Waste Management 77 (2018) 59–66

Contents lists available at ScienceDirect

Waste Management

journal homepage: www.elsevier.com/locate/wasman

Emission characteristics of a pyrolysis-combustion system for the co-production of biochar and bioenergy from agricultural wastes

Lewis Dunnigan [a], Benjamin J. Morton [a], Peter J. Ashman [a], Xiangping Zhang [b], Chi Wai Kwong [a,]*

[a] School of Chemical Engineering, The University of Adelaide, Adelaide, SA 5005, Australia
[b] Institute of Process Engineering, Chinese Academy of Sciences, Beijing, China

ARTICLE INFO

Article history:
Received 13 December 2017
Revised 29 April 2018
Accepted 2 May 2018

Keywords:
Pyrolysis
Biochar
Bioenergy
PM
Gaseous
Emissions

ABSTRACT

The co-production of biochar and bioenergy using pyrolysis-combustion processes can potentially minimize the emission problems associated with conventional methods of agricultural by-product disposal. This approach also provides significant added-value potential through biochar application to soil. Despite these advantages, variations in biomass composition, including sulfur, nitrogen, ash, and volatile matter (VM) content, may significantly influence both the biochar quality and the emissions of harmful particulate matter (PM) and gaseous pollutants (SO_2, H_2S, NO_2, NO). Using a laboratory-scale continuous pyrolysis-combustion facility, the influence of biomass composition (rice husk and grape pruning) and volatile production (pyrolysis) temperature (400–800 °C) on the biochar properties and emissions during combustion of the raw pyrolysis volatiles were evaluated. Utilization of grape pruning resulted in higher energy-based yields of PM_{10} than the rice husk, the majority of which consisted of the $PM_{1.1}$ fraction due to the elevated pyrogas content of the volatiles. The PM emissions were found to be independent of the feedstock ash content due to its retainment in the biochar. Greater volatilization of biomass sulfur and nitrogen during pyrolysis at higher temperatures resulted in higher yields of sulfurous and nitrogenous gaseous pollutants. The energy-based yields of NO and NO_2 were found to increase by 16% and 50% for rice husk and 21% and 189% for grape pruning respectively between 400 and 800 °C. The same trend was also observed for the emissions of H_2S and SO_2 for both feedstocks.

1. Introduction

Agricultural wastes generated within the horticulture industry present numerous challenging and financially burdensome disposal problems for food producers. At present, many of these wastes have little or no value and do not provide an additional revenue stream for producers. Rice husk is one such abundant waste, of which approximately 822 million tonnes are produced annually worldwide (Naqvi et al., 2014). Grape pruning, a common woody by-product from the growing of grape vines, is also an agricultural waste with few options for recycling. Current utilization rates for both feedstocks are low, with rice husk typically openly burnt or buried following detachment of the chaff from the grain after cultivation (Zhang et al., 2015). Grape pruning is typically disposed of by burning or landfilling, or on very few occasions used for composting (Liang et al., 2016). These conventional methods of disposal are not only providing little added-value to the wastes, but they are in many cases also resulting in significant emissions of harmful particulate matter (PM) and gaseous pollutants (SO_2, H_2S, NO_2, NO) when openly burned (Hays et al., 2005). This is of significant concern, as PM_{10} and $PM_{2.5}$ emissions are currently one of the primary causes of premature mortality in developing countries (Barron and Torero, 2017), while the negative health consequences of exposure to harmful gaseous pollutants is well documented (Zhang and Smith, 2007). One potential option for providing both added-value to these agricultural wastes and a method for reducing the harmful emissions associated with disposal is a combined pyrolysis and combustion process (hereafter called ‘pyrolysis-combustion’) for the co-generation of biochar and bioenergy.

Biochar, the solid carbonaceous product of pyrolysis, can provide value adding potential to the agricultural wastes given its ability to offset carbon emissions by long-term carbon sequestration with additional agricultural benefits (Taherymoosavi et al., 2017). The co-generation of energy through utilization of the heat liberated from combustion of the raw pyrolysis volatiles can also potentially reduce on-site energy costs. Alternative approaches for the generation of bioenergy from agricultural wastes, such as direct combustion and gasification (Xin et al., 2017), have often

* Corresponding author.
E-mail address: philip.kwong@adelaide.edu.au (C.W. Kwong).

https://doi.org/10.1016/j.wasman.2018.05.004
0956-053X/

been hampered by the wide-variability in the fuel qualities possessed by such feedstocks, including fuel ash contents (Gilbe et al., 2008). Pyrolysis-combustion processes have previously been shown to be tolerant of wide-ranging feedstock ash contents, as the majority of the biomass-bound ash is retained in the solid pyrolysis product (biochar) prior to combustion (Claoston et al., 2014; Dunnigan et al., 2018; Hung et al., 2017). The diversity of agricultural wastes also implies that attempts to increase utilization rates will result in broad variations in biomass properties, such as sulfur and nitrogen content. The volatilization and subsequent oxidation of these species has been shown to strongly influence the emission of gaseous pollutants (including SO_2, H_2S, NO_2, NO) during direct combustion (Moron and Rybak, 2015; Reddy and Venkataraman, 2002). Variations in the volatile matter (VM) content of feedstocks are also of concern, as this affects not only the higher heating value (HHV) for energy generation, but may also promote the formation of sub-micron PM (Mitchell et al., 2016). Fine particle emissions are considered a significant health concern as they cannot be cleared readily from the lung once inhaled (Salvi, 2007). Therefore, despite it being well documented that fuel sulfur, nitrogen, ash, and VM content strongly influences the yields of both gaseous and PM emissions during direct combustion, there is currently no information available regarding their influence on the emissions from the pyrolysis-combustion process.

In order to examine the influence of biomass composition on the emissions associated with waste utilization through the pyrolysis-combustion process, two abundant agricultural wastes with significantly different compositions were compared: rice husk, previously investigated by the authors (Dunnigan et al., 2018) and grape pruning. Grape pruning is considered more representative of the majority of biomass types, with low ash and sulfur contents, as well as a high VM and nitrogen content (Liang et al., 2016; Marshall et al., 2017; Nasser et al., 2014). Rice husk is abundant, but less typical of the majority of biomass, as it has high ash and sulfur content, along with low VM and nitrogen content (Claoston et al., 2014). Conventionally, the condensable (bio-oil) and non-condensable (pyrogas) fractions are separated from the raw (untreated) pyrolysis volatiles generated from biomass pyrolysis (Alper et al., 2010). However, it has been suggested that separation is not the most suitable approach for the distributed utilization of agricultural wastes as bio-oil contributes significantly to the heating value of the raw pyrolysis volatiles, especially at lower volatile production temperatures (<500 °C) (Dunnigan et al., 2018). Therefore, co-combustion of the bio-oil and pyrogas (i.e. combustion of raw pyrolysis volatiles) is a viable approach for distributed generation as it avoids the need for separation and energy intensive bio-oil upgrading processes (Mortensen et al., 2011). The composition of the raw pyrolysis volatiles are not only dependent on the feedstock used, but are also influenced by the volatile production temperature. These factors can therefore influence the emissions from the process associated with energy generation. High volatile production temperatures can lead to an increase in volatilization of feedstock sulfur and nitrogen (Claoston et al., 2014), as well as variations in the ratio of pyrogas and bio-oil in the raw pyrolysis volatiles (Dunnigan et al., 2018). To date there have been no studies investigating the role of volatile production temperature on the gaseous emissions from a pyrolysis-combustion process utilizing agricultural wastes. Therefore, the objectives of the study are to determine (1) the yields and HHV of the primary pyrolysis products from the grape pruning and compare them to those from the rice husk (Dunnigan et al., 2018), (2) the influence of feedstock ash and VM content on the energy-based yields of PM, and (3) the influence of biomass sulfur and nitrogen content, and volatile production temperature, on the energy-based yields of NO, NO_2, H_2S, and SO_2.

2. Materials and methods

2.1. Material characterizations

The grape pruning used in this study was grown in Urrbrae, South Australia, while the rice husk used was supplied by Beerbelly Brewing Equipment (Pooraka, South Australia). Both feedstocks were first ground in a rotary mill and then sieved to 420–500 μm. Drying was done in an oven at 105 °C for a minimum of 15 h, following ASTM D4442. The ultimate analysis of the grape pruning and biochar (produced following the method outlined in Section 2.2) was carried out using an Elemental Analyser (PerkinElmer, 2400 Series II CHNS/O) following ASTM D5373, with the oxygen content calculated by difference. The proximate analysis of the samples was carried out using a thermogravimetric analyser (TGA) (METTLER TOLEDO, TGA/DSC 2 STARe System) following ASTM D7582 with modifications. Approximately 65 mg of sample was loaded into the TGA and heated to 950 °C at 30 °C/min under N_2. The sample was then held until a constant weight was achieved, at which point the atmosphere was switched to oxygen to determine the ash content. The HHV of the feedstock and biochar were calculated using the Boie equation (Boie, 1953). The characterization results of the grape pruning were compared to those previously presented for rice husk in Dunnigan et al. (2018).

2.2. Lab-scale pyrolysis-combustion process

A schematic diagram of the experimental pyrolysis-combustion system is shown in Fig. 1. The dried and ground biomass samples were first loaded into a hopper and fed into the main screw at a rate of 1.3 g/min. The main screw reactor (length ≈ 1 m with residence time of 16 min) was heated externally with two electrical heaters to 400, 500, 600, 700, or 800 °C. These temperatures are referred to as "pyrolysis temperature" when discussing the properties of the biochar and bio-oil, and "volatile production temperature" when discussing the PM and gaseous emissions generated from combustion of the raw pyrolysis volatiles. The solid biochar was collected from a char collection pot at the exit of the main screw reactor. The yield of biochar was calculated using the mass ratio of biochar collected and the mass of feed (on a dry basis). Nitrogen carrier gas was supplied at two different gas inlets to purge the system. The first of these was located on the top of the hopper (0.25 L/min) and the second was above the char collection pot (0.1 L/min). The raw pyrolysis volatiles generated during the pyrolysis reaction were directed into a burner where they were then mixed with the HEPA filtered combustion air. The raw pyrolysis volatiles produced between 400 and 800 °C were combusted at 850 °C. The heat required for auto-ignition of the volatile/air mixture was provided by a vertical 3-zone tube furnace (Carbolite®, GVC 12/1050). This combustion temperature (850 °C) was fixed through the combustion experiments and was chosen as a typical temperature found in small-scale combustion systems (Obernberger, 1998). The flue gas generated from the combustion reaction was contained within a quartz tube (1.5 m height × 45 mm I.D.) where it was diverted into an air dilution tunnel (ADT) situated above the combustion furnace and mixed with HEPA filtered air.

2.3. Pyrogas and bio-oil sampling

The pyrogas (non-condensable) fraction of the raw pyrolysis volatiles was sampled by attaching a Teflon gas sampling bag to the burner outlet via a sampling pipe. Once the bag was full, the pyrogas was then passed through a coalescing filter before