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Citation for published version:

Cong, H, Yao, Z, Mašek, O, Meng, H, Sheng, C, Wu, Y & Zhao, L 2020, 'Co-combustion, co-densification, and pollutant emission characteristics of charcoal-based briquettes prepared using bio-tar as a binder', *Fuel*, pp. 119512. <https://doi.org/10.1016/j.fuel.2020.119512>

Digital Object Identifier (DOI):

[10.1016/j.fuel.2020.119512](https://doi.org/10.1016/j.fuel.2020.119512)

Link:

[Link to publication record in Edinburgh Research Explorer](#)

Document Version:

Peer reviewed version

Published In:

Fuel

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**Co-combustion, co-densification, and pollutant emission
characteristics of charcoal-based briquettes prepared using bio-tar as
a binder**

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ABSTRACT: In this study, charcoal-based briquettes prepared using bio-tar as a binder are proposed as a substitute for conventional coal in rural China; furthermore, the fuel properties of the blends of charcoal, semi-coke, and bio-tar and the co-densification and pollutant emission characteristics of charcoal-based briquettes are investigated. The addition of charcoal improved the heating value and combustion index; however, the addition of a small amount of bio-tar did not have any significant effect on the fuel properties of the charcoal and semi-coke blend. The blending of charcoal and semi-coke with a mass ratio of 1:1 increased the integrated combustion characteristics from 7.73×10^{-12} (only semi-coke) to $16.92 \times 10^{-12} \text{ K}^{-3}\text{min}^{-2}$. Results of the co-densification experiments suggest that the bio-tar effectively improved the physical stability of charcoal-based briquette. By increasing the addition of bio-tar from 0 to 9 wt% at the densification temperature of 20 °C, the strength compaction and drop resistance increased by 8.6% and 13.9%, respectively. Increasing the densification temperature from 20 to 50 °C resulted in negative effects on mechanical strength. Pollutant emissions were observed during the ignition, stable combustion, and burnout phases of the stove. The bio-tar addition had distinct negative influences, increasing the total suspended particle and VOC emissions, which could be effectively weakened or eliminated by adding 3 wt% of hydrated lime. Herein, the applied chain and technical chain of charcoal-based briquettes are summarized. The study provides technical support for the industrial application of charcoal-based briquettes in rural China, with replication potential elsewhere.

Keywords: charcoal-based briquette, bio-tar, fuel property, co-densification, pollutant emission

1. Introduction

Heating in rural area of north China faces prominent problems such as underdeveloped heating infrastructure, low energy efficiency, and high pollutant emission [1]. The annual consumption of heating coal is approximately 400 million tons of standard coal in China, of which approximately 200 million tons is accounted by scattered burning in rural areas [2]. The use of conventional coal with poor quality plays a crucial role in the formation of haze. Therefore, the “substituting conventional coal” project was proposed by the government [3]. To overcome this issue, an effective strategy is to partially replace or improve the conventional coal, as it is difficult to completely replace coal in short term.

Semi-coke and charcoal are cleaner than raw coal, and are therefore encouraged for use as heating fuel in China [4][5]. Semi-coke and charcoal are obtained from bituminous raw coal and tree branches, respectively, by using the slow pyrolysis technology [6]. Given the relationship between mass density, energy density, and mechanical strength, the use of densified fuels with raw solid fuels is critical [7][8]. The semi-coke and charcoal are generally densified individually or codensified with other solid fuels into briquettes to improve the fuel properties [9][10]. Small amounts of starch, sodium nitrate, limestone, borax, and sawdust are generally added to briquettes for improving ignition, promoting steady burning, and to achieve efficient

manufacturing [11]. A honeycomb briquette was developed from a blend of semi-coke and corn stover char, and the value-chain model analysis of the new fuel suggests that it has promising application prospects as a heating fuel in rural China [12]. However, difficulties in handling and using bio-tar presented a severe problem to the biomass pyrolysis project. Bio-tar is a complex organic mixture of condensable or non-condensable hydrocarbons comprising 1- to 5-ring aromatic compounds along with other oxygen-containing hydrocarbons and polycyclic aromatic hydrocarbons; it poses a severe risk of contamination if discarded [13]. In rural China, in addition to crop straws, many wooden materials are collected from orchards. Developing charcoal-based briquettes using bio-tar as a binder is an effective way to solve the abovementioned problem. Therefore, it is necessary to systematically investigate the fuel properties, co-densification characteristics, and pollutant emission characteristics of the blends of charcoal, semi-coke, and bio-tar.

From the viewpoint of fuel properties, such as alkali and alkaline earth metal content, bulk density, and heating value [14], charcoal, semi-coke, and bio-tar might have some complementary effects, which should be determined. In particular, the co-combustion characteristics of several kinds of blends, such as straw, municipal solid waste, raw coal, sludge, and oil shale, have been studied [15][16]; however, a considerable knowledge gap remains from the viewpoint of blends of charcoal, semi-coke, and bio-tar. From the viewpoint of co-densification, bio-tar is an effective lubricant and binder for the densification process owing to its high viscosity [10][17].

Although several studies have investigated the co-densification characteristics of blends of biomass, char, or raw coal with starch, clay, polymer plastic, or wood fiber as a binder [18][19], the co-densification characteristics of charcoal and semi-coke using bio-tar as a binder have rarely been investigated. In particular, most studies have focused on the material ratio, pressure, moisture content, and particle size in co-densification experiments [18][19][20]. The viscosity of bio-tar can be reduced and its fluidity can be improved by properly increasing its temperature (35–50 °C). In this study, the densification temperature was also considered as an influencing factor. The optimized co-densification condition of bio-tar addition ratio and co-densification temperature should be determined. Moreover, information pertaining to the flue emission from combustion of charcoal-based briquettes is limited. The addition of bio-tar to briquettes might affect pollutant emissions, especially particulate matter (PM) and volatile organic compound (VOC) emissions [21]. Therefore, the pollutant emission from combustion of charcoal-based briquettes, when bio-tar is used as a binder, should also be considered.

This study primarily aims to investigate the fuel properties, co-densification characteristics, and pollutant emission characteristics of blends of charcoal, semi-coke, and bio-tar. Herein, we aim to establish a method for using the bio-tar produced by biomass pyrolysis projects; furthermore, we aim to report results that are beneficial for developing a new clean fuel suitable for heating in rural China. This paper also summarizes the applied chain and technical chain of the charcoal-based briquettes to

provide basic support for industrial application of the charcoal-based briquettes using bio-tar as a binder in rural China.

2. Materials and methods

2.1 Materials

The raw materials primarily included charcoal, semi-coke, and bio-tar. Charcoal from tree branches was produced at a pyrolysis temperature of 550–600 °C and residence time of 30–35 min in the Qiannanyu Biomass Pyrolysis Demonstration Project of Hebei Province [22]. Semi-coke was produced via low-temperature pyrolysis of volatile bituminous coal from Shenmu County, Shaanxi Province. The bio-tar was also collected from the Qiannanyu Biomass Pyrolysis Demonstration Project as a by-product of tree branch pyrolysis at the abovementioned conditions. Heating values were measured using a bomb calorimeter (LECO AC-300) following the adiabatic method according to the China National Standard (GB T 213 2008) [23]. Ultimate analysis (carbon, hydrogen, nitrogen, and sulfur) was performed using a Vario ELIII Elemental Analyzer according to ASTM D5373 and ASTM D4239 [24][25]. The metal element contents were determined by inductively coupled plasma mass spectrometry (Thermo Fisher Scientific) according to the AOAC official method 975.03.

2.2 Experimental facility

2.2.1 Testing instrument setup

Thermogravimetric analyzer (DTG-6A) manufactured by Shimadzu Corporation

was used to analyze the combustion characteristics. The reactor had a diameter of 60 mm, and the reaction atmosphere was canned air. The initial test temperatures were set at 20 °C, which were increased to 1 000 °C at a heating rate of 10 °C/min; air flow rate was 100 mL/min. Single and blended samples were milled to less than 0.15 mm, and approximately 10 mg sample was used for each test.

Total suspended particles (TSP) were collected using an electrical low-pressure impactor (Dekati ELPI+) manufactured by DEKATI Ltd., which can collect particles from 6 nm to 10 µm in 14 size fractions. The mass size and number size distributions of TSP were estimated using ELPI software V12.0. To ensure that the collected PM was kept below saturation, flue gas was diluted 64 times using two Dekati diluters.

Flue gas analyzer (ECOM-J2KN) manufactured by RBR was used to test the NO_x and SO₂ emissions. In response to the ignition, stable combustion, and burnout phases of stove running, flue gases were tested four times, once every 3 min. The results were converted from ppm to mg/Nm³.

2.2.2 Densification platform and test setup

Densification experiments were conducted using a customized die with cooperation of a universal testing machine (Fig. 1a). The die mainly comprised a die piston, die sleeve, and heating jacket. The die sleeve had an inner diameter of 60 mm, outer height of 120 mm, and a cylinder with diameter of 25 mm on the central axis. A heating jacket connected to temperature control system was installed on the outside of die sleeve to heat the raw materials.

Particle size is a key factor influencing the densification performance [20]. To ensure consistent particle size distribution of raw materials for all the densification experiments, the semi-coke and charcoal were individually milled on a disintegrating mill once for all. The particle size distributions are shown in Fig. S1. Bio-tar was added to the blends of charcoal and semi-coke using a pipette according to the experimental design. During this process, the bio-tar was heated to approximately 50 °C in a water bath to enhance its fluidity for pumping with pipette. Under experimental conditions with heating, the raw material was filled into the die sleeve, and temperature of the heating jacket was increased to the desired level and held for 5 min. Finally, the die piston started to move for densification. Once the die piston reached desired pressure of 10 kN, it was maintained for 20 s for all the densification experiments.

Fig. 1. Schematic of the (a) densification testing platform and (b) emission testing

platform.

2.2.3 Pollutant emission platform and test setup

Herein, an NF9C household heating stove was used. The hearth height, outer length $\times$ width, and inner diameter of the stove are 650, 480×480 , and 230 mm, respectively (Fig. 1b). It features an air inlet with a diameter of 60 mm near the bottom. Air inlet was set up with an open ratio of 100% during pollutant emission tests, and the flue gas was sampled or real-time tested on ignition, stable combustion, and burnout phases. Charcoal-based briquettes with a height of 83–85 mm, outer diameter of 60 mm, and hole with a diameter of 25 mm were used herein. For contrast analysis, three kinds of briquettes were used in the emission experiments.

2.3 Determination of co-combustion, co-densification and pollutant emission characteristics

Combustion characteristics can be evaluated using several combustion parameters, such as ignition temperature, burnout temperature, burnout characteristics, and integrated combustion characteristics [27]. Burnout index C_b ($10^{-4}/\text{min}$) was used to characterize the burnout characteristics of samples; herein, large values represent better burnout characteristics.

$$C_b = \frac{f_1 \cdot f_2}{t_0}, \quad (1)$$

where f_1 (%) is the initial burnout rate, which characterizes the rate of loss of fuel weight on the ignition point of the TG curve, f_2 (%) is the late burnout rate, and t_0 (min) is the burnout time, representing the time from the initiation of combustion mass loss to

burnout (with a mass loss rate of 98%).

The integrated combustion characteristics of the sample are described by combustion index S_N ($10^{-12} \text{ K}^{-3} \text{ min}^{-2}$); herein, large values represent better combustion characteristics.

$$S_N = \frac{(dw/dt)_{\max} \cdot (dw/dt)_{\text{mean}}}{t_i^2 t_f}, \quad (2)$$

where $(dw/dt)_{\max}$ (%/min) and $(dw/dt)_{\text{mean}}$ (%/min) are the maximum and average burn rates, respectively, and t_i (K) and t_b (K) are the ignition and burnout temperatures, respectively.

The energy consumption is described by specific energy consumption (SEC). The index expresses the energy consumed during densification for unit mass of raw material.

$$SEC = \frac{W}{m} = \frac{\int f \cdot ds}{m}, \quad (3)$$

where SEC (J/kg) is the specific energy consumption, W (J) is densification energy, m (kg) is mass of the briquette, and f (kN) and s (mm) are the pressure and displacement, respectively. In addition, the drop resistance and compaction strength of briquettes were tested according to the method recommended by the China National Standard GB 34170-2017 [28].

Ozone formation potential (OFP) (mg/m^3) was calculated by considering VOC source profiles and maximum incremental reactivity of each species [22].

$$OFP = \sum_{i=1}^n MIR_i \times VOC_i, \quad (4)$$

where MIR_i and VOC_i represent the maximum incremental reactivity (gram O_3 per gram VOCs) and concentration of the i^{th} VOC species ($\mu\text{g}/\text{m}^3$).

3. Results and discussion

3.1 Fuel property

3.1.1 Basic physicochemical characteristics

As listed in Table 1, the LHV of charcoal was 9.4% higher than that of semi-coke. The addition of charcoal was beneficial for enhancing the heating value of briquettes. The bulk density of charcoal was considerably smaller than that of semi-coke, which had a negative effect on the mass density of briquettes. From the viewpoint of biomass utilization, the energy density was significantly increased by pyrolysis and densification. The atomic H/C and O/C ratio are commonly used to evaluate the energy quality of solid fuels. The smaller the index value, the better the fuel quality for solid fuels [29]. Based on the ultimate analysis results, the atomic O/C ratios of charcoal, semi-coke, and bio-tar were 0.20, 0.31, and 0.51, respectively, whereas H/C ratios were 0.02, 0.43, and 0.41, respectively. The O/C and H/C ratios of the charcoal were close to those of anthracite [30], suggesting that the performance of charcoal is better than that of semi-coke. The addition of charcoal and bio-tar negatively affected fuel-N content. The S contents of charcoal and bio-tar were distinctly lower than those of the semi-coke, and therefore, the clean levels of blend fuels are much higher.

Table 1

Characteristics of charcoal, semi-coke, and bio-tar.

Feedstocks	Charcoal	Semi-coke	Bio-tar
Bulk density [kg/m ³]	388	908	/

	LHV ^a [MJ/kg]	30.53	27.91	20.51
Proximate analysis	Moisture	2.46	7.41	3.07
[wt%, ad]	Volatile	12.02	9.01	53.57
	Ash	3.35	11.86	9.13
	Fixed carbon	82.03	71.72	34.23
Ultimate analysis	C	78.27	68.41	57.32
[wt%, daf]	H	0.11	2.47	1.97
	O ^b	20.72	28.42	38.68
	N	0.78	0.38	1.93
	S	0.12	0.32	0.10
Metal elements [mg/g]	Na	1.99	2.04	4.33
	K	3.32	0.27	0.79
	Ca	25.52	16.25	5.24
	Mg	22.58	0.52	1.00

ad: air-dry basis; daf: dry and ash-free basis.

^a LHV: lower heating value.

^b Calculated by the difference.

The thermogravimetry (TG) and differential thermogravimetry (DTG) curves of semi-coke, charcoal, and bio-tar are shown in Fig. 2. The weight losses of the three fuels were clearly different. When the samples were heated, water evaporated, which was accompanied by devolatilization, volatile flaming, and fixed carbon firing. During the first stage with heating temperatures below 100 °C, semi-coke experienced a high

reduction in weight owing to the loss of its relatively high moisture content (Table 1). Charcoal exhibited a more intense rate of weight loss during the second stage, and this occurred at a distinctly lower temperature than that of the semi-coke. The curve of bio-tar was the most tortuous, which indicated that the weight loss fluctuated throughout the process owing to its complex components consisting of phenols, amides, and lipids with >20C atoms [31]. The burnout indices and integrated combustion indices of the materials are listed in Table. 2. Bio-tar had the highest reaction rate, whereas charcoal had the lowest ignition and burnout temperatures. The combustion index value depends on the above indices. Charcoal presented the best combustion performance, with burnout and combustion index of $57.41 \times 10^{-4}/\text{min}$ and $36.28 \times 10^{-12} \text{ K}^{-3} \cdot \text{min}^{-2}$, respectively. Semi-coke showed the poorest combustion performance, with burnout and combustion index of $13.49 \times 10^{-4}/\text{min}$ and $7.73 \times 10^{-12} \text{ K}^{-3} \cdot \text{min}^{-2}$, respectively. These results highlight the distinctly different combustion characteristics of the three fuels.

Fig. 2. Combustion characteristic curves, i.e., thermogravimetry (TG) and differential thermogravimetry (DTG) of charcoal, semi-coke, and bio-tar.

3.1.2 Co-combustion characteristics of charcoal and semi-coke

Blends of charcoal and semi-coke were prepared with mass ratios of 3:7, 5:5, and 7:3, which were denoted as CS37, CS55, and CS73, respectively. The TG and DTG curves of CS37, CS55, and CS73 are shown in Fig. 3. With an increase in the proportion of semi-coke, the TG curves of CS73, CS55, and CS37 gradually shifted to the right. This tendency is consistent with the theoretical results. An intense rate of weight loss was achieved for the blends during the second stage than that achieved for semi-coke. Accordingly, the burnout index and integrated combustion index of CS37, CS55, and CS73 increased gradually. A higher burnout index implies lesser time for the fuel to burn out, thereby minimizing unburnt carbon loss [32]. The weighted average values of these indices corresponding to the blending ratio were larger than the test results, suggesting that the interactions occurring between the components of blends slightly lowered the reactivity. The interactive effects varied with the characteristics of components in blends, such as heterogeneity, nature, and distribution of reacting species [33]. However, the integrated combustion index of CS55 reached $16.92 \times 10^{-12} \text{ K}^{-3} \cdot \text{min}^{-2}$, which was more than two times higher than that of the semi-coke alone, indicating that the blend of charcoal and semi-coke is a promising solid fuel.

Fig. 3. Co-combustion characteristic curves, i.e., TG and DTG of blends CS37, CS55, and CS73.

3.1.3 Influence of bio-tar addition on co-combustion characteristics

To analyze the influence of bio-tar addition on the combustion characteristics, 3, 6, and 9 wt% bio-tar were added to sample CS55, which were denoted as T03, T06, and T09, respectively. TG and DTG curves of the different blends are shown in Fig. 4. These curves were almost coincident, indicating that the addition of bio-tar had little influence on the combustion characteristics of the samples. In fact, the combustion characteristic parameters of the bio-tar and CS55 were also quite proximate, as shown in Table 2. The integrated combustion characteristics of T03, T06, and T09 were $14.46\text{--}17.20 \times 10^{-12} \text{ K}^{-3}\text{min}^{-2}$, which is close to those of CS55. From the viewpoint of combustion characteristics, the effect of a small amount of bio-tar addition on CS55 could be ignored, suggesting that co-combustion is a feasible method for handling and using bio-tar.

Fig. 4. Co-combustion characteristic curves, i.e., TG and DTG of blends T03, T06, and T09.

Table 2

Combustion indices of different samples.

Sample	Ignition temperature (°C)	Burnout temperature (°C)	Burnout characteristics $C_b/(10^{-4}/\text{min})$	Maximum reaction rate $(dw/dr)_{\text{max}}/(\%/ \text{min})$	Average reaction rate $(dw/dr)_{\text{mean}}/(\%/ \text{min})$	Integrated combustion characteristics $S_N/(10^{-12}\text{K}^{-3}\text{min}^{-2})$
Charcoal	349.10	511.80	57.41	-5.53	-4.09	36.28
Semi-coke	415.61	695.65	13.49	-3.23	-2.88	7.73
Bio-tar	391.81	554.94	43.87	-6.31	-3.09	22.87
CS37	405.20	623.15	21.91	-4.03	-3.34	13.15
CS55	382.55	587.09	27.35	-4.25	-3.42	16.92
CS73	380.83	548.72	35.87	-4.95	-3.76	23.37
T03	383.10	598.12	27.69	-3.96	-3.21	14.46
T06	378.30	582.21	29.25	-4.31	-3.32	17.20
T09	368.67	595.96	28.93	-3.93	-3.12	15.15

3.2 Co-densification experiments

Based on the conclusions of co-combustion characteristics detailed in Section 3.1, a blend with 5:5 mass ratio of charcoal to semi-coke (CK) was selected for further

analyzing the co-densification characteristics of charcoal and semi-coke using bio-tar as a blinder.

3.2.1 Mechanical strength

In this study, the mechanical strength includes compaction strength and drop resistance, representing the stability of briquettes under the influence of different types of external forces [20][34]. To investigate the influence of densification temperature and bio-tar addition on the mechanical strength of briquettes, densification temperatures of 20, 35, and 50 °C were applied. Briquettes with 3, 6, and 9 wt% bio-tar were prepared and denoted as 3T, 6T, and 9T, respectively. In addition, 1 wt% cellulose was added to the blends as a basic binder for all co-densification experiments.

The compaction strength and drop resistance of the charcoal-based briquettes (CK, 3T, 6T, and 9T) are shown in Fig. 5. The compaction strength and drop resistance increased distinctly as the proportion of bio-tar addition increased at densification temperatures of 20 and 35 °C. The strength compaction and drop resistance increased by 8.6% and 13.9%, respectively, when the bio-tar addition was increased from 0% to 9% at the densification temperature of 20 °C. These results indicate that bio-tar addition had a strong positive effect on densification quality at densification temperatures of 20–35 °C. This trend could be explained by the fact that increasingly more binding compounds are introduced into the briquettes and stronger adhesion bonds of the material particles develop as the proportion of bio-tar addition is increased [10]. SEM images (Fig. S2) with 500× magnification of cross sections show that many

interweaving structures of bio-tar and material particles present in T9 densified at temperatures of 20 and 35 °C. The filamentous form of bio-tar and abundant fine charcoal particles embedded in the binders were found, which might effectively strengthen the binding force of the particles. When the temperature was increased to 50 °C, the overall mechanical strength was reduced, indicating that increasing the temperature from 20 to 50 °C had an adverse effect on the co-densification performance. The water vapor formed under strong pressure and higher densification temperature might weaken the binding force between the particles [35].

Fig. 5. Compaction strength and drop resistance of charcoal-based briquettes, herein, briquettes with 0, 3, 6, and 9 wt% bio-tar were denoted as CK, 3T, 6T, and 9T.

3.2.2 Energy density and specific energy consumption

Mass density is also an important index of densification quality, as it affects the transportation and use cost of the solid fuels. In general, a higher mass density means a higher energy density for single-material briquettes. The energy density and mass density were not linearly related herein because various proportions of bio-tar were

added to the blends, and its heating value was significantly lower than that of charcoal and semi-coke.

The mass density, energy density, and specific energy of the charcoal-based briquettes densified under different conditions are shown in Fig. 6. An increase in the proportion of bio-tar addition positively contributed to mass density, and this trend was more obvious at relatively low densification temperatures. Herein, T9 and T6 represent the sample with the largest energy density at the densification temperature of 20 and 35 °C. The contribution of bio-tar addition from 6% to 9% to the mass density was insufficient to offset the negative influence of the bio-tar on heating values. Increasing the proportion of bio-tar and densification temperature would help to reduce the SEC. This might be explained as a reduction in friction among the particles and between the particles and die as a result of properly increasing the bio-tar addition and densification temperature [36]. Specific energy affects the production costs; however, the energy consumption of heating the materials at densification temperatures of 35 and 50 °C was not considered herein.

Fig. 6. Energy density and specific energy consumption of various charcoal-based briquettes at different densified temperatures.

3.2.3 Parameter optimization

The effects of bio-tar addition and densification temperature on the compaction strength, drop resistance, mass density, energy density, and specific energy, which represent densification quality and production cost, were investigated. To evaluate the comprehensive performance of different experimental conditions, a gray relation projection, as a comprehensive evaluation method applying the gray system theory and vector projection principle, was adopted herein [37].

Based on the index vector (I) shown in equation (5), a decision matrix of gray correlation projection (Y) was constructed by combining the test values of each experiment (equation (6)). The first line represents the optimal case of all tests, meaning that the maximum and minimum values were taken for the benefit and cost index, respectively. To eliminate the incommensurability caused by index magnitude and units, the decision matrix (Y') was initialized using percentage conversion (equation (7)). The correlation coefficients between each vector point in space and optimal vector point were calculated, and a gray correlation judgment matrix (F) was established as equation (8).

$$I = (\text{compaction strength, drop resistance, energy density, specific energy}), \quad (5)$$

$$Y = \begin{bmatrix} 414.0 & 89.9 & 21499.4 & 526.3 \\ 307.4 & 85.6 & 19965.5 & 627.9 \\ 331.6 & 87.8 & 20890.8 & 598.3 \\ 352.4 & 89.0 & 21252.7 & 569.3 \\ 414.0 & 89.4 & 21499.4 & 526.3 \\ 283.3 & 83.9 & 19874.4 & 637.6 \\ 328.4 & 86.5 & 20327.1 & 614.8 \\ 362.9 & 86.3 & 20904.6 & 570.3 \\ 379.9 & 89.9 & 20776.6 & 568.9 \\ 315.6 & 81.8 & 20244.5 & 609.2 \\ 328.7 & 83.0 & 20396.0 & 598.2 \\ 326.9 & 80.0 & 21311.1 & 541.5 \\ 332.4 & 86.7 & 21153.0 & 542.1 \end{bmatrix}, \quad (6)$$

$$Y' = \begin{bmatrix} 1.00 & 1.00 & 1.00 & 1.00 \\ 0.74 & 0.95 & 0.93 & 0.84 \\ 0.80 & 0.98 & 0.97 & 0.88 \\ 0.85 & 0.99 & 0.99 & 0.92 \\ 1.00 & 0.99 & 1.00 & 1.00 \\ 0.68 & 0.93 & 0.92 & 0.83 \\ 0.79 & 0.96 & 0.95 & 0.86 \\ 0.88 & 0.96 & 0.97 & 0.92 \\ 0.92 & 1.00 & 0.97 & 0.93 \\ 0.76 & 0.91 & 0.94 & 0.86 \\ 0.79 & 0.92 & 0.95 & 0.88 \\ 0.79 & 0.89 & 0.99 & 0.97 \\ 0.80 & 0.91 & 0.98 & 0.97 \end{bmatrix}, \quad (7)$$

$$F = \begin{bmatrix} 1.00 & 1.00 & 1.00 & 1.00 \\ 0.38 & 0.77 & 0.69 & 0.49 \\ 0.44 & 0.87 & 0.85 & 0.57 \\ 0.51 & 0.94 & 0.93 & 0.68 \\ 1.00 & 0.96 & 1.00 & 1.00 \\ 0.33 & 0.70 & 0.68 & 0.47 \\ 0.43 & 0.81 & 0.74 & 0.52 \\ 0.56 & 0.80 & 0.85 & 0.67 \\ 0.66 & 1.00 & 0.82 & 0.68 \\ 0.40 & 0.64 & 0.73 & 0.54 \\ 0.43 & 0.67 & 0.75 & 0.57 \\ 0.43 & 0.59 & 0.95 & 0.85 \\ 0.44 & 0.63 & 0.91 & 0.84 \end{bmatrix}. \quad (8)$$

The weight coefficient vector of each index according to the expert scores was $W = (0.13, 0.37, 0.21, 0.29)$, and the gray correlation projection weight coefficient vector was $W' = (0.03, 0.26, 0.08, 0.16)$. Gray relational projection values (Table 3) of different experimental conditions were obtained based on equation (8) and W' . The larger the

value, the better the comprehensive performance of corresponding briquettes. The best comprehensive performance was achieved with high bio-tar addition under low and medium densification temperatures, average performance was observed with medium and high bio-tar addition at high temperature, and poorest performance was observed at medium and high densification temperatures without bio-tar addition.

Table 3

Gray relational projection values of briquettes under different experimental conditions

Conditions	20 °C	20 °C	20 °C	20 °C	35 °C	35 °C	35 °C	35 °C	50 °C	50 °C	50 °C	50 °C
	CK	3T	6T	9T	CK	3T	6T	9T	CS	3T	6T	9T
Values	0.35	0.40	0.44	0.52	0.32	0.37	0.40	0.46	0.32	0.34	0.38	0.39

3.3 Pollutant emission experiments

Based on the conclusions of co-densification characteristics detailed in Section 3.2, regarding the potential pollutant emission risk of excessive bio-tar addition, charcoal-based briquettes, densified at 20 °C using 6 wt% bio-tar as a binder, were selected to investigate the pollutant emission characteristics. Three samples of briquettes were prepared for a controlled trial study. Herein, CK represents briquettes produced with the blend of charcoal and semi-coke (mass ratio 1:1), 6T represents briquettes produced with 6 wt% bio-tar as a binder based on CK, and 6Tp represents briquettes produced with 3 wt% hydrated lime as a catalyst based on 6T.

3.3.1 Conventional gaseous pollutants

NO_x and SO₂ emissions, which are regarded as the main conventional gaseous pollutants, are shown in Fig. 7. NO_x emission concentrations for CK, T6, and T6p were

113.2–225.6, 133.1–234.4, and 120.8–208.9 mg/Nm³, respectively. For these three charcoal-based briquettes, the changing trends in NO_x emissions during different combustion phases were similar, indicating that bio-tar addition had no distinct influence on the NO_x emission. NO_x emissions are mainly affected by the fuel-N content and combustion temperature [38][39]. During the entire process, the stove temperatures showed a trend of initial increase and subsequent decrease for the three briquettes (Fig. S3), and the NO_x emissions varied among different combustion phases. According to the ultimate analysis results in Table. 1, the addition of 6 wt% bio-tar had a little influence on the fuel-N content of the briquettes.

The changing trends of SO₂ emissions in the three phases were similar. The SO₂ emission concentrations for CK, T6, and T6p were 22.4–33.1, 20.7–32.6, and 7.8–11.3 mg/Nm³, respectively. In the stable combustion phase, the relatively higher stove temperature resulted in a higher conversion rate of fuel sulfur to gaseous SO₂ [40]. Overall, the amount of SO₂ emission from T6p was significantly lower than that from CK and T6, and the desulfurization efficiency was more than 70%. These results indicate that the addition of hydrated lime was effective for reducing the SO₂ emission of charcoal-based briquettes. This can be explained by the fact that Ca(OH)₂ can easily capture SO₂ and volatile emissions during the combustion process [41].

Fig. 7. Emission concentrations of NO_x (a) and SO₂ (b) of CK, 6T, and 6Tp at different combustion phases (CK, 6T, and 6Tp represent briquettes produced with the blend of charcoal and semi-coke (mass ratio 1:1), briquettes produced with 6 wt% bio-tar as a binder based on CK, and briquettes produced with 3 wt% hydrated lime as a catalyst based on 6T, respectively).

3.3.2 Total suspended particles

The distributions of mean emissions of TSP, including mass size and number size concentrations, for different briquettes during the entire combustion process are shown in Fig. 8. The changing trends of the three mass size concentration curves were similar, indicating that the particle masses were mainly distributed within grades 11–14 with particle sizes of 2.5–10 μm for the three kinds of briquettes. Meanwhile, the three number size concentration curves were also similar, indicating that the particle numbers were mainly distributed within grades 1–7 with particle sizes of $<0.16 \mu\text{m}$. The mass and number size of particles within grades 1–7 increased by 2.83 and 2.78 times,

respectively, after the addition of bio-tar, whereas after the addition of hydrated lime, these values were significantly reduced by 46.0% and 53.1%, respectively. These results indicate that controlling particulate emission is a key concern for bio-tar addition, and the mechanism of reducing particle emission through the addition of hydrated lime should be further explored [42].

a. Particle mass size distribution

b. Particle number size distribution

Fig. 8. Total suspended particles (TSP), mean particle number size, and mean mass size distributions of CK, 6T, and 6Tp throughout the process.

The particle concentrations during different combustion phases for CK, 6T, and 6Tp are shown in Fig. 9. The total suspended particles (PM10) and fine suspended particles (PM2.5), which are known to adversely affect the human health, were separately studied. Overall, the concentration of particulate emission during the ignition phase was the largest, regardless of mass size or number size, whereas that in the burnout phase was the smallest. In the three combustion phases, the addition of bio-tar increased the particle emissions, whereas the further addition of hydrated lime significantly decreased it again. Particulate emissions were the highest during the ignition phase, which might be associated with unstable combustion [43]. The bio-tar addition increased the PM10 emission from 12.2 to 33.3 mg/m³, and then hydrated lime addition decreased it to 21.3 mg/m³. For PM2.5, the bio-tar addition increased it from 5.1 to 13.8 mg/m³, and then the further addition of hydrated lime decreased it to 7.6 mg/m³. These results indicate that the combined addition of bio-tar and hydrated lime is effective for the application of charcoal-based briquettes.

a

b. Particle number size concentrations

Fig. 9. TSP, particle number size, and mass size concentrations of CS, 6T, and 6Tp during different combustion phases.

3.3.3 VOCs emission

A total of 98 VOC species were detected using GC/MS according to the China Environmental Industry Standard HJ 759-2015 [44]. The VOC chemical profiles emitted from CK, 6T, and 6Tp are shown in Fig. 10. These profiles presented some similarities, all containing mainly aromatic hydrocarbons, alkanes, and alkenes. Toluene and m-xylene were the two largest volatile organic pollutants. A small amount of alkene was detected only for 6T in the ignition and stable combustion phases. This can be attributed to the presence of an active functional group as an unsaturated bond that reacts easily with oxygen to form carbon dioxide and water [45].

The concentrations of VOCs fluctuated widely under different experimental conditions, and these were reflected in the OFP index. The OFP of the CK, 6T, and 6Tp were 0.18–0.37, 0.24–48.4, and 0.23–37.4 mg/m³ during the entire combustion process.

The bio-tar addition distinctly increased VOC emissions. Comparing 6T with CS, the emission of VOCs increased to 131.3 and 4.5 times during ignition and stable combustion phases, respectively, whereas comparing 6Tp with 6T, VOC emissions decreased to 0.77 and 0.29 times, respectively. The influence of bio-tar addition on VOC emission was the strongest during the ignition phase, but the addition of hydrated lime had only a minor effect. The hydrated lime had a distinct effect during the stable combustion and burnout phases, resulting in similar VOC emissions of 6Tp and CK. This indicates that a higher combustion temperature and hydrated lime addition are key factors for reducing the VOC emission of 6T [46].

Fig. 10. Mass percentage of VOC species and ozone formation potential for CK, 6T, and 6Tp during different combustion phases.

3.3.4 Comprehensive properties of the briquettes

To utilize charcoal and bio-tar more efficiently and to develop a new heating fuel, a charcoal-based briquette prepared using bio-tar as a binder was investigated systematically from the viewpoint of the fuel properties, co-densification characteristics,

and pollutant emission characteristics. The applied chain and technical chain of the charcoal-based briquette are shown in Fig. 11. Regarding fuel properties, the addition of charcoal improved the heating value, S content, atomic ratios of H/C and O/C, and combustion index. There was no distinct influence on the fuel properties, when small amounts of bio-tar were added to the blend CS55. Regarding the co-densification characteristics, the bio-tar addition improved the compaction strength, drop resistance, and specific energy, although some negative effects were observed when the densification temperature was increased from 20 to 50 °C. Regarding the pollutant emission characteristics, the bio-tar addition had a distinctly negative influence on TSP and VOC emissions; however, the adverse influence could be effectively weakened by further adding hydrated lime. Overall, the charcoal-based briquettes prepared with bio-tar as binder are technically feasible.

From the viewpoint of application chain, approximately 1.5 billion tons of agroforestry residues are produced annually in China [1]. Some of these resources could be converted to fuel gas, char, bio-tar, and vinegar-like fractions using slow pyrolysis technology. Fuel gas is mainly used as coking energy, and the vinegar-like fraction is purified and used as a pesticide. The remaining char and bio-tar are utilized as heating fuel in rural China with the technical route provided by this investigation. The Chinese government proposed the implementation of projects for clean energy production via biomass pyrolysis technology in the main grain-producing provinces (districts) of North China [47]. Therefore, the promotion and application of new charcoal-based briquettes

are timely and fulfill the actual requirements in China.

Fig. 11. Applied chain and technical chain of the charcoal-based briquettes

4. Conclusion

Herein, charcoal-based briquettes prepared using bio-tar as a binder were proposed as a substitute for conventional coal for heating in rural China. The fuel properties of the blends of charcoal, semi-coke, and bio-tar and the co-densification and pollutant emission characteristics of the proposed charcoal-based briquettes were determined. The addition of charcoal improved the heating value and combustion index of the blends. The combustion characteristics of blend CS55 could be improved from $7.73 \times$

10⁻¹² (only semi-coke) to 16.92 × 10⁻¹² K⁻³min⁻². The co-densification test indicated that bio-tar could enhance the physical stability of charcoal-based briquettes. The strength compaction and drop resistance increased by 8.6% and 13.9%, respectively, when the bio-tar addition was increased from 0 to 9 wt% at the densification temperature of 20 °C. However, some negative influences were found, when the densification temperature was increased from 20 to 50 °C. Pollutant emissions characteristics were monitored during ignition, stable combustion, and burnout phases of the stove. The bio-tar addition had distinct negative influences, increasing the total suspended particle and VOC emissions. However, this problem was easily overcome by the addition of common additive (hydrated lime) at 3 wt%

Acknowledgements

The study benefited from the technical support provided by the Key Laboratory of Energy Resource Utilization from Agriculture Residue, MARA, China. The authors gratefully appreciate the research funding provided by the China Agriculture Research System [Grant number: CARS-02] and the National Key Research and Development Project [Grant number:].

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668 **COMPETING INTERESTS**

669 The authors declare that they have no competing interests.