the same volume, less biomass sample (15 g) was used to fill the same cell, with coal at about 25 g. Given also that SPONCOM occurs due to a chemical reaction between the sample and oxygen, it could be that the samples are not exposed to similar airflow due to the difference in their bulk density and porosity. Given the mass difference of the samples used in a fixed standard Wits-Ehac cell, the SPONCOM results obtained from the Wits-Ehac cannot be compared with those of the TGA.

Conclusion

This research aimed to determine whether the Wits-Ehac and TGA thermal analysis tests are suitable techniques for predicting the SPONCOM potential of coal, biomass and hydrochar/coal blends. The main findings of this study are as follows:

- (i) The conversion of biomass into hydrochar by hydrothermal carbonization improves the energy quality of the fuel. The hydrochar had reduced ash and volatile matter content, as well as higher fixed carbon and calorific values compared to the biomass.
- (ii) The physicochemical analysis conducted on discard coal, biomass, hydrochar and their blends revealed that biomass and discard coal have the lowest energy content. Blending hydrochar with coal discard at different ratios provides better quality blends. The 75% hydrochar + 25% discard coal is of the best quality, with a fixed carbon, volatile matter, moisture content and calorific value of 48.56%, 39.51%, 2.04% and 26.97 MJ/kg, respectively.

- (iii) The Wits-Ehac indices indicated that there was no direct link between the results of the Wits-Ehac SPONCOM test and those of the physicochemical properties of the six samples. However, the Wits-Ehac technique proved to be reproducible for all samples tested, even though it is not a reliable test for biomass, hydrochar, and the hydrochar/coal blends due to the difference in their bulk density from that of coal.
- (iv) The TGA revealed that biomass was largely susceptible to SPONCOM related to other fuels with a TG_{spc} index of 0.1457%/°C.min at lower heating rates. The results further revealed that discard coal had the lowest SPONCOM liability with a TG_{spc} index of 0.0135%/°C.min at lower heating rates. The physicochemical properties of these samples correlate the findings from the TGA.
- (v) The biomass had a high moisture content of 8.01%, a high volatile matter content of 60.52%, a low fixed carbon content of 19.54%, and a low calorific value of 17.28%. The combination of these high physicochemical properties resulted in higher SPONCOM in biomass compared to discard coal with lower physicochemical properties.
- (vi) According to the FTIR analysis, the high susceptibility of biomass to SPONCOM was caused by functional groups (C=O) and poor silicate (mineral) transmission. Whereas the presence of a pronounced silicate peak in coal for discard coal may be the reason why it is least liable to SPONCOM.

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Author Contribution

Ethel Matsobane: Methodology, Investigation & interpretation of data, Formal analysis, Writing – original draft, Writing – review & editing. Moshood Onifade: Investigation & interpretation of data, Writing – review & editing. Bekir Genc: Methodology, Resources, supervision, Writing – review & editing. Samson Bada: Conceptualization, Methodology, Funding acquisition, Resources, Supervision, Writing – review & editing.

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Appendix A

A. Thermogravimetric analysis derivative profiles of the samples

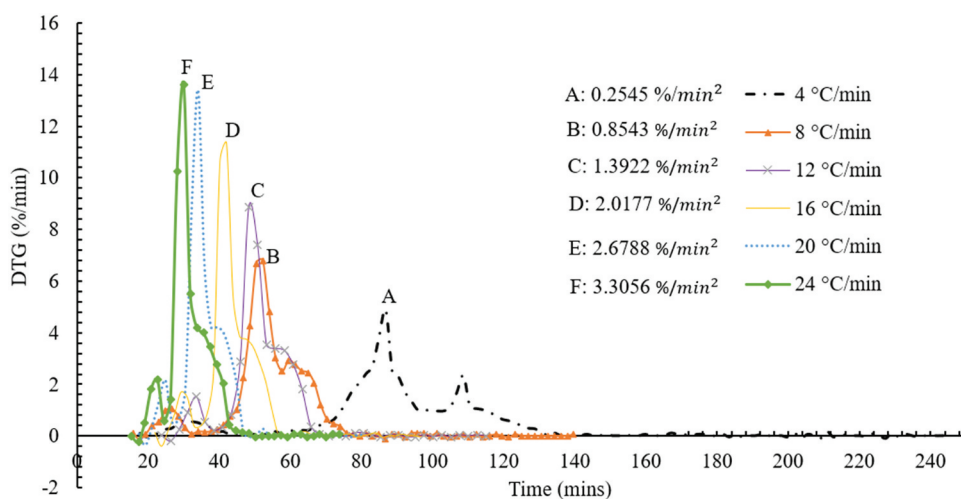


Figure A1. Derivative profile of biomass.

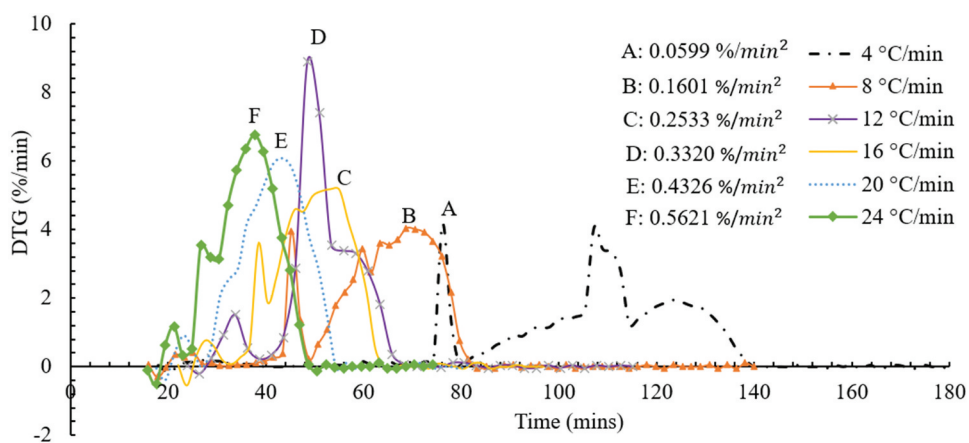


Figure A2. Derivative profile of hydrochar.

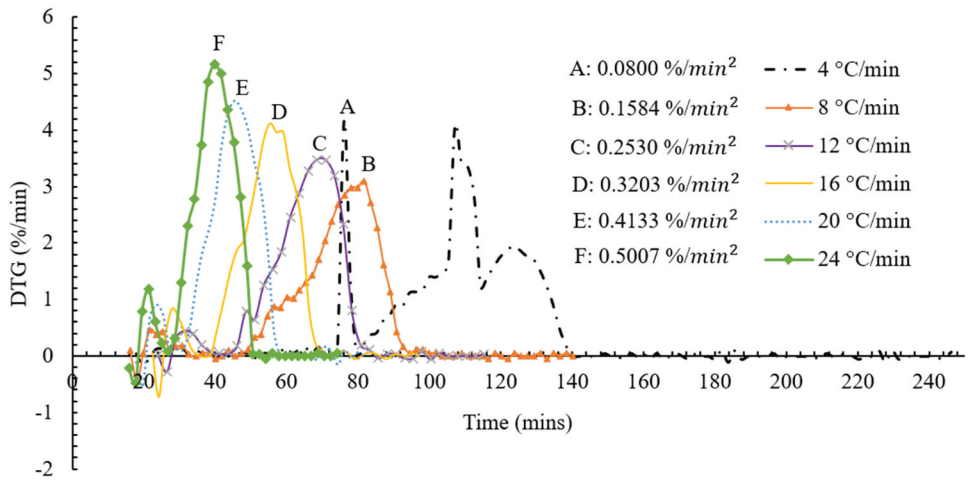


Figure A3. Derivative profile of 25% hydrochar + 75% discard coal.

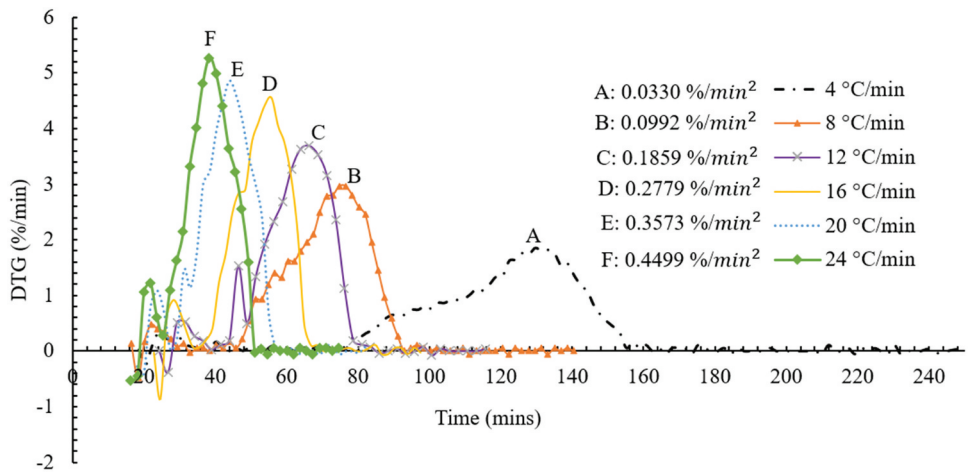


Figure A4. Derivative profile of 50% hydrochar + 50% discard coal.

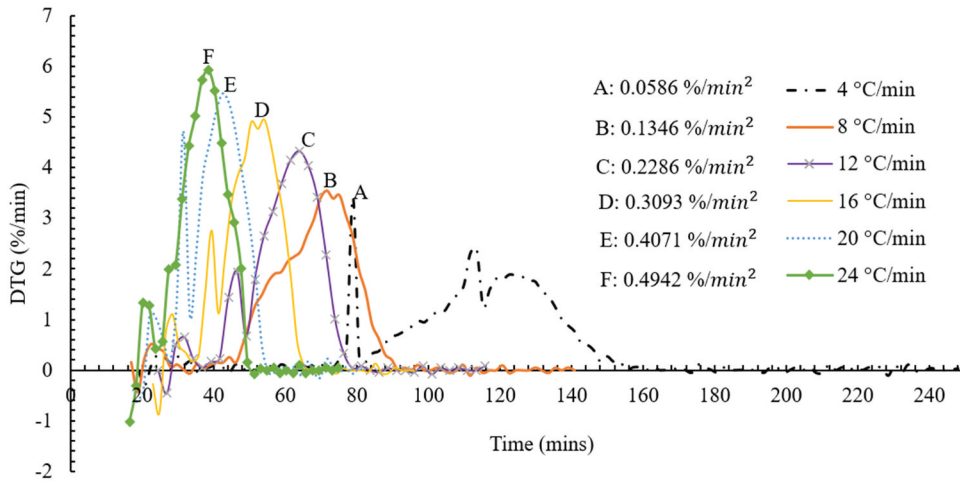


Figure A5. Derivative profile of 75% hydrochar + 25% discard coal.

B. Heating rates used against the slopes of the derivative curve for the samples

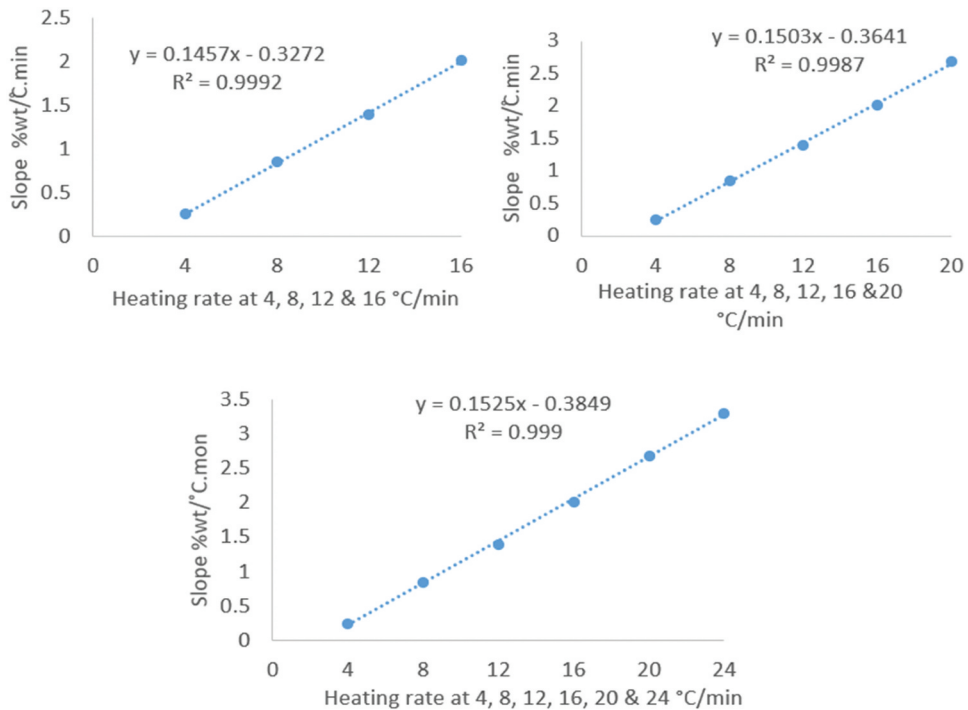


Figure B1. Heating rates against the slopes of the derivative curve for biomass.

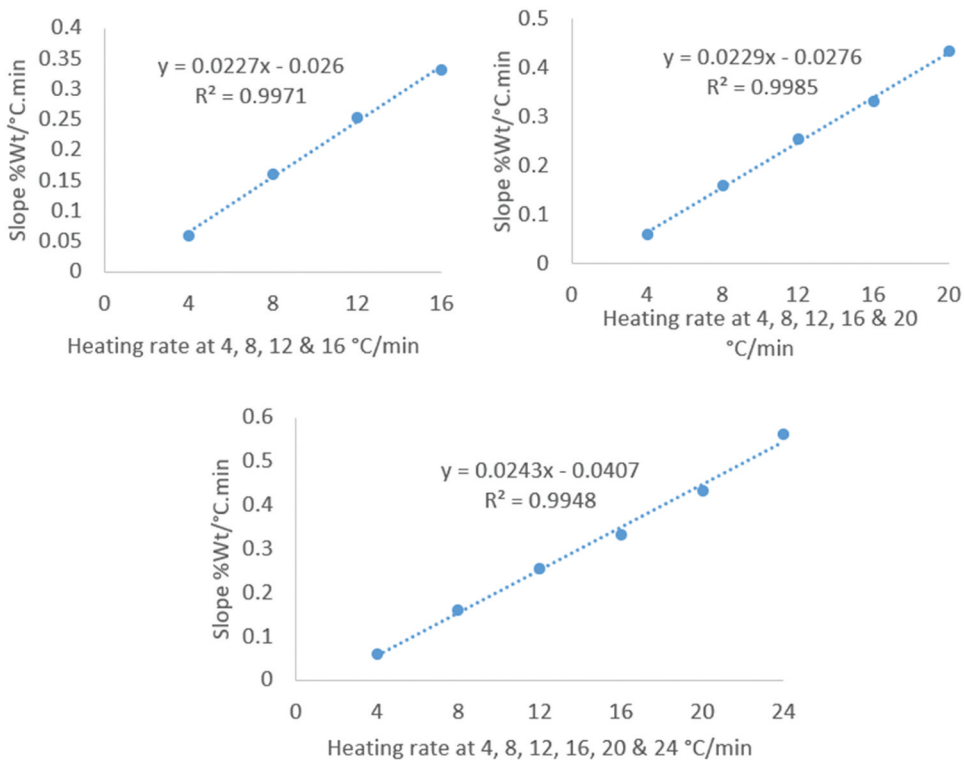


Figure B2. Heating rates against the slopes of the derivative curve for hydrochar.

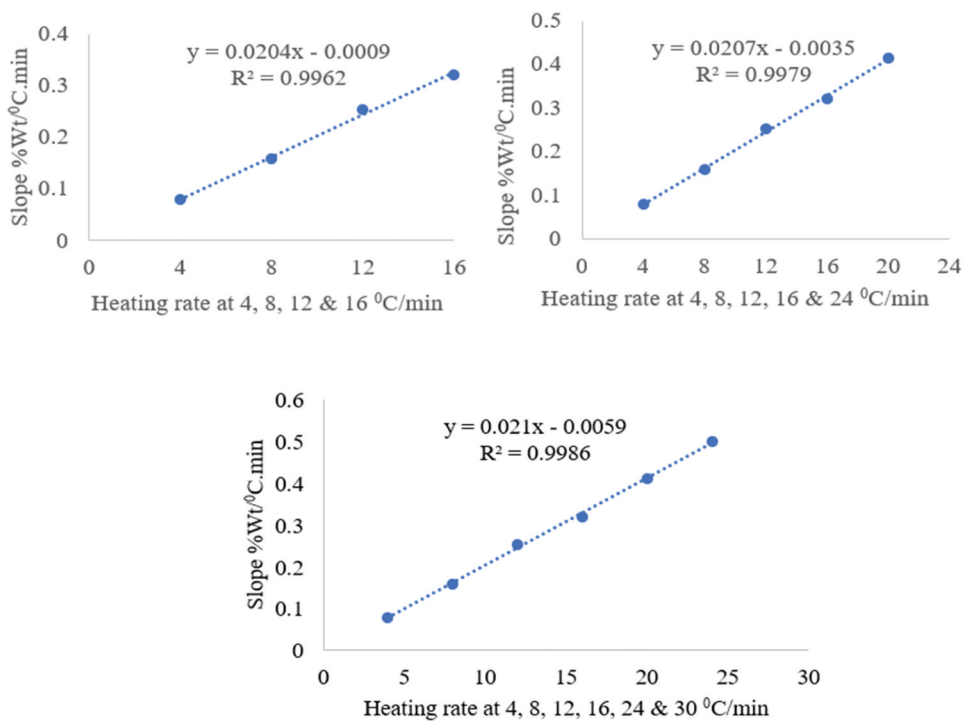


Figure B3. Heating rates against the slopes of the derivative curve for 25% hydrochar + 75% discard coal.

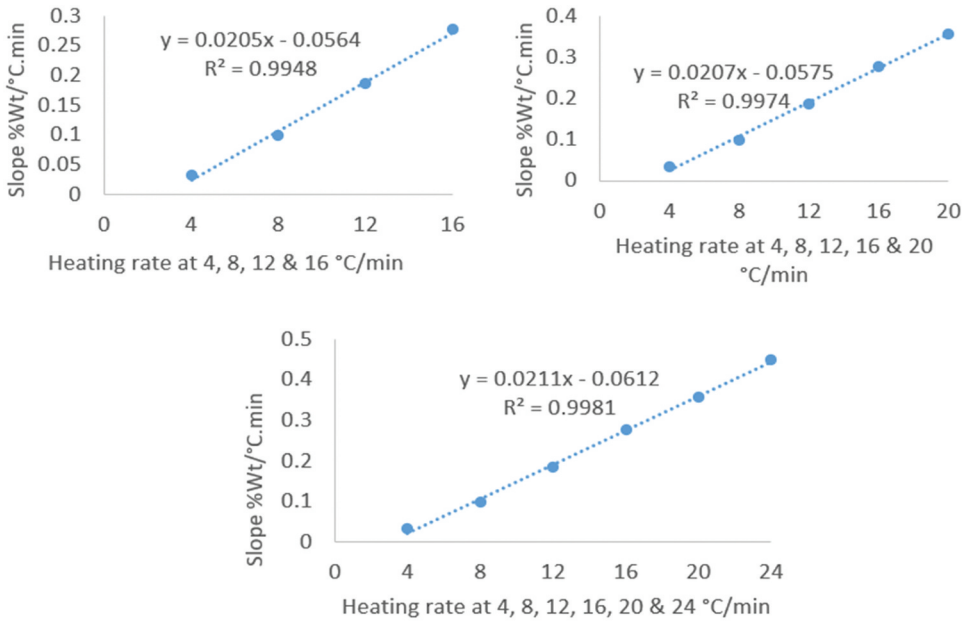


Figure B4. Heating rates against the slopes of the derivative curve for 50% hydrochar + 50% discard coal.

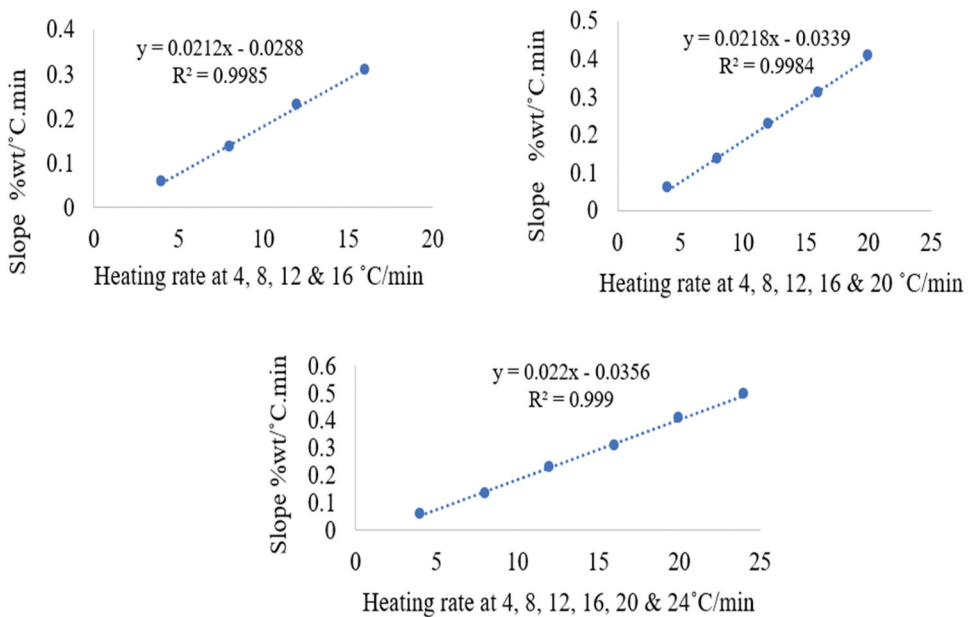


Figure B5. Heating rates against the slopes of the derivative curve for 75% hydrochar + 25% discard coal.