

Thermal Behaviour, Reactivity and Synergistic Effect Analyses of Petroleum Coke and Sugar Cane Bagasse Chars During Co-gasification

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Abstract

Petroleum coke (PC) is found to have lower reactivity, high sulphur and vanadium oxide contents; hence, mixing with other biomass is required. In this study, PC, sugar cane bagasse (SCB) and their blends at temperatures of 500, 800 and 900 °C (denoted as PC: SCB5, PC:SCB8 and PC: SCB9) were investigated using a thermogravimetric analyser (TGA) under non-isothermal conditions at a heating rate of 20 °C min⁻¹. The derivatives thermogravimetric (DTG) curves were obtained experimentally. The results showed that pyrolysis temperature and blending ratio significantly affect the behaviour and the reactivity of PC. The DTG for PC was 15.44%, but for SCB chars, PC: SCB5, PC: SCB8 and PC: SCB9 were in the range of 6.28– 6.66, 4.63– 8.48, 4.97– 8.58 and 4.97–12.10% respectively. The thermal behavior of PC, SCB char and the blends was found to be completed in a single stage. PC reactivity improved by 26.72% with the addition of SCB chars. Synergistic effect was found to be varied, but among the samples, PC: SCB5 at a blend ratio of 1:1 showed the most synergistic effect.

Key words-*Petroleum coke, sugar cane bagasse char, co-gasification, reactivity, synergistic effect.*

ABBREVIATIONS

PC: Petroleum coke.
SCB: Sugar cane bagasse.
TGA: Thermogravimetric analyzer
DTG: derivatives thermogravimetric
SCB5, SCB8, SCB9: Sugar cane bagasse at 500, 800 and 900 °C.
PC: SCB5, PC: SCB8, PC: SCB9: PC and SCB blend at 1:1, 1:3 and 3:1 blend ratio and temperature of 500, 800 and 900 °C.
V₂O₅ : Vanadium oxide.
RMS: Root mean square.

I. INTRODUCTION

Sugar cane bagasses have been used as a biomass source for fuel utilization[1]. In Sudan their about 2.7610⁶ tons of sugar cane bagasses produced annually[2]. This huge amount of SCB could be used for energy generation. Besides, it has the advantages of good ignition characteristics, cheap and low pollution[3]. Petroleum coke is by product of delayed coking of residual oil. Petroleum coke has low ash content and high calorific value found attractive fuel source[4]. However, low reactivity, high sulphur and vanadium oxide (V₂O₅) contents are some drawback hindering its exploration as a fuel. This drawback causes boiler pipes clogging and environmental problems[5]. To overcome PC problems, techniques such as blending, high temperature and longer reaction time or catalyst additives were used[5][6]. Gasification is an attractive technique for solving PC problems. This is performed by converting the combustible PC into syngas and other chemical production[7]. However, due

to lower reactivity, the use of PC gasification was limited. Therefore, longer residence time and higher gasification temperature were used. Co-gasification techniques achieved high thermal conversion, stable operation, and eliminated tar reduction issues[8][9][10][11][12][13][14][15]. However, co-process technologies needs better understanding of biomass kinetics reaction, physical and thermal properties. The chars are produced by decomposing the biomass in an oxygen-free environment[16]. Char gasification goes through various carbon reactions with the gasification agent and produces gases. The reactivity is an important parameter because it determines gasification design, fuel suitability, and conversion rate[15][17]. Edreis et al[12] conducted co-gasification of PC and various types of biomass wastes (rice husk and stalk, sawdust, cotton straw, and sugar cane bagasse) using a thermogravimetric analyzer. The results showed that PC gasification was improved with the addition of biomass wastes. The reactivity of the samples was in the range of 1.97–2.47%/min °C. Whereas for the blends was 1.22–1.82%/min°C. Among the blends, PC: sawdust was the most reactive. Synergistic effect was observed in the order of PC: cotton straw, PC: sawdust, PC: rice husk, and PC: sugar cane bagasse, respectively. In another study, Edreis et al.[18] studied co-gasification of PC, SCB, and their blends using thermogravimetric and Fourier transform infrared spectroscopy (FTIR). Sugar cane bagasse gasification occurred in two steps: volatiles releasing (150–430°C) and char gasification (700– 1220°C). But for PC, the gasification occurred in one step between 800– 1220°C. A synergistic effect was observed in all samples, and it increased as the ratio of PC decreased. Wei et al.[19] studied rice straw, bituminous coal, and their blends using a thermogravimetric

analyzer. Gasification temperatures of 800, 850, 900, 950 and 1000 °C were used. The results showed that as conversion increased, the reactivity decreased. Liang et al.[20] investigated bamboo reed char, semi-coke and, their blends using a non-isothermal thermogravimetric analyser. The reactivity of semi-coke was found to be lower than that of bamboo reed char. This was due to the carbonaceous structure and lower alkali content. The addition of bamboo reed char improved the reactivity. The synergistic effect was found between the blends. Wang et al.[21] studied co gasification of semicoke and bituminous coal at higher temperatures in a CO_2 atmosphere. High temperature drop tube furnace was used. The interactions between the fuels found to release NO_x and it's increased as the content of semicoke increases. Motta et al.[22] investigated CO_2 gasification of sugar cane bagasse char. Gasification temperatures range of 750 to 900°C at isothermal conditions were tested using a thermogravimetric analyser. Random pore and extended random pore models were examined. Increasing gasification temperature did not affect the surface composition and the char morphology but reduced the initial gasification rate, surface area and the fitted rate constants. The extended random pore model effectively modelled the gasification behaviour. Lahuri et al.[23] investigated CO_2 gasification of high-sulphur petroleum coke. A thermogravimetric analyser under isothermal conditions was used. K_2CO_3 was used as the catalyst at a loading rate of 0- 20 wt.%. The shrinking core model and volume reaction model were used to investigate the reaction rate and activation energy. Activation energy was reduced significantly from 150.66 kJ/mol to 96.96kJ/mol at a 5wt.% catalyst loading. Sudanese SCB has low emission, high activity, high alkali and alkaline earth metal (AAEM) content. This alkali and alkaline perform as a natural catalyst and boost PC gasification[5][6]. It is found that SCB release large amount of volatiles at the beginning of gasification; therefore, blending it with PC could provide more energy[18][24]. Hence, understanding the synergistic effect of PC and SCB is crucial for thermal studies. Nemanova et al.[25] studied the co-gasification of PC and biomass. Blending ratios of 0/100, 20/80, and 50/50 (PC: Biomass) were investigated. Atmospheric bubbling fluidized bed reactor, and thermogravimetric analyzer at operation temperature of 800 and 900°C were used. The conversion and the total conversion were found to increase in the range of 47.6-75.3% and 85.2-90.8%, respectively. The tar decreased from 1.2 to 0.8g/ Nm^3 . Increasing biomass content in the mixture decreased gasification time. This is due to alkali and alkaline earth metals from biomass ash, hence increasing the reactivity of PC gasification. Synergetic effect was observed; this was evidenced by the reduction of the activation energy, which was 121.5, 96.3 and -83.5 kJ/mol for PC, 50:50 and 20/80 blends respectively. Zhang et al.[26] investigated CO_2 co-gasification behaviour and synergistic effect of PC, biomass and their blends. They report synergistic at the char gasification stage due to the presence of alkali and alkaline compounds. Wei et al.[5] investigated petroleum coke-biomass and char using co-gasification. A synergistic effect was observed at the complete generation of biomass ash. However, the effect was weakened as the temperature increased from 1000 to 1100 °C. Similar synergistic behaviour

was reported by Liu et al.[27] using CO_2 co-gasification of PC and straw char. Wang et al.[28] investigated PC, corncob and their blends (80/20, 60/40, 40/60 and 20/80) by mass. A thermogravimetric analyser at non-isothermal conditions was used. Corncob was found to be more reactive than PC; this is due to pore structure, low degree of order and high content of alkali metals. The reactivity of PC was found to improve with the addition of corncob up to 20% by mass. Synergetic effect was observed with the addition of corncob. The activation energy of PC and corncob was 203.2 and 243.3 kJ/mol, but for the blends was in the range of 197.8-238.3kJ/mol. From the literature investigation, gasification behaviour, reactivity and synergistic effects of Sudanese SCB chars and PC during co-gasification have not been studied. Therefore, in this study, PC, SCB char and their blends were evaluated using a thermogravimetric analyzer. Further, the fuel synergistic effect during co-gasification was investigated. Comparison between the theoretical and experimental DTG was also included.

II. MATERIALS AND METHODS

A. Materials and samples preparation

In this study petroleum coke and sugar cane bagasse char were used. The samples were dried (remove the moisture to less than 10%), crushed and sieved. The sizes of the particles were in range of 180 – 450 μm .

B. Chars preparation

SCB chars were devolatilization in a horizontal tube furnace using pure nitrogen. Nitrogen flow rate was 1 L/min for 20 min at a temperature of 500, 800 and 900°C, respectively. When the furnace reach the desired temperature, ceramic container content 1g of SCB sample was placed outside the tube and nitrogen was purged. The oxygen was eliminated and the container was pushed into tube heating zone. After the specified time, the char was removed and cooled to a room temperature using nitrogen. The SCB char produced at a temperature of 500, 800 and 900°C were denoted by SCB5, SCB8 and SCB9, respectively. The char's samples were stored in desiccator to prevent moisture. The mixture of PC and SCB char were prepared at ratio of 1:1, 1:3 and 3:1 by mass.

C. Proximate and ultimate analysis

Proximate analysis of the samples was performed using TGA-2000 (Navas Instruments, Spain) following D5142 standard method. For ultimate analysis Euro-CA 3000 elemental analyser was used. Table(I), shows the proximate and ultimate analyses of PC and SCB chars samples.

^a As received basis

^b Dry ash basis

^c Calculated by differences

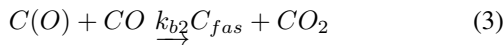
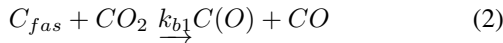
D. Thermogravimetric analysis

Pure and co-gasification of PC and SCB chars were performed using TGA NETZSCHSTA 449/F3 model thermogravimetric analyser under non-isothermal condition. Gasification experiments were conducted at a temperature range of

TABLE I: Proximate and ultimate analyses of PC and SCB char samples.

Sample	Proximate analysis (w%) ^a				Ultimate analysis (w%) ^b				
	M	Ash	VM	FC	C	H	O ^c	N	S
PC	2.15	2.01	9.32	86.52	91.04	3.76	3.03	1.06	1.08
SCB	7.317	4.411	76.93	11.33	46.95	6.06	46.78	0.13	0.08
SCB 5	2.30	10.15	23.04	54.28	47.39	4.51	47.54	0.46	0.1
SCB 8	1.14	13.50	9.72	75.48	51.52	1.96	46.06	0.35	0.11
SCB 9	0.70	15.61	6.90	74.62	73.45	1.74	23.57	1.01	0.23

25 to 1300 °C under heating rate of 20 °C/min. Pure CO₂ was used as a gas agent at a flow rate of 100 mL/min. A 8- 9 mg of sample was used for each experiment and it was dispersed flatly on a crucible. A small amount of sample and a slow heating rate were used to avoid heat transfer limitation and to minimize mass transfer effects. All the experiments were repeated three times to test the reproducibility of the results and the average values was used for the analysis. The mass loss (TG) and derivative curve (DTG) of the samples were presented as a function of temperature. For the active site, Boudouard reactions mechanism Equation(1, 2, 3 and 4) were considered for char gasification in carbon dioxide domain[29][30].



Where k_i is the rate of the i th reaction.

E. Reactivity measurement

The TG data were expressed as a function of conversion (X), which is defined as:

$$X = \frac{w_i - w_t}{w_i - w_f} \quad (5)$$

where w_i , w_t and w_f are initial, at given time t (min) and final samples masses (mg). The reactivity index $R_{0.5}$ defined in Equation(6) was applied to quantify gasification reactivity of the samples[31][32][33], and further was used to investigate the synergy effect during the co- gasification of the fuel blends.

$$R_{0.5} = \frac{0.5}{t_{0.5}} \quad (6)$$

where $t_{0.5}$ is the time required for 50% carbon conversion. $R_{0.5}$ is the higher reactivity.

F. Synergistic effects analysis

To find out the occurrences of synergistic effect between PC and SCB chars; theoretical DTG curve was calculated and compared with the experimental DTG using Equation(7)[6].

$$\frac{dw}{dt} = x_{PC} \left(\frac{dw}{dt} \right)_{PC} + x_B \left(\frac{dw}{dt} \right)_{SCB} \quad (7)$$

where $\left(\frac{dw}{dt} \right)$ is the corresponding weight loss rate (%/min) of the individual material. The x_{PC} and x_{SCB} are the mass fractions of PC and SCB, respectively. For further characterization of synergistic effect during co-gasification, Root Mean Square (RMS) was used as presented in Equation(8) and the higher RMS the high the synergistic effect[34].

$$RMS = \sqrt{\frac{\sum_{i=1}^N \left(\frac{(dw/dt)_{exp}^i - (dw/dt)_{cal}^i}{(dw/dt)_{cal}^i} \right)^2}{N}} \quad (8)$$

Where $(dw/dt)_{exp}$ and $(dw/dt)_{cal}$ are experimental and calculation weight loss rate, respectively and N is the number of data points.

III. RESULTS AND DISCUSSIONS

A. Gasification behaviour and reactivity analysis

Figure(1) shows the variation of DTG against the temperature for PC, SCB chars, and the blends. The gasification found occurred in one stage at a temperature higher than 600 °C. Edreis et al.[35] also claimed the same trend with SCB and biomass blends. From Table(II), it is found that, the DTG max for SCB chars increases when the pyrolysis temperature increases. At a blending ratio of 1:3, the DTG max for SCB chars was higher than that of the blends. While the DTG max for SCB8 and SCB9 char at 1:3 blend ratio were similar. Also it's found that, DTG max for PC, SCB chars blends increases when the PC content in the blend increases. The T max values were 848, 903 and 906 °C for SCB5, SCB8 and SCB9 char, respectively, but for PC was 1116 °C. For PC: SCB5, PC: SCB8 and PC: SCB9, the T max values were in the range of 855-1031, 909-1060 and 910-1098 °C, respectively. This indicates that as PC content in the blends increases, the Tmax increases. It was observed that DTG max and T max were proportional to pyrolysis temperature and blending ratio. The gasification of SCB5 occurred at a temperature range of 600 - 975 °C with a DTG max of 6.28 %/min compared to SCB 8, SCB 9 and PC, which occurred at a temperature range of 700- 1010 °C, 700 - 1020 °C and 840- 1200 °C with DTG max of 6.37, 6.66 and 15.44 %/min, respectively. This indicates that, chars prepared at lower temperatures were

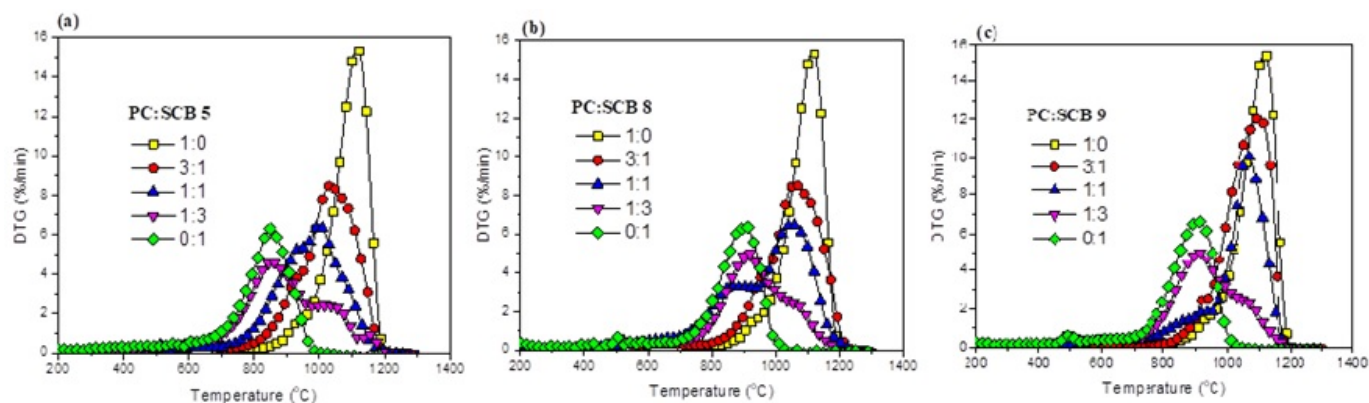


Fig. 1: DTG for the blends against temperature.

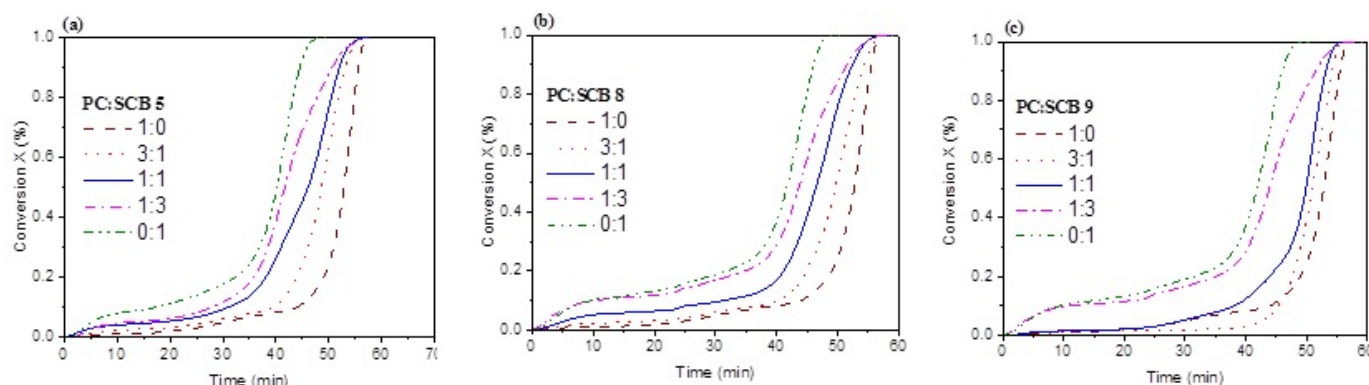


Fig. 2: Samples conversion rate against time.

more reactive compared to those of higher temperatures. This is due to the higher volatile matter and lower fixed carbon content in the SCB5 sample compared to SCB 8, SCB 9 and PC. For the blends, PC: SCB5 showed lower values of DTGmax and T max compared to PC: SCB 8 and PC: SCB 9. Figure(2) shows blends conversion rate against the time. The conversion rate was found to be significantly influenced by pyrolysis temperature and blending ratio. The variations of the reactivity for PC, SCB and the blends are presented in Figure(3). PC reactivity was found to be lower than SCB and its increase as the content of SCB char in the blend increased. Similar results were obtained by Wang et al.[36], who used PC and millet husk char using a thermogravimetric analyser at isothermal conditions. Compared to SCB chars, SCB 9 showed lower reactivity individually and as a blend with PC. This is due to the crushing of the surface structure during the pyrolysis at high temperature, hence affected the char reactivity. It's concluded that pyrolysis temperature and blending ratio have a significant effect on gasification behaviour and reactivity. The reactivity of PC was significantly improved with the addition of SCB char during the co-gasification. The same results were reported in the literature by different researchers[21][23].

DTGmax: maximum mass loss rate (% min⁻¹), Tmax: The temperature (°C) when the gasification rate reaches the maximum

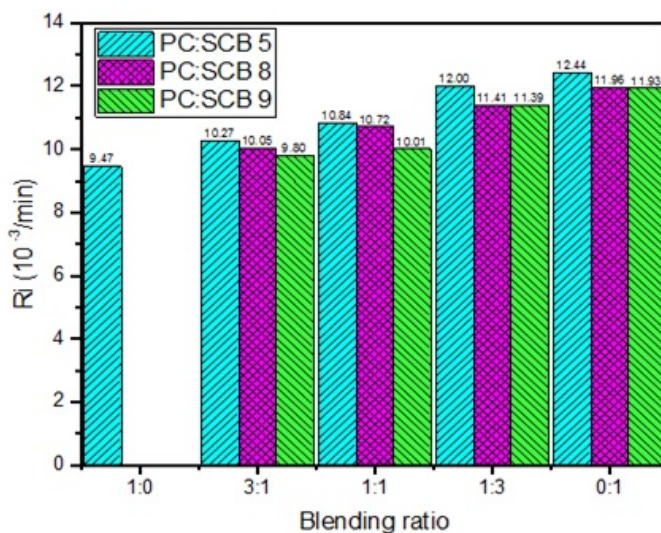


Fig. 3: Reactivity index as a function of blend ratio

B. Synergistic effects analysis

The comparison between the experimental and the calculated DTG curve of the co-gasification is depicted in Figure(4). From the figure, the synergistic effect was found dominate for PC: SCB5 at a blending ratio of 1:1. This is due to the interaction between PC and SCB5. For the blends of PC: SCB5 and PC: SCB8, the calculated DTG curves were over the

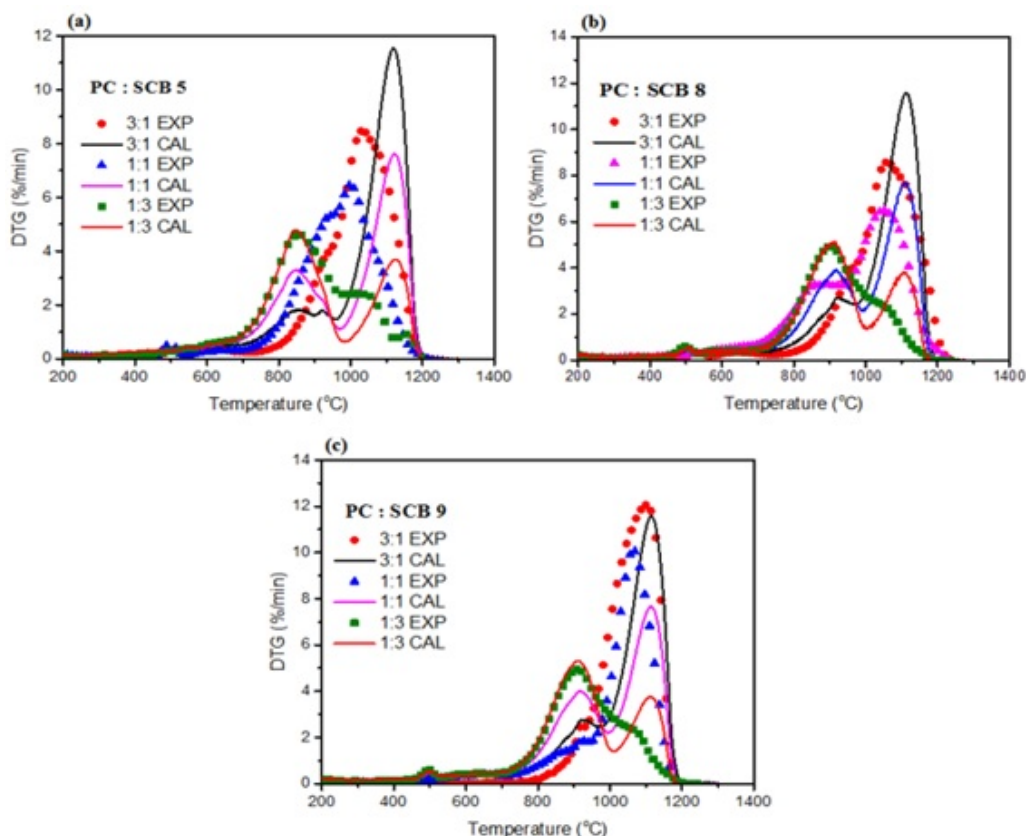


Fig. 4: DTG for the blends as function of temperature.

TABLE II: Thermogravimetric characteristics analysis

Characteristics	Blending ratio (PC:SCB Char)				
	1:0	3:1	1:1	1:3	0:1
For PC: SCB 5					
Tmax	1116	1031	1000	855	848
DTGmax	15.44	8.48	6.46	4.63	6.28
For PC: SCB 8					
Tmax	1116	1060	1049	909	903
DTGmax	15.44	8.58	6.48	4.97	6.37
For PC: SCB 9					
Tmax	1116	1098	1064	910	906
DTGmax	15.44	12.10	10.11	4.97	6.66

experimental curves; hence, negative synergistic effect was observed. But for the blend of PC with SCB9 positive synergistic effect was found at a blending ratio of 3:1 and 1:1. Figure(5) shows the results of RMS versus the blending ratio. It was observed that, for PC: SCB char blends, the RMS value was maximum at blending ratio of 1:1 and minimum at blending ratio of 1:3. Compared to PC: SCB5, PC: SCB8 and PC: SCB9 showed significant synergistic effect at blending ratio of 1:1. This is due to the decomposition of biomass during the gasification and forms CO₂, H₂O, and CH₄, hence enhanced PC gasification[33][37][38]. This also could be because of the catalytic effect in SCB chars[5][6][38]. Similar results were found in the literature by different researchers[7][39]. We concluded that the gasification process was non-spontaneous and the reaction was endothermic. PC: SCB5 showed the most synergistic effect with a blending ratio of 1:1.

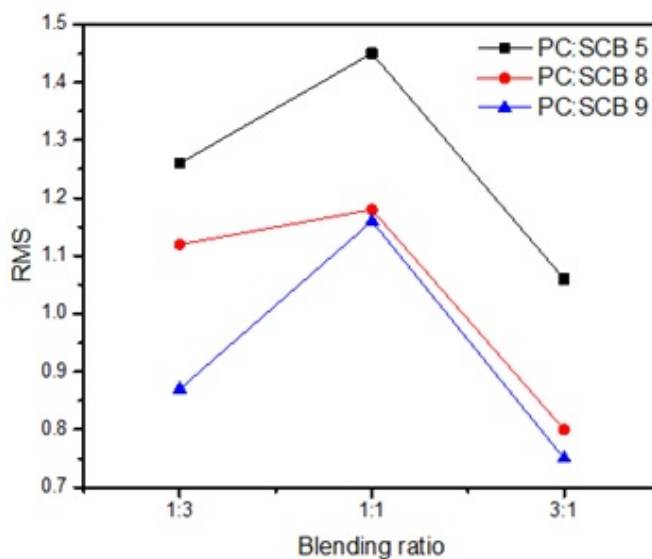


Fig. 5: Root mean square as a function of blending ratio.

IV. CONCLUSIONS

The effect of pyrolysis temperature and blending ratio on the reactivity, behavior, and synergistic during co-gasification of sugar cane bagasse chars and petroleum coke were studied using TGA. The results indicate that the reactivity and behavior were significantly affected by the pyrolysis temperature

and blending ratio. PC reactivity was significantly improved upon blending with SCB chars. SCB chars prepared at a lower temperature or blended with PC showed higher reactivity. Significant synergistic effect between PC and SCB chars was observed in the blend of PC: SCB5 at a blending ratio of 1:1.

ACKNOWLEDGEMENT

The authors would like to acknowledge Prof. Hong Yao for the financial support and the staff at key laboratory of coal combustion Huazhong University of Science and Technology for their assistance on conducting the experiments.

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