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Review

Relationships between cellulosic biomass particle size and enzymatic hydrolysis sugar yield: Analysis of inconsistent reports in the literature

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ABSTRACT

Cellulosic ethanol made from cellulosic biomass is a promising alternative to petroleum-based transportation fuels. Enzymatic hydrolysis is a crucial step in cellulosic ethanol production. In order to better understand the mechanisms of enzymatic hydrolysis, relationships between cellulosic biomass particle size and enzymatic hydrolysis sugar yield have been studied extensively. However, the literature contains inconsistent reports. This paper presents an analysis of the inconsistent reports on the relationships in the literature. It discusses the differences in the reported experiments from five perspectives (biomass category, particle size definition, sugar yield definition, biomass treatment procedure, and particle size level). It also proposes future research activities that can provide further understanding of the relationships.

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1. Introduction

During the last three decades, consumption of petroleum-based liquid transportation fuels (including gasoline, diesel, and jet fuels) in the U.S. has increased by more than 30% [1]. These fuels account for about 70% of total petroleum consumption in the U.S. [1]. In 2010, more than 3 billion liters of petroleum were consumed in the U.S. every day, and over 60% of them were imported [1]. Also, the price of petroleum in the U.S. has almost doubled during the last ten years [1]. In addition, use of petroleum-based fuels contributes to accumulation of greenhouse gas (GHG) in the atmosphere [2]. Therefore, it is critically important to explore alternatives to petroleum-based liquid transportation fuels [3–5]. One such alternative is cellulosic ethanol.

An essential step in cellulosic ethanol production is enzymatic hydrolysis which converts cellulose into soluble sugars. A high sugar yield in enzymatic hydrolysis is crucial to the cost-effectiveness of cellulosic ethanol production. Several characteristics of cellulosic biomass have been identified as key factors affecting sugar yield in enzymatic hydrolysis, including biomass particle size, degree of polymerization, crystallinity, and accessible surface area [6–10]. In order to understand the mechanisms of enzymatic hydrolysis and to increase sugar yield in enzymatic hydrolysis, many studies have been done to investigate the relationships between cellulosic biomass particle size and enzymatic hydrolysis sugar yield. However, three different relationships have been reported: negative (smaller particle size produces higher sugar yield), neutral (particle size has no effects on sugar yield), and positive (larger particle size produces higher sugar yield).

This paper, for the first time, presents an analysis of the inconsistent reports on the relationships. It discusses the differences in the reported experiments from five perspectives (biomass category, particle size definition, sugar yield definition, biomass treatment procedure, and particle size level). It also proposes future research activities that can provide further understanding of the relationships.

2. Two categories of cellulosic biomass

The cellulosic biomass used to investigate relationships between particle size and sugar yield can be classified into two categories: pure cellulose and lignocellulosic biomass. As summarized in Fig. 1 and Table 1,when pure cellulose was used, negative

Abbreviations: AFEX, Ammonia fiber explosion; ASABE, American society of agricultural and biological engineers; BET, Brunauer Emmet and Teller; MCC, Microcrystalline cellulose; NREL, National renewable energy laboratory; SSA, Specific surface area; UV-A, Ultrasonic vibration-assisted.

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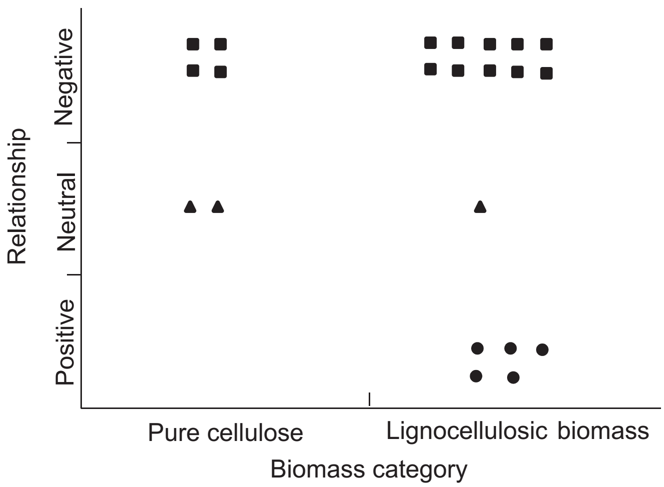

Fig. 1. Reported relationships between particle size and sugar yield for two categories of cellulosic biomass.

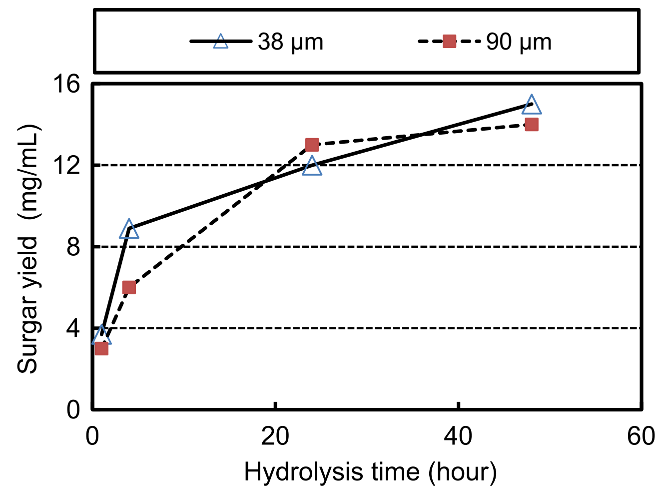

Fig. 2. Relationships between particle size and sugar yield based on the data reported by Shewale and Sadana [15].

relationships were reported in a majority of the related studies. When lignocellulosic biomass was used, three different relationships were reported by different researchers.

Shewale and Sadana [15] reported a neutral relationship between particle size and sugar yield of pure cellulose MCC (microcrystalline cellulose). They used two particle sizes, 38 and 90 μm in average. Their results are plotted in Fig. 2. It is noted that the relationships between particle size and sugar yield were different for different hydrolysis time. Sugar yield of smaller particles (38 μm) was always higher except when hydrolysis time was 24 h. Because they did not provide any information on variations in the sugar yield data, it was not clear whether the differences were statistically significant or not. Peters et al. [16] reported that sugar yield was identical for three particle sizes (38–46, 46–63, and 74–105 μm) of MCC. However, they obtained their different particle sizes by screening a commercial brand of MCC (Avicel PH 102) with screens of different mesh sizes. In most related studies, the different particle sizes were obtained by milling followed by screening.

In the following, several observations will be discussed. Based on these observations, hypotheses (or speculations) are formulated to explain why different biomass categories might cause different relationships between particle size and sugar yield. When appropriate, future research activities are also proposed to test these hypotheses.

2.1. Ball milling had different effects on specific surface areas of pure cellulose versus lignocellulosic biomass

One crucial step in enzymatic hydrolysis is the binding of enzyme molecules to susceptible sites on cellulose surfaces [33–35]. As specific surface area (SSA) of cellulosic biomass increases, the number of enzyme–cellulose bonds will increase, causing sugar yield to increase [34,36–38]. Ball milling has been the most commonly used method to reduce particle size for pure cellulose. The extent to which particle size is reduced depends on milling time. Longer milling time would produce smaller particle sizes. It has been reported that, when ball milling was used to reduce particle size of pure cellulose, SSA increased as particle size decreased (or milling time increased). However, when ball milling was used to reduce particle size of lignocellulosic biomass, SSA did not always increase as particle size decreased (or milling time increased).

For pure cellulose, Sinitsyn et al. [12] reported that SSA of cotton linter doubled as particle size was reduced from 32 to 17 μm by ball milling. Ouajai and Shanks [39] reported that SSA of MCC increased by more than four times after 5-h ball milling.

For lignocellulosic biomass, Mikushina et al. [40] reported that SSA of wood sawdust was increased by five times after 5-h ball milling. However, Gharpuray et al. [41] reported that there was no difference in SSA of wheat straw after4-h versus 24-h ball milling. In addition, even though the SSA of lignocellulosic biomass was increased by milling, the SSA of cellulose (inside the lignocellulosic biomass) might not increase as much due to the complex structure of lignocellulosic biomass. In the literature, there are no investigations about the SSA of cellulose inside lignocellulosic biomass for different particle sizes.

Based on the above observations, it is hypothesized that, when pure cellulose is used, ball milling can increase SSA (while reducing particle size) and thereby increase sugar yield. When lignocellulosic biomass is used, ball milling does not change SSA (while reducing particle size) and thereby does not increase sugar yield. It is noted that other milling methods (such as knife milling and hammer milling) have also been widely used for lignocellulosic biomass. These milling methods will be discussed in Section 3.

2.1.1. Proposed future work

The hypothesis can be tested by investigating effects of ball milling on SSA and sugar yield of pure cellulose and lignocellulosic biomass. Various types of pure cellulose (such as cotton linter, powdered cellulose, and MCC) and lignocellulosic biomass (such as switchgrass, corn stover, and wheat straw) will be milled by ball milling. For each biomass type, the milling process will be stopped every 2 h (up to 48 h). Particle size, SSA, and sugar yield of milled particles will be measured. Particle size will be determined according to ASABE standard S424.1 [42]. SSA will be measured by applying the BET equation to nitrogen adsorption data (this method has been used by many researchers [33,43–45]). Sugar yield will be

Table 1
Reported relationships between particle size and sugar yield for two categories of cellulosic biomass.

Biomass category	Relationship	Reference
Pure cellulose	Negative	[11–14]
	Neutral	[15,16]
Lignocellulosic biomass	Negative	[17–26]
	Neutral	[27]
	Positive	[28–32]

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