



Hydrothermal carbonization of agricultural residues: A case study of the farm residues -based biogas plants

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ABSTRACT

Hydrothermal carbonization (HTC) of biomass is a promising method to produce carbonaceous materials. The work presented in this article addresses the application of hydrothermal carbonization (HTC) to produce a solid fuel named HTC-Biochar, whose characteristics are comparable to lignite coal. Biogas sludge (SD), maize silage (MS), and barley silage (BS) as a substrates were hydrothermally carbonized in a 1.5 L batch reactor at 200 °C for 6 h. The effect of mixing ratios of different substrates on HTC was investigated. Chemical compositions and combustion characteristics of hydro-chars obtained from mono- and co-carbonization were evaluated. Result showed that HTC increased carbon contents and higher heating values (HHV) by 1.4–14.4% and 13–36%, respectively. The evolution of the H/C and O/C atomic ratios indicated that dehydration and decarboxylation occurred during hydrothermal carbonization for all samples. Furthermore, a significant synergistic enhancement was observed for HHV and carbon content. A mixing ratio of 1:1 for BS and SD showed the best performance for co-HTC. In summary, hydrothermal co-carbonization is a promising strategy to tailor high-performance hydro-char for energy applications. © 2018 Published by Elsevier B.V. on behalf of KeAi Communications Co., Ltd. This is an open access article under the CC BY-NC-ND license (<http://creativecommons.org/licenses/by-nc-nd/4.0/>).

Introduction

According to the European Technology Platform for Zero Emission Fossil Fuel Power Plants [1], policy makers, industries, and any committee responsible for fuel and energy production must follow guidelines and apply environmentally friendly techniques to control emissions and secure energy supplies sustainably. Thus, researchers have begun to concentrate efforts on the use of natural materials instead of fossil fuels, which had been successful in 20th century [2]. Since biomass has a high potential for energy production, it would support sustainable development if solids, liquids, and gases released during controlled combustion were recovered, treated, and refined [3].

Moreover, the utilization of anaerobic treatment of biomass for biogas production is particularly promising. Several countries have recommended biogas as the preferred option for a renewable and sustainable energy source [4]. Furthermore, use of biogas sludge as fertilizer on arable land can provide nitrogen (N), phosphor (P), and other nutrients, thereby recycling nutrients for agricultural production, which is consistent with development of a circular economy and sustaining soil organic carbon (SOC) concentrations [5]. On the other hand, it is worth noting that most plants cannot uptake and use all of the applied N. Therefore, N fertilizers, as well

as direct application of large amount of sludge on farmlands, may lead to N emissions, with residual N potentially leaching into groundwater as nitrate [6].

Optimization of biogas sludge management is needed. In this context, the treatment of biogas sludge for biochar production could be useful. Recently, the conversion of waste biomass into biochar using hydrothermal carbonization (HTC) (yielding hydro-char) attracted widespread attention. HTC is a thermo-chemical process that takes place in subcritical water under a moderate temperature and pressure, which improves the properties of raw biomass for subsequent use [3,7,8]. A major advantage is the fact that the hydro-char is hydrophobic in comparison to the feedstocks and therefore the water content can be significantly reduced [9]. During HTC, both the oxygen and hydrogen content of the feed (described by the molecular O/C and H/C ratio) are lowered by five primary reactions, namely hydrolysis, dehydration, decarboxylation, polymerization, and aromatization [10]. This method was invented to derive the greatest advantage from by-products for soil amendment and power production. Several studies [3,7,8,10,11] have been done to improve HTC, investigating different types of biological input materials for HTC, changing parameters and lab conditions, and using different reactors and autoclaves, in order to reach a better quality hydro-char that is more effective and is more similar to lignite. Using hydro-char as a renewable energy source would have worldwide environmental benefits. Furthermore, previous studies indicated that hydro-char from lignocellulosic biomass via HTC

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had a high hydro-char yield and was rich in oxygen-containing functional groups [12]. In contrast, the hydro-char obtained from biogas sludge had a relatively low hydro-char yield. But other, now beneficial outcomes are expected, because biogas sludge has a high nutrient level compared to lignocellulosic biomass, including N, P, and potassium [13]. Another benefit of hydrothermal co-carbonization is that the functionality of hydro-char could be tailored based on the type of feedstocks and mixing ratios used. For example, woody biomass is rich in organic components, especially lignin that could provide a skeleton for the growth of small particles, and nitrogen in biogas sludge could help in the functionalization of carbon materials.

Recently, two studies have been reported using biogas sludge for HTC [10,13]. However, those studies used different experimental conditions than the work presented here. On the basis of the above considerations, this study aims to explore possible synergistic effects of hydrothermal co-carbonization of biogas sludge (SD), maize silage (MS), and barley silage (BS) on hydro-char production, as compared to mixing the separately carbonized materials.

Material and methods

Raw materials

Three biomass substrates were selected for this study, which was conducted in the laboratory of the Institute of Chemical and Energy Engineering (IVET), University of Natural Resources and Life Sciences, Vienna (BOKU). Two agricultural residues, barley and maize silage (BS and MS), as well as sludge (SD) from a biogas plant were used as substrates. Substrates were collected from a farm-based biogas plant located in the town of Utzenaich, Austria. Barley and maize silage then shredded into pieces of 3–5 cm. Each substrate was stored in glass bottles until HTC experiments were performed.

Table 1a
Experimental Conditions and Substrate Mixtures (All experiments were run with a retention time of 6 h and at a reaction temperature of 200 °C).

Trial	HTC test	Substrate (%)		
		SD	BS	MS
Trial I	T 1	100	–	–
	T 2	–	100	–
	T 3	–	–	100
Trial II	T 4	20	80	–
	T 5	50	50	–
	T 6	20	–	80
	T 7	50	–	50
	T 8	33	34	33

Hydrothermal carbonization procedure

HTC experiments using SD, MS, BS, and mixtures (co-HTC) of those materials were carried out in a 1.5 L batch reactor. All experiments were run with a retention time of 6 h and at a reaction temperature of 200 °C. Comprehensive test campaigns were done at the institute with variation of the process parameters (pressure, temperature, pH, residence time, etc.) which lead to the decision to choose 200 °C for this test series. Although lignin is hardly converted at this temperature it has been chosen to gain the maximum amount of solid product (hydro-char). However, the hydro-char can be easily separated from the water phase when applying these conditions. Eight experiments were carried out, including three with a single substrate and five with mixtures of substrates. In each test with a mixed substrate (T4–T8), SD was always used as one of the contributions, since the goal is to find a waste management for the sludge (Table 1a).

Abbreviations that are used throughout the following sections are summarized with a brief explanation in Table 1b.

For each experiment, the substrate was loaded into the reactor mixed with a water in a ratio of dry mater to water of 1:4. Then the reactor was sealed and purged with nitrogen. The reactor was then heated to the desired temperature of 200 °C for a fixed time of 6 h. After the reaction, the process was stopped and the reactor was allowed to cool naturally.

Analytical methods

Elemental analyses for all samples (pure substrates and hydro-chars) were carried out with a PerkinElmer Element-Analyzer according to EA 1108 CHNS-O, Carlo Erba. All analyses were performed by the Microanalytical Laboratory of the University of Vienna according to standard procedures [14]. Oxygen (O) percentages were calculated as 100% minus the measured values of carbon (C), hydrogen (H), N, and sulphur (S). All tests were performed in triplicate. A modified van-Soest method using the ANKOM A200 Filter Bag Technique (FBT) was used to determine the contents of hemicellulose, cellulose, lignin, and aqueous soluble compounds in solid samples [16].

The heating values of the samples (pure substrates and hydro-chars) were determined experimentally using a bomb calorimeter (IKA C200 Oxygen) at the Institute of Agricultural Engineering, University of Natural Resources and Life Sciences, Vienna. This test was at least done twice on each sample, including raw materials and hydro-chars. The carbon content and the higher heating value (HHV) of the pure feed substrates (T_{1f}–T_{3f}) were measured, whereas these values were calculated for the mixed feeds (T_{4f}–T_{8f}) from the pure substrate values.

Table 1b
Experimental notation.

Notations		
HTC test	Substrate	Hydro-char
T1 = 100% SD	T1 _f = pure feed with SD	T1 _h = hydro-char of T1
T2 = 100% BS	T2 _f = pure feed with BS	T2 _h = hydro-char of T2
T3 = 100% MS	T3 _f = pure feed with MS	T3 _h = hydro-char of T3
T4 = 80% BS + 20% SD	T4 _f = mixed feed for T4	T4 _h = hydro-char of T4
T5 = 50% BS + 50% SD	T5 _f = mixed feed for T5	T5 _h = hydro-char of T5
T6 = 80% MS + 20% SD	T6 _f = mixed feed for T6	T6 _h = hydro-char of T6
T7 = 50% MS + 50% SD	T7 _f = mixed feed for T7	T7 _h = hydro-char of T7
T8 = 34% BS + 33% MS + 33% SD	T8 _f = mixed feed for T8	T8 _h = hydro-char of T8

Note: Properties of the mixed feed streams were calculated from pure substrate properties which were measured. Properties of the hydro-char was measured and compared to calculated values.

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