

Co-Pyrolysis Characteristics of Torrefied Pine Sawdust with Different Rank Coals

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Torrefaction of sawdust has the advantages of increasing its heating value, lowering its O/C ratio, and making it more convenient to transport and crush. Torrefied sawdust has characteristics that are more similar to coal than to sawdust. The initial pyrolysis temperature and residue content of torrefied sawdust are higher than those of sawdust. With the increase of coalification degree of three different coals, the initial pyrolysis temperature and residue contents are raised. The initial pyrolysis temperatures and remaining contents of lignite, bituminous coal, and anthracite are 330, 380, and 500 °C and 61.3, 75.1, and 89.5%, respectively. The torrefied sawdust also has a synergistic effect on the conversion of anthracite and bituminous coal, but it has an inhibitory effect on lignite. The composition of gaseous products was also measured after pyrolysis. The results demonstrate that with the addition of torrefied sawdust to anthracite and bituminous coal, the gaseous products contain more combustible components, such as H₂, CO, and CH₄, which increase the heating value. Moreover, the effect is more obvious with the co-pyrolysis of torrefied sawdust and anthracite. However, the co-pyrolysis of torrefied sawdust and lignite leads to decreasing CO and light hydrocarbons (C_nH_m (n=1, 2)) in the resulting gaseous products, which has a negative effect on the quality of the gaseous products. The co-pyrolysis characteristics of torrefied sawdust with different rank coals are discussed in this paper.

Keywords: Torrefaction; Sawdust; Coal; Lignite; Bituminous coal; Anthracite; Co-pyrolysis

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INTRODUCTION

Increasing consumption of fossil fuels and their impacts on the environment are forcing scholars to investigate innovative ways to utilize these energy resources. Compared with petroleum and natural gas, coal has the advantage of being an abundant resource (Zhang *et al.* 2007). However, the combustion of coal also has the disadvantage of emitting pollutants such as NO_x, SO_x, and particles, which have negative repercussions on the environment. Biomass is the third most abundant energy resource in the world and is one of the cleanest and most sustainable energy resources (Kirubakaran *et al.* 2009), despite the problems of low heating value, high moisture content, and high transport cost, *etc.* Currently, investigations are focused on the effect of the co-pyrolysis of coal and biomass. Researchers have revealed that co-firing of coal and biomass fuels (*e.g.*, wood waste, wheat straw, peat, and municipal solid waste) can reduce CO₂ emissions (Werther *et al.* 2000; Williams *et al.* 2001; Sami *et al.* 2001). Co-combustion

of coal with biomass is also considered to be an effective method to decrease the amount of NO_x, SO_x, Polycyclic Aromatic Hydrocarbons (PAHs), Volatile Organic Compounds (VOCs), and Total Organic Compounds (TOCs) (Chao *et al.* 2008). A handful of studies (Nikkhah *et al.* 1993; Rüdiger *et al.* 1995; Pan *et al.* 1996; Collot *et al.* 1999) have investigated the mechanism of the gas-phase species product that the biomass with high hydrogen content was pyrolyzed earlier than coal, acting as H₂-donors to coal pyrolysis (Straka *et al.* 2004; Ishaq *et al.* 2006; Sharypov *et al.* 2007). Biomass was shown to have synergistic effects on coal in producing more gas products.

In those studies, the investigators mainly focused on the co-pyrolysis of coal and biomass without pretreatment. Studies on the co-pyrolysis of coal and torrefied biomass are relatively rare. Torrefaction technology could improve the combustion efficiency of biomass and reduce its transportation and storage costs (Deng *et al.* 2009). Torrefied biomass also has some favorable properties, such as high heating value, low moisture content, high grindability, and high similarity to coal (Bridgeman *et al.* 2010; Gilbert *et al.* 2009; Repellin *et al.* 2010; Phanphanich and Mani 2011; Prins *et al.* 2006; Chen *et al.* 2011). Kiel *et al.* (2004) reported that torrefied biomass could reduce tar byproducts by lowering the moisture and lowering the concentration of hemicellulose and lignin in the feedstock. Torrefied biomass has a lower O/C ratio (Pentananunt *et al.* 1990), which improves its gasification or pyrolysis efficiency compared to untreated biomass. A high O/C ratio can lead to over-oxidizing of biomass in the reactor when undergoing pyrolysis at high temperatures (Prins *et al.* 2006). Due to these advantages of torrefied biomass, this study focused on the comparison of torrefied sawdust and sawdust. The study also investigated the effects of torrefied biomass to different rank coals by analyzing the conversion of samples, the components of the gaseous products, the composition of tar, and the yield of products. The goal is to check whether the use of torrefied sawdust is better than that of sawdust and whether torrefied sawdust also has beneficial effects when used with coals. For this purpose, pine sawdust was selected as the biomass sample, while different rank coals, such as anthracite, bituminous coal, and lignite, were also used in this work.

MATERIALS AND METHODS

Materials

The pine sawdust biomass used in this study was from Nanjing (Jiangsu Province), while the anthracite, bituminous coal, and lignite were from Shanxi Province. All the raw materials were milled and sieved to less than 125 μm in diameter. The sawdust was torrefied at 250 °C for 30 min. Blends of torrefied sawdust with the different coals were prepared with sawdust-to-coal ratios of 0:10, 2:8, 5:5, 8:2, and 10:0, which was used to investigate the co-pyrolysis characteristics. Simple physical method was used to make torrefied sawdust well-distributed in coals. The main characteristics of the sawdust, torrefied sawdust, and three different rank coals are presented in Table 1.

Torrefaction

Sawdust torrefaction was done in a quartz tube reactor surrounded by a heater band. Thermocouples were inserted into the sample and the heater band, respectively, for torrefaction temperature control and data acquisition. A nitrogen carrier gas at 50 mL/min

was passed through the sample to eliminate the presence of oxygen (to avoid oxidation and ignition). A diagram of the quartz tube torrefaction reactor is shown in Fig. 1.

Table 1. Proximate, Ultimate, and Higher Heating Value (HHV) Analyses of Sawdust, Torrefied Sawdust, and Three Different Rank Coals

	Proximate analysis (%)				Ultimate analysis (%)					HHV MJ/kg
	M _{ad}	A _{ad}	V _{ad}	F _c	C	H	N	S	O	
Sawdust	9.05	2.90	72.25	15.80	45.95	7.37	0.36	1.54	42.04	17.71
Torrefied sawdust	2.67	9.03	70.04	18.26	49.66	6.93	0.42	1.24	39.36	18.23
Anthracite	1.30	18.84	10.15	69.71	80.15	3.97	1.35	0.84	—	28.26
Bituminous coal	2.28	27.95	27.18	42.59	60.5	4.565	1.22	0.65	—	23.32
Lignite	12.6	17.30	32.94	37.16	46.60	4.30	0.37	0.54	—	21.54

*Note: M_{ad}, A_{ad}, V_{ad}, and F_c are presented for moisture, ash, volatile, and fixed carbon, respectively. All the materials were air dry basis except torrefied sawdust, which was torrefied at the condition of 250 °C for 30 min.

The sample was charged into the reactor at room temperature. The N₂ flow was started and maintained. The reactor was heated to 110 °C and maintained for 10 min for drying the sawdust samples. Then, the reactor was heated to the target temperature at the heating rate of 10 °C/min. The temperature was maintained for 30 min at 250 °C. Finally, the sample was cooled to room temperature under the protection of the nitrogen.

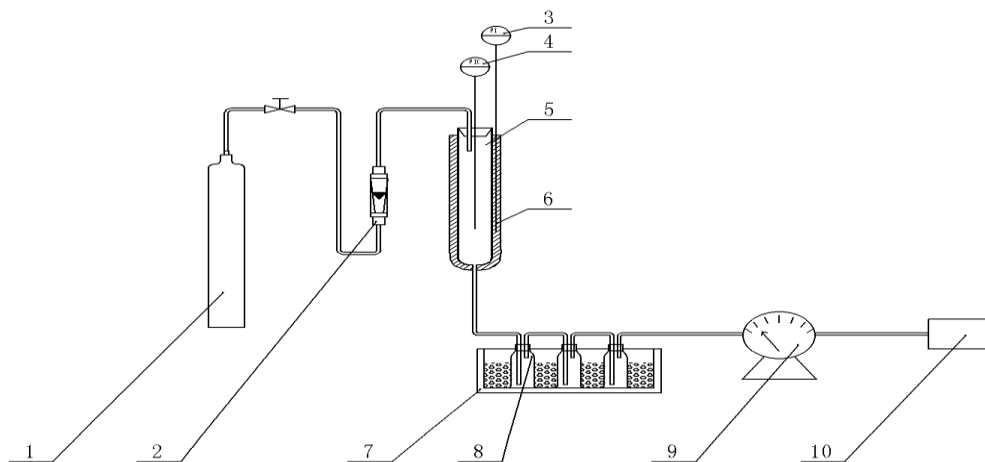


Fig. 1. Bench-scale set-up of sawdust torrefaction reactor: 1. Nitrogen; 2. Gas flow meter; 3. Temperature controller; 4. Temperature indicator; 5. Quartz reactor; 6. Heater band; 7. Ice container; 8. Condenser; 9. Wet-type gas flow meter; and 10. Gas holder

Thermal Gravimetric Analysis (TGA)

The pyrolysis experiments were performed with a thermogravimetric (TG) analyzer (NETZSCH TG 209 F1 Iris) utilizing a nitrogen flow rate of 20 mL/min and a heating rate of 10 °C/min to the final temperature of 900 °C. The experiment was carried out twice under each condition.

Thermogravimetry-Fourier Transform Infrared Spectrometry (TG-FTIR)

The samples were pyrolyzed in a TG analyzer (NETZSCH STA-409-PC) with the protection of N₂ and heating rate of 10 °C/min to final temperature of 1000 °C. Then the gas flowed into FTIR (Thermo Scientific Nicolet iZ10) to be analyzed. The scan time was 10, resolution ratio was 4, and the wavenumber region was 800 to 4000 cm⁻¹.

Co-Pyrolysis Experiments in a Tubular Reactor

A steel tube was chosen as the reactor. A 5-g sample was pyrolyzed in the tubular reactor, held in a horizontal position, with a nitrogen flow of 20 mL/min. Before conducting the experiments, the reactor was heated to the target temperature (900 °C); nitrogen was introduced into the reactor for 10 min to purge any traces of oxygen gas. For each experimental run, the sample was prepared in a quartz container and quickly pushed to the reaction zone, where it was maintained for 30 min. The volatile products passed through a two-stage condenser, which contained small iron hoops that were cooled by ice for removing tar. The gaseous products were metered by a wet-type gas flow meter and were collected in an aluminum gas bag. Figure 2 shows the set-up for the tubular pyrolysis reactor. The solid product was extracted from the test chamber after each experiment and cooled to room temperature for weighing. The gas composition was analyzed by a Shimadzu GC-2014 gas chromatograph following the procedure described elsewhere (Nilsson *et al.* 1999). The experiments were all repeated for twice.

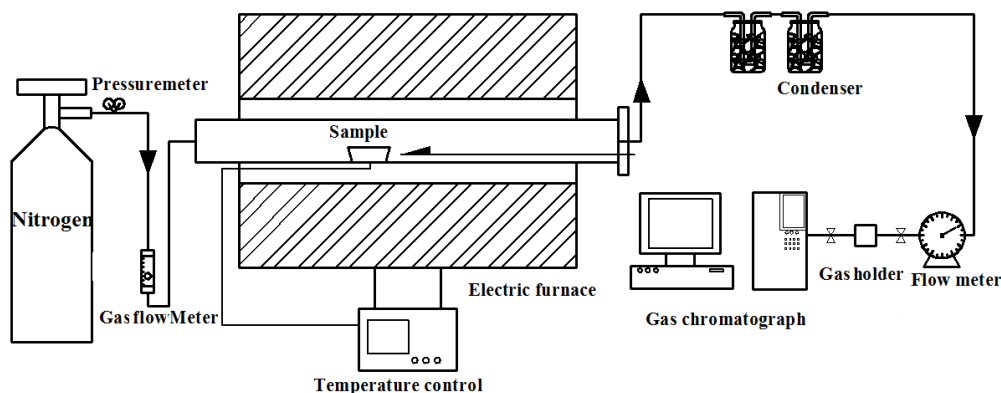


Fig. 2. Experimental set-up for tubular pyrolysis reactor

RESULTS AND DISCUSSION

TGA Characterization of Coal and Biomass

Figure 3 shows the thermogravimetric (TG) and differential thermogravimetric (DTG) curves of the samples. The main weight loss shown in the TGA and DTG curves at 110 °C was due to dehydration. In this stage, sawdust had higher weight loss than torrefied sawdust, which indicated that torrefaction removed water from the sawdust. Another weight loss stage of sawdust started at approximately 200 °C; the main loss at this stage could be due to the loss of the reactive hemicelluloses, which begin to decompose at this temperature (Prins *et al.* 2006). However, the main weight loss stage for the torrefied wood started at approximately 250 °C. This shift was caused by the modification of hemicelluloses due to the torrefaction process. When the temperature was

increased above 270 °C, the cellulose started to decompose and volatilize (Gaur and Reed 1998). The highest weight loss rates for both sawdust samples occurred at 360 °C. When the temperature was higher than 800 °C, there was negligible weight loss for both samples. From the TGA curves, the residue contents of the sawdust and the torrefied sawdust were 25.6% and 30.1%, respectively. Also, there was more carbon residue from the torrefied sawdust than from the sawdust, which was in agreement with results of other biomass torrefaction studies (*e.g.*, Almeida *et al.* 2010). The low water and low active volatile content contained in the torrefied sawdust could decrease the transportation cost and crushing energy of raw material. The higher fixed carbon content in the torrefied sawdust increased the heating value of the raw material. These characteristics of torrefied sawdust made it more competitive than sawdust. The anthracite was much harder than bituminous coal and lignite to be pyrolyzed; it had the highest initial pyrolysis temperature of 500 °C for the start of weight loss and had the least weight loss (10.5%). Bituminous coal and lignite is of lower coalification degree. Coalification is the process from burial of plants to coal formation. The early stage is the low-temperature biogeochemical stage, and the later stage is high-temperature physical and chemical processes, that consist of initial pyrolysis temperatures of 380 °C and 330 °C, respectively, and residue contents of 75.1% and 61.3%, respectively (Stach *et al.* 1982).

The DTG curves demonstrated that the characteristics of samples of biomass and coals were different. After about 150 °C, the three different rank coals only had one peak, and that peak shifted to low temperatures as their coalification degree decreased. However, there were three pyrolysis peaks for torrefied sawdust and sawdust. The first shoulder peak occurred at about 310 °C, which was indicative of the pyrolysis of hemicelluloses. The second peak, which was the most obvious, occurred at 355 °C and was attributable to the pyrolysis of cellulose. The last peak was a trailing peak at about 420 °C that was attributed to the pyrolysis of lignin. The DTG curves also showed that the pyrolysis reactivities of cellulose and lignin from torrefied sawdust were higher than those of sawdust.

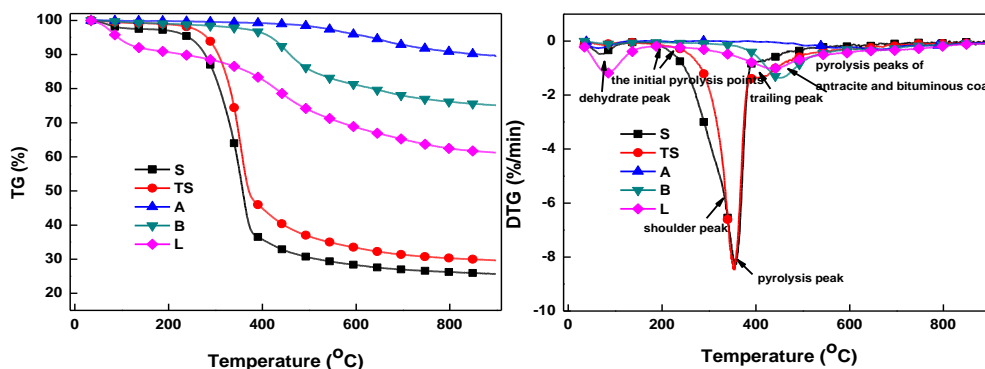


Fig. 3. TGA and DTG curves of the samples: S: sawdust, TS: torrefied sawdust, A: anthracite, B: bituminous coal, and L: lignite. The same abbreviations are used in the subsequent figures.

TG and DTG Analysis of Co-Pyrolysis of Torrefied Sawdust and Different Rank Coals

Figure 4 shows the TGA curves of the torrefied sawdust and coal blends. With the proportion of torrefied sawdust increasing in the blends, the weight loss increased and the curves shifted to lower temperature region. This phenomenon was much more obvious for anthracite blends and the least obvious for lignite blends. The reason for this

observation was that the pyrolysis temperature of torrefied sawdust was much lower than that of anthracite, and there is a huge disparity of conversion between anthracite and torrefied sawdust when mono-pyrolyzed, so the addition of little torrefied sawdust in the coal could raise the co-pyrolysis conversion quickly. However, bituminous coal and lignite are of lower coalification degree, which made their characteristics more similar to torrefied wood; thus, the addition of torrefied sawdust did not remarkably increase the conversion.

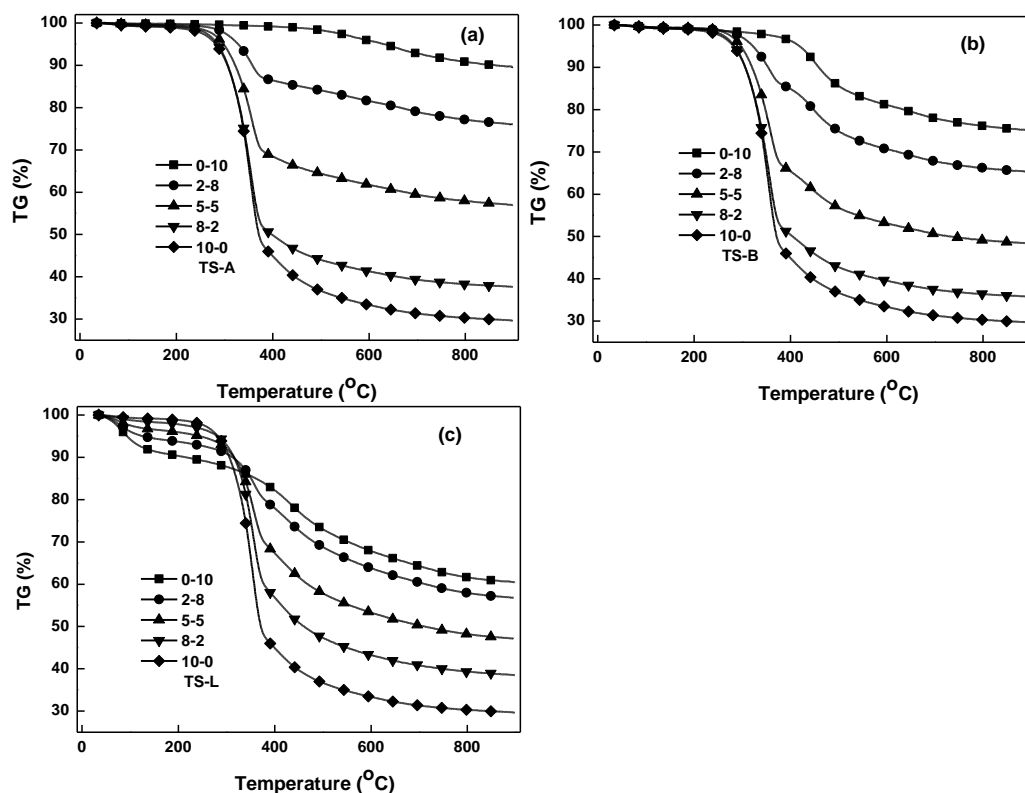


Fig. 4. TGA curves of the blended samples

The effect of torrefied sawdust on the coals can be analyzed using Fig. 5. As shown, the calculated (cal) curves were determined using data from the blends at different ratios. The equation used was as follows,

$$X_{\text{cal}} = X_{\text{TS}} \times r + X_{\text{coal}} \times (1-r) \quad (1)$$

where X is the remaining weight at different temperatures and r (20%) is the proportion of torrefied sawdust in blended samples (torrefied sawdust was the material after torrefaction and the three kinds of coals were just received basis after air dry). Figure 5 demonstrates that the calculated curves were higher than the observed experimental curves for the co-pyrolysis of torrefied sawdust and anthracite when the temperature was higher than 400 °C. This indicated that there was a synergistic interaction of co-pyrolysis of torrefied sawdust and anthracite. However, the reduced rate of final experimental residue content was not very high. This effect also existed for the co-pyrolysis of

torrefied sawdust and bituminous coal blends. The torrefied sawdust was almost completely pyrolyzed prior to 400 °C, then the torrefied sawdust had both a positive and negative effect on the pyrolysis of coal. The high content of hydrogen, alkali metal and alkali earth metal in torrefied sawdust boosted the pyrolysis of coal. However, the residue of pyrolyzed torrefied sawdust could block the pore of coal and then inhibit the pyrolysis of coal. Therefore, regarding both the beneficial and negative effects of torrefied sawdust on anthracite and bituminous coal, the experimental conversion was not higher than the calculated one. The figure also shows that the torrefied wood had an inhibitory effect on the conversion of lignite. A possible reason for this observation is that the pyrolysis temperatures of torrefied sawdust and lignite were so close that the negative effect of torrefied sawdust blocking the pore of lignite exceeded the positive effects of hydrogen donating and catalyzing.

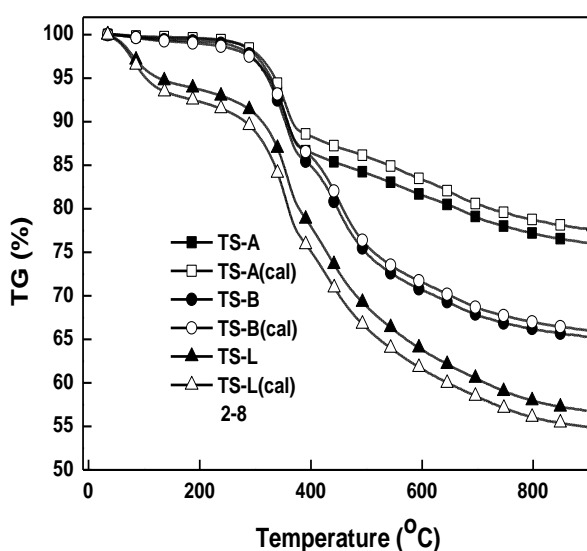


Fig. 5. TGA curves of calculated and experimental blends

The TG-FTIR Analysis of Co-Pyrolysis of Torrefied Sawdust and Coals at Different Blend Ratios

There was some noncondensable gas such as CO, CO₂, and CH₄ released when torrefied sawdust and coals were pyrolyzed at different blend ratios. In light of the fact that different compounds had different infrared absorbance, data was selected under a certain infrared band with different temperatures, and variations were observed for pyrolysis products with respect to temperature change. The corresponding curves are shown in Fig. 6. With decreasing coalification degree of the three coals, the release temperature of noncondensable gas was lowered and there was more CO released. For anthracite and bituminous coal, the addition of torrefied sawdust had the effect of increasing CO. There were obvious influences of the torrefied sawdust when blended with anthracite or bituminous coal at ratios of 5:5 and 8:2; the release of CO was higher with the pyrolysis of sawdust and anthracite or bituminous coal blends. However, there was no such effect for lignite; the release of CO for mono-pyrolysis of lignite was higher than the co-pyrolysis of torrefied sawdust and lignite. The infrared absorption curves for CO₂ demonstrated that the pyrolysis of the torrefied sawdust resulted in high emissions of

CO₂, and co-pyrolysis with coals led to much less CO₂ being released. The figures of CH₄ infrared band absorbance with temperature change indicated that the addition of torrefied sawdust in coals had an effect of discharging CH₄ within a lower temperature region. The addition of torrefied sawdust to anthracite had a synergistic effect to increase CH₄ emissions, especially at blend ratios of 2:8 and 5:5. This synergism was not observed for the co-pyrolysis of torrefied sawdust and bituminous coal or lignite. Therefore, a synergistic effect existed when torrefied sawdust was co-pyrolyzed with anthracite and bituminous coal. On the other hand, the addition of torrefied sawdust had an antagonistic effect on lignite.

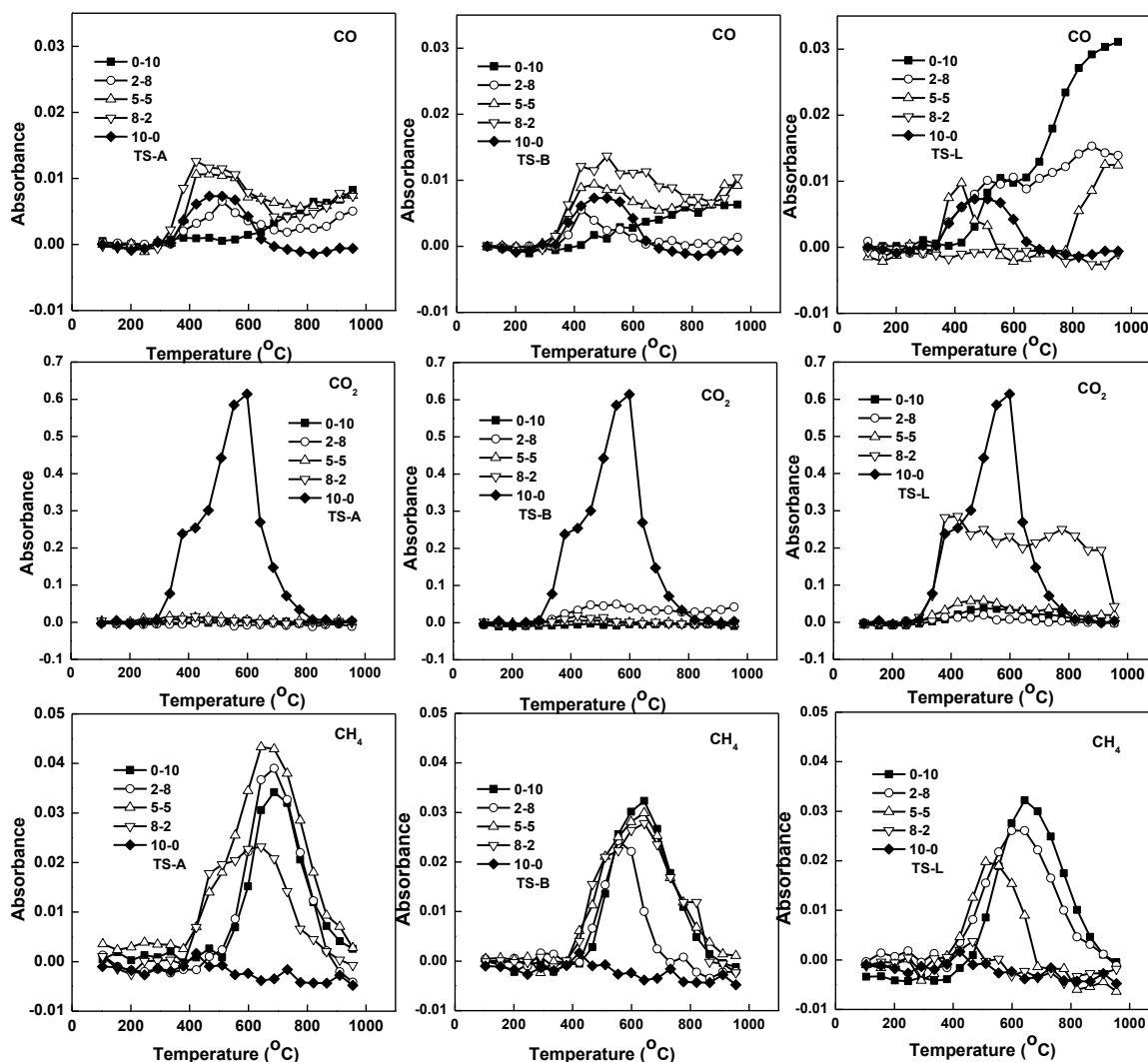


Fig. 6. Variation of pyrolysis products with temperature change under different mixed ratios of torrefied sawdust

Co-Pyrolysis Characteristics of Torrefied Sawdust and Different Rank Coals at High Temperature

The analysis of gaseous product

Figure 7 represents the gaseous components from calculations and experimental measurements of co-pyrolyzed torrefied sawdust and coals. The experimental data are shown as black symbols, and the calculated values are indicated as white symbols. For example, the calculated value of H₂ (X_{H₂}) at any blending ratio was obtained using the following equation,

$$X_{H_2} = r \times X_{TS,H_2} + (1-r) \times X_{Coal,H_2} \quad (2)$$

where r is the proportion of torrefied sawdust in blend, X_{TS,H_2} is the H₂ content from torrefied sawdust pyrolysis, and X_{Coal,H_2} is the H₂ content from coal pyrolysis. There were some uncertainty organic gases in the gaseous products, such as hydrocarbon compounds with more than three carbons, but these kinds of compounds were just of trace amounts, and therefore these compounds were not presented in Fig. 7.

Figure 7(a) shows the co-pyrolysis gas composition of anthracite coal and torrefied sawdust. The main components were H₂ and CO with the advantage of increasing the heating value of the gaseous product; the experimental values of H₂ and CO were also higher than the calculated values. One of the possible reasons was that anthracite had more fixed carbon than bituminous coal, lignite, and torrefied sawdust to react with the oxygen in torrefied sawdust and with the CO₂ produced by pyrolysis of torrefied sawdust in the reactor. The other possible reason was that the alkali metals and alkaline-earth metals in the torrefied sawdust residue had a catalytic effect on the pyrolysis of anthracite. With the increase of torrefied sawdust in the blends, the H₂ decreased and the CO increased. The experimental and calculated results for light hydrocarbons (C₂H_m) from co-pyrolysis of torrefied sawdust and anthracite were almost equal.

The experimental and calculated heating value data are shown in Table 2. The equation used for calculating the lower heating value is as follows,

$$HV_g = (126.36 \times CO + 107.98 \times H_2 + 358.18 \times CH_4 + 629.09 \times C_2H_m) \times 10^{-1} \quad (3)$$

where CO, H₂, CH₄, and C₂H_m are the contents of gaseous products.

Table 2. Lower Heating Value (LHV) of Gaseous Products

Ratio of torrefied sawdust-to-anthracite	Experimental LHV / MJ·m ⁻³	Calculated LHV / MJ·m ⁻³
0:10	7.68	7.68
2:8	11.66	9.01
5:5	13.29	11.01
8:2	13.41	13.01
10:0	14.34	14.34

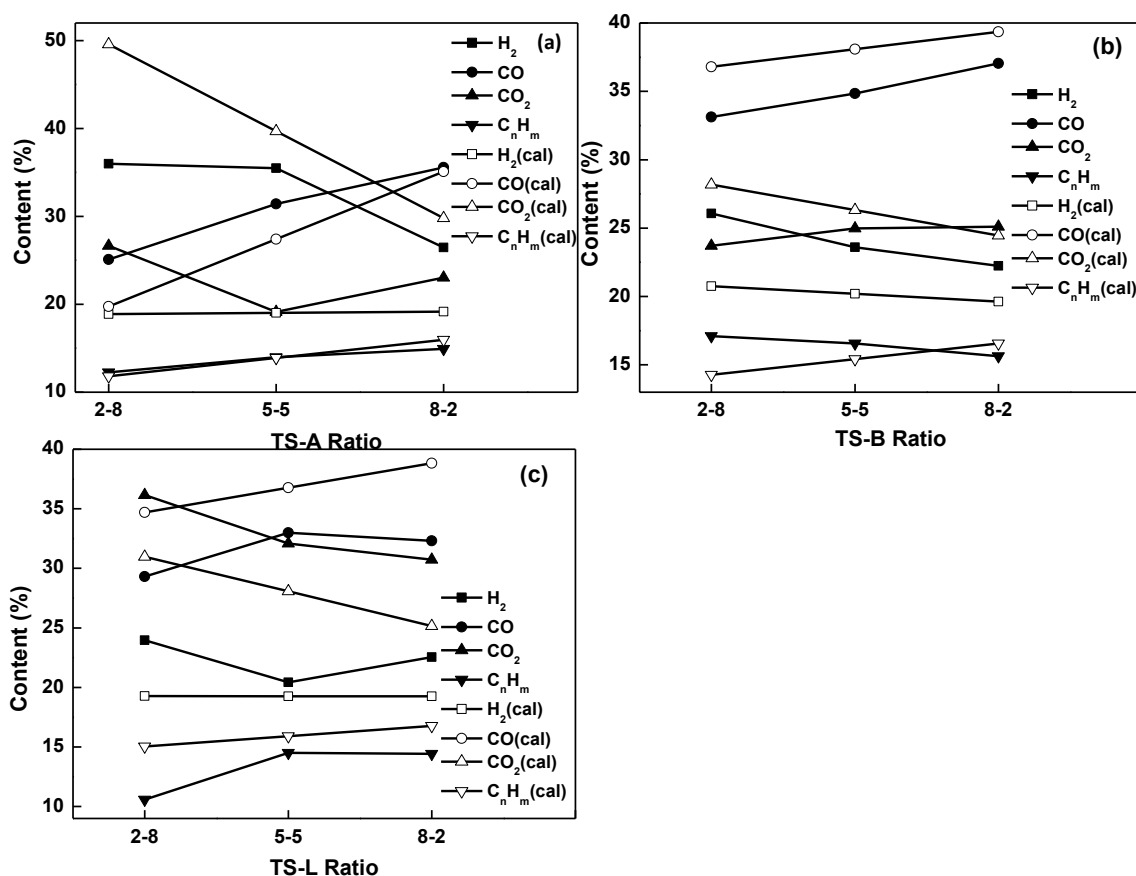
**Fig. 7.** Calculated and experimental gaseous components of pyrolysis blends

Figure 7(b) illustrates the characteristics of co-pyrolysis of torrefied sawdust and bituminous coal. CO was the main gaseous product and contributed 33 to 37% of the total yield. As the torrefied sawdust to bituminous coal ratio was increased, the CO yield increased and the H₂ yield decreased. The experimental yields of H₂ and C₂H_m were higher than the calculated yields. In addition, the CO and CO₂ experimental values were lower than the calculated values. These results showed that co-pyrolysis of bituminous coal and torrefied biomass slightly increased the combustible contents of the gaseous products. Figure 7(c) highlights the co-pyrolysis gaseous products from torrefied sawdust and lignite. The experimental H₂ and CO₂ contents were higher than the calculated ones.

However, the CO and C₂H_m data showed the opposite trend. These characteristics of the gaseous products decreased their heating value. Figure 7(c) also shows that CO yields were increased when the blending ratio increased, and the H₂ component slightly decreased.

In conclusion, the co-pyrolysis of torrefied sawdust and different rank coals resulted in higher CO yields and in lower H₂ yields as the torrefied sawdust percentage increased. The experimental H₂ yields were higher than the calculated values, which showed that co-pyrolysis had the advantage of increasing the amount of H₂ yield. Moreover, the blend of torrefied sawdust with high rank coal (anthracite) had the effect of increasing the combustible components, which resulted in higher heating values of the gaseous products. The synergistic effect of torrefied wood on the middle rank coal (bituminous coal) was not significant, and there was an opposite effect on the low rank coal (lignite).

The analysis of tar

Figure 8 shows the analysis of gas chromatograph-mass spectrometer (GC/MS) of tar. The tar from the pyrolysis of torrefied sawdust had much less compounds than that of tar from pyrolysis of sawdust. At about 23.090 min, the absorption peak represented butylated hydroxytoluene, which is a good anti-oxygen compound. The most abundant compound in the tar of torrefied sawdust pyrolysis was butylated hydroxytoluene (BHT). The other compounds were present as trace amounts in the tar. It follows that BHT may be conveniently separated for the binding of oxygen and for other uses. There were many more compounds in tar which resulted from pyrolysis of lignite than from anthracite and bituminous coal. After the addition of torrefied sawdust in the coals, the tar had fewer kinds of compounds, especially in the case of lignite.

The yield of gaseous, liquid and solid products

Figure 9 shows the yield of gaseous, liquid, and solid products. Figure 9 (a) shows the yield of liquid and solid products. The most important part of the liquid product was tar, and it also contained a bit of water. The solid product was ash and some fixed carbon that did not fully react. Figure 9 (a) shows that the solid product decreased and the liquid product increased slightly with the increase ratio of torrefied sawdust to coals. The results were similar to TGA curves in Fig. 4.

Figure 9 (b) shows the yield of gaseous product, which includes the N₂ that was used for removing oxidizing gases. However, the flow of N₂ was always kept for 20 mL/min so that it would not affect the comparison of different samples. As one can see from Fig. 9 (b), with the increase of the ratio of torrefied sawdust to coals, the yield of gaseous product increased, especially after the co-pyrolysis of anthracite and bituminous coals with torrefied sawdust.

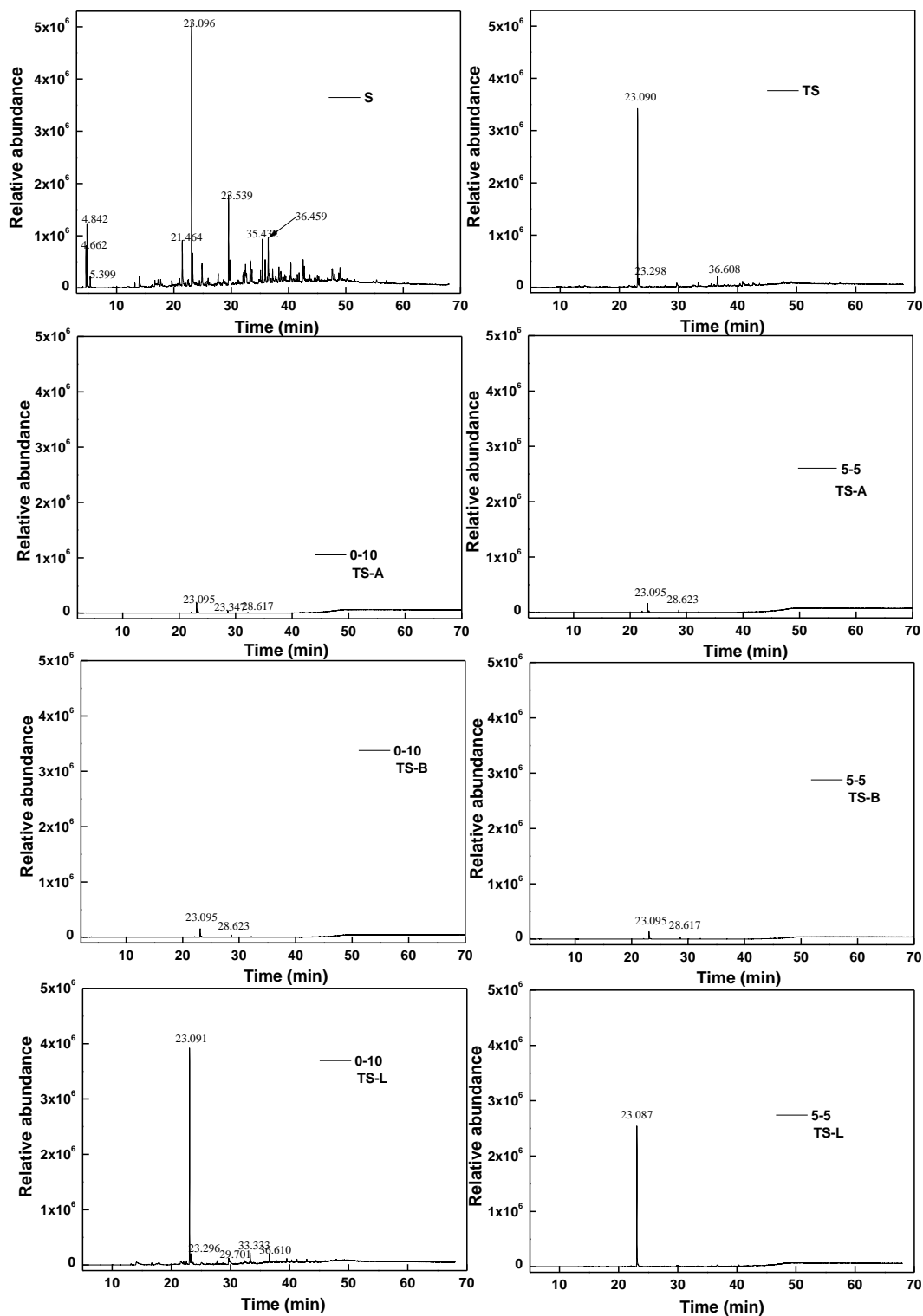


Fig. 8. The GC/MS analysis of tar from pyrolysis of different samples

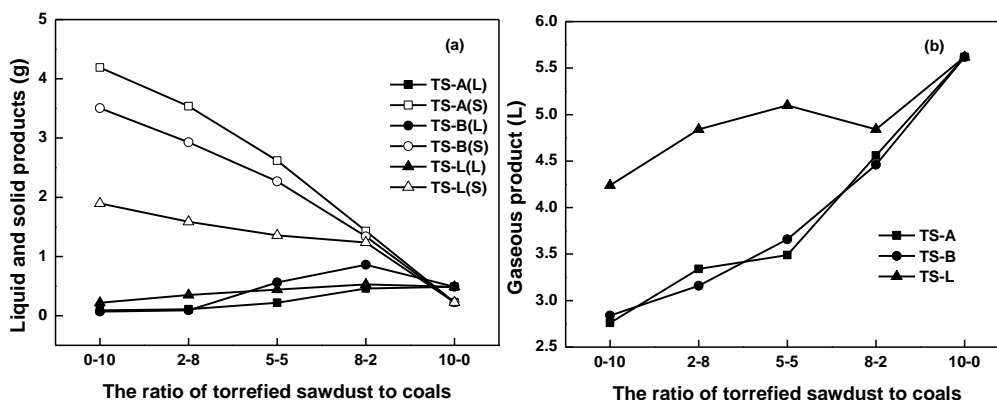


Fig. 9. The yield of gaseous, liquid, and solid products (L-liquid, S-solid)

CONCLUSIONS

1. Torrefaction has the effect of lowering the water content and increasing the fixed carbon content of torrefied sawdust; it also made the characteristics of the torrefied sawdust more similar to those of coal.
2. From the co-pyrolysis of torrefied sawdust and different rank coals, there was a synergistic effect of torrefied sawdust on the conversion of blends, the proportion of combustible gases in gaseous product, and the heating value of gaseous product. There was only a slight effect for bituminous coal. However, the torrefied sawdust has an inhibitory effect on lignite. With the difference of coals, the effect of torrefied sawdust to coals was different.
3. According to the infrared absorption of CO, CO₂, and CH₄, the co-pyrolysis of torrefied sawdust and anthracite had the effect of increasing CO and CH₄ while decreasing CO₂. With the decrease of coalification degree of the three coals, the synergistic effect of increasing CO and CH₄ and decreasing CO₂ lowered. In addition, increasing the percentage of torrefied sawdust of the blends had the effect of accelerating the CO content and decreasing the H₂ content at high pyrolysis temperatures.
4. The process of torrefaction had the effect of simplifying tar and the addition of torrefied sawdust also had the same effect on coals.

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