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Research paper

Mechanical pretreatment of biomass - Part I: Acoustic and hydrodynamic cavitation

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ABSTRACT

Acoustic and hydrodynamic cavitation were examined as suitable mechanical pretreatments for lignocellulosic biomass. Microcrystalline cellulose and lime-treated sugarcane bagasse were subjected to acoustic cavitation, whereas raw and lime-treated sugarcane bagasse were subjected to hydrodynamic cavitation. Acoustic cavitation successfully increased microcrystalline cellulose enzymatic digestibility by 37% compared to no acoustic cavitation treatment; however, there was no significant effect on limetreated sugarcane bagasse. Hydrodynamic cavitation increased the enzymatic digestibility of both raw and lime-treated sugarcane bagasse. Best results were obtained using cavitation treatment of bagasse followed by lime treatment; the 3-d enzymatic digestion increased by 46% when compared to lime treatment only.

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1. Introduction

Increasing interest in alternative energy sources is occurring for multiple reasons: (1) concern over dependence on foreign petroleum; (2) rising oil prices; (3) increasing public interest in environmental preservation; and (4) global warming from the buildup of greenhouse gases, primarily carbon dioxide [1].

Biological conversion of lignocellulosic materials to liquid biofuels addresses each of these concerns. Processes that convert lignocellulose into usable products have been studied for many years [2-6]. These processes are especially attractive because they can convert a variety of feedstocks (e.g., crop wastes, municipal solid waste, sewage sludge, energy crops, woody biomass) into liquid fuels. Additionally, the combustion of these alternative liquid fuels will not contribute to global warming because there is no net addition of carbon dioxide into the atmosphere.

Lignocellulosic biomass is the world's most abundant biological material and is composed primarily of cellulose, hemicellulose, and lignin. Cellulose is a linear, unbranched polymer of β -glucose that

Corresponding author. E-mail address: liang.chao@tamu.edu (C. Liang). provides structure to plants [7,8]. There are two configurations of cellulose: crystalline and amorphous. Amorphous sections are more disordered and allow water to penetrate, thereby increasing susceptibility to enzymatic hydrolysis. In contrast, crystalline sections have hydrogen bonds between the polymers that make it more resistant to enzymatic hydrolysis [9].

Hemicellulose is more readily hydrolyzed than cellulose because of its amorphous structure. It is primarily composed of pentoses (xylose and arabinose) and hexoses (glucose, galactose, and mannose). Hemicellulose polymers are shorter than cellulose. Hemicellulose has a degree of polymerization (DP) of 50-200, whereas cellulose has a DP of 500-15,000.

Lignin is a phenyl-propane polymer that acts as a glue to hold hemicellulose and cellulose together. Its monomers are *p*-coumaryl alcohol, coniferyl alcohol, and sinapyl alcohol. In plant cell walls, cellulose provides the cell structure and strength and is enclosed in a hemicellulose matrix surrounded by lignin, which holds the entire framework together [7].

The crystalline structure of cellulose hinders enzymatic hydrolysis by limiting the number of enzyme adsorption sites [10–12]. Successful pretreatments should open the biomass structure to make it more accessible to enzymes [13]. To increase the extent of biomass digestion, Chang and Holtzapple [14] found the two main





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(a)

contributors are the lignin content and the degree of crystallinity.

Crystallinity measures the relative amounts of the amorphous and crystalline regions of cellulose [11]. It is often described by the crystallinity index (CrI); a higher CrI denotes a more crystalline material. Segal et al. [15] developed the following crystallinity index:

$$CrI = \frac{I_{002} - I_{am}}{I_{002}} \times 100$$
 [1]

where,

 $I_{002} = \text{intensity at } 2\theta \text{ of } 22.5^{\circ}$ $I_{am} = \text{intensity at } 2\theta \text{ of } \sim 18.7^{\circ}$

Crystallinity is measured using an X-ray diffractometer (XRD). High lignin content and high CrI reduce enzymatic digestibility [14,16]; therefore, the most effective pretreatment is one that both delignifies and decrystallizes the biomass.

Lime (Ca(OH)₂) pretreatment is an effective, inexpensive, and safe delignification method. Long-term lime pretreatment involves mixing the biomass and calcium hydroxide into a large pile at low temperatures (25-70 °C). Then, for about one month, air is purged through the pile while the pile is wetted with water. This pre-treatment removes sufficient lignin from the biomass to enhance enzymatic digestion [17]. Short-term lime pretreatment treats biomass for a short time (1-6 h) at elevated temperatures (100-160 °C) with oxygen [18,19] or without oxygen [20,21].

Cellulose decrystallization is typically performed using physical pretreatment methods, such as ball-milling or two-roll milling [22,23]. In this work, cavitation is used as a physical pretreatment to determine if it enhances the enzymatic digestibility of lignocellulose.

Cavitation is the formation, growth, and rapid collapse of gas- or vapor-filled bubbles. There are two main types of cavitation: acoustic and hydrodynamic.

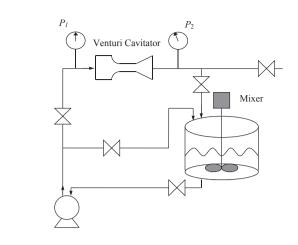
Acoustic cavitation is caused by pressure variations from ultrasonic waves passing through a fluid. Acoustic waves create microcavities where gas bubbles grow and then collapse. The collapse generates "shock waves" that cause mechanical effects, such as particle erosion [24]. This high-power, low-frequency ultrasound is usually used to create a permanent chemical or physical change in a substance [25]. High-power acoustic cavitation is used for cleaning and welding [26]. For this study, a laboratory-scale sonicator was used to decrystallize both microcrystalline cellulose and limetreated biomass via acoustic cavitation.

Acoustic cavitation reduces particle size, likely resulting from erosion caused by collapsing gas bubbles. Additionally, acoustic cavitation lowers crystallinity. Preliminary studies have shown an increase in biomass enzymatic digestibility when cavitation is used as a pretreatment step [27,28].

Hydrodynamic cavitation occurs when a moving fluid encounters a sudden change in velocity that results in a localized pressure drop. For example, this can occur in a venturi (Fig. 1). Cavities form at the throat when the pressure falls below the fluid vapor pressure. The bubbles collapse when pressure is recovered.

Relative to cavitation, there are two main features of bubble dynamics: (1) the maximum bubble size and (2) the distance traveled by the bubble before collapse, i.e., bubble life.

The maximum bubble size defines the cavitation intensity. Bubbles grow at low pressure or high temperature [15]. Larger bubbles implode with a higher intensity and can cause greater effects on a substance than smaller bubbles. For example, a large number of exploding bubbles can alter the structure of biomass. Bubble life is a measure of the active volume where cavitation effects are observed. Generally, bubble life decreases as the region of



(b)

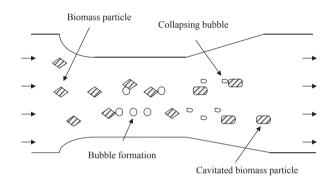


Fig. 1. Scheme of cavitation in a venturi cavitator showing (a) cavitation system including the mixing vessel, centrifugal pump, cavitator, valves, and pressure gauges; and (b) biomass particles inside the cavitator.

active cavitation decreases and vice versa. Fig. 1 shows hydrodynamic cavitation of biomass in a venturi.

Many factors affect cavitation. To create cavitation, highly viscous fluids require higher energy input. A fluid with a low vapor pressure requires more energy to produce cavitation. Also, the size and geometry of the cavitator affects the efficiency [24].

The cavitation number C_v measures the resistance to cavitation. It is a dimensionless parameter and is given by the following equation:

$$C_{\nu} = \frac{P_f - P_{\nu}}{0.5\rho U^2}$$
[2]

where P_f is the downstream pressure, P_v is fluid vapor pressure, ρ is the fluid density, and U is the average velocity near the orifice. A high cavitation number indicates cavitation will not likely occur and vice versa. If cavitation is already occurring, lowering the cavitation number by decreasing the pressure or by increasing the flow rate will increase the cavitation intensity. Raising the cavitation numbers, bubbles may combine to form larger bubbles or bubble clusters, which are carried away with the liquid and thereby reduce cavitation effectiveness [29].

Current hydrodynamic cavitation applications include water treatment [30], biogas production [31], cell disruption [32], refining of wood pulp, and creating agitation in chemical reactors [33]. Hydrodynamic cavitation is easier and more economical to scale-up than acoustic cavitation [28,34]. In addition, the numerous local

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