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Codensification of *Eucommia ulmoides* Oliver stem with pyrolysis oil and char for solid biofuel: An optimization and characterization study

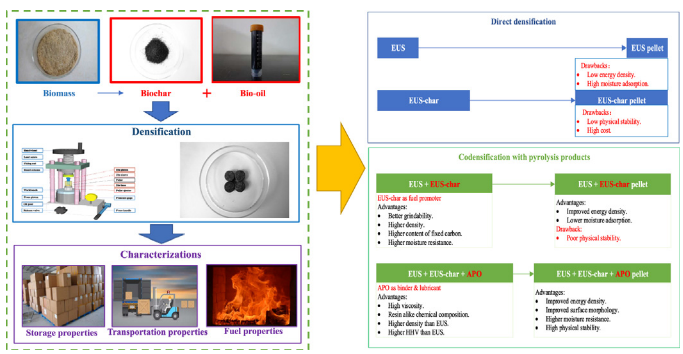
Kang Kang[a,*], Mingqiang Zhu[a], Guotao Sun[a], Ling Qiu[a], Xiaohui Guo[a], Venkatesh Meda[b], Runcang Sun[c]

[a] College of Mechanical and Electronic Engineering, Northwest A&F University, Yangling, Shaanxi 712100, China
[b] Department of Chemical and Biological Engineering, University of Saskatchewan, 57 Campus Drive, Saskatoon, SK S7N 5A9, Canada
[c] Beijing Key Laboratory of Lignocellulosic Chemistry, Beijing Forestry University, Beijing 100083, China

HIGHLIGHTS

- *Eucommia ulmoides* Oliver stem was codensified with pyrolysis oil and char.
- Densification parameters and pellet formula were optimized.
- *Eucommia ulmoides* Oliver char is an effective fuel promoter.
- Apple tree branch pyrolysis oil functioned as an effective binder.
- Codensification generated durable pellets with promising fuel properties.

GRAPHICAL ABSTRACT



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ABSTRACT

This paper presents the results regarding the parametric optimization and characterizations on codensification of *Eucommia ulmoides* Oliver stem using biochar and bio-oil as additives. The results reveal that the relative importance of different parameters contributing to the pellet physical stability follows the order of: particle size > bio-oil content > biochar content > pressure > bio-oil type, and smaller particles size (0.1–0.3 mm) is critical for the formation of durable pellets. The biochar functioned as the fuel promoter and improved the higher heating value and energy density, however, caused poor physical stability by secondary size reduction during densification. The apple tree branch pyrolysis oil acted as an effective binder, improved the surface morphology and enabled strong interlocking of the particles, therefore, enhanced the physical stability. Above all, codensification of *Eucommia ulmoides* Oliver stem with biochar and bio oil generated pellets with excellent physical stability and moisture resistance. The pellets also showed better fuel characteristics than the *Eucommia ulmoides* Oliver stem pellets, and the cost is significantly lower than the char pellets. With comprehensiveness, these results can guide the future industrial implementation of codensification technology for the production of clean, sustainable, and efficient fuel with low cost.

* Corresponding author.
E-mail address: kknwafu@hotmail.com (K. Kang).

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1. Introduction

Fuels produced from biomass is attracting growing interest because they can reduce the dependence on the fossil fuels, their effect on the atmosphere is more carbon-neutral, and they are less toxic in the environment [1,2]. In industry, biomass cannot be directly utilised due to its poor energy density, unstable combustion performance, high particle emissions, and difficulties in storage and transportation [3]. Densified solid bio-fuel produced via the densification of lignocellulosic biomass is highly practical for industrial implementation due to its advantages in reducing the cost of handling, transportation, and storage in the biomass supply chain [4,5]. In China, the dramatic growth of the solid biofuel market started since 2007, with predicted annual utilization mass reaching 50 million ton by 2020 [6]. In 2010, the collectible output of agricultural waste in China was 711 million ton, and about 280 million ton per year is burned in the field, causing severe issues of energy lose, air contamination and drives the demand for the fuel conversion [7,8]. Biomass fuel pellets play a critical role in the energy sector as they are widely utilized for power generation, residential heating appliances such as boilers and furnaces, and showed huge potential in the synthesis of high value chemicals and liquid fuels [9,10]. Although novel technologies for biomass conversion are under development, densification into solid fuel will still be one of the most practical options in constructing the sustainable energy system due to dramatic advantage in terms of process simplicity and cost-effectiveness.

Pyrolysis has been extensively studied as its capability for converting a range of lignocellulosic biomass into renewable energy [11]. Pyrolysis refers to the thermal decomposition in the absence of oxygen, which converts lignocellulosic biomass into products including biochar, bio-oil, and syngas [12]. The direct utilization of all pyrolysis products as low grade fuel is not economically feasible and the upgrading is still quite expensive and difficult [13]. Specifically, the poor fuel characteristics, high viscosity, and chemical complexity created a significant gap between the bio-oils and conventional liquid fuels [14]. Whereas for biochar, despite its higher hydrophobicity, grindabilty, and heating value than the biomass precursor, the main issues for fuel utilization lies in the low density and poor physical strength. The poor physical strength is due to the lack of chemically bonded water and low-melting point compounds which can function as natural binders during typical biomass densification [15].

Based on the issues of raw biomass and its pyrolysis products, the codensification process is a viable route for converting these materials simultaneously into sustainable fuel with satisfying performance and low cost. The basic concept of codensification is to make full use of distinct properties and the complementary effects of different materials, to minimize undesirable properties and maximize the cost-effectiveness of the process and product. In the case of codensification, the raw biomass, which is abundant and cheaper, can be used as the main feedstock, whereas the biochar and bio-oil, due to higher cost, could be used as additives to enhance the performance of the pellet. So far, codensification of biomass with various materials have been carried out. For example, mix with bean cakes, polymer plastic, carboxymethyl cellulose, lignin, starch, calcium hydroxide, sodium hydroxide, and bio oil, etc., as the binders, to improve the physical stability of the pellets [15–20]. In addition, with sewage sludge, torrefied agriculture residues and biomass, and carbonized wood char were used as auxiliary fuel to achieve better combustion characteristics [21–26].

However, very little information exists regarding the individual effect as well as interaction effects of biomass, char, and bio oil on the quality of the prepared pellets. In addition, there is a lack of knowledge regarding the parametric optimization of process conditions including pressure, feedstock particle size, and formulation factors including biochar content, bio–oil content and type for the preparation of strong pellets with promising fuel properties.

In this study, we describe the codensification of *Eucommia ulmoides* Oliver stem with both biochar and bio-oil. The main objectives of this study are (1) revealing the unique roles of biochar and bio-oil in the formation of fuels pellets via systematic characterizations including physicochemical properties, surface morphology, moisture adsorption behavior, and combustion characteristics, and evaluate the feasibility of utilizing pyrolysis products as additives to improve the quality of the solid biofuel; (2) understanding the individual as well as complementary effects of densification conditions and the pellet formulations on the physical stability of the pellets, and achieve the maximum stability through the parametric optimization.

In this study, two biomass precursors were selected for producing pyrolysis products and densified pellets, including stem of *Eucommia ulmoides* Oliver (Gutta-percha tree) and branche of *Malus pumila Mill* (apple tree). These two materials are abundant agriculture wastes in the northwest part of China, urgently need to be recycled, and represent waste from the cultivation of Chinese medicinal crop and orchard, respectively. Specifically, for *Eucommia ulmoides* Oliver, the annual production of stems can reach 18.0–22.5 tons/hectare, and the planting area is increasing due to the high value of secondary metabolites in the leaves and barks, however, the utilization of the stems has rarely been reported [27]. Similar to the case of *Malus pumila*, after fruit harvesting, huge quantity of branches was left as waste, causing severe issues of space occupation and energy dissipation [28].

Based on the above, one novelty of this study is the raw material used for the pellet production, as the densification characteristics of *Eucommia ulmoides* Oliver stem, combined with its pyrolysis char and the bio-oil produced from apple tree branch have rarely been reported. Since these materials are all abundant wastes, the simultaneous utilization of them via codensification has great potencial in producing high quality soild fuel with low cost. Another contribution of this paper lies in its comprehensiveness. Specifically, through the Taguchi optimization, the optimum combination of pelleting parameters including pressure, feedstock particle size, and formulation factors including biochar content, bio-oil content and type could be established, which is critical for the future industrialization of the process. In addition, via systematic characterizations on the physicochemical properties, surface morphology, moisture adsorption behavior, and combustion characteristics of both the feedstocks and the pellets, deeper understanding on the unique roles of different pellet components can be achieved.

2. Materials and methods

2.1. Materials

The stems of *Eucommia ulmoides* Oliver (EUS) were harvested from an experimental field at the Northwest A&F University, China. The EUS pyrolysis oil (EUO) and EUS pyrolysis char (EUS-char) were simultaneously prepared using the EUS samples mentioned above with a lab-scale pyrolysis reactor located in the college of forestry at Northwest A&F University. Apple tree branch pyrolysis oil (APO) were provided by Yixin Biological Energy Science and Technology Development Co., Ltd. (Shaanxi, China). For the preparation of both APO and EUO samples, the slow pyrolysis process was applied. Specifically, the reactor was heated to 500 °C with a heating rate of 2 °C/min and held for 1 h, during which a nitrogen flow of 100 mL/min was applied. Basic properties of the APO and EUO samples are provided in Table S1 [29]. The water used in this study was produced by an ultrapure water generator (Molecular, Shanghai, China).

2.2. Material pre-treatment before densification

Before pelleting, the solid samples were treated with a super centrifugal grinder (ZM 200, Retsch, Germany) to reduce and unify the particle size to less than 0.9 mm. The particle size distribution of the EUS and EUS-char were determined before the densification. For each test, a total amount of 1500 g of sample was passed through a series of steel sieves with different mesh sizes to obtain the particle size

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