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Thermal Analysis of Co-Utilization of Empty Fruit Bunch and Silantek Coal Under Inert Atmosphere Using Thermogravimetric Analyzer (TGA)

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Agricultural residues have been traditionally used as energy resources for many years. In light of current environmental and fossil fuel supplies for energy applications, agricultural residues are regarded as sustainable supplies for energy production. However, the suitability to be renewable fuel and as a co-fuel in coal combustion facilities has to be investigated. A thermal analysis was conducted to investigate the effect of the blending and heating rate of the thermal behavior of Malaysian bituminous coal (Silantek), oil palm biomass (empty fruit bunch), and their blends using thermogravimetric analysis. The investigation was done in an inert atmosphere at the heating rate of 10, 20, and 40°C/min. Characteristics including proximate, ultimate, and calorific analyses were also examined. Six different mass ratios were selected from both samples to study the effect of blending of the two materials. The results showed that thermal degradation of empty fruit bunch (EFB) occurred in three stages while Silantek coal (SC) only involved two regions due to their different fuel properties. The blending of both SC/EFB did not follow their individual samples, which showed non-additive behavior suggesting that there is an interaction between coal and biomass. The outcome of this research provides insight on the behavior of Malaysian bituminous coal and oil palm biomass, which enhances knowledge for the future of energy generation.

Keywords: pyrolysis, Silantek coal, empty fruit bunch, thermogravimetric analysis, co-utilization, coal/biomass blends

INTRODUCTION

Concerns over the environment associated with emissions due to the use of fossil fuels as a main energy resource has been the main agenda in most countries around the world. The combustion of fossil fuels has contributed to the emission of greenhouse gases such as carbon dioxide. From 2010 to 2017, the combustion of fossil fuels has released carbon dioxide emission of about 4,188.5 million tonnes recorded in Asia alone, but the amount is increasing in 2018 due to an increase in energy demand, in line with the robust economy growth plus (IEA, 2020). The emissions have caused global warming and are indirectly increasing the need of either heating or cooling requirements globally. Currently, natural gas is the main energy source for power production in Malaysia. In 2018, about 73,352 GW Wh of the electricity generation from the total of 168,897 G Wh energy produced was fueled by coal followed by natural gas (Energy Commission, 2019). However, the depletion of petroleum energy reserves over the years has called for the industry to increase the share of coal in the total energy mix for power production. Alternative solutions to replace fossil fuels have been sought after to ensure a reduction in emission, particularly in electricity generation. This situation has led the Malaysian government to introduce the use of other energy resources to meet the increasing energy demand.

Agricultural wastes are essentially a biomass resource that has the ability to be used as fuel, as its components consist of carbon, hydrogen, and oxygen, coming from the major biomass components of hemicellulose, cellulose, and lignin. The combustion of biomass has been promoted because of the net zero carbon dioxide emission due to biomass's ability to consume carbon dioxide during its growth, while releasing carbon dioxide during combustion. Its utilization is able to reduce fossil fuel consumption because of higher fuel flexibility, higher combustion efficiency, higher heat transfer and other environmental benefits (Vuthaluru, 2004; Jayaraman et al., 2017).

From a Malaysian perspective, the tropical climate conditions that prevail around the year are favorable for oil palm plantation. This has led the country to become one of the major palm oil producers in the world (Norhidayu, et al., 2017). The palm oil milling process allows an extraction of palm oil from fresh fruit bunches. However, the processes have contributed to production of solid residues. With an increment of the residues of 5.6% from 2016 to 2017, dry solid residues (biomass) are expected to reach 85–110 million dry tonnes by 2020 (Chow, 2008; Agensi Inovasi Malaysia, 2013). It is estimated that, by the year 2020, about 8 million tonnes of empty fruit bunches (EFB) will have been produced and the amount is increasing with an increase in oil palm production every year (Agensi Inovasi Malaysia, 2013). According to Hasanuzzaman et al. (2014), based on 10 million ton/year of dry EFB, approximately 700 MW of electricity will be generated. It is predicted that the amount of EFB generated to date is sufficient to be used for electricity generation. Traditionally, empty fruit bunches have been sent to plantations for mulching, and mostly left unattended since they contain high moisture, in comparison to the palm kernel shell and palm mesocarp fiber, which are used as fuel for boilers. However, application of single biomass for energy production may lead to several problems such as low heating value, high moisture content, excess smoke during combustion and low energy density (Tumuluru et al., 2011). High ash content in these biomass materials causes major problems in a boiler furnace, as it causes slagging hence reducing heat transfer efficiency and requires frequent maintenance (Saidur et al., 2011). Nevertheless, with proper pre-treatment, and available conversion technology, such limitations can be overcome.

Much research has emerged on the co-utilization of biomass with coal for power production, which is advantageous with respect to cost, sustainability, and reduced release of CO_2, SO_x, and often NO_x emissions (Sahu et al., 2014). Coal is available at different ranks namely, anthracite, bituminous, sub-bituminous, and lignite; whereby bituminous coal is the most widely used for the thermal industry (Grammelis et al., 2016). Although much research has been reported on the importance of the co-firing of coal and biomass, limited work has proved the synergistic effect between the utilization of raw biomass and coal. Synergistic effect between the two fuels is important as it indicates interaction between the components in both of the fuels. As coal and biomass are distinct in their properties, it is important to understand the behavior of their co-utilization, prior to commercial application during combustion.

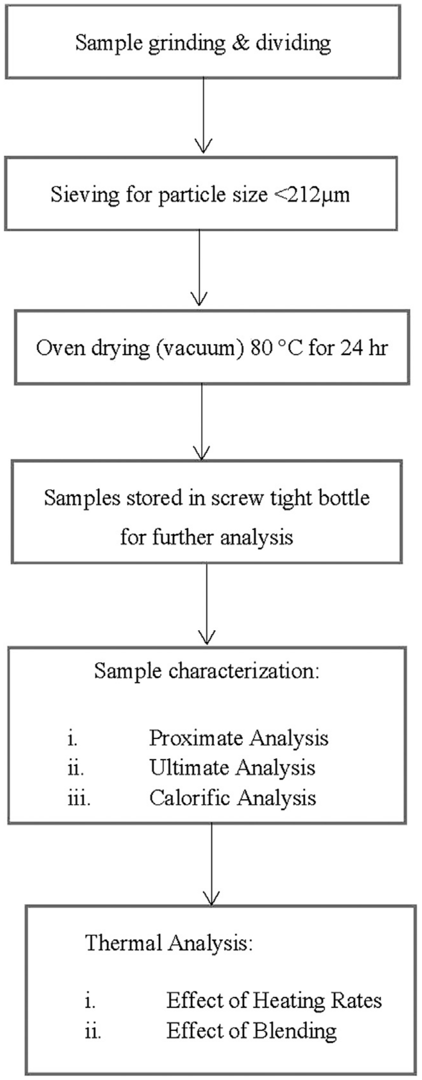

FIGURE 1 | Flow diagram for research methodology.

Previously, Silantek coal's utilization has been performed by Matali et al. (2016). However, in this work, the investigation focuses on single utilization rather than co-utilization with raw and torrefied biomass (*Leucaena Leucocephala* and oil palm frond). Jamaluddin et al. (2011) reported the co-combustion of the palm kernel shell (PKS) and its produced char from themicrowave pyrolysis technique. It was reported that there was a lack of synergistic effect during co-combustion of Silantek coal with PKS. Nevertheless, a synergistic effect was reported when the coal was co-combusted with PKS char. Thus far, limited work has been reported on using empty fruit bunch (EFB) with Silantek coal. Therefore, the aim of the current work is to investigate the thermal behavior of Malaysian bituminous coal which is Silantek coal blended with EFB by varying the heating rate and blending ratio. The significance of this work is that it provides an insight into the suitability of utilizing agriculture residue such as oil palm residue along with a high rank coal for a thermal application, which in turn could reduce the impact on the environment as a result of the waste disposal and combustion of fossil fuel (coal).

MATERIALS AND METHODS

Empty fruit bunch (EFB) was collected from Malaysia Palm Oil Board (MPOB), Palm Mill Technology, located in Labu, Negeri Sembilan, Malaysia and Silantek coal (SC) used in this study was originated from Silantek, Sarawak. Both coal and biomass were air dried for 2–3 days to remove moisture. The samples were ground and sieved to a size of <212 μm and further dried in a vacuum oven at 105°C for 24 h to remove excess moisture before being stored in screw-capped bottle. For chemical characteristics, proximate analyses of SC and EFB were done based on the standard method of ASTM D 5142–02a using a thermogravimetric analyzer model TGA/SDTA51^e (Mettler Toledo, United States) (ASTM 5142–02a). As for ultimate analysis, data on percentage of nitrogen, carbon, hydrogen, and sulfur were obtained using a Thermo Finnigan Flashed 1,112 analyzer which followed ASTM 5373–02 (ASTM D5373–02). For calorific data, both SC and EFB was analyzed using a bomb calorimeter model IKAWORKS calorimeter System C500 Control. The methodology is summarized in a process flow as in **Figure 1**. The result of the characterization of the raw sample is presented in **Table 1**.

Thermal analyses of SC, EFB, and their blends were evaluated by a thermogravimetric analyzer TGA/SDRA51^e (Mettler Toledo, United States). TGA provides simultaneous thermogravimetric (TG) and derivative thermogravimetric (DTG) data of raw and blending sample. In this experiment, approximately 20 mg of sample was placed in 150 μL alumina crucible and heated under nitrogen atmosphere of flowrate 50 ml/min, at temperatures from 25 to 900°C. All samples were heated at three different heating rates which were 10, 20, and 40°C/min. To study the effect of blending, biomass and coal were blended, at five different weight ratios which were 0:100, 20:80, 50:50, 80:20, and 100:0. Each pyrolysis was run at least twice, but more repetitions were carried out in case some inconsistencies were observed.

Synergistic investigation between SC and EFB was analyzed using a summation of the weighted average of the two fuels present in the blends given by **Eq. 1** (Vuthaluru, 2004; Vamvuka et al., 2003; Xie et al., 2018; Merdun and Laougé, 2021):

$$W_{blend} = x_{sc} w_{sc} + x_{EFB} w_{EFB} \tag{1}$$

where x_{SC} and x_{EFB} are mass fractions of SC coal and EFB in the blend, respectively and W_{SC} and W_{EFB} are normalized weight loss of SC and EFB, respectively, which were obtained from individual experiment data under the same process conditions.

TABLE 1 | Proximate, ultimate, and calorific data for EFB and SC.

	Silantek coal (SC)	Empty fruit bunch (EFB)
Proximate analysis (^{ad}) (wt%)		
Moisture	1.48 ± 0.29	7.47 ± 0.71
Volatile matter	22.05 ± 5.69	58.52 ± 5.02
Fixed carbon	62.98 ± 2.18	22.0 ± 2.71
Ash	13.49 ± 3.81	12.01 ± 3.60
Ultimate analysis (^{db}) (wt%)		
Carbon	74.7 ± 3.0	42.7 ± 0.89
Hydrogen	4.6 ± 0.5	6.16 ± 0.37
Nitrogen	1.95 ± 0.01	1.92 ± 0.18
Sulfur	0.5 ± 0.1	0.11 ± 0.10
Oxygen^a	18.25 ± 2.7	49.11 ± 1.34
Calorific value (MJ/kg)	30.12 ± 0.4	17.8 ± 0.5

^{a}Calculated by difference, ^{ad} as determined basis, ^{db} dry basis.

In a similar manner, the same equation is applied on the thermogram as shown on **Eq. 2**

$$TG_{blend} = x_{sc} TG_{sc} + x_{EFB} TG_{EFB} \tag{2}$$

where x_{SC} and x_{EFB} are mass fractions of SC coal and EFB in the blend, respectively and TG_{SC} and TG_{EFB} are thermogram curve of pure SC and EFB, respectively, which were obtained from individual experiment data under the same process conditions.

RESULTS AND DISCUSSION

Thermal Behavior of Individual Sample

Generally, coal and biomass characteristics differ widely due to their individual characteristics. From **Table 1**, it can be observed that Silantek coal (SC) contains high fixed carbon (FC), low volatile matter (VM), unlike EFB which has high VM and low fixed carbon. As for elemental characteristics, SC has a carbon content of 74.7%, while EFB only contained 42.7% of carbon. Accordingly, the calorific value of SC was observed to be almost double that of EFB. In terms of thermal behavior, **Figure 2** shows thermograms (TG) and derivative thermograms (DTG) for both individual samples of SC and EFB at a heating rate of 10°C/min. Thermal activity on a thermogram can be observed based on changes in the gradient. The changes are depicted as peaks on a derivative thermogram (DTG). It can be observed that the thermal degradation of EFB occurred in three different stages where the first stage was due to the moisture drying process followed by main devolatilization and end with a slight devolatilization process. During the moisture removal region, EFB experienced a loss of excessive moisture from 25°C to 109°C. The devolatilization stage started at a temperature range of 131°C up to 478°C. A shoulder peak at around 200°C prior to the main peak (at peak temperature 320°C) indicates the degradation of hemicellulose, while the main peak was attributed to the decomposition of cellulose (Carrier et al., 2011). The proceeding non-observable peak (curve) at about 360°C–500°C can be regarded due to degradation of lignin. This finding was evident by a work reported by Wu et al. (2014). The last stage also involved the

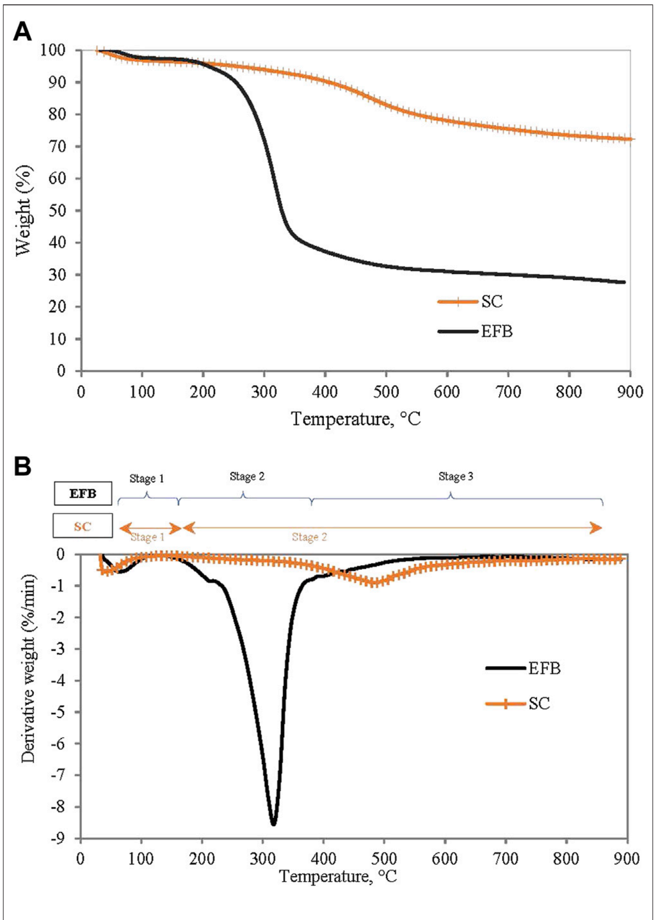

FIGURE 2 | **(A)** TG curves and **(B)** DTG curve for Silantek coal and EFB material at heating rate of 10°C/min.

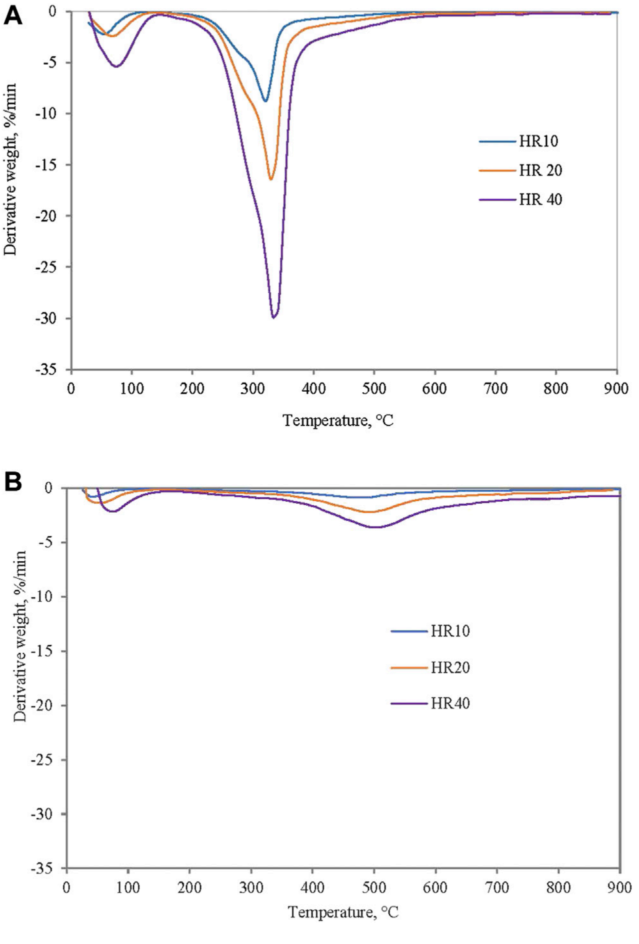

FIGURE 3 | DTG profiles of **(A)** EFB and **(B)** Silantek coal (SC) at heating rates of 10, 20, and 40°C/min under pyrolysis condition.

charring of the remaining solid char of the EFB until the end of the pyrolysis (Toptas et al., 2015; Wang et al., 2017). A similar study of EFB done by Omar et al. (2011) also reported the same thermal degradation profile which consists of the moisture drying region, cellulose and hemicellulose decomposition, and lastly lignin decomposition. As for the SC, the degradation only involved two major stages where the first stage corresponds to moisture removal, followed by volatile matter released at much a higher temperature as compared to that of EFB. Similar findings were also reported by other researchers on the pyrolysis of bituminous coal (Othman and Boosroh, 2009; Vhathvarothai et al., 2014).

From the DTG curve, reactivity of both SC and EFB can be determined based on maximum peak temperature and peak height. Reactivity is important as it indicates how reactive the fuel is or vice versa (Miranda et al., 2008). It appears that the peak temperature of EFB is lower than SC at 320°C with the peak height of 1.73 mg/min indicating that EFB is more reactive than SC. Similar findings from other researchers on comparison of biomass and coal reactivity were observed (Kastanaki et al., 2002; Omar et al., 2011; Jayaraman et al., 2017). This is because EFB contains more volatile matter than that of SC, thus reducing the reactivity of SC. The two fuels have distinct properties; hence it is important to understand the behavior during co-utilization.

Effect on Heating Rate

Pyrolysis parameters such as temperature, residence time, and heating rate are among the important factors that affect yield and desired product quality (de Jong et al., 2007). **Figure 3** shows trends of pyrolysis of EFB and SC at three different heating rates of 10, 20, and 40°C/min. It can be observed that derivative curves show similar profiles as heating rate increases for both samples. Parameters of initial, peak, and final temperatures during pyrolysis of EFB at different heating rates are shown in **Table 2**. Initial reaction temperatures of EFB at a heating rate of 10, 20, and 40°C/min are 131.31, 164.75, and 189.24°C, respectively. It is observed that the peak temperature increases with increasing heating rate. Corresponding peak temperatures for heating rates at 10, 20, and 40°C/min are 318.28, 331.29, and 335.31°C, respectively.

Final reaction temperature for a heating rate of 10°C/min is lower as compared to a heating rate of 20 and 40°C/min. Temperature shifts as heating rate is increased can be explained by ineffective heat transfer when fast heating rates was applied. In heating the same sample mass, with higher heating rates, heat transfer inside inner portion and particle of biomass is reduced, hence more time is needed to achieve the

TABLE 2 | The characteristic temperatures of EFB and SC pyrolysis at heating rate of 10, 20, and 40°C/min.

	EFB			SC		
Heating rate (°C/min)	**Initial temperature, T_i (°C)**	**Maximum peak temperature, T_m (°C)**	**Final temperature, T_f (°C)**	**Initial temperature, T_i (°C)**	**Maximum peak temperature, T_m (°C)**	**Final temperature, T_f (°C)**
10	131.31	318.28	478.08	362	450	868
20	164.75	331.29	603.63	363	495	850
40	189.24	335.31	647.92	378	504	900

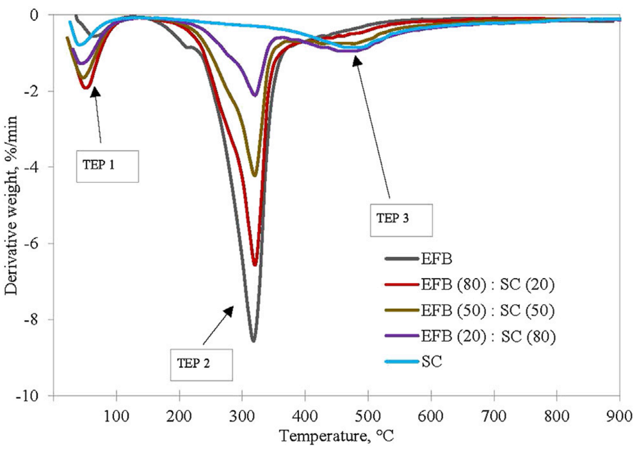

FIGURE 4 | EFB blended with SC at ratio of 100:0, 80:20, 50:50, 20:80, and 0:100 under pyrolysis condition at heating rate of 10°C/min.

same heating, which can be achieved at a later time, hence at a much higher temperature (El-Sayed and Khairy, 2015). This data agrees reasonably well with the work carried out by other researchers on effect of heating rate on thermal degradation of EFB in inert atmosphere (Mohammed et al., 2013; Idris et al., 2010). However, it can be observed that, for SC, characteristic temperatures (Ti, Tp and Tf) were not affected as much as that of biomass.

Effect of Blending in Inert Condition

It is important to understand the thermal degradation behavior of the two fuels by performing thermal analysis under inert conditions. Thermal degradation of EFB blended with SC at ratio of 100:0, 80:20, 50:50, 20:80, and 0:100 is shown in **Figure 4**. Three decomposition profiles are observed for SC/EFB blends where the first peak (TEP 1) appears at a temperature below 150°C, second peak (TEP 2) between temperatures of 274 and 400°C, and third peak (TEP 3) is between 434 and 566°C. The first peak can be associated with water removal from both samples, while the second peak indicates the devolatilization of biomass and the last were assigned to decomposition of SC. As can be seen, both samples followed the same thermal evolution profile as their parent fuel. Maximum rate of devolatilization (also known as peak height) gradually increases with an increasing amount of biomass in the blends due to high volatile quantities released (Idris et al., 2010; Vamvuka et al., 2003; Panwar et al., 2020)). This also shows improved reactivity of the sample where biomass increment will increase the maximum rate of mass loss (Jayaraman et al., 2017). Moreover, the position of the maximum peak was shifted to a lower temperature as the biomass percentage increased, while the peak temperature was also found to be at a lower temperature as compared to SC. From **Table 1**, char yield of SC and EFB is 76.47% and 34.01%, respectively. This indicates the remaining solid particles in the main body mass upon devolatilization. Apparently, the char yield of the blends decreases with increasing biomass. This can be seen on the TG curve of the EFB:SC at the mixture of 20:80, 50:50, and 80: 20 where the final residues are 55.35%, 45.85%, and 33.9%, respectively. The main reason for this finding is because of the differences in the coal and biomass chemical structures. Coal is known to have a strong C=C bond energy structure of 1,000 kJ/mol that makes it difficult to rupture under thermal treatment as compared to a weak energy bond of 380–420 kJ/mol for biomass. Thus, it is easier for the biomass to decompose when subjected to heating (Quan & Gao, 2016). Similar findings have also been reported by Vuthaluru (2004) on pyrolysis of wood waste and wheat straw, He et al. (2018) on co-pyrolysis of bituminous coal and fermented cornstalk, and (Panwar et al., 2020) on co-pyrolysis of raw/torrefied biomass and coal blends, and (Quan et al., 2014) on white pine and bituminous coal using thermogravimetric analysis.

The synergistic interaction between the SC and EFB can be observed from the additive or non-additive behavior based on the sum of weighted average of the mass loss profile, as indicated in **Eq. 1** (Vamvuka et al., 2003; Merdun and Laougé, 2021; Xie et al., 2018). An equation was applied on the char yield of the blends as shown in **Figure 5**. The trend line represents data as calculated by using **Eq. 1**, while experimental data were represented by the data points. It appears that the char yield profile does not show an additive behavior as the data point for the experimental result lies away from the summation of weighted average line (trendline). Such a behavior is indicative of synergistic effect between fuels in the blend. Similar observation was reported by Quan et al. (2014) on the co-pyrolysis of white pine with bituminous coal using TGA. It was reported that the interaction exists due to the formation of volatiles and residues of biomass that took place at a much lower temperature than that of coal, and could easily be deposited on the coal surface. At a low heating rate, there is a

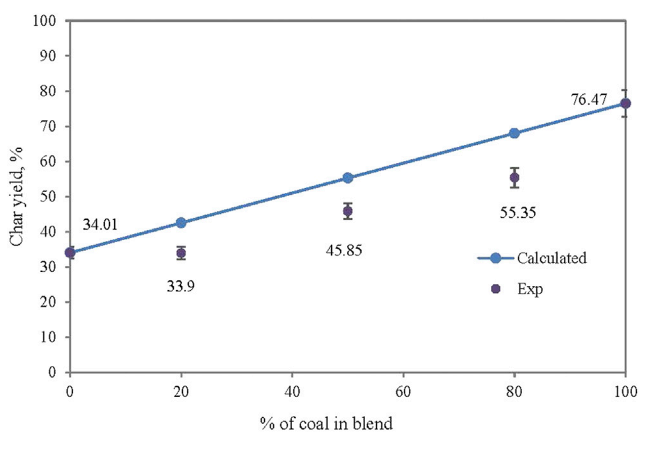

FIGURE 5 | Char yield from pyrolysis of coal/biomass blend at of 10°C/min.

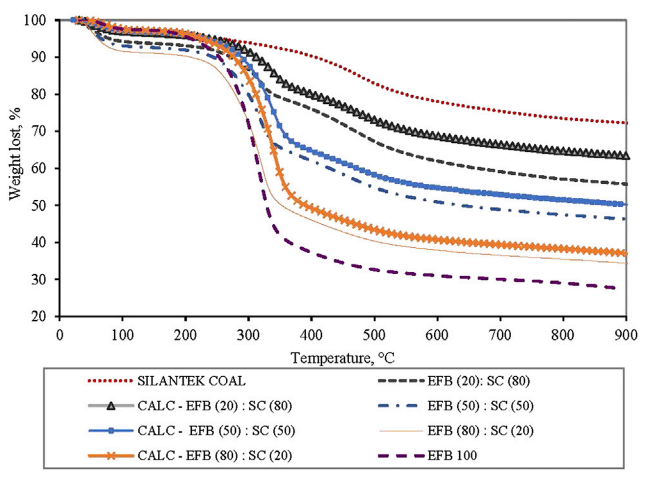

FIGURE 6 | Experimental and calculated data of EFB and SC heating rate of 10°C/min.

longer release time of volatiles from both materials, hence allowing secondary reaction of intraparticle volatiles.

Using **Eq. 2**, an additional plot was made for calculated and experimental thermograms (TG) of the blends as shown in **Figure 6** (Xie et al., 2018). It appears that the calculated data are not able to represent the experimental data by a simple weighted average, indicating the existence of interaction between the fuels (Vuthaluru, 2004; Quan and Gao, 2016). It is also observed at the beginning of the process up to 300°C, the experimental data for the blends appear to be much lower than that of the parent fuels. This is also evidence of the synergistic effect of the two fuels. This could be due to lignocellulosic materials being primarily transformed into laevoglucose with the breakdown of glycosidic bonds, which can provide a hydrogenous donor for bituminous coal pyrolysis (Wu et al., 2014). Interaction between coal and biomass during co-pyrolysis, although uncommon, has been reported by some researchers involving different ranks of coal and various types of biomass (Haykiri-Acma and Yaman, 2008; Aboyade et al., 2013; Xie et al., 2018)). Similar work performed but on different coal by Idris et al. (2010), however, showed the additive behavior when Mukah Balingian coal (lower rank coal) and EFB were co-utilized during pyrolysis.

It has been demonstrated that, synergistic effect exists for the two fuels during co-pyrolysis. Its application for co-combustion is much influenced by the understanding of both behaviors in this condition i.e., in the absence of oxidative gas. Ideally, the best ratio could be selected so as to overcome limitations of both fuels during combustion. It is already highlighted that biomass has high volatile matter and low characteristic temperature, meaning its reactivity is high as compared to coal which has a high initial temperature. A high char yield would translate into a char combustion phase at a much higher temperature and longer duration could be maintained and is desirable (Sotannde et al., 2010; Jamaluddin et al., 2011; Matali et al., 2016; Abdullah et al., 2019) Thus it can be said that the blend of 50% coal and biomass, has an ignition temperature of about 172°C, intermediate reactivity, with a long reaction time. Nevertheless, further combustion analysis of the co-blend materials shall be carried out in our future research.

CONCLUSION

As a conclusion, thermal decomposition of Silantik Coal (SC), Empty Fruit Bunches (EFB), and their blends under inert conditions has been determined using a thermogravimetric analysis technique. Degradation of EFB occurred at a lower temperature compared to SC and reactivity of the EFB is higher than SC. Heating rate has an effect on the thermal behavior of the fuel causing temperature shifts to higher temperatures as the heating rate increases. Blended fuels showed a degree of interactions during co-pyrolysis, observed from the sum of the weighted average of the thermograms. The interactions between the biomass and coal are of paramount importance with the hope to further reduce NO_x or SO_x during combustion, hence sustaining the environment. Outcomes from this work provide an insight and prompts further research into reducing utilization of coal by increasing the share of biomass materials as fuel toward obtaining energy for the benefit of future generations.

DATA AVAILABITY STATEMENT

The original contributions presented in the study are included in the article, further inquiries can be directed to the corresponding author.

AUTHOR CONTRIBUTIONS

NH: Writing-Original Draft, methodology, data curation SI: Corresponding author, supervision, conceptualization, grant acquisition and administration, review and editing NA: Review and editing SM: Data curation NA: Supervision, review and editing.

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Conflict of Interest: The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.