



Full Length Article

Control of pollutants in the combustion of biomass pellets prepared with coal tar residue as a binder



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ABSTRACT

This study involved comprehensively investigating pollutant emissions and control methods of NO, SO₂, polycyclic aromatic hydrocarbons (PAHs) and dioxins (PCDD/Fs) in the combustion of biomass pellets prepared with hazardous waste of coal tar residue (CTR) as a binder. The NO emissions from moso bamboo pellets and SO₂ emissions from wheat straw pellets corresponded to the highest emissions among the three biomass pellets. In contrast, NO and SO₂ emissions from the sawdust pellet corresponded to the lowest emissions among three biomass pellets. When the biomass pellets were prepared with 30 wt% CTR binder, the pollutant emissions of NO, SO₂, PAHs and PCDD/Fs were significantly lower than those in the direct combustion of only CTR. The SO₂ emissions of wheat straw pellets with 30 wt% CTR binder gradually increased when the furnace temperature increased from 800 °C to 1300 °C. Conversely, the NO emission gradually decreased because more volatiles derived from biomass pellets locally generated stronger reducing atmospheres at a higher temperature to restrict the NO production. The SO₂ emission of wheat straw pellets with 30 wt% CTR binder decreased by 55.6%–71.0% when limestone was added with a molar ratio of Ca/S at 2, while emission factors of PAHs and total I-TEQ of PCDD/Fs decreased by 13.3% and 59.9%, respectively, at 1200 °C.

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1. Introduction

Coal tar residue (CTR) is a type of toxic hazardous industrial solid waste generated in the process of coal gasification or coking and is composed of heavy tar oil, pulverised coal and other solid particles entrained in gases produced by coal pyrolysis. Previous studies indicate that CTR contains 2–6 aromatic rings, some of which are carcinogenic polycyclic aromatic hydrocarbons (PAHs), such as naphthalene and benzo[a]pyrene, and its direct discharge would cause serious pollution to soil, groundwater and the surrounding air [1–4]. Recently, the coal–chemical industry of China is characterised by continuous expansion. However, most companies ignore the treatment and utilisation of CTR in the production process, thereby disregarding serious environmental pollution resulting from the direct combustion of CTR [3]. Additionally, CTR is a secondary energy source with high calorific value and should be handled with care. Several researchers have carried out the treatment of CTR, which is typically classified into the following two models: 1) separating tar and solid particles by physical or chemical methods and then using and/or treating them

further; and 2) directly using CTR as a raw material for coal blending, fuel and resources [2,3]. Shi et al. [5] utilised CTR as an additive for coke making and as an adhesive for fabricating briquettes. Good experimental results were obtained in both cases. Specifically, CTR has high viscosity at room temperature and high calorific value, and thus the use of CTR as a binder to prepare biomass pellets and then utilising a high temperature combustion process to realise clean disposal of CTR constitutes a very meaningful topic to study.

Biomass is a type of biofuel that is advantageous because it is clean, sustainable and carbon-neutral. However, biomass is not directly utilised because of its low energy density, unstable combustion rate, high particle emissions and difficulties in storage and transportation [6,7]. Pelletisation is considered as an effective method of improving storage and transport properties of biomass. Generally, in the production of densified biomass pellets, high pressures (70–250 MPa) and hot-pressing temperatures (100–250 °C) are employed to increase durability and soften the point components to increase the attractive forces between small biomass particles. However, pelletising at high pressures and hot-pressing temperatures consumes a significant amount of energy. Therefore, specific binders are added to improve the mechanical durability of biomass pellets under conditions of

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ambient temperature and low pressure [8–14]. Hu et al. [13] added four binders (i.e. lignin, starch, calcium hydroxide and sodium hydroxide) to prepare rice husk char pellets. The results indicated that lignin and $\text{Ca}(\text{OH})_2$ pellets exhibited considerably low compression energy consumption and moisture uptake, enhanced the mechanical strength and promoted combustion performance. Lu et al. [14] investigated the use of crude glycerol, bentonite, ligno-sulphonate and softwood residues as binders in biomass fuel pellets used for thermochemical conversion to enhance the quality of the pellets for transportation and storage. The results showed that specific energy consumption in the pelletisation of wheat straw decreased significantly given the addition of ligno-sulphonate, bentonite, wood residues and wood residues pre-treated with crude glycerol. It is technically possible to use CTR as a binder in the preparation of the biomass pellets due to the high viscosity and high calorific value of CTR, although this is rarely investigated by extant literature.

Recent studies examined the combustion, pyrolysis and pollutant emission characteristics of various biomass wastes. A study by Roy et al. [15] revealed that NO_x emissions under steady-state conditions correlated well with nitrogen content of pellets and a minimum was observed in the case of grade 1 wood pellets. However, grass pellets exhibited higher NO_x than that of wood pellets due to high fuel bound N_2 . The share of the NO emissions in the total NO_x corresponded to the range of 95%–97.5% for pellet combustions. Bartocci et al. [16] studied the pyrolysis kinetic parameters for a pellet made with sawdust and glycerol. Through evolved gas analysis it was demonstrated that the thermal degradation of glycerol contained in the pellet can increase hydrogen content in pyrolysis gases. Chen et al. [17] studied the emission characteristics in the combustion of microalgae biomass blended with different oil shale contents by using a tube furnace. The co-combustion of microalgae biomass and oil shale impacted the NO_x and SO_2 emission profiles, and this could be attributed to the inhibitory effects on mass transfer that are caused by the extremely high ash content present in the used oil shale samples. Moroñ et al. [18] investigated the NO_x and SO_2 emission characteristics of coals, biomass and their blends under different oxy-fuel atmospheres. The NO_x and SO_2 emissions were significantly reduced by the addition of biomass to the coal. However, the addition of biomass also increased the NO emissions based on the fuel nitrogen content. Qiu et al. [19] investigated flue gas emissions and particularly pollutants including CO , NO_x and particle emissions of a domestic biomass boiler under various operating conditions. If primary and secondary air supplies for the biomass furnace are provided by factory-set openings and vent tubes, then the measured flue gas emissions correspond to NO_x 54.92 ppmv, CO_2 11.98 vol%, CO 0.24 vol%, O_2 8.19 vol% and $\text{PM}_{0.1-10}$ concentrations of $72.7 \text{ mg}/\text{Nm}^3$ by burning wood pellets. Shen et al. [20] measured the emission factors of oPAHs (EF_{oPAHs}) for two types of pellets composed of corn straw and pine wood. With respect to the pellets composed of pine wood, EF_{oPAHs} corresponded to $77.7 \pm 49.4 \text{ } \mu\text{g kg}^{-1}$ and $348 \pm 305 \text{ } \mu\text{g kg}^{-1}$ in combustion mode I (without secondary side air supply) and mode II (with secondary air supply), respectively. With respect to the pellets composed of corn straw, EF_{oPAHs} corresponded to $189 \pm 118 \text{ } \mu\text{g kg}^{-1}$ and $396 \pm 387 \text{ } \mu\text{g kg}^{-1}$ in modes I and II, respectively. Large variations were observed in the two types of pellets and two combustion modes due to the differences in fuel properties and combustion conditions.

The direct combustion of CTR causes serious pollution in the environment. Nevertheless, the use of CTR with high viscosity and high calorific value as a binder to prepare biomass pellets can improve the mechanical strength and calorific value of the pellets and reduce energy consumption involved in the granulation process. However, there is a paucity of research examining pollutant emission performance of biomass pellets with CTR as a binder.

Therefore, the present study involves a comprehensive investigation of the pollutant emissions and control methods of NO , SO_2 , PAHs and dioxins (PCDD/Fs) in the combustion of biomass pellets prepared with the hazardous waste of CTR as a binder.

2. Materials and methods

2.1. Properties of CTR

Gas chromatography–mass spectrometry (GC–MS) (Finnigan, San Jose, CA) was used to investigate the components of CTR. Chromatographic separation was performed using a DB-5 ms capillary column ($30 \text{ m} \times 0.25 \text{ mm} \times 25 \text{ } \mu\text{m}$). The inlet temperature was $250 \text{ }^\circ\text{C}$ and the oven temperature was increased from $50 \text{ }^\circ\text{C}$ (held for 1 min) to $300 \text{ }^\circ\text{C}$ (held for 10 min) at a heating rate of $5 \text{ }^\circ\text{C}/\text{min}$. High-purity helium was used as the carrier gas with a flow rate of $1.7 \text{ ml}/\text{min}$. The mass spectrometer was operated in electro-spray ionisation mode at 70 eV and the ion source temperature was $250 \text{ }^\circ\text{C}$.

2.2. Preparation of the biomass pellets

In the present study, wheat straw, sawdust and moso bamboo are used as raw materials. Additionally, CTR produced from coal gasification is used as an additional binder to prepare biomass pellets. Table 2 presents the proximate and ultimate analyses of the

Table 1
Detailed organic compositions of coal tar residue detected on GC–MS.

Residence time	Organic compositions	Molecular formulas	Relative content (%)
6.1645	3,4-Dimethylpyridine	$\text{C}_7\text{H}_9\text{N}$	0.6794
6.7143	Mesitylene	C_9H_{12}	0.7913
6.7919	Phenol	$\text{C}_6\text{H}_6\text{O}$	1.1943
6.9407	Benzonitrile	$\text{C}_7\text{H}_5\text{N}$	0.5363
7.096	Benzofuran	$\text{C}_8\text{H}_6\text{O}$	1.9479
7.4259	1,2,3-Trimethylbenzene	C_9H_{12}	1.1299
7.62	Indan	C_9H_{10}	5.8753
7.7493	Indene	C_9H_8	5.8614
8.0339	4-Methyl-phenol	$\text{C}_9\text{H}_8\text{O}$	2.4122
8.4673	Cinnamic aldehyde	$\text{C}_9\text{H}_8\text{O}$	1.0104
8.8813	2,4-Dimethylphenol	$\text{C}_9\text{H}_{10}\text{O}$	1.7509
9.0818	2-Ethyl-phenol	$\text{C}_8\text{H}_{10}\text{O}$	0.937
9.1077	3,5-Dimethylphenol	$\text{C}_8\text{H}_{10}\text{O}$	0.8241
9.4312	Naphthalene	C_{10}H_8	6.0464
9.5023	Benzo[thiophene]	$\text{C}_8\text{H}_6\text{S}$	0.1512
9.981	Quinoline	$\text{C}_8\text{H}_7\text{N}$	0.112
10.5567	2-Methyl-naphthalene	$\text{C}_{11}\text{H}_{10}$	1.3426
10.6602	2-Methyl-quinoline	$\text{C}_{10}\text{H}_9\text{N}$	0.1609
10.712	2-Methyl-naphthalene	$\text{C}_{11}\text{H}_{10}$	0.9731
10.8737	6-Fluoro-2-methyl-quinoline	$\text{C}_{10}\text{H}_8\text{FN}$	0.0827
11.1001	7-Methyl-quinoline	$\text{C}_{10}\text{H}_9\text{N}$	0.2116
11.6952	1,3-Dimethyl-naphthalene	$\text{C}_{12}\text{H}_{12}$	0.7564
11.8828	1,4-Dimethyl-naphthalene	$\text{C}_{12}\text{H}_{12}$	0.7002
12.5685	Dibenzofuran	$\text{C}_{12}\text{H}_8\text{O}$	1.2022
13.1248	Fluorene	$\text{C}_{13}\text{H}_{10}$	1.1922
14.1727	2-Methyl-fluorene	$\text{C}_{14}\text{H}_{12}$	0.7134
15.0201	Phenanthrene	$\text{C}_{14}\text{H}_{10}$	1.0232
15.13	Anthracene	$\text{C}_{14}\text{H}_{10}$	1.0591
15.5246	2,5-Dichloro-3-acetyl-thiophene	$\text{C}_6\text{H}_4\text{Cl}_2\text{OS}$	0.2752
15.8739	Nonadecane	$\text{C}_{19}\text{H}_{40}$	0.9057
16.8442	Eicosyl	$\text{C}_{20}\text{H}_{42}$	0.9839
17.7369	Fluoranthene	$\text{C}_{16}\text{H}_{10}$	0.7296
17.7951	<i>n</i> -Heneicosane	$\text{C}_{21}\text{H}_{44}$	0.9982
18.2609	Pyrene	$\text{C}_{16}\text{H}_{10}$	0.9431
18.7137	Docosane	$\text{C}_{22}\text{H}_{46}$	1.094
19.0436	Retene	$\text{C}_{18}\text{H}_{18}$	1.2984
19.5934	Eicosane	$\text{C}_{20}\text{H}_{42}$	1.2579
20.4343	Lignocerane	$\text{C}_{24}\text{H}_{50}$	1.3081
21.2494	Pentacosane	$\text{C}_{25}\text{H}_{52}$	1.3304
23.3775	Erucamide	$\text{C}_{22}\text{H}_{43}\text{NO}$	0.3406

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