

INVESTIGATIONS ON POWER CONSUMPTION, PELLETING TEMPERATURE, PELLET  
QUALITY, AND SUGAR YIELD IN PELLETING OF CELLULOSIC BIOMASS

by

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B.S., University of Wollongong, 2005

AN ABSTRACT OF A DISSERTATION

submitted in partial fulfillment of the requirements for the degree

DOCTOR OF PHILOSOPHY

Department of Industrial and Manufacturing Systems Engineering  
College of Engineering

KANSAS STATE UNIVERSITY  
Manhattan, Kansas

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## **Abstract**

The U.S. economy has been depending on petroleum-based liquid transportation fuels (such as gasoline, diesel, and jet fuels). Currently, about 50% of petroleum used in the U.S. is imported. Petroleum is a finite and non-renewable energy source and its use emits greenhouse gases. Therefore, it is extremely important to develop domestic sustainable alternatives for petroleum-based liquid transportation fuels. Ethanol produced from cellulosic biomass can be such an alternative. However, several technical barriers have hindered large-scale, cost-effective manufacturing of cellulosic ethanol. One such barrier is related to the low density of cellulosic feedstocks, causing high cost in their transportation and storage. Another barrier is low efficiency in conversion of cellulose to fermentable sugar (pretreatment and enzymatic hydrolysis are two major conversion processes), causing high cost in pretreatment and enzymatic hydrolysis of cellulosic biomass. Ultrasonic vibration-assisted (UV-A) pelleting increases both density and sugar yield of cellulosic feedstocks. Incorporating UV-A pelleting into cellulosic ethanol manufacturing may help realize cost-effective manufacturing of cellulosic ethanol.

This PhD dissertation consists of 13 chapters. An introduction is given in Chapter 1. Chapter 2 presents a literature review on related topics. Experimental studies regarding effects of input parameters (such as particle size, pressure, and ultrasonic power) on output parameters (density, durability, stability, and sugar yield) are presented in Chapters 3–4. In Chapters 5–6, comparisons are made between UV-A pelleting and ring-die pelleting (a traditional pelleting method) in terms of pellet properties (density and durability), power consumption, and sugar yield under different conditions. Next, effects of input parameters (such as biomass type, particle size, moisture content, pelleting pressure, and ultrasonic power) on power consumption are studied in Chapters 7–9. Chapter 10 presents an investigation on biomass temperature in UV-A pelleting. Chapter 11 presents an investigation on effects of UV-A pelleting on sugar yield and chemical composition of cellulosic biomass. Chapter 12 presents an investigation on influence of UV-A pelleting on biomass characteristics (such as crystallinity index, thermal properties, and morphological structure). Finally, conclusions are presented in Chapter 13.

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Dr. Donghai Wang

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# Chapter 1 - Introduction

## 1.1 Significance of cellulosic ethanol

The consumption of liquid transportation fuels (including gasoline, diesel, and jet fuels) in the U.S. has increased by 50% during the last three decades [1]. Conventional liquid transportation fuels are derived from petroleum and account for 70% of total petroleum consumption in the U.S [1,2]. Petroleum is a non-renewable resource and about half of the petroleum consumed in the U.S. is imported [1]. Also, use of conventional liquid transportation fuels contributes to the accumulation of greenhouse gas (GHG) in the atmosphere. In this context, it is extremely important to develop domestic sustainable energy sources to replace conventional liquid transportation fuels.

Cellulosic ethanol produced from cellulosic biomass (the fibrous, woody, and generally inedible portions of plant matter) can be such an alternative [3]. It reduces the U.S. dependence on other countries, and has the potential to reduce GHG emissions by more than 86%, while continuing to meet the domestic need of liquid transportation fuels [4,5]. In addition, cellulosic ethanol industry would create jobs, increase farmer's income, and benefit economy [4].

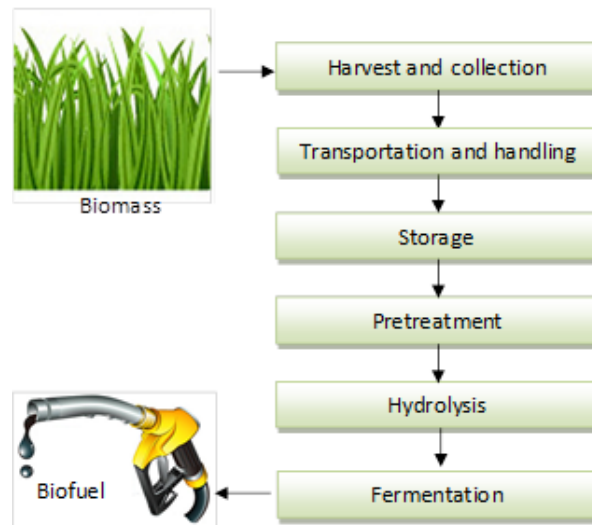
Cellulosic biomass is an abundant renewable resource. An investigation in 2005 jointly supported by DOE and USDA shows that land resources in the U.S. are sufficient to sustain production of enough cellulosic biomass (about 1.3 billion dry tons) annually to replace 30% or more of the nation's current consumption of liquid transportation fuels [4,6]. Unlike grain-based feedstocks for biofuels (such as corn, sugar cane, and beans), cellulosic biomass does not directly compete for limited agricultural land with food and feed industries. Also, cellulosic biomass crops require less energy, fertilizer, and pesticide and improve soil fertility [7]. Furthermore, cellulosic ethanol may become economically feasible with advances in agriculture and biotechnology [8].

## 1.2 Challenges in cellulosic ethanol manufacturing

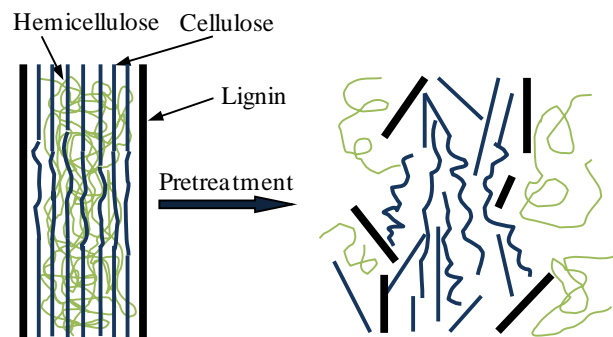
Figure 1.1 shows major steps for manufacturing of cellulosic ethanol. After harvesting and collection, cellulosic biomass is transported and stored for future use. The purpose of pretreatment of cellulosic biomass is illustrated in Figure 1.2. Pretreatment can break the lignin

seal and disrupt the crystalline structure of cellulose, making it more accessible to enzymatic hydrolysis [3,4]. Hydrolysis breaks down cellulose into its component sugars that are convertible to ethanol by fermentation [3,4].

**Figure 1.1 Major steps in biofuel manufacturing (after [3,4])**



**Figure 1.1 Purpose of pretreatment (after [4])**



Total annual capacity of large-scale cellulosic biofuel plants being (or to be) built in the U.S. will be less than 4 million gallons [9-14]. In order to meet the U.S. government's mandate of 16 billion gallons of cellulosic ethanol annually by 2022 [15], many more large-scale plants need to be built. However, several technical barriers have hindered large-scale, cost-effective manufacturing of cellulosic ethanol. One such barrier is related to the low density of cellulosic

feedstocks, causing high cost in biomass transportation and storage [8,16,17]. Another barrier is low efficiency in conversion of cellulose to fermentable sugar (pretreatment and enzymatic hydrolysis are two major conversion processes), causing high cost in pretreatment and enzymatic hydrolysis of cellulosic biomass [18].

### **1.3 Ultrasonic vibration-assisted pelleting of cellulosic biomass**

Pelleting of cellulosic biomass can increase its density, resulting in reduced transportation and storage costs of cellulosic biomass [19]. Furthermore, pellets with uniform size and shape can be handled and transported with existing grain-handling equipment, leading to increased handling efficiency [19].

Preliminary studies show that ultrasonic vibration-assisted (UV-A) pelleting can produce pellets whose density is up to  $1200 \text{ kg/m}^3$  [20]. Furthermore, experiments show that the biomass feedstocks treated by UV-A pelleting can produce higher sugar yield and ethanol than those without UV-A pelleting [21].

### **1.4 Objectives and scope of this research**

The objectives of this research on UV-A pelleting are as the following:

(1) To study effects of input parameters (such as particle size, pressure, and ultrasonic power) in UV-A pelleting process on pellet quality and sugar yield.

(2) To compare pellets produced by UV-A pelleting and by ring-die pelleting in terms of pellet quality and sugar yield (under different conditions).

(3) To investigate effects of input parameters (such as biomass type, moisture content, particle size, pressure, and ultrasonic power) on power consumption in UV-A pelleting.

(4) To investigate the temperature of biomass in UV-A pelleting and effects of input variables (ultrasonic power, pelleting pressure, and pellet weight) on pelleting temperature.

(5) To study mechanisms through which UV-A pelleting increases sugar yield by examining effects of UV-A pelleting on biomass characteristics (such as chemical composition, crystallinity index, thermal properties, and morphological structure).

This dissertation contains 12 chapters, most of which either have been published as technical papers or will be submitted to Journals.

Chapter 1 is an introduction providing the background and the objectives of this work.

Chapter 2 is a literature review on effects of treatments on cellulosic biomass structure in ethanol manufacturing.

Chapters 3 and 4 report experimental investigations on effects of input parameters on pellet properties and sugar yield. Chapter 3 investigates effects of biomass particle size on pellet density and stability. Chapter 4 investigates effects of ultrasonic power and particle size on pellet quality (density, durability, and stability) and sugar yield.

Chapters 5 and 6 present comparisons between UV-A pelleting and ring-die pelleting in terms of pellet properties, power consumption, and sugar yield. In Chapter 5, comparisons of pellet density, durability, and power consumption between the two pelleting methods are conducted. In Chapter 6, sugar yield comparison between the two pelleting methods is presented with different particle size and under different pretreatment conditions with varying acid concentration, solid content, pretreatment temperature, and pretreatment time.

Chapters 7-9 investigate power consumption in UV-A pelleting. Chapter 7 investigates effects of pelleting pressure, ultrasonic power, particle size, and biomass types on power consumption in UV-A pelleting with four types of cellulosic biomass (big bluestem, corn stover, sorghum stalk, and wheat straw). Chapter 8 studies effects of moisture content, pelleting pressure, ultrasonic power, and particle size on power consumption and power consumption rate. Chapter 9 employs a 24 full factorial design to investigate main and interaction effects of four input variables (biomass moisture content, particle size, pelleting pressure, and ultrasonic power) on power consumption.

Chapter 10 investigates temperature of biomass in UV-A pelleting and effects of input variables (ultrasonic power, pelleting pressure, and pellet weight) on pelleting temperature.

Chapter 11 investigates effects of UV-A pelleting on sugar yield and composition of biomass.

Chapter 12 investigates effects of UV-A pelleting biomass characteristics (such as crystallinity index, thermal properties, and morphological structure).

Chapter 13 summarizes the conclusions and contributions of this research.

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## Chapter 2 - Literature review

### Paper Title:

Effects of treatments on cellulosic biomass structure in ethanol manufacturing: a literature review

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## **Abstract**

Ethanol made from cellulosic biomass is an alternative to petroleum-based liquid transportation fuels. Enzymatic hydrolysis uses enzymes to convert cellulosic biomass into sugars that are fermented into ethanol. In order to increase sugar yield, various treatments (such as biomass size reduction and pretreatment) are applied to cellulosic biomass before enzymatic

hydrolysis. These treatments will alter structure parameters of cellulosic biomass, such as crystallinity index, degree of polymerization, particle size, pore volume, and specific surface area. There are currently no review papers on these structure parameters of cellulosic biomass in ethanol manufacturing. This paper reviews experimental investigations in the literature about effects of various treatments on the structure parameters of cellulosic biomass.

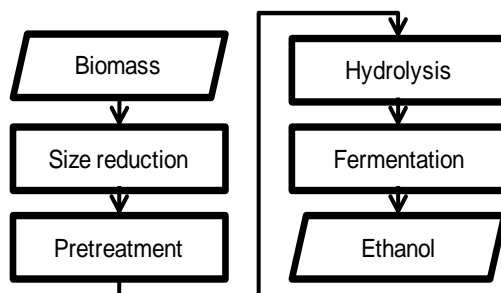
## **2.1 Introduction**

70% of petroleum used in the U.S. is for transportation fuels [1]. Demands for transportation fuels vastly exceed the nation's production capacity, forcing the U.S. to rely on imported petroleum. They are likely to continue rising in the future [2]. Considering this and other factors, such as finite resources, non-uniform distribution, contribution to greenhouse gas (GHG) emission, and volatile prices of petroleum, it is desirable to find alternative fuels. One such alternative is ethanol produced from cellulosic biomass (fibrous, woody, and generally inedible portions of plant matter).

In the U.S., ethanol is produced primarily from corn [3]. However, corn-based ethanol competes with food or feed production for limited agricultural land. Cellulosic ethanol will not have this problem [4]. Land resources in the U.S. are sufficient to sustain production of enough cellulosic biomass annually to replace 30% of current annual consumption of liquid transportation fuels [5]. Cellulosic ethanol can also reduce GHG emissions by 85% compared to petroleum-based fuels [6].

Major steps in manufacturing of cellulosic ethanol are shown in Figure 2.1. In order to increase sugar yield in enzymatic hydrolysis, various treatments have been applied to cellulosic biomass, as summarized in Table 2.1. These treatments will alter structure parameters of cellulosic biomass, such as crystallinity index, degree of polymerization, particle size, pore volume, and specific surface area. Many investigations have been conducted on effects of various treatments on structure parameters of cellulosic biomass. However, there are currently no review papers in the literature that cover these investigations.

**Figure 2.1 Major steps in manufacturing of cellulosic ethanol (after [14])**



This paper reviews experimental investigations in the literature about the effects of various treatments on structure parameters (including crystallinity index, degree of polymerization, particle size, pore volume, and specific surface area). Firstly, background information is provided about size reduction and pretreatments of cellulosic biomass. Secondly, the reported experimental results on structure parameters are presented. Materials and conditions used in the reported experiments are tabulated. Finally, concluding remarks are given.

**Table 2.1 Summary of investigations on cellulosic biomass structure**

Sample material	Treatment	Structure	Ref.
Carboxymethylcellulose, wood shavings	\	CI	[8]
Avicel, fibrous cellulose	Phosphoric acid	CI	[9]
Cotton linter, Foley Fluffs wood pulp, Southern pine kraft pulp	Ball milling	CI	[10]
Mixed hardwood	Acid	SSA, pore volume	[11]
Poplar wood	Ball milling	CI	[12]
Sulfite pulp, MCC	Ball milling, water swollen, solvent drying	SSA	[13]
Textile cotton waste	X-ray, NaOH, ZnCl <sub>2</sub>	CI, DP	[14]
Textile cotton waste	ZnCl-HCl, ZnCl-acetone	CI, DP, SSA, pore volume	[14]
Aspen wood	Ball milling	Particle size	[15]
Wheat straw	Milling, peracetic acid, ethylene glycol	CI, SSA	[16]
White pine, mixed hardwood	Partial acid hydrolysis, steam explosion	Pore volume	[17]
Poplar, pine, mixed hardwood	Dilute acid, steam	CI, pore volume	[18]
Poplar wood	Steam explosion	Pore volume	[19]
Corn stover	Integrated wet-milling and alkali	CI	[20]
Wood pulp	Wiley milling, ball milling	CI, DP	[21]
Corn stover	Milling, lime	Particle size	[22]
Cellulose powder, newsprint, white pine wood, black cotton wood	Ball milling, and wet milling	CI	[23]
Corn stover	Lime	CI	[24]
Absorbent cotton, Avicel, Solka floc	Ball milling, hammer milling, NaOH, H <sub>3</sub> PO <sub>4</sub>	CI, DP, SSA	[25]
Coastal bermuda grass	Hot water, ammonia fiber explosion	CI	[26]
Corn stalk, solka floc	NaOH	Pore volume	[27]
Sawdust of aspen wood	Vibratory centrifugal milling, planetary milling, ball milling	CI, particle size, SSA	[28]
Cotton linter, newsprint, douglas fir, red oak	Ball milling	CI	[29]
Hemp cellulose fiber	Ball milling	CI, SSA, pore volume	[30]
Wheat straw	Milling, wet oxidation	Particle size	[31]
Bagasse, wheat straw, eucalyptus regnants, pinus radiata, and cotton linters	Ball milling, CO <sub>2</sub> explosion, alkali explosion, ozonation, and NaCl delignification	CI, DP	[32]
Avicel, bagasse, rice straw, sludge, newspaper, cardboard, mill waste	Wet and dry ball milling	CI, particle size	[33]
Microcrystalline cellulose	-	Particle size	[34]
Cotton cellulose powder	Acid dissolution	CI	[35]
Cotton linter, sugar cane bagasse	Ball milling, X-ray irradiation, acid	CI, DP, SSA, particle size	[36]
Black spruce wood pulps	Acid sulfite process, half Milled	Pore volume, SSA	[37]
Mixed hardwood	Acid, organosolv, alkaline peroxide	SSA	[38]
MCC, noncrystalline cellulose, cotton	Milling	CI, pore volume	[39]
Microcrystalline cotton cellulose	Media milling	Particle size	[40]
Miscanthus sinensis	Ball milling, sodium chlorite	CI	[41]
Corn stover	Hot water	Particle size	[42]
Hardwood	Pan milling	CI, particle size, SSA	[43]

## **2.2 Background information on size reduction and pretreatment of cellulosic biomass**

### ***2.2.1 Size reduction***

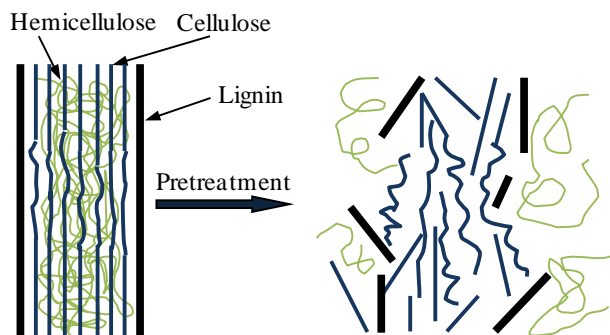
Size reduction of cellulosic biomass can be performed using a variety of mechanical methods, such as ball milling [44], cutting milling [45], compression milling [46,47], hammer milling [48], fluid energy milling [49], and colloid milling [50]. More information about these methods can be found in a review paper [51]. Size reduction can increase density and uniformity of biomass feedstock, reduce costs of feedstock storage and transportation, and improve feedstock bioconversion rate [52].

### ***2.2.2 Pretreatment***

In cellulosic biomass, cellulose exists within a matrix of hemicelluloses and lignin. Hemicellulose and lignin block the contact of enzymes with cellulose by absorbing enzymes or blocking access of enzymes to the cellulose surface [53], leading to low sugar yields in enzymatic hydrolysis. The goal of pretreatment is to break the lignin seal and remove hemicellulose [54], making cellulose more accessible to enzymes in enzymatic hydrolysis, as illustrated in Figure 2.2.

A number of different pretreatment approaches have been investigated, including biological (such as enzymes and bacteria), chemical (such as sulfuric acid, sulfur dioxide, alkali, oxidizing agents, ammonia, and organic solvents), and physical (such as heat and ultrasonic) means. So far, only chemical pretreatments offer the high sugar yields vital to economic success [56]. More information about cellulosic biomass pretreatment can be found in some review papers [27,45,57].

**Figure 2.2 Purpose of pretreatment in manufacturing of cellulosic ethanol (after [55])**

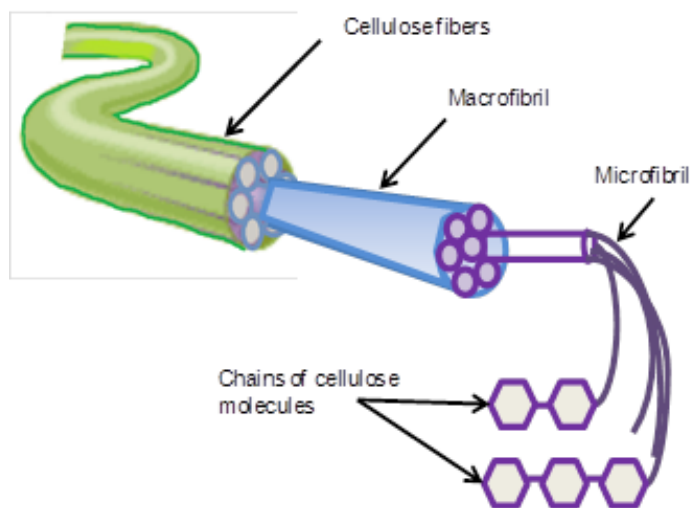


### 2.3 Crystallinity index (CI)

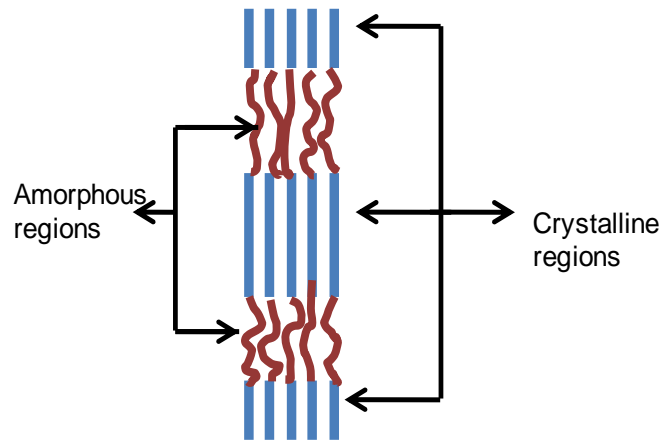
As illustrated in Figure 2.3, cellulose fibers contain macrofibrils that contain microfibrils. A microfibril consists of crystalline regions (with highly ordered molecule arrangement) and amorphous regions (with less ordered molecule arrangement) [59], as shown in Figure 2.4. In the literature, there are two ways to define CI. One is the percentage (ratio) of the amount of cellulose in crystalline regions versus that in amorphous regions [61]. The other is the percentage of the amount of crystalline material in the biomass [24,62].

The technologies used to determine CI include X-ray diffraction [24,32,43,62], density measurement [63], Raman spectroscopy [64], infrared spectroscopy, and nuclear magnetic resonance.

**Figure 2.3 Illustration of cellulose fibers and molecular chains (after [58])**



**Figure 2.4 Crystalline and amorphous regions in cellulose microfibril (after [60])**



CI can be significantly affected by milling processes (used to reduce the particle size of cellulosic biomass). As shown in Table 2.2, most milling processes can decrease CI when the cycle time is sufficiently long [15,33]. It is believed that intensive mechanical action on cellulosic biomass can cause destruction of crystalline regions and lead to reduced CI [28]. Recrystallization may happen at the end of milling processes [40, 41], because the mechanical disruption of the crystalline regions and formation of molecular irregularities (less ordered molecule arrangement) are partially reversible. Dry ball-milled particles can regain crystallinity when exposed to moisture [66].



**Table 2.2 Effects of milling processes on CI**

Material	Method	Time (h)	CI before milling	CI after milling	Ref.
Cotton linter	Ball milling	10	74	32	[10]
Foley fluff	Ball milling	6	56	29	[10]
Southern pine	Ball milling	7	53	35	[10]
Poplar	Ball milling	112	55	12	[12]
Wood pulp	Ball milling	96	74.2	4.9	[13]
Wheat straw	Ball milling	24	69.6	19.4	[16]
Aspen wood	Ball milling	5	81	0	[28]
Newsprint	Ball milling	0.5	55	10	[29]
Red Oak	Ball milling	0.5	38	6	[29]
Hemp fiber	Ball milling	6	62	32	[30]
Avicel	Ball milling	\	82.8	48.1	[33]
Wheat straw	Ball milling	12	80	40	[65]
Cotton	Compression milling	147	81.7	46.2	[25]
Cotton	Media milling	2	93	80	[40]
Hardwood	Pan milling	40 cycles	65	22	[43]
Aspen wood	Planetary	0.03	81	100	[28]
Aspen wood	Vibratory centrifugal	0.25	81	100	[28]

Results are different with wet milling processes, in which cellulosic biomass is milled in water. Reductions of CI are much smaller in wet ball milling [33,66]. It is suggested that dry material is more friable and more readily fracture upon impact during milling [33,66].

He et al. [20] investigated an integrated wet-milling and alkali pretreatment on corn stover. Corn stover was wet-milled for 1 hour using a ball mill with a rotational speed of 500 rpm. Sodium hydroxide (NaOH) solution was used as the wet-milling medium. The crystalline structure was destroyed completely in one hour. He et al. [20] believed that the integrated pretreatment can reduce both milling time and NaOH concentration compared with the reported individual dry milling or individual alkali pretreatment. It disrupts the crystalline structure of cellulose due to the combined physical and chemical effects. NaOH solution can dissolve lignin by breaking the ester bonds cross-linking lignin and xylan [67], thus increasing the porosity of biomass and causing the milling process to disrupt the crystalline region easily.

Most aqueous pretreatments (such as those involving acid, alkali, and hot water) will increase CI, as summarized in Table 2.3. Water soaking (soaking biomass in water) may cause recrystallization on milled cellulose fibers [60,66]. Some amorphous regions of milled cellulose absorb water and recrystallize into crystalline regions. The solubilization of lignin and

hemicellulose (both are amorphous components) results in an increased percentage of crystalline regions [24,32].

**Table 2.3 Effects of aqueous pretreatments on CI**

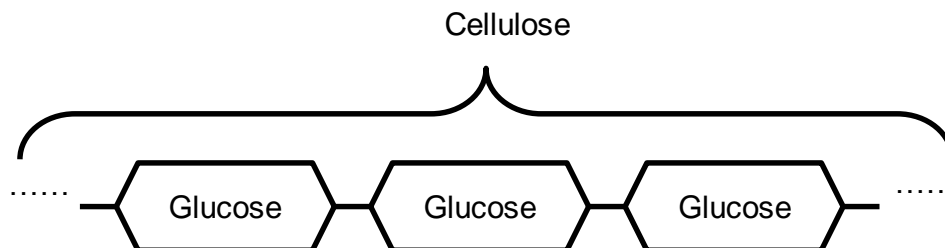
Material	Method	CI after pretreatment	Ref.
Textile cotton waste	NaOH	Increase	[14]
Textile cotton waste	ZnCl <sub>2</sub>	Increase	[14]
Poplar	Dilute acid	Increase	[18]
Pine	Dilute acid	Increase	[18]
Mixed hardwood	Dilute acid	Increase	[18]
Corn stover	Lime in water	Increase	[24]
Bermuda grass	Hot water	Increase	[26]
Bermuda grass	Ammonia	Increase	[26]
Bagasse	Alkali	Increase	[32]
Bagasse	Carbon dioxide	Increase	[32]
Wheat straw	Alkali	Increase	[32]
Wheat straw	Carbon dioxide	Increase	[32]
Mixed hardwood	Acid	Increase	[38]
Mixed hardwood	Alkaline hydrogen peroxide	Increase	[38]
Mixed hardwood	Organosolv	Increase	[38]

Focher et al. [14] investigated effects of X-ray irradiation on CI of cotton waste. The cotton waste was irradiated in a water suspension by a Co radiation source at a dose of 50 Mrad. They found that x-ray at this level did not significantly affect CI. Lee et al. [26] used ammonia explosion to treat coastal Bermuda grass, causing CI to increase from 50.2% to 59.5%.

## 2.4 Degree of polymerization (DP)

A cellulose molecule is made up of many glucose units [63], as illustrated in Figure 2.5. The chain length of cellulose molecules can differ widely. Cellulose has the empirical formula of (C<sub>6</sub>H<sub>10</sub>O<sub>5</sub>)<sub>n</sub> [67]. n designates the number of glucose units linked together in the long chain of cellulose molecules. The value of n is commonly known as degree of polymerization (DP).

**Figure 2.5 Chain-like molecular structure of cellulose (after [63])**



Different methods have been used to determine DP of cellulose, such as osmotic pressure measurement [68], sedimentation in an ultracentrifuge [68], light scattering [69], and viscosity measurement [14,32,36].

Table 2.4 lists typical cellulosic materials and their DPs before any treatments. Table 2.5 summarizes DPs after treatments. The results clearly indicate that all reported treatment techniques result in a reduction in DP.

**Table 2.4 Typical cellulosic materials and their dps without any treatments (after [70])**

Cellulosic material	DP
Native cellulose	3500-10000
Chemical cotton	500-3000
Wood pulp	500-2100

**Table 2.5 Effects of treatments on DP**

Material	Treatment	DP before treatment	DP after treatment	Ref.
Textile cotton wastes	X-ray	2075	60	[14]
Textile cotton wastes	NaOH	2075	1000	[14]
Textile cotton wastes	ZnCl <sub>2</sub>	2075	1430	[14]
Wood pulp	Ball milling	1205	642	[21]
Cotton linter	Compression milling	2240	1880	[21]
Solka floc	NaOH	1210	1010	[25]
Solka floc	H <sub>3</sub> PO <sub>4</sub>	1210	1090	[25]
Bagasse	Ozone	925	800	[32]
Bagasse	CO <sub>2</sub> explosion	925	572	[32]
Bagasse	Alkali	925	550	[32]
Wheat straw	Ozone	1045	908	[32]
Wheat straw	CO <sub>2</sub> explosion	1045	698	[32]
Wheat straw	Alkali	1045	662	[32]
Eucalyptus regnans wood	Ozone	1510	1065	[32]
Eucalyptus regnans wood	CO <sub>2</sub> explosion	1510	815	[32]
Pinus radiata	Ozone	3063	2900	[32]

## 2.5 Particle size

Particle size refers to the size of milled biomass particles. It is controlled by using sieves with different screen sizes. The screen size of the sieve is also referred to as particle size. Using standard stainless steel wire sieves, milled biomass particles can be classified into different size ranges [22]. The average particle size can also be estimated by observation with scanning electronic microscopes [43,60], video-displays [36] and laser diffraction particle size analyzers [40,43].

During milling processes, biomass materials endure impacting, compression, shearing, and stretching [43]. Large particles are broken into small ones. The volume percentage of small particles increases with milling time [28]. In general, the average particle size decreases with the milling time [40]. The distribution curve of the milled particle size is believed to follow normal distribution [43]. With longer milling time, the curves have smaller mean values but larger standard deviations [43].

## 2.6 Pore volume

There are numerous pores and pore arrays distributed on the surface of cellulose fibers [60]. The total volume of pores distributed in the porous cellulose fiber is regarded as pore volume [60]. Pore volume, expressed by  $\text{cm}^3/\text{g}$ , is related to the surface area of cellulose fibers as well as pore size (the size of the pore opening). The total volume of pores in cellulosic biomass can be used to estimate its internal surface area. A higher pore volume indicates a larger internal surface area of cellulosic biomass. Burns et al. [11] estimated the surface area of the wall of pores from the pore volume. The actual pore geometry is not quite clear, but it is suggested that pores in wood pulp are parallel slits [71]

Pore volume and pore size are determined by different ways for dry or wet cellulosic biomass. Adsorption of liquid nitrogen and the BET equation are often used to determine the pore volume of dry cellulosic biomass [60]. Pore volume and pore size in wet cellulosic biomass may be characterized by using the solute exclusion technique [37].

The solute exclusion technique is a preferred method for measuring pore volume [6,11,39] because it uses a wet sample submerged in an aqueous environment (the same environment present during enzymatic hydrolysis). In the dry environment, the pore structure may collapse or shrink during nitrogen adsorption [18].

Steam explosion and water swelling are two major approaches to increase pore volume [17,19,37,38,72,73]. In steam explosion, high-pressure steam heats and penetrates the cellulosic biomass. After a desired reaction time, the biomass is quickly exposed to atmospheric pressure. The water inside the biomass vaporizes and expands rapidly, causing great increase in the porosity of biomass [19]. Both the number and volume of pores increase after steam explosion [17,19,72].

In water swelling, acid or alkali causes the capillary structure of cellulose to expand, increasing pore volume and surface area [37,38,73]. Lee [74] believes that the degree of swelling is the most important factor to pore volume. However, when cellulosic biomass is dried from the water-swollen state, the pore structure collapses and shrinks due to the surface tension forces [75]. The average pore volume and pore size will decrease [37]. Some pores even disappear [60]. However, both pore volume and pore size of water-swollen cellulose are largely retained when the cellulose is dried using a solvent drying technique [13] This drying process can increase pore volume by 50% [60].

Knappert et al. [76] reported that the most responsive biomass to pretreatment is biomass in its natural state (such as oak and corn stover) but not highly purified and delignified cellulosic biomass. Removal of hemicelluloses with dilute acid or hot water results in a more porous or swollen structure in the residue.

Pore volume can also be increased by ball milling [30]. The rate of the increase in pore volume declines after a certain time. The internal pore sites are opened during early stages of ball milling.

## **2.7 Specific surface area (SSA)**

Specific surface area (SSA) is the amount of surface area per unit mass of cellulose and is expressed as  $\text{m}^2/\text{L}$  [77]. SSA is usually referred to as the sum of the external and internal surface area of cellulosic substrate [37,78]. The external surface area is closely related to the shape and size of the cellulose biomass. The internal surface area depends on the volume and size of the pores [13].

SSA can be measured using various approaches, such as surface catalytic method, argon adsorption [28], chymotrypsin adsorption, nitrogen adsorption [13,25], solute exclusion technique [37,38], microscope [74], mercury porosimetry, and dry adsorption.

SSA can be affected by various treatments, as summarized in Table 2.6. Many milling methods can increase SSA [28]. The shapes of tools and containers affect SSA during milling. Zhang et al. [43] found that SSA increased almost linearly with milling cycles, indicating that breakage of bulky cellulosic materials into small particles could produce more surface area. Similar effects were reported by Gharpuray et al. [16], Kelsey and Shafizadeh [23], Lee et al. [25] and Ouajai and Shanks [30]. Fan et al. [13] compared SSA of sulfite pulps before and after 96 hours of ball milling. They found that SSA was slightly changed from 2.09 to 1.91 m<sup>2</sup>/g. Water soaking dramatically increases SSA of cellulose fibers [13]. The expanded capillary structure of water-soaked cellulosic biomass may cause a significant increase in surface area, and the total area of the soaked material may be as much as 100 fold greater than that in a dry state [75].

**Table 2.6 Treatments with significant effect on SSA (after [79])**

<b>Treatment</b>	<b>Effect on SSA</b>
AFEX	Increase
ARP	Increase
Dilute acid	Increase
Flow-through acid	Increase
Flow-through liquid hot water	Increase
Lime	Increase
Liquid hot water	Increase
pH controlled hot water	Increase
Uncatalyzed steam explosion	Increase

## **2.8 Concluding remarks**

This paper reviews effects of various treatments (including size reduction and pretreatments) on structure parameters of cellulosic biomass. There are various treatment methods on biomass. Some treatments have significant effects only on one of structure parameters. Some treatments, e.g., water soaking, have effects on more than one structure parameters.

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## Chapter 3 - Effects of particle size on pellet quality

### Paper Title:

Ultrasonic vibration assisted pelleting of wheat straw: effects of particle size

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### **Abstract**

Cellulosic biomass is an important feedstock source for biofuel manufacturing. Nevertheless, there are several barriers for large-scale and cost-effective manufacturing of cellulosic biofuels. One barrier is the high cost caused by the transportation and storage of cellulosic biomass due to its low density. Ultrasonic vibration-assisted pelleting is proposed to increase the density of biomass feedstock without using high-temperature steam, high pressure, and binder materials. Wheat straw biomass is milled into particles with different sizes before

ultrasonic vibration-assisted pelleting. Effects of the wheat straw particle size on pelleting density and stability (spring-back) are experimentally investigated.

### **Keywords**

Biofuel, biomass, density, particle size, stability, ultrasonic vibration-assisted pelleting

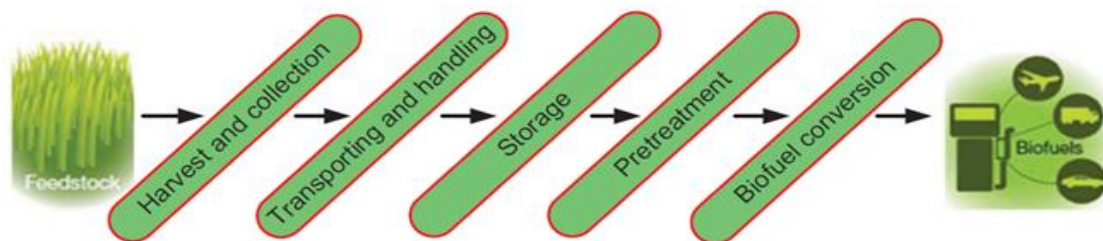
## **3.1 Introduction**

In the last decade, the consumption of petroleum for transportation in the U.S. increased dramatically [1]. In 2008, the daily consumption of the liquid transportation fuels was about 13.7 million barrels in the U.S. [1]. Traditional liquid transportation fuels (including gasoline, ethane, diesel fuel, and other types of fuel) are distilled from petroleum [2]. The rapidly increasing demand of liquid transportation fuels makes the U.S. more dependent on the imported petroleum [3]. Additionally, the volatile prices of the liquid transportation fuels could impact the economic growth of the U.S. significantly [4]. It is important to promote the development of sustainable energy sources to replace the traditional liquid transportation fuels.

Among all renewable energy sources, cellulosic biofuels (including biodiesel, ethanol, and other types of biomass-derived fuels) represent an attractive alternative source that could replace the traditional liquid transportation fuels. Ethanol fuel, which is an alcohol made by fermenting sugar components of plant material, is the most common biofuel [5]. Although ethanol, in its pure form, can be used as a fuel for vehicles, it is commonly used as a gasoline additive [5]. Cellulosic biomass, such as wheat straw, switchgrass, or sorghum stalks, is a very good candidate for the feedstock for manufacturing of biofuels [6].

Using cellulosic biomass can also reduce greenhouse gas emissions by 85% over petroleum-based fuel [7]. Land resource in the U.S. is sufficient to sustain production of enough biomass annually to displace 30% of current consumption of liquid transportation fuels [8]. Thus, cellulosic ethanol production can reduce petroleum imports from other countries and improve national energy security [6]. Furthermore, unlike ethanol from corn and other feedstocks (e.g. sugar cane, and soybeans), cellulosic biofuel has no effects on food supply, and does not result in high food price [9]. However, there are only pilot biofuel plants using cellulosic feedstock in the U.S. currently [10]. Figure 3.1 shows major steps for manufacturing of cellulosic biofuels.

**Figure 3.1 Major Steps for manufacturing of cellulosic biofuels (after [10,11])**



There are several barriers for large-scale and cost-effective manufacturing of cellulosic biofuel [12,13]. One of such barriers is the high cost of the transportation and storage of biomass due to its low density [12,14,15].

Pelleting could improve the density of biomass feedstock [16]. Traditional pelleting methods include using a screw extruder, a briquetting press, or a rolling machine. These methods generally require high-temperature steam, high pressure, and binder materials [17,18]. With these methods, it is difficult to achieve cost-effective pelleting on or near the field where cellulosic biomass is available. Ultrasonic vibration-assisted (UV-A) pelleting process, a new pelleting method without using high-temperature steam and binder materials, is proposed to increase the density of biomass feedstock. Preliminary studies show that UV-A pelleting can produce biomass pellets whose density is comparable to that processed by traditional pelleting methods [19,20].

Biomass is milled into particles of different sizes before UV-A pelleting. The particle size of biomass is an important parameter in UV-A pelleting. The investigations on the effects of particle size can help to get better understanding of the mechanism of UV-A pelleting. In this paper, for the first time, the effects of biomass particle size on the characteristics of pellets in UV-A pelleting are investigated.

Smaller particle sizes tend to increase not only the density but also the surface area of cellulosic biomass [21]. The increased surface area allows more access by enzymes and chemicals, resulting in higher yields of biofuel. However, it usually requires more energy to mill biomass into smaller particles. According to the National Renewable Energy Laboratory, reducing the particle size took about 1/3 of the total energy needed to convert cellulosic biomass to ethanol [22]. In other words, smaller particle sizes are desired from the viewpoint of biofuel yield, but are undesirable from the viewpoint of energy consumption in milling. In order to find



the optimum particle size, it is important to know the relationships between particle size and biofuel yield and between particle size and energy consumption in milling. In addition to these the above viewpoints, this paper adds a third viewpoint: effects of particle size on characteristics of pellets. A comprehensive understanding about the effects of particle size from all these viewpoints is needed in order to recommend the optimum particle size/combination for most efficient biofuel production.

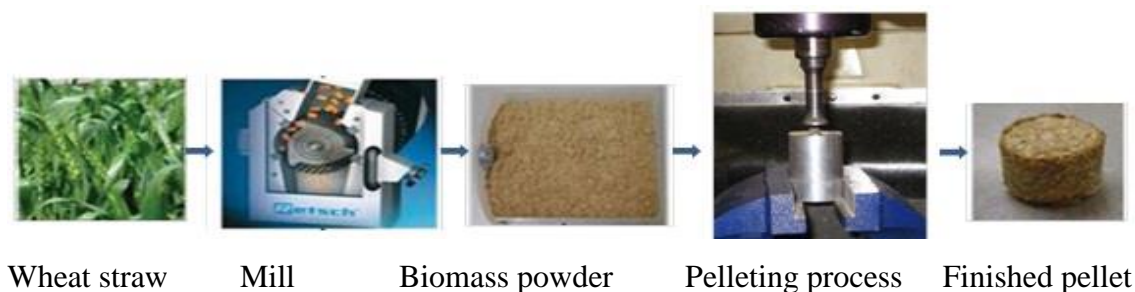
## 3.2 Experimental Procedure and Parameters

### 3.2.1 Ultrasonic vibration-assisted pelleting

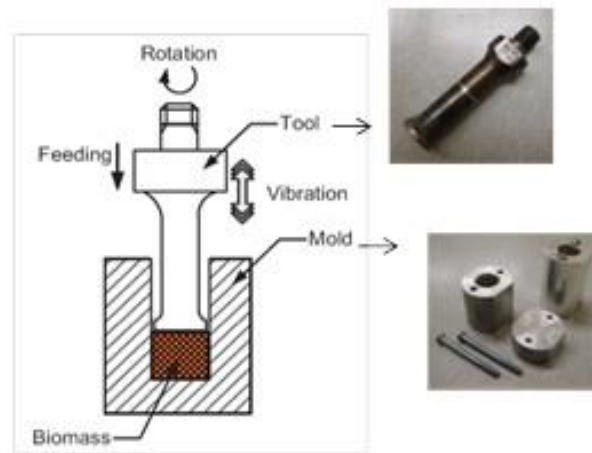
Major steps of ultrasonic vibration-assisted (UV-A) pelleting are shown in Figure 3.2. Wheat straw, the raw material used in the experiments, was milled into particles of different sizes. The moisture content of the biomass particles after milling was measured and adjusted to the desired levels. UV-A pelleting was performed on a Sonic Mill Series 10 machine (Sonic-Mill, Albuquerque, NM, U.S.).

Figure 3.3 illustrates UV-A pelleting. An aluminum mold with a cylindrical cavity in the center was used to hold the biomass particles. A tool with a flat end was mounted to a rotary spindle and fed towards the biomass at a preset feedrate. Meanwhile, an ultrasonic vibration (at a frequency of 20 kHz) was applied to the tool during the pelleting process. The diameter of the tool (17.4 mm) was slightly smaller than that of cavity in the mold (18.6 mm). Once the tool reached a selected depth inside the cavity of the mold, it would be automatically retracted. Then the pellet would be removed from the mold.

**Figure 3.2 Major Steps for UV-A pelleting**



**Figure 3.3 Illustration of UV-A pelleting**



### ***3.2.2 Experimental parameters***

#### ***3.2.2.1 Biomass particle size***

A cutting mill, model SM 2000 (Retsch, Newtown, PA, USA), was used to mill the wheat straw into particles. The particle sizes were controlled by sieves with preset hole sizes. For example, if a sieve with 8 mm hole size was put inside the mill, 8 mm particle size was obtained, as illustrated in Figure 3.4. By choosing sieves with different hole sizes, different sizes of particles would be obtained. In these experiments, five particle sizes (0.25, 1, 1.5, 2, and 8 mm) were produced. Two individual particle sizes were also mixed together to investigate whether the combination of the particle sizes could improve the pelleting results. Table 3.1 shows the mixed-size particles used in the experiments.

**Figure 3.3.4 Using a sieve with a certain hole size to control particle size**



(a)Wheat straw as received    (b) Sieve    (c) Sieve inside the mill    (d) Wheat straw particles

**Table 3.1 Mixed-size particles**

Mixed-size particles	Weight percentage
0.25 + 8 mm	50% for each particle size
0.25 + 1.5 mm	50% for each particle size
1.5 + 8 mm	50% for each particle size

**3.2.2.2. Biomass moisture content**

Moisture content (MC) means the percentage of the moisture contained in a certain amount of biomass. Lower MC means less moisture in the biomass.

$$MC = \frac{\text{Weight of moisture}}{\text{Weight of biomass including moisture}} * 100\%$$

In these experiments, the MC of the wheat straw particles before pelleting was adjusted to 13%.

**3.2.2.3. Other important parameters**

Table 3.2 shows other important parameters in the experiments. Ultrasonic power controls the amplitude of vibration. A higher ultrasonic power means a larger amplitude.

**Table 3.2 Values of main UV-A pelleting parameters**

Parameter	Value
Feedrate (mm/s)	0.267
Spindle speed (rpm)	50
Weight of biomass for each pellet (g)	3
Ultrasonic power (%)	35

**3.2.3 Measurement methods****3.2.3.1. Pellet density**

Five sample pellets for each particle size were made. The density of each pellet was recorded daily up to ten days after the pellets were taken out of the mold. Height and diameter of the pellet were measured with a vernier caliper. The volume of each pellet was calculated from its height and diameter. Weight of the pellet was measured with an electronic scale (Ohaus, Pine Brook, NJ, U.S.). The density of pellets was computed by the following formula:

$$\text{Pellet density} = \frac{\text{Weight of the pellet}}{\text{Volume of the pellet}}$$

### 3.2.3.2. Pellet spring-back

Spring-back of a pellet is employed to evaluate the change in a pellet's volume over time. Once the tool was retracted in UV-A pelleting, the pellet would expand immediately inside the mold. After it was taken out of the mold, the pellet would continue expanding for a few days until becoming stable. For each sample pellet, its spring-back was recorded daily up to ten days. The spring-back of a pellet can be expressed as:

$$\text{Spring back} = \frac{\text{Volume} - \text{Original volume}}{\text{Original volume}}$$

where, volume = the volume of the pellet obtained on the measurement day, and original volume = theoretical volume of the pellet in the mold obtained by measuring the diameter of the mold cavity and the tool stop position.

## 3.3 Experimental Results and Discussion

### 3.3.1 Results on one-sized particles

Figure 3.5 shows the results of pellet density for one-sized particles. There was a dramatic decrease in pellet density during the first two days before becoming stabilized. Smaller particles produced higher density.

**Figure 3.5 Effects of particle size on pellet density**

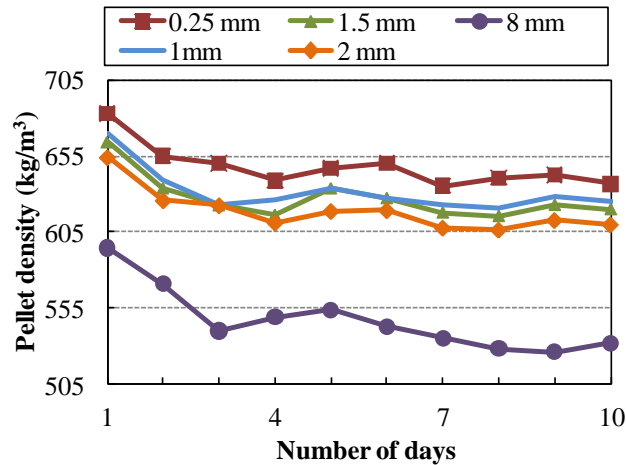
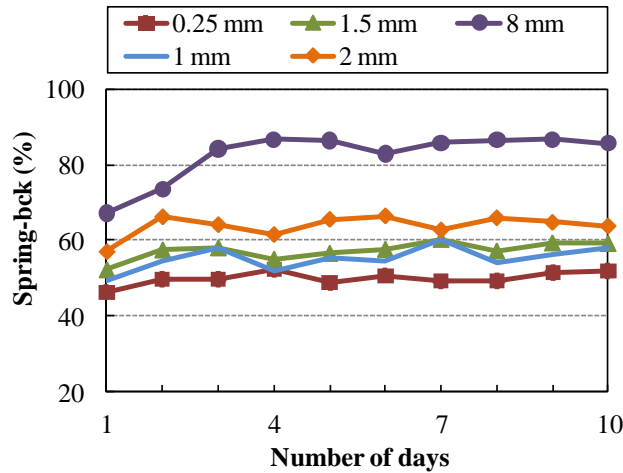


Figure 3.6 shows the results of pellet spring-back for one-sized particles. Spring-back increased during the first two days and became stabilized afterward. Smaller particles produced smaller spring-back.

**Figure 3.6 Effects of particle size on pellet spring-back**

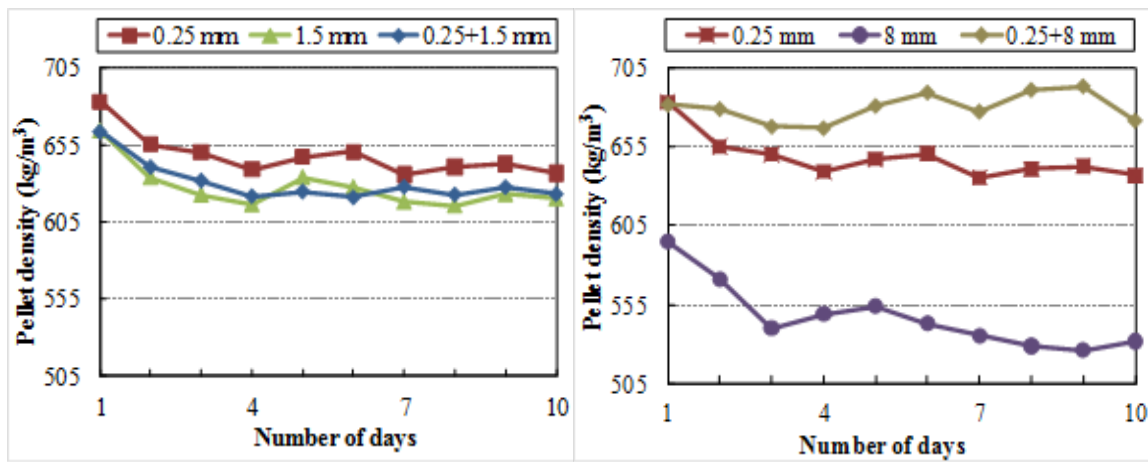


### 3.3.2 Results on mixed-sized particles

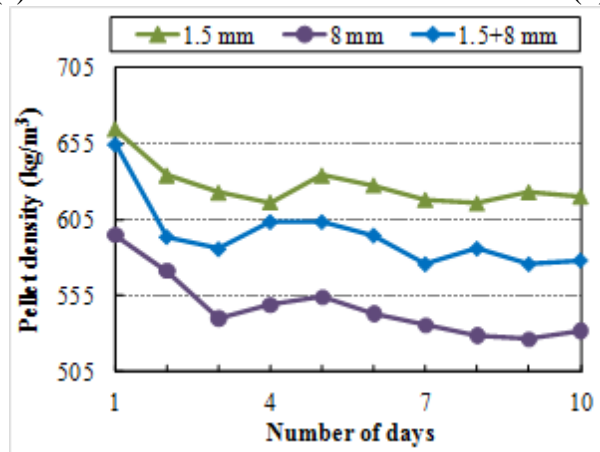
The results of pellet density for mixed-size particles are shown in Figure 3.7. In some conditions, the pellet density generated by the mixed-size particles was higher than those generated by the one-size particles. For example, the pellet density of 0.25 mm particle size mixed with 8 mm particle size was higher than that of either individual particle size. In other conditions, the pellet density generated by the mixed-size particles was between those generated by the one-size particles.

The results of pellet spring-back for mixed-size particles are shown in Figure 3.8. The spring-backs of the pellets with mixed-size particles were between those of the pellets with one-size particles.

Figure 3.7 Effects of particle size on pellet density for mixed-size particles

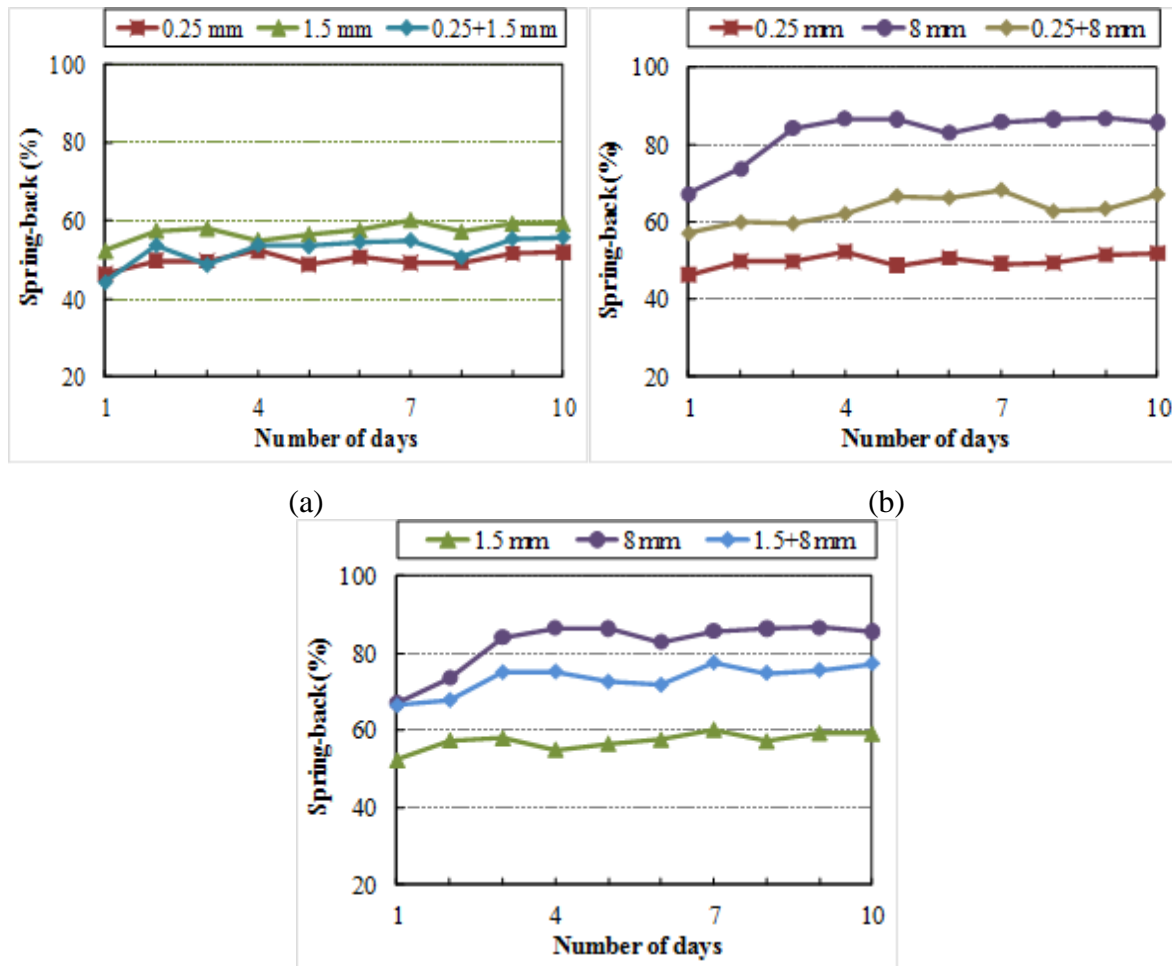


(a) (b)



(c)

**Figure 3.8 Effects of particle size on pellet spring-back for mixed-size particles**



### 3.4 Conclusions and future work

This paper investigates the effects of biomass particle size on pellet density and spring-back in ultrasonic vibration assisted (UV-A) pelleting of wheat straw. The following conclusions can be drawn from the study:

1. Particle size has significant effects on pellet density and spring-back. Smaller particles can produce higher density and smaller spring-back.
2. In some conditions, mixed-size particles can produce higher density than one-size particles. The spring-back of the pellets with mixed-size particles are between those of the pellets with one-size particles.

Besides the effects of particle size on pellet characteristic, the author's group are (or will be) conducting research on other aspects of UV-A pelleting of cellulosic biomass. The research

includes analysis of the energy required for pelleting versus the energy saving in subsequent processes (such as transportation, pretreatment, and biofuel conversion), analysis of total costs in UV-A pelleting versus conventional pelleting methods (such as those using high-temperature steam), and experimental investigations into effects of UV-A pelleting on pretreatment and biofuel conversion as well as the mechanism of these effects. The results of these research activities will be published in separate paper.

### **Acknowledgements**

The authors would like to thank Daniel Nottingham and Emily Jones for their help on experiments, and measurements. The authors also would like to thank Sonic-Mill, Inc. for the technical support.

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## **Chapter 4 - Effects of pressure and ultrasonic power on pellet quality and sugar yield**

### Paper Title:

Ultrasonic vibration assisted pelleting of sorghum stalks: effects of pressure and ultrasonic power

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## **Abstract**

Cellulosic biofuels can be used to replace traditional liquid transportation fuels. Cellulosic biomass is feedstock in manufacturing of cellulosic biofuels. However, the low density of cellulosic biomass feedstock hinders large-scale and cost-effective manufacturing of

cellulosic biofuels. Another bottleneck factor in manufacturing of cellulosic biofuels is the low efficiency of the enzymatic hydrolysis of cellulosic biomass materials resulting in a low sugar yield. Ultrasonic vibration-assisted (UV-A) pelleting can increase the density of cellulosic biomass feedstocks via combined effects of mechanical compression and ultrasonic vibration of the tool on the cellulosic biomass. Meanwhile ultrasonic vibration may act as a beneficial pretreatment for enzymatic hydrolysis, which can possibly increase the efficiency of hydrolysis and obtain a higher sugar yield. The pressure and the ultrasonic power are important parameters in UV-A pelleting. Their effects on pellet quality (density, durability, and stability) and sugar yield (after hydrolysis) are experimentally investigated.

### **Keywords**

Biofuel, cellulosic biomass, durability, pellet, ultrasonic vibration

## **4.1 Introduction**

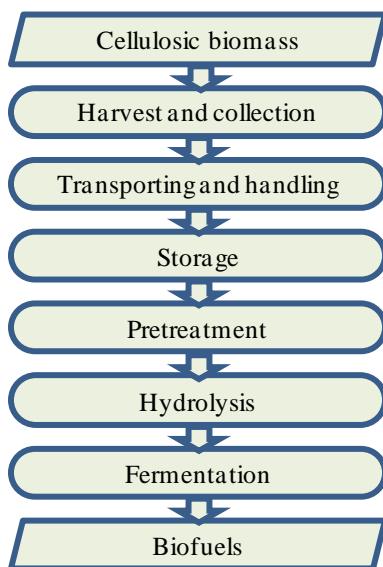
In the last decade, the consumption of petroleum for transportation in the U.S. increased dramatically [1]. In 2008, the daily consumption of the liquid transportation fuels was about 13.7 million barrels in the U.S. [1]. Traditional liquid transportation fuels (including gasoline, ethane, and diesel fuel) are distilled from petroleum [2]. The rapidly increasing demand of liquid transportation fuels makes the U.S. more dependent on imported petroleum [3]. The volatile prices of petroleum and need of reducing imports from foreign countries necessitate the development of renewable energy sources to replace traditional liquid transportation fuels [3,4].

Among all renewable energy sources, cellulosic biofuels (including biodiesel and ethanol) represent an attractive alternative source that could partially replace traditional liquid transportation fuels.

Using cellulosic biomass can reduce greenhouse gas emissions by 85% over petroleum-based fuels [6]. The cellulosic biomass resource is abundant, 1.3 billion dry tons of cellulosic biomass could be produced each year in the U.S., from which enough ethanol could be made to replace more than 50% of the gasoline [7]. Furthermore, unlike ethanol from corn and other feedstock (e.g. sugar cane, and soybeans), cellulosic biofuels have no effects on food supply, and do not result in higher food price [8]. Thus, cellulosic ethanol production can reduce petroleum imports, create jobs, and improve national energy security [5].

However, there are only pilot biofuel plants using cellulosic feedstock in the U.S. currently [9]. Figure 4.1 shows major steps for manufacturing of cellulosic biofuels. There are several barriers for large-scale and cost-effective manufacturing of cellulosic biofuel [11,12]. One of such barriers is the high cost of transportation and storage of biomass due to the low density of cellulosic biomass feedstocks [11,13,14]. Another is the low efficiency of the enzymatic hydrolysis [16-19].

**Figure 4.1 Major steps for manufacturing of cellulosic biofuels (after [9,10])**



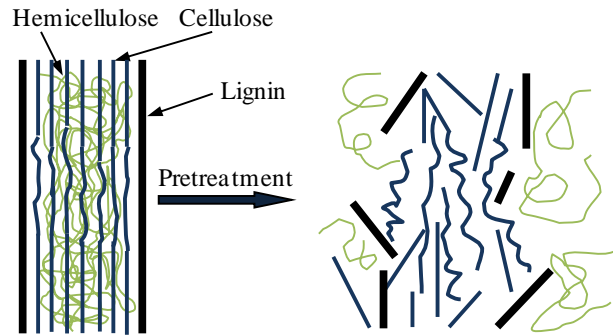
Pelleting can improve the density of biomass feedstocks [19]. Traditional pelleting methods (e.g. using a screw extruder, a briquetting press, or a rolling machine) usually require high-temperature steam, high pressure, and binder materials [18,19]. With these methods, it is difficult to achieve cost-effective pelleting on or near the field where cellulosic biomass is available. Ultrasonic vibration-assisted (UV-A) pelleting does not use high-temperature steam and binder materials, and can be used to increase the density of biomass feedstock via the combined effects of mechanical compression and ultrasonic vibration of the tool on the cellulosic biomass. Preliminary studies show that UV-A pelleting can produce biomass pellets whose density is comparable to that processed by traditional pelleting methods [18,19].

UV-A pelleting can also act as a pretreatment process. The purpose of pretreatment in cellulosic biofuel manufacturing is shown in Figure 4.2. Pretreatment can break the lignin seal and disrupt the crystalline structure of cellulose, increasing its surface area and making it more

accessible to enzyme hydrolysis [6]. In UV-A pelleting, the ultrasonic vibration of the tool might disrupt lignin and crystalline structure of cellulose and subsequently lead to higher sugar yield in hydrolysis. Pilot tests show that, without any other pretreatment, UV-A pelleting can increase the sugar yield by more than 70% compared with pelleting without ultrasonic vibration [19]. With pretreatment following the NREL LAP procedure [20], sugar yield of pellets produced by UV-A pelleting is more than 20% higher than that of the pellets produced by pelleting without ultrasonic vibration [19].

The pressure and the ultrasonic power are important parameters in UV-A pelleting. In this paper, their effects on pellet quality (density, durability, and stability) and sugar yield (after hydrolysis) are experimentally investigated. Various experiments have been conducted and it is very difficult to report all experiment results in one paper. Experiments to compare pelleting results with versus without ultrasonic vibration have been conducted and the results will be reported in a separate paper.

**Figure 4.2 Purpose of pretreatment in cellulosic biofuel manufacturing (After [6])**



## **4.2 Experimental procedure and parameters**

### ***4.2.1 UV-A pelleting***

Major steps of ultrasonic vibration-assisted (UV-A) pelleting are shown in Figure 4.3. Sorghum stalks, the cellulosic biomass material used in this experiment, were milled into particles using a cutting mill (model SM 2000, Retsch, Inc., Haan, Germany) with the sieve size of 1.5 mm, as shown in Figure 4.4. The moisture content of the biomass particles after milling was measured and adjusted to 9% according to ASABE Standard S358.2 [21].

UV-A pelleting was performed on a Sonic Mill Series 10 machine (Sonic-Mill, Albuquerque, NM, U.S.). A schematic illustration of UV-A pelleting is shown in Figure 4.5. A pneumatic cylinder, on top of the converter, is controlled by a pressure regulator and connected to an air compressor. It applies a compressive pressure to the biomass material through the tool. Meanwhile, an ultrasonic vibration, provided by the ultrasonic converter, is applied to the tool during the pelleting process. A tool with a flat end was fed towards the biomass at preset pressures and ultrasonic power. An aluminum mold with a cylindrical cavity in the center is used to hold the sorghum particles (3 grams) and fixed by a fixture. The diameter of the tool (17.4 mm) is slightly smaller than that of the cavity in the mold (18.6 mm).

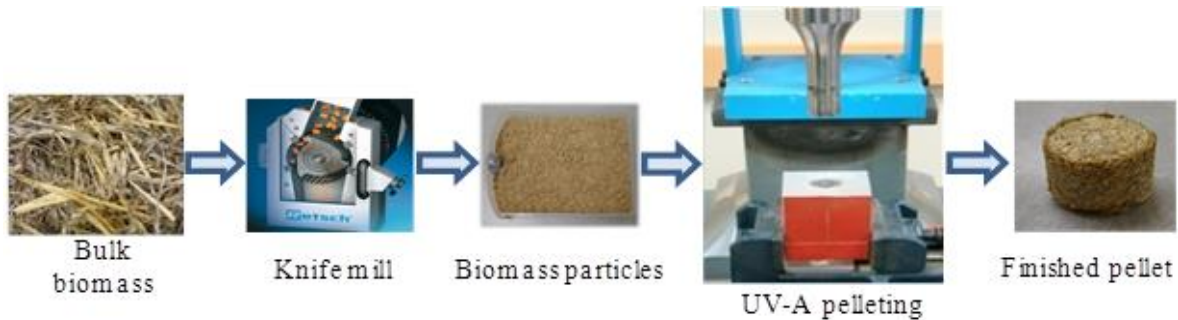
#### ***4.2.2 Important parameters in UV-A pelleting***

The pressure represents the air pressure in the pneumatic cylinder. A higher air pressure in the pneumatic cylinder means a higher compressive pressure applied on the sorghum particles. Four levels of pressures were used. They were 20, 30, 40, and 50 psi while ultrasonic power was fixed at 40%.

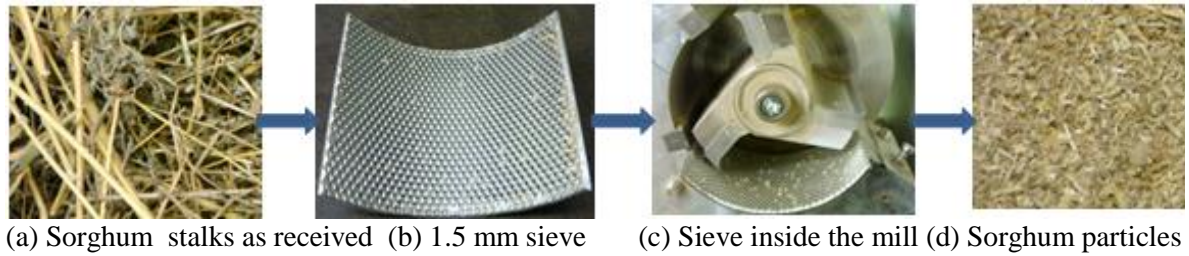
Ultrasonic power, supplied by a power supply, controlled the vibration amplitude of the tool. A higher ultrasonic power means higher amplitude of vibration. The ultrasonic power was adjusted and ranged from 0 (no ultrasonic power) to 100% (the maximum ultrasonic power). In this experiment, four ultrasonic powers (30%, 40%, 50%, and 55%) were used while pressure was fixed at 40 psi.

The frequency of the ultrasonic vibration is 20 kHz. For each pellet, three grams of sorghum particles was compressed for 2 minutes.

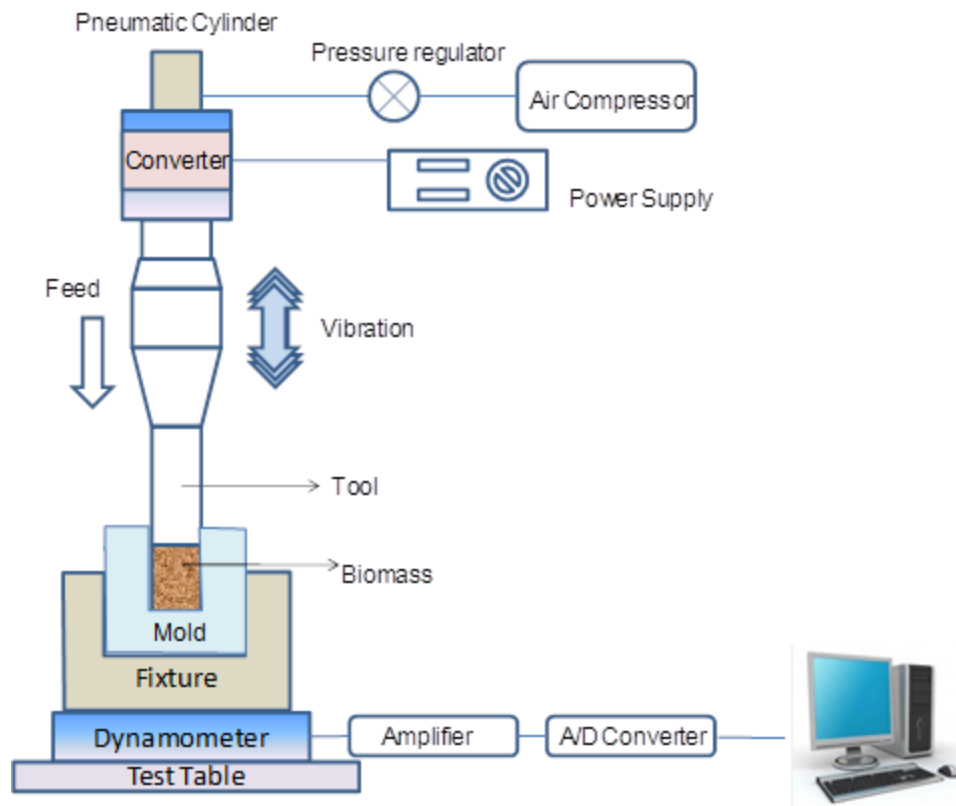
**Figure 4.3 Major steps for UV-A pelleting experiments**



**Figure 4.4 Control of particle size**



**Figure 4.5 Illustration of UV-A pelleting**



### **4.3 Measurement methods for the output variables**

#### **4.3.1 Pellet density**

The density of a pellet is determined by the ratio of its weight to its volume. The weight of a pellet was measured with an electronic scale (Ohaus, Pine Brook, NJ, U.S.). The volume of a

pellet was determined by its diameter and height measured with a vernier caliper. More than 55 pellets were made and five pellets were measured for each level of pressure and ultrasonic power. The density of each pellet was measured once a day for 10 days and the average of the five pellets was taken as the value of pellet density for each condition.

#### ***4.3.2 Pellet durability***

Pellet durability is the ability of the pellet to withstand impact and other forces encountered during handling and transportation [22]. Pellet durability was measured based on the ASABE standard S269.4 [22] with some modifications. Fifty grams of pellets were kept tumbling inside a pellet durability tester (Seedburo Equipment, Des Plaines, IL, USA) for 10 minutes and then sieved through a U.S. No. 6 sieve. The pellet durability was calculated as the ratio of the weight of the remaining pellets (that did not fall through the No. 6 sieve) after tumbling to the weight of the pellets before tumbling.

#### ***4.3.3 Pellet stability***

Stability of a pellet was determined by evaluating changes in its dimensions (or volume) with time. A general trend was observed: the volume of the pellets would increase (spring-back) with time after they were taken out from the mold. The stability of the pellets was measured by the spring-back of the pellets which was calculated by

$$\text{Spring-back} = \frac{\text{Stable pellet volume} - \text{Initial pellet volume}}{\text{Initial pellet volume}}$$

where, stable pellet volume is the volume of pellets measured when their volume become stable, and initial pellet volume is the volume of the pellet measured right after the pellet is taken out from the mold. Five pellets were measured for each level of pressure and ultrasonic power. The volume of each pellet was measured once a day for 10 days. Pilot tests showed that the pellet volumes became stable after three days since pelleting [19]. The stable volume for a pellet was then determined by taking the average value of the volumes of each pellet from day 4 to day 10.

#### ***4.3.4 Sugar yield***

Before hydrolysis, pretreatment was carried out in a pressure reactor apparatus (Parr Instrument Company, Moline, IL, USA) equipped with impeller mixers and a pressurized



injection device. Dissociated sorghum pellets were mixed with a sulfuric acid solution to obtain 10% dry matter (weight/volume). The slurry was loaded into a 1-L reactor and treated at a temperature of 180°C for 15 minutes with diluted sulfuric acid (at a ratio of 20 g per liter of distilled water). Enzyme hydrolysis of pretreated biomass was conducted, following the NREL LAP procedure [20], in sealed serum bottles in a 50°C reciprocal water bath shaker running at 100 rpm for 72 hours. The enzyme used in the hydrolysis was Accellerase 1500 from Genencor (Rochester, NY, USA). After hydrolysis, a high performance liquid chromatography (HPLC) was used to measure the sugar yield.

## 4.4 Experimental results

### 4.4.1 Results on pellet density

Figure 4.6 shows effects of different pressures levels (20, 30, 40, and 50 psi) on pellet density. There was a dramatic increase on pellet density as the pressure increased from 20 to 30 psi and to 40 psi. However, the pellet density was almost the same for pressure levels of 40 psi and 50 psi.

**Figure 4.6 Effects of pressure on pellet density**

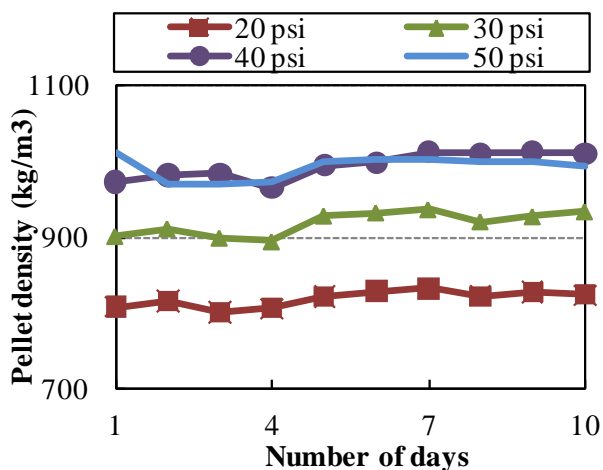
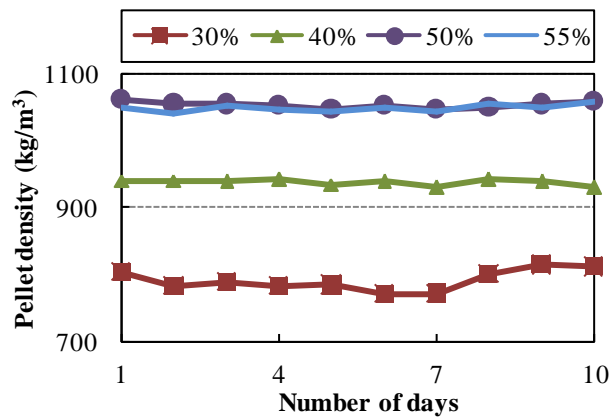


Figure 4.7 shows effects of ultrasonic power on pellet density. The pellet density increased significantly as the ultrasonic power increased from 30 to 50%. When the ultrasonic power was increased from 50 to 55%, the pellet density did not change much.

**Figure 4.7 Effects of ultrasonic power on pellet density**



#### ***4.4.2 Results on pellet durability***

Effects of pressure on pellet durability are shown in Figure 4.8. As the pressure increased from 20 to 40 psi, the durability of pellets increased. However, the pellet durability had a dramatic drop when the pressure was 50 psi.

**Figure 4.8 Effects of pressure on pellet durability**

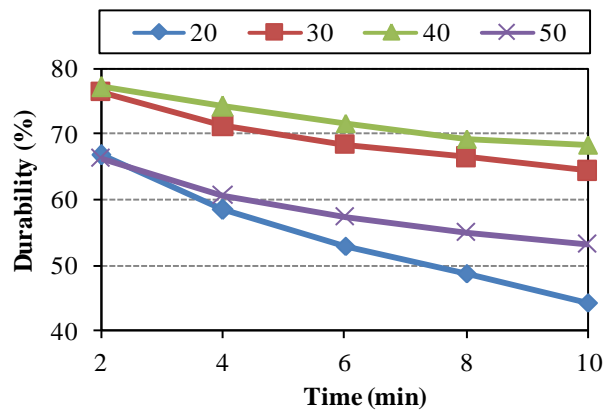
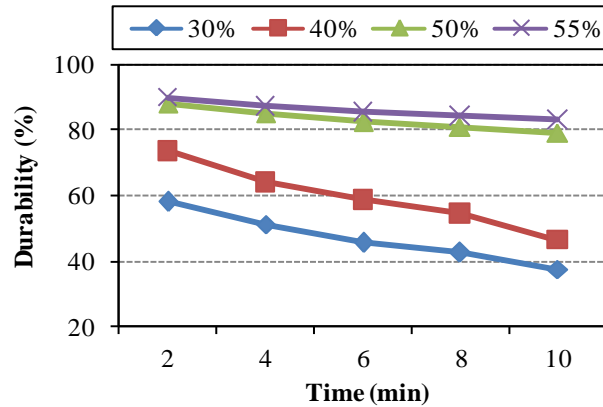


Figure 4.9 shows effects of ultrasonic power on pellet durability. The durability of pellets increased as ultrasonic power increased. Ultrasonic power level of 50% and 55% resulted in almost the same pellet durability.

**Figure 4.9 Effects of ultrasonic power on pellet durability**



#### ***4.4.3 Results on pellet stability***

Figure 4.10 shows effects of pressure on pellet stability (spring-back). As the pressure increased from 20 to 50 psi, there was no significant difference in spring-back between these four pressure levels. In other words, effects of pressure on pellet stability were not obvious.

**Figure 4.10 Effects of pressure on pellet stability**

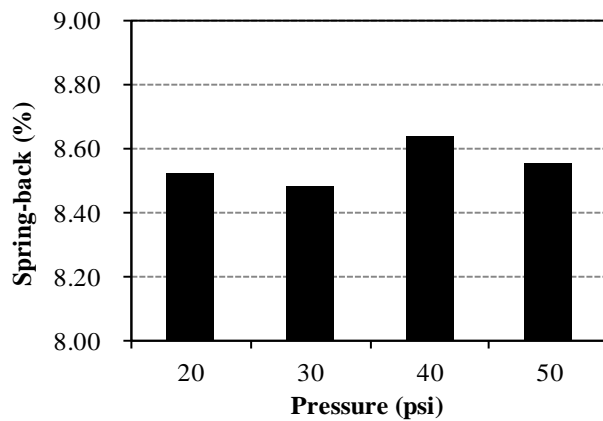
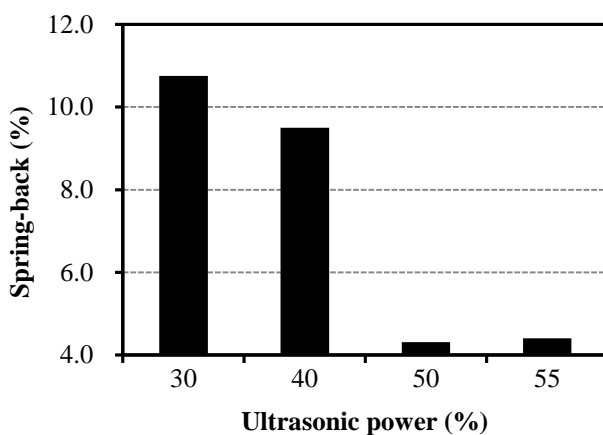


Figure 4.11 shows effects of ultrasonic power on pellet stability (spring-back). As the ultrasonic power increased from 30% to 40%, the spring-back decreased slightly. When the ultrasonic power increased from 40% to 50%, the spring-back of the pellets underwent a dramatic decrease. There was little difference in spring-back values between 50% and 55% ultrasonic power levels. In summary, spring-back decreases as the ultrasonic power increases from 30% to 50%.

#### 4.4.4 Results on sugar yield

Figure 4.12 shows effects of pressure on sugar yield. As the pressure increased from 20 to 50 psi, the sugar yield decreased first and then increased. The lowest sugar yield was obtained at the pressure of 40 psi. There was not much difference between the sugar yields with the pressure of 20 and 50 psi.

**Figure 4.11 Effects of ultrasonic power on pellet stability**



**Figure 4.12 Effects of pressure on sugar yield**

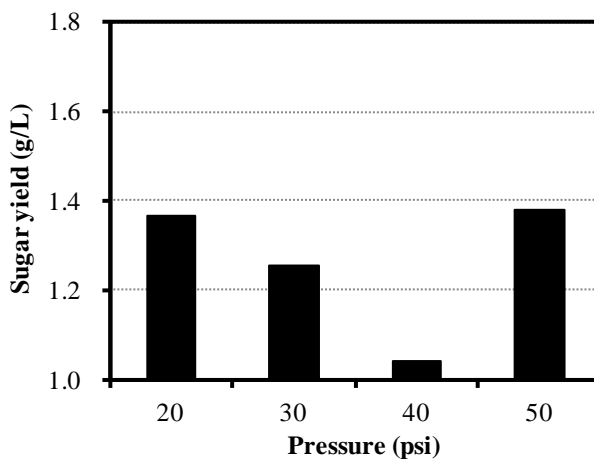
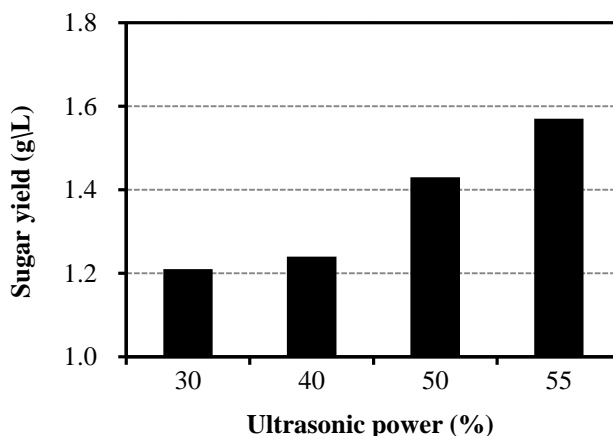


Figure 4.13 shows effects of ultrasonic power on sugar yield. The sugar yield increased about 20% as the ultrasonic power increased from 30% to 55%.

**Figure 4.13 Effects of ultrasonic power on sugar yield**



## **4.5 Conclusions**

This paper presents the results of experimental investigations into the effects of pressure and ultrasonic power on pellet quality (density, durability, and stability) and sugar yield in ultrasonic vibration-assisted (UV-A) pelleting of sorghum stalks. A higher pellet density could be obtained by increasing the pressure or ultrasonic power. As the pressure increased, the pellet durability increased first and then decreased. The pellet durability increased as ultrasonic power increased. Pressure had little effects on pellet stability. Pellet stability increased as ultrasonic power increased from 30% to 50%. Sugar yield decreased first and then increased with an increasing pressure. A higher ultrasonic power led to higher sugar yield.

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## **Chapter 5 - Comparison of pellet quality and energy consumption in two pelleting methods**

### Paper Title:

An experimental comparison of two pelleting methods for cellulosic ethanol manufacturing

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## **Abstract**

Ethanol produced from cellulosic biomass is an alternative to petroleum-based transportation fuels. However, manufacturing costs of cellulosic ethanol are too high to be competitive. Low density of cellulosic feedstocks increases their handling and transportation costs, contributing to high overall costs of cellulosic ethanol manufacturing. Pelleting can increase density of cellulosic feedstocks, reduce transportation and storage costs, and make cellulosic ethanol production more competitive. UV-A (ultrasonic vibration-assisted) pelleting is a new pelleting method available only in lab scale now. Preliminary research showed that UV-A pelleting could significantly increase pellet density and pellet durability but it has never been compared with other pelleting methods (e.g., using an extruder, a briquetting press or a ring-die pelleting). The objectives of this research are to compare UV-A pelleting with ring-die pelleting in terms of pellet density, pellet durability, energy consumptions of pelleting. The results will be useful to find a better pelleting method for cellulosic ethanol manufacturing.

### **Keywords**

Cellulosic biomass, Density, Durability, Energy consumption, Ethanol, Pellet, Sugar yield, Ultrasonic vibration

## **5.1 Introduction**

Petroleum-based liquid transportation fuels (including gasoline, diesel, and jet fuels) account for 70% of U.S. oil consumption [1]. The increasing demand of liquid transportation fuels in the U.S. has been far beyond its domestic production capacity, making the nation rely on foreign supplies [2]. In 2009, the imported oil was about 15 million barrels per day [3]. Use of petroleum-based liquid transportation fuels contributes to the accumulation of GHG (greenhouse gas) in the atmosphere. Therefore, there is an urgent need for alternative fuels that can reduce GHG emissions and U.S dependence on imported oil [4,5].

Ethanol, used as a substitute for gasoline or a gasoline additive, is an alternative fuel in the transportation sector. Total ethanol production capacity of 13.1 billion gallons are estimated in 2010 [6], of which majority is produced from corn. Using 100% of the corn crop in the U.S. for ethanol production would meet about 17% of the nation's needs [7]. Furthermore, a dramatic increase in ethanol production using current grain-starch-based technology may be limited by the

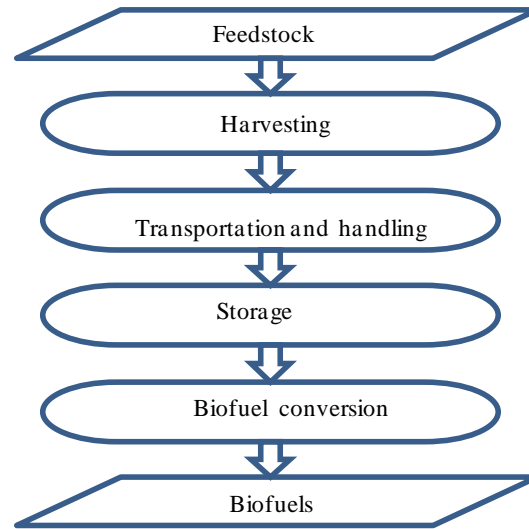
fact that grain production of ethanol will compete for limited agricultural land with food and feed production.

Ethanol made from cellulosic biomass (herbaceous, woody, and generally inedible portions of plant matter) is promoted by the U.S Department of Energy (DOE) [8]. Cellulosic biomass is abundant. Land resources in the U.S. is sufficient to produce 1.3 billion dry tonne of cellulosic biomass annually, from which enough ethanol can be made to replace over 50% of gasoline used currently in the U.S. [10]. Using cellulosic ethanol can reduce GHG emissions by 85% over petroleum-based fuels [9,11].

Low density of cellulosic biomass materials results in high costs in transportation and storage. Figure 5.1 shows major steps in manufacturing of cellulosic ethanol [12,13]. After harvesting, biomass bales are transported from fields to warehouse and stored for future use. The bulky characteristic of biomass feedstocks makes it difficult to transport and storage them, resulting in high cost [4,14-15].

Densification of cellulosic biomass into pellets can significantly increase pellet density, resulting in low transportation and storage costs [16]. Density of pelleted feedstocks can be (600-800 kg/m<sup>3</sup>), much higher than that of loose biomass (40-250 kg/m<sup>3</sup>) [17,18]. Meanwhile, pellets with the uniform size and shape are easier to handle using existing handling and storage equipment. Furthermore, transportation and handling costs of pelleted cellulosic biomass are much lower than those of baled or chopped cellulosic biomass, as shown in Table 1. Physical properties of pellets (including pellet density and pellet durability) are considered when selecting equipment and processes for cellulosic ethanol production [21]. In addition, energy consumption is also a concern which directly impacts cost of ethanol production.

**Figure 5.1 Major steps in manufacturing of cellulosic ethanol (after [12,13])**



**Table 5.1 Transportation and handling costs of cellulosic biomass [19, 20]**

	Transportation cost (\$/dry tonne)	Handling cost (\$/dry tonne)
Bales	9.98	24.64
Chops	16.33	21.76
Pellets	4.49	13.38

UV-A pelleting has the combined effects of mechanical compression and ultrasonic vibration of the tool on the cellulosic biomass. It can significantly increase pellet density and pellet durability. There are many traditional pelleting methods (e.g., using an extruder, a briquetting press or a ring-die pelleting mill). The objective of this research is to compare two pelleting methods: UV-A pelleting and ring-die pelleting mill in terms of pellet density, pellet durability and energy consumptions of pelleting. This is the first paper to compare UV-A pelleting with traditional pelleting methods.

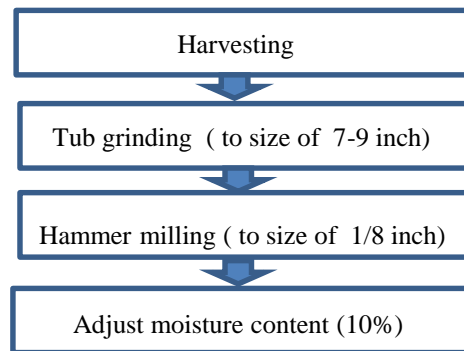
## **5.2 Experimental conditions**

### ***5.2.1 Preparation of biomass materials***

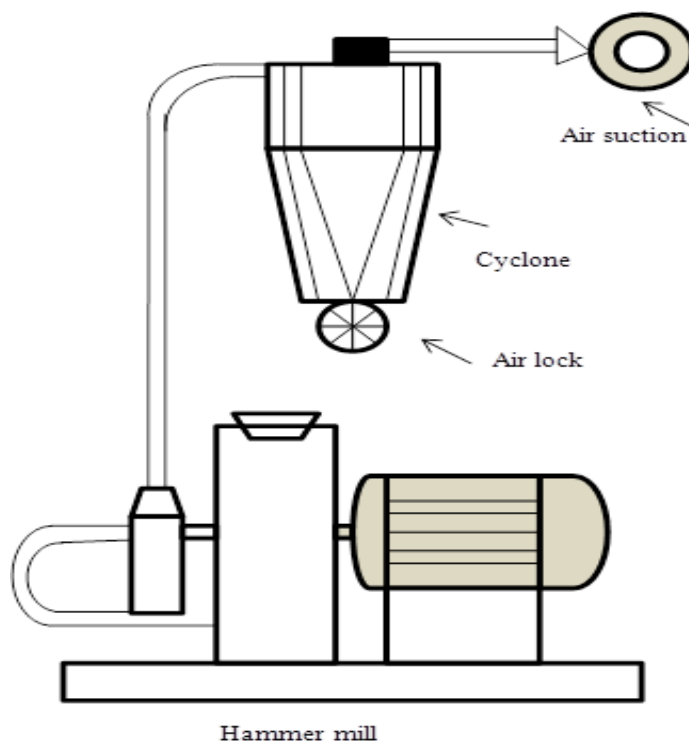
Sorghum stalks were harvested by the Kansas State Agronomy Farm in December of 2009. Figure 5.2 shows the material preparing steps before pelleting. After harvesting, sorghum stalks were chopped to particle size to approximately 17.8-22.9 cm (7-9 inches) in length using a large tub grinder (Haybuster H-1150 series, DuraTech Industries International Inc., Jamestown,

ND). The tub grinder was powered by a diesel engine and capable of grinding a large round bale (1.8 m x 1.2 m, height x diameter) in less than 30 seconds. Those bales were then transported to Bioprocessing and Industrial Value Added Program (BIVAP) building located in Kansas State University. The feed materials were manually loaded onto a belt conveyor which fed into a 7.4 kW (10 hp) hammer mill (Schutte-Buffalo Hammer mill Model 18-7-300, Schutte Pulverizer Co., New York, NY). The particles of sorghum stalks were obtained using the hammer mill with a sieve size of 3.2 mm (1/8 inch). While milling, an air suction system and a cyclone were attached to the hammer mill to remove the milled particles, as shown in Figure 5.3. The purpose of cyclone was to separate the dust particles from air by centrifugal force. The dust particles were left on bottom because of higher density. The milled sorghum stalks were collected and kept in sealed paper bags at room temperature. Before pelleting, the moisture content of sorghum stalks was adjusted to 10% (dry basis) by mixing tap water with sorghum stalk particles at room temperature for 2 mins. The procedure of adjusting moisture content was based on ASABE Standard S385.2 [22]. No external binding agents were added in both pelleting experiments.

**Figure 5.2 Steps of prepare sorghum stalks for pelleting**



**Figure 5.3 Illustration of hammer mill (after [27])**



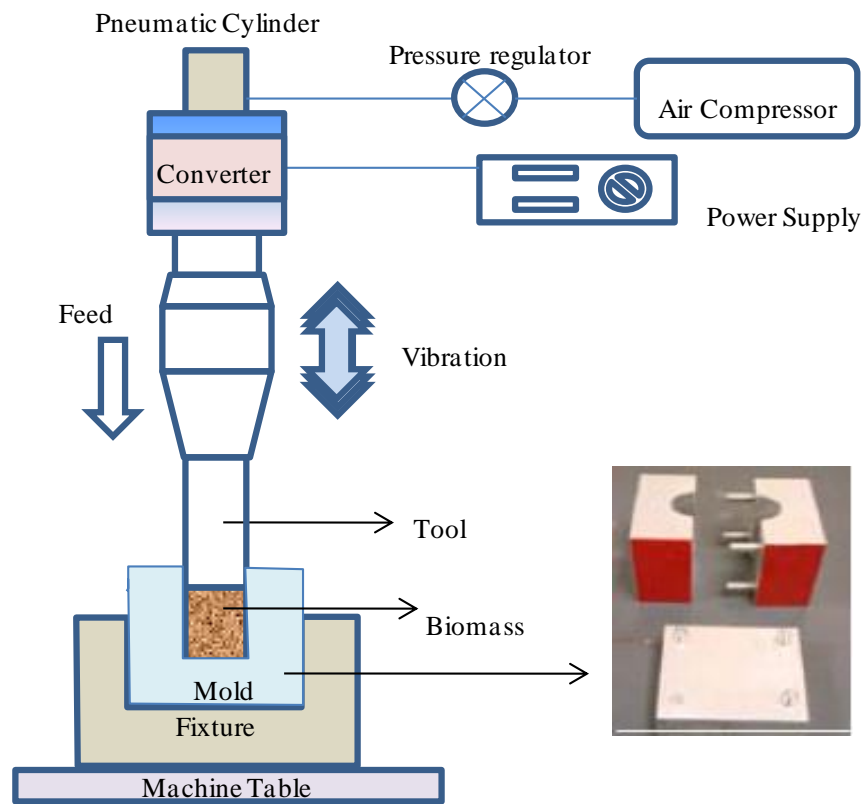
### ***5.2.2 UV-A Pelleting***

Figure 5.4 is a schematic illustration of the experimental set-up for UV-A pelleting. Pelleting is performed on an ultrasonic machine (Sonic-Mill, Albuquerque, NM, U.S.). An aluminum mold was made in three separate parts that were assembled together with pins. The top two parts formed a cylindrical cavity (18.6 mm in diameter) and the bottom part served as a base. Biomass was loaded into the center cavity of the mold and the mold was clamped by a fixture. The tool was connected to an ultrasonic converter. The tip of the tool was a solid cylinder with a flat end (17.4 mm in diameter). The tool was fed into the biomass in the mold. Meanwhile, the pneumatic cylinder applied a compressive pressure to biomass through the tool and the ultrasonic converter provided ultrasonic vibration to the tool. The pneumatic cylinder, on top of the converter, was controlled by a pressure regulator and connected to an air compressor. After a predetermined period of time during which the tool was in contact with the biomass, the tool was retracted and the mold was disassembled to unload the cylinder-shaped pellet.

Table 5.2 shows some experimental parameters and their values. Pelleting time was recorded with a stop watch. The recording began when the tool started dropping and stopped

when the tool started retreating. The pressure represented the air pressure in the pneumatic cylinder. A higher pressure meant a higher compressive pressure applied on the biomass in the mold. Ultrasonic power, supplied by a power supply, controlled the vibration amplitude of the tool. A higher ultrasonic power meant a higher vibration amplitude. The ultrasonic power could be adjusted from 0 (no ultrasonic power) to 100% (the maximum ultrasonic power).

**Figure 5.4 Illustration of experimental set-up for UV-A pelleting**



**Table 5.2 Values of pelleting parameters**

Parameter	Value
Pelleting time (s)	6.5
Pressure (psi)	40
Ultrasonic power (%)	100
Weight of each pellet (g)	1
Vibration frequency (kHz)	20

### ***5.2.3 Ring-die pelleting***

Ring-die pelleting experiments were conducted using a 22.1 kW (30 hp) ring-die pelleting mill with 1.5 ton capacity (CPM Master Model Series 2000, California Pellet Mill Co., Crawfordsville, IN), as shown in Figure 5.5. Each test run lasted for 10-11 minutes and used 25.025 lbs of biomass. The die size was 6.35 mm x 31.75 mm (0.25 inch x 1.25 inch), hole diameter x effective thickness. The main shaft was operated at 10,650 rpm. The feeder screw was rotating at 7 rpm. As shown in Figure 5.5 (c), the die was turning clockwise, causing two rollers to turn in the same direction. Cylinder-shaped pellets,  $(18.9 \pm 1.6 \text{ mm}) \times (6.4 \pm 0.1 \text{ mm})$ , height x diameter, were produced by extruding the sorghum stalk particles through the channels of the ring die. Frictional heating during pelleting resulted in increasing of pellet temperature. The temperature of pellets exiting the die was between 74°C - 84°C.

## **5.3 Evaluation parameters and their measurement procedures**

### ***5.3.1 Pellet density***

Pellet density, or true density of the pellets, means the density of an individual pellet and was computed by the following equation:

$$\text{Pellet density} = \frac{\text{Weight of the pellet}}{\text{Volume of the pellet}} \quad (1)$$

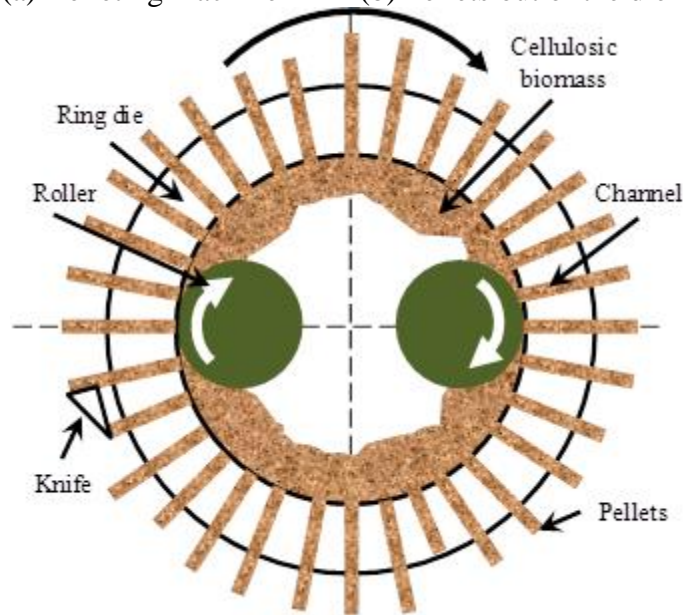
Ten samples of each kind were randomly selected. Weight of the pellet was measured by an electronic scale (Ohaus, Pine Brook, NJ), with four replications. Height and diameter of the cylinder-shaped pellet was measured using a vernier caliper, with four replications as well. Volume of the pellet was calculated by cylinder volume equation ( $\pi r^2 h$ ). The average value of the results was calculated and used in calculating pellet density.

**Figure 5.5 Experimental set-up for traditional pelleting**



(a) Pelleting machine

(b) Pellets out of the die



(b) Illustration of pelleting process

### **5.3.2 Pellet durability**

Pellet durability measures the ability of pellets to withstand impact and other destructive forces during transportation and storage. A higher durability is desirable in biofuel manufacturing because pellets of low durability can cause dust emissions resulting in a health hazard and an increased risk of fire explosion during pellet handling and storage [23,24]. Above 80% durability is considered high and below 70% durability is low [25].



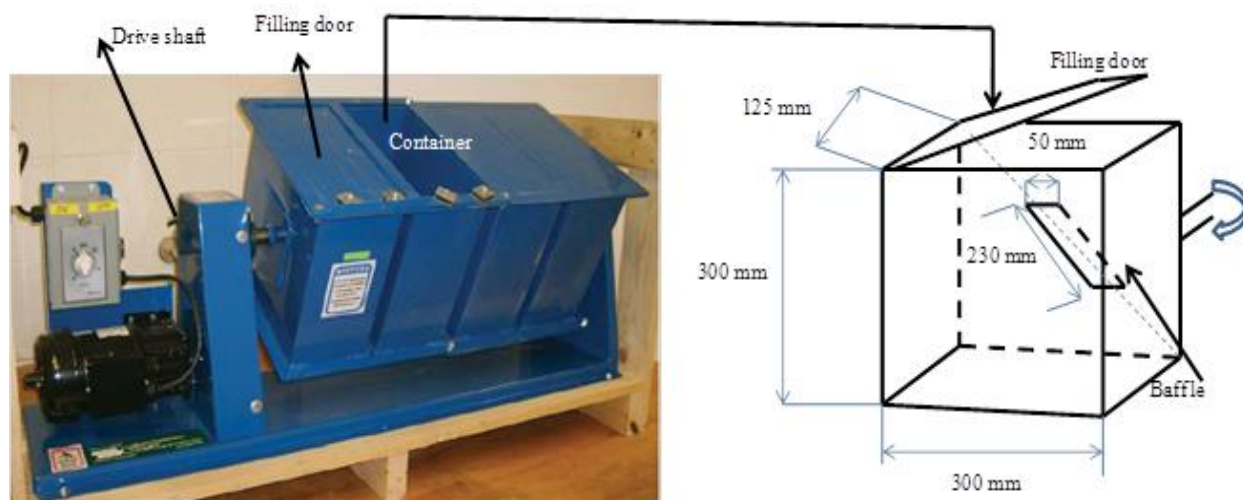
ASABE S269.4 [26] specifies a procedure to measure pellet durability. Pellet durability was determined with a durability tester (Seedburo Equipment, Des Plaines, IL) designed according to ASABE standard S269.4. The durability tester, as shown in Figure 5.6, had four rectangular containers, each with inner dimensions of 300 mm x 300 mm x 125 mm. A 230 mm long baffle, extended 50 mm inside each container, was affixed symmetrically to the diagonal of one side (300 mm x 300 mm) of the container. A drive shaft powered by a motor was used to rotate the containers around the axis centered and perpendicular to the sides (300 mm x 300 mm).

Fifty grams of each type of pellets were put in one of the containers and sealed by a filling door. While the containers rotated at 50 rpm, pellets were kept tumbling inside the container. After tumbling for 10 minutes, both types of pellets were taken out and sieved through a U.S. No. 6 sieve with a hole diameter of 3.15 mm. The weight of the remaining pellets that did not fall through the No. 6 sieve was measured with an electronic scale. Pellet durability was calculated by the following equation:

$$\text{Durability} = \frac{\text{Weight of pellets after tumbling}}{\text{Weight of pellets before tumbling}} * 100\% \quad (2)$$

In this study, durability tests were conducted with four replications for each type of pellets. The average value and standard deviation were calculated and plotted in Figure 5. 9.

**Figure 5.6 Pellet durability tester**



### ***5.3.3 Energy consumption in pelleting***

Energy consumption in this paper refers to electricity consumption since no hot steam was applied in both pelleting process. A power analyzer (AEMC 2010.86 PowerPad Jr. Model 8230, AEMC-Instruments, Foxborough, MA) was used to measure the electricity consumption, as shown in Figure 5.7 (a).

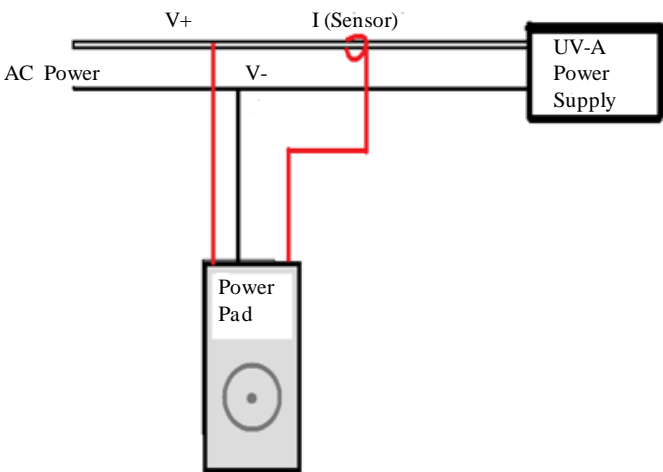
Figure 5.7 (b) and (c) illustrate electricity consumption measurements. Figure 5.7 (b) shows the measuring method for UV-A pelleting where a single phase power was used. A stripped power cable was connected in series with the UV-A machine. Voltage probe leads were connected to the 120 Volt AC source lines and a current sensor was clamped around the positive AC cable. This will allow the measurement device to take voltage/current readings. The power analyzer began recording data when the tool started dropping and stopped recording data when the tool started retreating. A set of ten pellets were made and electricity consumption data was collected for each pellet.

Figure 5.7 (c) shows the measuring method for ring-die pelleting where three phase power was used. There were L1, L2, and L3 cables which were connected in series with ring-die pelleting mill and all of them were connected to the AC power. The current sensor was clamped on L3. A positive wire was connected to L1 and negative wire was connected to L2. The power analyzer started recording data when the mill was turned on and stopped recording data when the mill was turned off. The measurements were conducted with three replications and measurement time was 11 minutes each run. For electricity consumption of both pelleting methods, the average value of measurements was calculated and used as source data of Figure 5.10. Results were obtained from the instruments as Wh/g and converted to kWh/ton in order to be used for industry comparison.

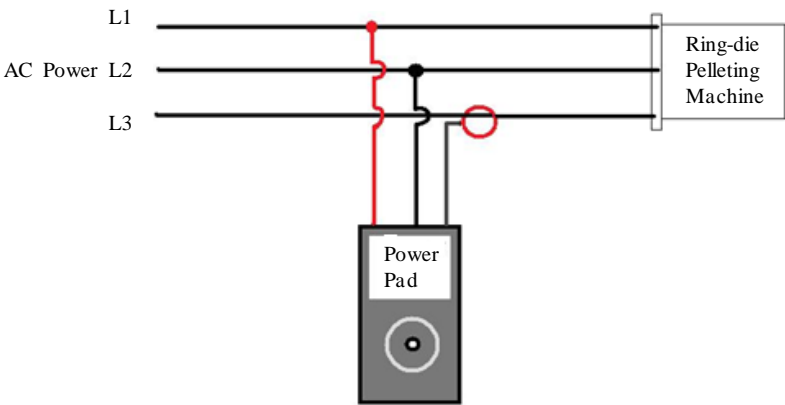
Figure 5.7 Illustrations of electricity consumption measuring methods



(a) Power quality analyzer (After [28])



(b) UV-A pelleting



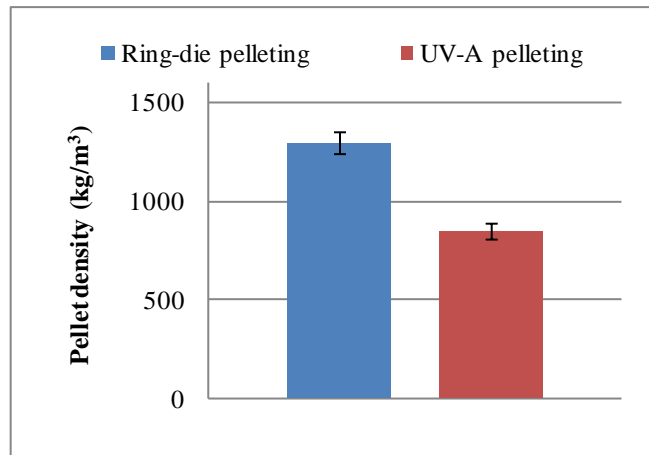
(c) Ring-die pelleting

## 5.4 Experimental results

### 5.4.1 Results on pellet density

Figure 5.8 shows results on pellet density. Although both pelleting methods could significantly increase pellet density, pellets produced by ring-die pelleting had higher density than those by UV-A pelleting. The error bars represent the standard deviation of the data.

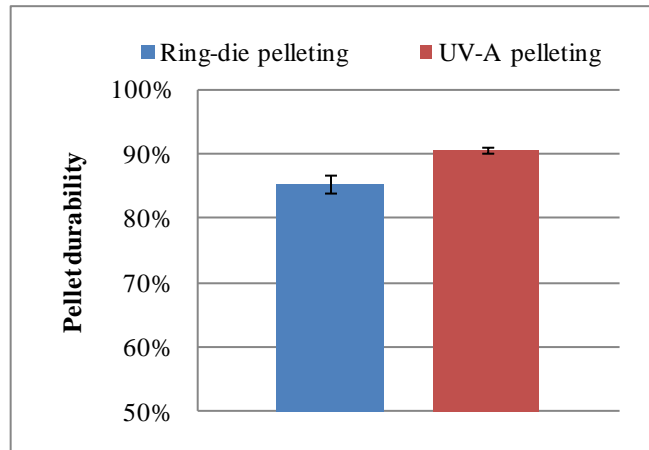
**Figure 5.8 Results on pellet density**



### 5.4.1 Results on pellet durability

Figure 5.9 shows results on pellet durability. Pellets produced by both pelleting methods had a durability value of above 80%, indicating that they were highly durable. Pellets produced by UV-A pelleting were about 5.9% higher than those by ring-die pelleting. Both standard deviations were very small, 0.5% for UV-A pelleting and 1.5 % for ring-die pelleting.

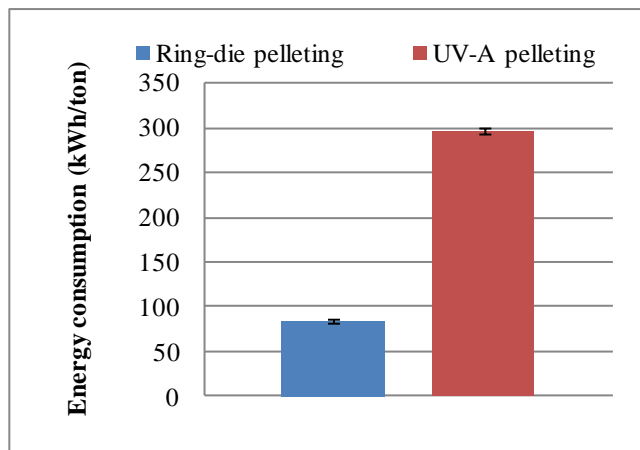
**Figure 5.9 Results on pellet durability**



#### ***5.4.2 Results on energy consumption***

Figure 5.10 shows the results on energy consumption. Energy consumption of UV-A pelleting (298 kWh/ton) was 3.5 times higher than that of ring-die pelleting (84.6 kWh/ton). The lab scale of UV-A pelleting setup limited its efficiency. With a production-scale setup, the efficiency should be greatly improved.

**Figure 5.10 Results on pellet durability**



### **5.5 Conclusions and remarks**

In this paper, data were obtained on pellet physical properties (i.e., pellet density and pellet durability) and energy consumption in two pelleting methods: ring-die pelleting and UV-A pelleting. The following conclusions can be drawn from the study:

Pellets produced by ring-die pelleting had higher density than those produced by UV-A pelleting.

Both pelleting methods could significantly increase pellet durability. Pellets produced by both pelleting methods were very durable with durability above 80 %.

To produce the same amount of pellets, ring-die pelleting consumed less electricity than UV-A pelleting.

Above conclusions shows that ring-die pelleting method is superior to UV-A pelleting method in terms of pellet density and electricity consumption. However, sugar yield comparison has not been done yet. The sugar yield is proportional to ethanol yield and directly related to overall costs of ethanol manufacturing. It is desirable to compare the effects of pelleting methods.

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## **Chapter 6 - Sugar yield comparison of wheat straw processed by two pelleting methods**

### Paper Title:

Sugar yield comparison of wheat straw processed by two pelleting methods for cellulosic biofuel manufacturing

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### **Abstract**

Biofuels made from cellulosic biomass are an alternative to petroleum-based liquid transportation fuels. However, low density of cellulosic biomass causes high costs in biomass transportation and handling in cellulosic biofuel manufacturing. Such costs can be reduced by pelleting processes that can densify cellulosic biomass. Ultrasonic vibration-assisted (UV-A) pelleting and ring-die pelleting are two pelleting methods. A previous study has compared the

two pelleting methods in terms of pellet quality and pelleting power consumption. This paper reports an experimental comparison on sugar yields of wheat straw processed by the two pelleting methods under different combinations of pretreatment variables. Results show that wheat straw processed by UV-A pelleting has higher sugar yield than that processed by ring-die pelleting when higher temperature and longer time are applied in pretreatment. Under other combinations of pretreatment variables, wheat straw processed by UV-A pelleting has lower sugar yield than that processed by ring-die pelleting.

## **6.1 Introduction**

Biofuels are an alternative to petroleum-based liquid transportation fuels (gasoline, diesel, and jet fuels). Biofuels can reduce the U.S.'s dependence on foreign petroleum, and cut GHG emissions while continuing to meet the nation's needs for liquid transportation fuels [1-3]. The U.S. government has called for an annual production of 36 billion gallons of biofuels by 2022 [4]. Ethanol is the most widely used biofuel [5]. Cellulosic biomass (such as wood, waste paper, and crop residues) has become more and more attractive as feedstocks for ethanol manufacturing because cellulosic biomass is abundant, relatively inexpensive, and outside the human food chain [6].

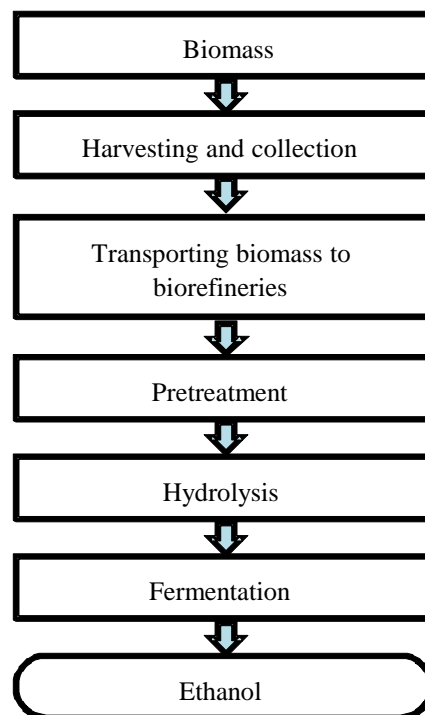
Major manufacturing processes for cellulosic ethanol are shown in Figure 6.1. Different harvesting and collection systems are used to harvest biomass and bale it into rectangular or round bales [9]. The bales are transported to biorefineries where biomass is converted to ethanol. The density of the bales is low (range from 40 to 250 kg/m<sup>3</sup>) [10,11]. The low density and bulky characteristic of biomass bales make it costly to transport and handle the bales. Densification of biomass by pelleting can significantly increase biomass density to more than 1000 kg/m<sup>3</sup> [12] and thus reduce transportation costs. In addition, the uniform size and shape of biomass pellets make them easier to be handled using existing handling equipment, resulting in lower handling costs [6].

In biorefineries, biomass is converted into ethanol by three major steps (pretreatment, hydrolysis, and fermentation). The purpose of pretreatment is to break the lignin seal and disrupt the crystalline structure of cellulose, increasing its surface area, and making it more accessible to enzyme hydrolysis [2,8], as shown in Figure 6.2. Hydrolysis is performed to break the polymeric structure of cellulose into its component sugars which are converted into ethanol in fermentation

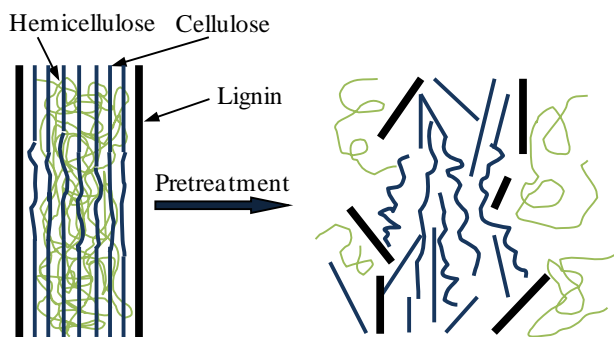
processes. The ethanol yield in fermentation is approximately proportional to the sugar yield in hydrolysis [13].

Ultrasonic vibration-assisted (UV-A) pelleting and ring-die pelleting are two pelleting methods. A previous study shows that the density of UV-A pellets (pellets produced by UV-A pelleting) is comparable to that of ring-die pellets (pellets produced by ring-die pelleting) [14]. UV-A pellets have higher durability than ring-die pellets [14]. The power consumptions of these two pelleting methods are also compared. This paper reports an experimental comparison on sugar yields of wheat straw processed by the two pelleting methods under different combinations of pretreatment variables.

**Figure 6.1 Major steps in manufacturing of cellulosic biofuel (after [7,8])**



**Figure 6.2 Purpose of pretreatment in biofuel manufacturing (after [8])**



## **6.2 Experimental conditions**

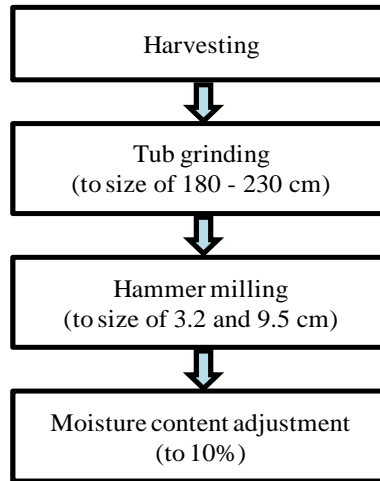
### ***6.2.1 Preparation of biomass materials***

The cellulosic biomass used in this study is wheat straw harvested by the Kansas State Agronomy Farm in December of 2010. Figure 6.3 shows the steps of wheat straw preparation for pelleting. After harvesting, wheat straw bales were chopped to particles using a large tub grinder (Haybuster H-1150 series, DuraTech Industries International Inc., Jamestown, ND). The size of the chopped particles was approximately 180 - 230 mm. The tub grinder was powered by a diesel engine and capable of grinding a large round bale (1.8 m  $\times$  1.2 m, height  $\times$  diameter) in less than 30 seconds.

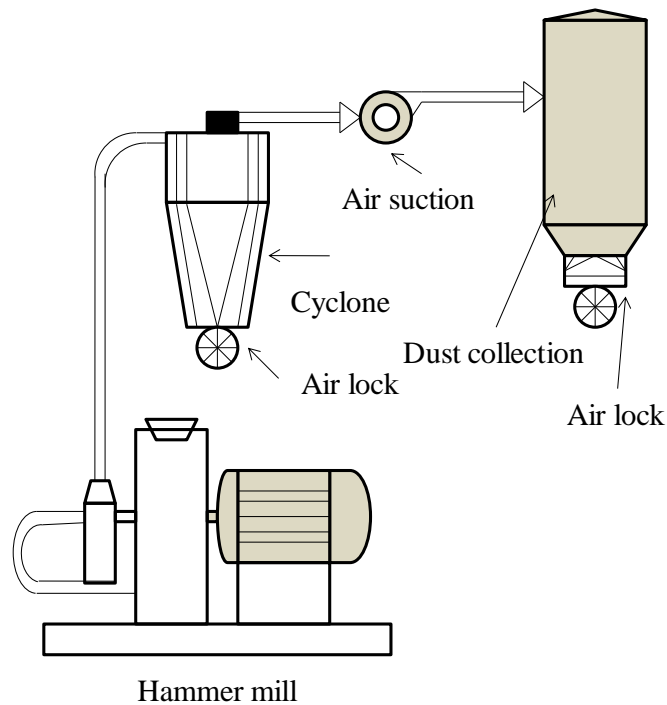
The chopped wheat straw was manually loaded onto a belt conveyor which fed into a 7.4 kW (10 hp) hammer mill (Schutte-Buffalo Hammer mill Model 18-7-300, Schutte Pulverizer Co., New York, NY). Particle size of the milled wheat straw particles was controlled by a sieve inside the hammer mill. Fig 6.4 illustrated the process. An air suction system was attached to the hammer mill to collect the milled particles and transferred biomass particles to a cyclone. Biomass particles in hammer mill had a wide distribution. Cyclone was used to separate the dust particles from air by centrifugal force and kept rest milled particles. The dust particles were stored in a dust collector (a plastic bag). In this study, two sieves with different screen sizes (3.2 and 9.5 mm) were used to control particle size. The milled wheat straw particles were collected and kept in sealed paper bags at room temperature.

Before pelleting, moisture content of wheat straw particles was adjusted to 10% (dry basis) following the ASABE Standard S385.2 [16]. No external binding agents were added in either of the pelleting methods.

**Figure 6.3 Preparation steps of wheat straw for pelleting**



**Figure 6.4 Illustration of the air suction system and cyclone attached to the hammer mill (after [15])**



### 6.2.2 UV-A Pelleting

Figure 6.5 is a schematic illustration of the experimental set-up for UV-A pelleting. UV-A pelleting was performed on a modified ultrasonic machine (Sonic-Mill, Albuquerque, NM, U.S.). An aluminum mold was made in three separate parts that were assembled together with pins. The top two parts formed a cylindrical cavity (18.5 mm in diameter) and the bottom part served as a base. Biomass was loaded into the center cavity of the mold and the mold was clamped by a fixture.

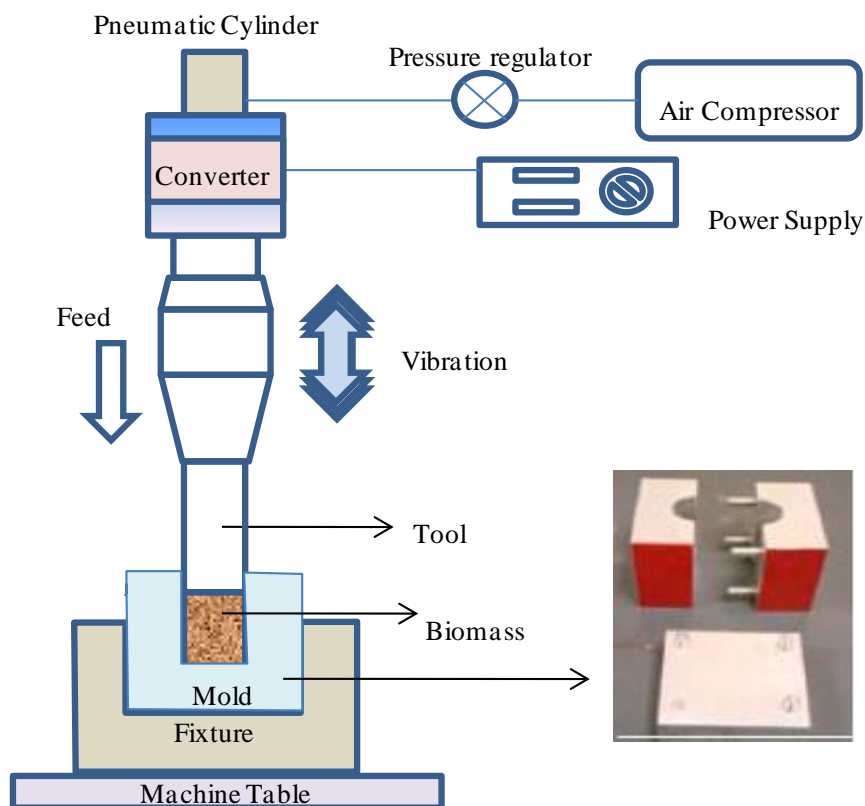
The pelleting tool was connected to an ultrasonic converter. The tip of the tool was a solid cylinder with a flat end (17.4 mm in diameter). The tool was fed into the biomass in the mold. A pneumatic cylinder applied a pressure to biomass through the tool and the ultrasonic converter provided ultrasonic vibration to the tool. The pneumatic cylinder, on top of the converter, was controlled by a pressure regulator and connected to an air compressor. Pelleting pressure represented the air pressure in the pneumatic cylinder. A higher pelleting pressure meant a higher compressive pressure applied on the biomass in the mold. Ultrasonic power, supplied by a power supply, controlled the vibration amplitude of the tool. A higher ultrasonic power meant a larger vibration amplitude. The ultrasonic power could be adjusted from 0 (no ultrasonic power) to 100% (the maximum ultrasonic power). After a predetermined period of time during which the tool was in contact with the biomass, the tool was retracted and the mold was disassembled to unload the cylinder-shaped pellet.

Table 6.1 shows major process parameters in UV-A pelleting and their values. Pelleting time was controlled using a stop watch. The recording began when the tool started descending. When the preset pelleting time (6 s) passed, the tool started retreating.

**Table 6.1 Values of major process parameters in UV-A pelleting**

<b>Parameter</b>	<b>Value</b>
Pelleting time (s)	6
Pressure (psi)	40
Ultrasonic power (%)	100
Weight of each pellet (g)	1
Vibration frequency (kHz)	20

**Figure 6.5 Illustration of experimental set-up for UV-A pelleting**



### **6.2.3 Ring-die pelleting**

Ring-die pelleting was performed on a ring-die pelleting mill (CPM Master Model Series 2000, California Pellet Mill Co., Crawfordsville, IN), as shown in Figure 6.6. The die was turning clockwise, causing the two rollers to turn in the same direction. The diameter of the channels on the ring die was 6.35 mm. The effective thickness of the die was 31.75 mm. Cylinder-shaped pellets,  $(18.9 \pm 1.6 \text{ mm}) \times (6.4 \pm 0.1 \text{ mm})$ , height  $\times$  diameter, were produced by extruding the wheat straw particles through the channels of the ring die. Frictional heating during pelleting resulted in increasing of pellet temperature. The temperature of pellets exiting the die was between 74 - 84°C.

### **6.2.4 Pretreatment**

Pretreatment was carried out in a pressure reactor (Parr Instrument Company, Moline, IL, USA). As illustrated in Figure 6.7, the reactor had a 600-mL reaction vessel. Wheat straw and 300 mL of diluted sulfuric acid were loaded into the vessel. The weight of wheat straw loaded

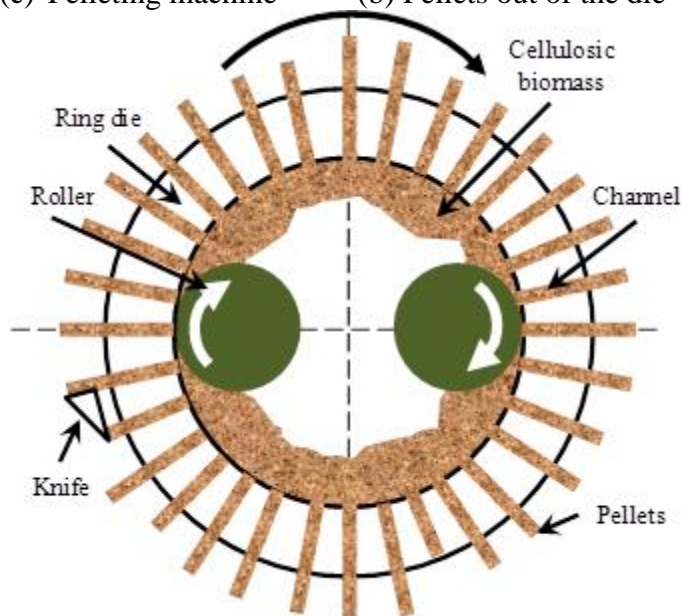
was referred to as solid content. The concentration of diluted sulfuric acid was referred to as acid concentration. The mixture of wheat straw and acid was heated by a heater to a predetermined temperature (referred to as pretreatment temperature). Meanwhile, the mixture was stirred by two impeller mixers to ensure an evenly distributed pretreatment temperature. The period of time during which wheat straw was treated in the reactor was referred to as pretreatment time. In this study, two levels of four pretreatment variables (acid concentration, solid content, pretreatment temperature, and pretreatment time) were investigated. Their values are listed in Table 6.2.

**Figure 6.6 Experimental set-up for ring-die pelleting**



(c) Pelleting machine

(b) Pellets out of the die



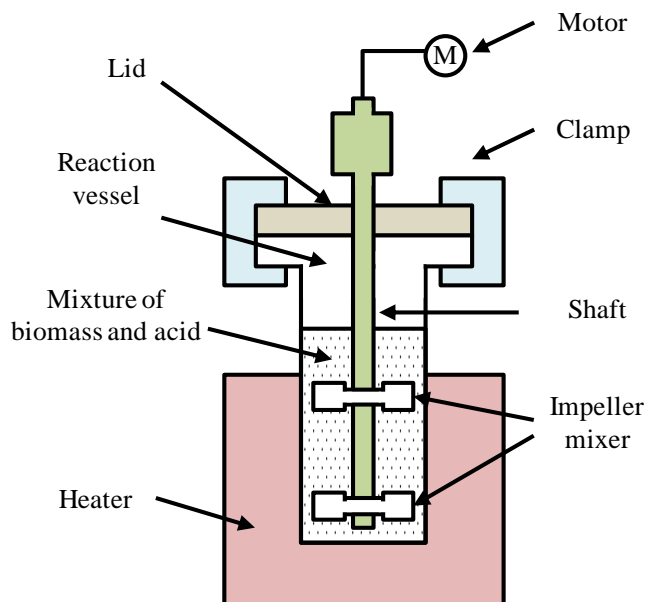
(d) Illustration of pelleting process



**Table 6.2 Variables in pretreatment and their levels**

Variable	Unit	Low level	High level
Acid concentration	%	1	2
Solid content	g	5	15
Pretreatment temperature	°C	140	160
Pretreatment time	min	30	50

**Figure 6.7 Schematic illustration of pretreatment reactor**



### **6.2.5 Hydrolysis**

Enzymatic hydrolysis was carried out in 125 mL flasks in a 50°C water bath shaker (Model C76, New Brunswick Scientific, Edison, NJ, USA) for 72 hours. Ten grams of pretreated wheat straw was loaded into the flasks together with 50 mL solution of 1.36% (w/v) sodium acetate and 0.02% (w/v) sodium azide. The pH value of the solution was adjusted to 4.8 by adding acetic acid into it. 1 mL enzyme (Accellerase 1500, Danisco US, Inc., Genencor Division, Rochester, NY) was loaded in the solution.

After enzymatic hydrolysis, samples were taken from the hydrolysis slurries by withdrawing 1 mL of slurry from each flask into 1.5 mL vials. The samples were heated in boiling water for 15 min to deactivate the enzyme. After the enzyme was deactivated, samples were centrifuged at 10,000 rpm for 15 minutes. 0.05 ml of supernatant was withdrawn from each

sample and was further diluted with 0.95 mL double-distilled water. The diluted sample (1 mL) was filtered into 1.5 mL autosampler vials through 0.2  $\mu\text{m}$  syringe filters (Millipore, Billerica, MA, USA).

#### ***6.2.6 Sugar yield measurement***

In this study, sugar yield means the concentration of glucose in the prepared samples. It was determined by an HPLC (high-performance liquid chromatography) (Shimadzu, Kyoto, Japan) equipped with an RCM-monosaccharide column ( $300 \times 7.8$  mm; Phenomenex, Torrance, CA, USA) and a refractive index detector (RID-10A, Shimadzu, Kyoto, Japan).

### **6.3 Experimental results**

For each pelleting method, three replicates of sugar yield measurements were conducted under each combination of wheat straw particle size and pretreatment variables. The final results were obtained by multiplying 20 and measured results. The average values were plotted in Figures 6.7 to 6.11. The error bars in the figures represent the standard deviation of the sugar yield data.

#### ***6.3.1 Effects of particle size***

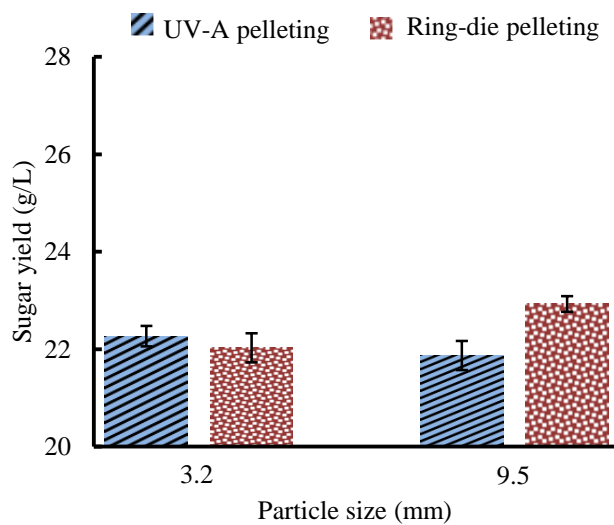
The comparison of sugar yield at two levels of particle size is shown in Figure 6.8. When small wheat straw particles (3.2 mm) were used as feedstocks for pelleting, the average sugar yield of UV-A pellets was slightly higher than that of ring-die pellets. However, the difference between the average sugar yields (0.24 g/L) was relatively small in comparison to the standard deviations (0.21 for UV-A pellets and 0.3 for ring-die pellets), indicating that there was no significant difference in sugar yield between the two pelleting methods. When large wheat straw particles (9.5 mm) were used, the difference between average sugar yield was 1.06 g/L, much larger than the standard deviations (0.3 for UV-A pellets and 0.16 for ring-die pellets). It suggests that sugar yield of UV-A pellets was lower than that of ring-die pellets.

#### ***6.3.2 Effects of acid concentration***

The comparison of sugar yield at two levels of acid concentration is shown in Figure 6.9. It can be seen that sugar yields of UV-A pellets are lower than those of ring-die pellets at both

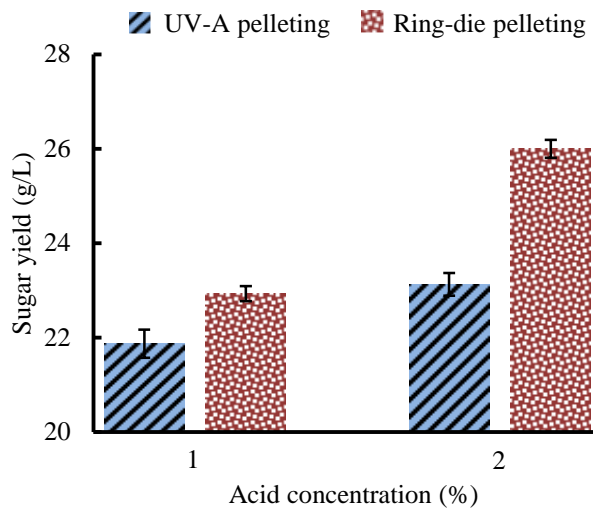
levels of acid concentration. The difference in sugar yield becomes larger when a higher acid concentration is used in pretreatment.

**Figure 6.8 Comparison of sugar yield at different levels of particle size**



Acid concentration: 1%  
Solid content: 15 g  
Pretreatment temperature: 140 °C  
Pretreatment time: 30 mins

**Figure 6.9 Comparison of sugar yield at different levels of acid concentration**

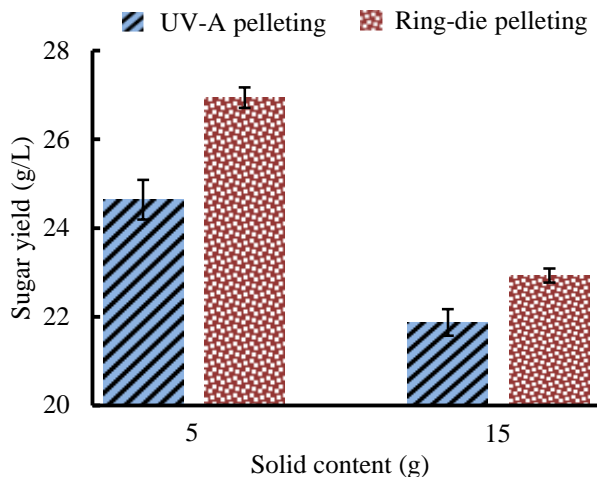


Wheat straw particle size: 9.5 mm  
Solid content: 15 g  
Pretreatment temperature: 140 °C  
Pretreatment time: 30 mins

### ***6.3.3 Effects of solid content***

The comparison of sugar yield at two levels of solid content is shown in Figure 6.10. Sugar yields of UV-A pellets are lower than those of ring-die pellets at both levels of solid content. The difference in sugar yield becomes larger when less solid content is used in pretreatment.

**Figure 6.10 Comparison of sugar yield at different levels of solid content**



Wheat straw particle size: 9.5 mm  
Acid concentration: 1%  
Pretreatment temperature: 140 °C  
Pretreatment time: 30 mins

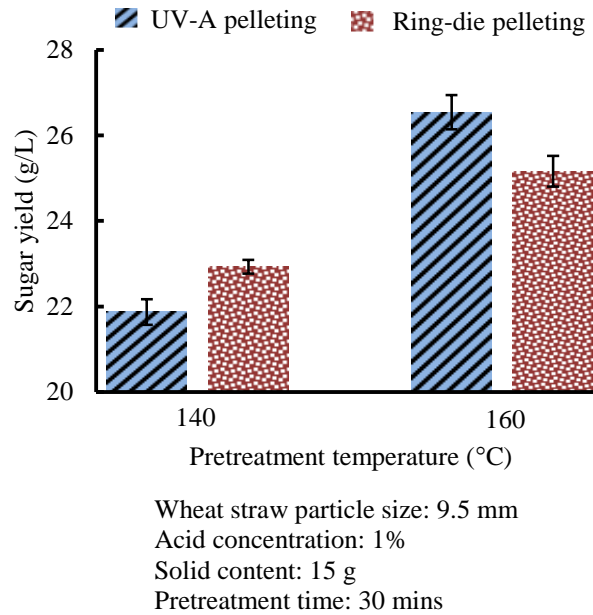
### ***6.3.5 Effects of pretreatment temperature***

The comparison of sugar yield at two levels of pretreatment temperature is shown in Figure 6.11. Sugar yield of UV-A pellets is lower than that of ring-die pellets at the lower level of pretreatment temperature (140°C). At the higher level of pretreatment temperature (160°C), UV-A pellets generate higher sugar yields than ring-die pellets.

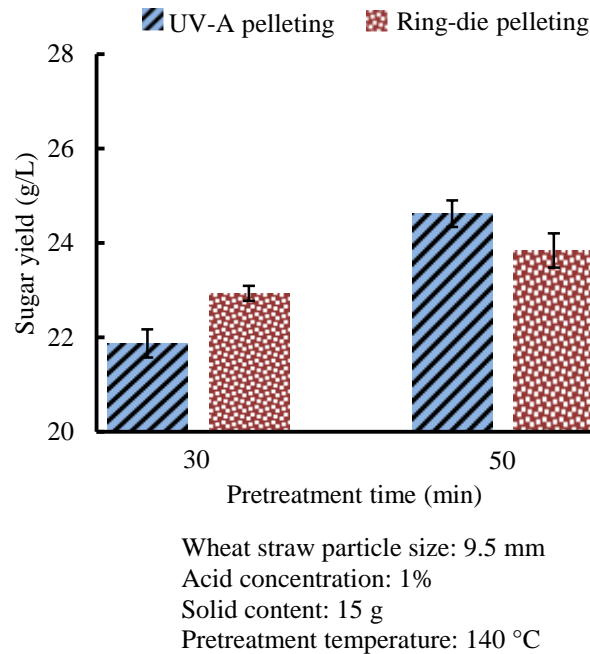
### ***6.3.6 Effects of pretreatment time***

The comparison of sugar yield at two levels of pretreatment time is shown in Figure 6.12. Sugar yield of UV-A pellets is lower than that of ring-die pellets at the lower level of pretreatment time (30 mins). At the higher level of pretreatment time (50 mins), UV-A pellets generate higher sugar yields than ring-die pellets.

**Figure 6.11 Comparison of sugar yield at different levels of pretreatment temperature**



**Figure 6.12 Comparison of sugar yield at different levels of pretreatment time**



## 6.4 Conclusions

This paper reports an experimental comparison on sugar yields of wheat straw processed by two pelleting methods, ultrasonic vibration-assisted (UV-A) pelleting and ring-die pelleting,

under different combinations of wheat straw particle size and pretreatment variables. The following conclusions can be drawn from this study.

There is no significant difference in sugar yield between the two pelleting methods when small wheat straw particles are used to make pellets. For pellets made of large wheat straw particles, sugar yield of UV-A pellets is lower than that of ring-die pellets.

The sugar yields of UV-A pellets are lower than those of ring-die pellets at both levels of acid concentration and solid content. The difference in sugar yields becomes larger when a higher acid concentration or a less solid content is used in pretreatment.

The sugar yield of UV-A pellets is lower than that of ring-die pellets at the lower level of pretreatment temperature or pretreatment time. At the higher level of pretreatment temperature or pretreatment time, UV-A pellets generate higher sugar yields than ring-die pellets.

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## **Chapter 7 - Effects of pressure and ultrasonic power on power consumption**

### Paper Title:

Ultrasonic-vibration assisted pelleting for cellulosic ethanol manufacturing: an experimental investigation on power consumption

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## **Abstract**

Ethanol produced from cellulosic biomass is an alternative to petroleum-based transportation fuels. However, its manufacturing costs are too high for cellulosic ethanol to be competitive. Cellulosic feedstocks have low density, causing their transportation and storage expensive, contributing to high manufacturing costs of cellulosic ethanol. Pelleting can increase the density of cellulosic feedstocks and reduce their transportation and storage costs. Ultrasonic vibration-assisted (UV-A) pelleting is a new pelleting method. Effects of input pelleting parameters (ultrasonic power, pelleting pressure, and particle size) on pellet quality and sugar yield have been studied. However, the effects of these parameters on power consumption in UV-A pelleting have not been studied. Since power consumption directly affects ethanol manufacturing cost, lower power consumption is desirable. The objective of this paper is to study effects of different input parameters (biomass material, particle size, ultrasonic power, and pelleting pressure) of UV-A pelleting on power consumption. Four types of biomass materials (big bluestem, corn stover, sorghum stalk, and wheat straw) were studied. Sorghum stalk consumed the least power. Pelleting pressure, particle size and ultrasonic power significantly affected power consumption of all four materials. Higher ultrasonic power and pelleting pressure resulted in lower power consumption. In addition, this paper also compares power consumption between UV-A pelleting and ring-die pelleting (a traditional pelleting method).

## **KEYWORDS**

Biofuel; Cellulosic biomass; Ethanol; Pellet, Power consumption, Ultrasonic vibration

## **7.1 Introduction**

Liquid transportation fuels (including gasoline, diesel, and jet fuels) account for 70% of the U.S. petroleum consumption [1]. The increasing demand for liquid transportation fuels in the U.S. has been far beyond its domestic production capacity, making the nation rely on foreign supplies [2]. In 2010, the U.S. transportation sector consumed about 19.1 million barrels of petroleum every day, and about half of them were imported [3]. Use of petroleum-based liquid

transportation fuels contributes to the accumulation of GHG (greenhouse gas) in the atmosphere. Considering its environmental impact and other factors (finite reserves, non-uniform distribution, and volatile prices of petroleum), there is an urgent need for alternative fuels that can reduce GHG emissions and the U.S dependence on imported petroleum [4,5].

Ethanol, used as a substitute for gasoline or as a gasoline additive, is a viable alternative fuel for transportation sector. A total ethanol production capacity of 13.1 billion gallons in 2010 in the U.S was estimated [6], a majority of which was produced from corn. Using 100% of the corn crop in the U.S. for ethanol production would meet about 17% of the nation's needs [7]. Furthermore, a dramatic increase in ethanol production using current corn-based technology may be limited by the fact that corn production for ethanol will compete with food and feed production for limited agricultural land.

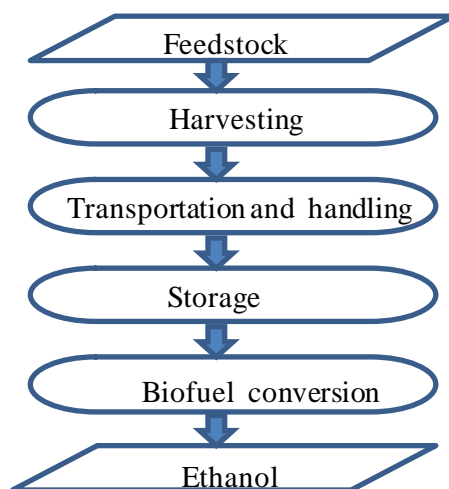
Ethanol made from cellulosic biomass (herbaceous, woody, and generally inedible portions of plant matter) is promoted by the U.S Department of Energy (DOE) [8]. Cellulosic biomass is abundant. Land resources in the U.S. are sufficient enough to produce 1.3 billion dry tonne of cellulosic biomass annually, from which enough ethanol can be made to replace over 50% of gasoline used currently in the U.S. [9]. Furthermore, using cellulosic ethanol can reduce GHG emissions by 85% over petroleum-based fuels [8,10].

Figure 7.1 shows major steps in manufacturing of cellulosic ethanol. After harvesting, biomass bales are transported from fields to warehouse and stored for future use. Cellulosic feedstocks have low density, causing their transportation and storage expensive, contributing to high manufacturing costs of cellulosic ethanol [4,13-14]. Densification of cellulosic biomass into pellets can significantly increase pellet density and reduce transportation and storage costs [15].

Pelleting is generally described as “the agglomeration of small particles into larger particles by the means of a mechanical process, and in some applications, thermal processing” [16]. If cellulosic biomass is pelleted, the density can be 600 - 800 kg/m<sup>3</sup>, much higher than that of loose cellulosic biomass (40-250 kg/m<sup>3</sup>) [17,18]. Meanwhile, pellets with uniform size and shape are easier to manage using existing handling and storage equipment. Table 7.1 compares

transportation and handling costs between pellets and bales or chops. It has been reported that, for biorefineries with annual capacities that are larger than 277 million liters, pelletizing is the lowest-cost choice [19].

**Figure 7.1 Major steps in manufacturing of cellulosic ethanol (after [11,12])**



**Table 7.1 Comparison of transportation and handling costs [19,20]**

	Transportation cost (\$/dry tonne)	Handling cost (\$/dry tonne)
Bales	9.98	24.64
Chops	16.33	21.76
Pellets	4.49	13.38

Ultrasonic Vibration-Assisted (UV-A) pelleting is a new pelleting method. Preliminary studies showed that input pelleting parameters (such as pelleting pressure, ultrasonic power, and particle size) had significant effects on pellet quality (density, durability, and stability) and sugar yield. However, the effects of input parameters on power consumption in UV-A pelleting have not been studied. Power consumption directly affects ethanol manufacturing cost. In order to reduce manufacturing costs of cellulosic ethanol, lower power consumption is preferred.

The objective of this paper is to study effects of input parameters (biomass material, pelleting pressure, ultrasonic power, and particle size) on power consumption in UV-A pelleting

using four types of cellulosic biomass. In addition, this paper also compares power consumption between UV-A pelleting and ring-die pelleting (a traditional pelleting method).

## **7.2 Experimental conditions and procedures**

### ***7.2.1 Preparation of biomass materials***

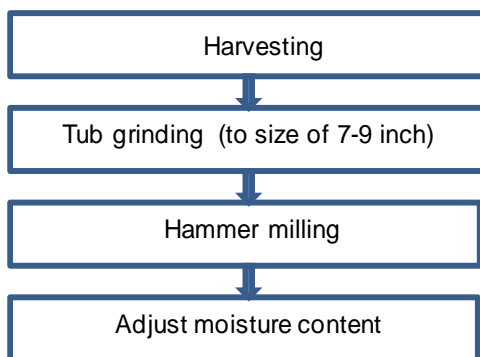
Four types of cellulosic biomass materials were used: big bluestem, corn stover, sorghum stalk, and wheat straw. The big bluestem was obtained from Star Seed in Beloit, Kansas, in January 2009. Corn stover and wheat straw were harvested by the Kansas State Agronomy Farm in November of 2009. Sorghum stalk was harvested by the same farm in December 2011. Big bluestem, corn stover and wheat straw were obtained in the form of 1.8 x 1.8 x 1.2 m (length x height x width) square bales and sorghum stalk was obtained in the form of a round bale with a diameter of 1.8 m.

Figure 7.2 shows material preparing steps before pelleting. After harvesting, all biomass materials were chopped to particles with a size of approximately 17.8-22.9 cm (7-9 inches) in length using a large tub grinder (Haybuster H-1150 series, DuraTech Industries International Inc., Jamestown, ND). The tub grinder was powered by a diesel engine and capable of grinding a large round bale in less than 30 seconds. All four types of biomass materials were then transported to the Bioprocessing and Industrial Value Added Program (BIVAP) building located on the campus of Kansas State University. Each type of materials was separately fed into a 7.4 kW (10 hp) hammer mill (Schutte-Buffalo Hammer mill Model 18-7-300, Schutte Pulverizer Co., New York, NY). Smaller particle sizes of each biomass material were obtained using the hammer mill with two different sieves of size 3.2 and 9.5 mm (1/8 and 3/8 inch), respectively, as shown in Table 7.2. The milled biomass materials were collected and kept in sealed paper bags at room temperature.

Before pelleting, initial moisture content (MC) of biomass materials was determined by drying about 25 g of each sample in an oven at 103 °C for 24 h [20]. The desired MC of biomass materials was adjusted to 15% (wet basis) by mixing tap water with biomass particles at room

temperature for 2 mins. The weight of water added was calculated according to ASABE Standard S385.2 [20].

**Figure 7.2 Steps of preparing cellulosic biomass for pelleting**



**Table 7.2 Particle size and moisture content of biomass materials**

Material	Particle size (inch)	Moisture content (%)
Corn stover	1/8; 3/8	15%
Big bluestem	1/8; 3/8	15%
Sorghum stalk	1/8; 3/8	15%
Wheat straw	1/8; 3/8	15%

### **7.2.2 UV-A Pelleting**

Figure 7.3 is a schematic illustration of the experimental set-up for UV-A pelleting. Pelleting is performed on an ultrasonic machine (Sonic-Mill, Albuquerque, NM, U.S.). An aluminum mold was made in three separate parts that were assembled together with pins. The top two parts formed a cylindrical cavity (18.6 mm in diameter) and the bottom part served as a base. Biomass was loaded into the center cavity of the mold and the mold was clamped by a fixture. The tool was connected to an ultrasonic converter. The tip of the tool was a solid cylinder with a flat end (17.4 mm in diameter). The tool was fed into the biomass in the mold. Meanwhile, the pneumatic cylinder applied a compressive pressure to the biomass through the tool and the ultrasonic converter provided ultrasonic vibration to the tool. The pneumatic cylinder, on top of

the converter, was controlled by a pressure regulator and connected to an air compressor. After a predetermined period of time during which the tool was in contact with the biomass, the tool was retracted and the mold was disassembled to unload the cylinder-shaped pellet.

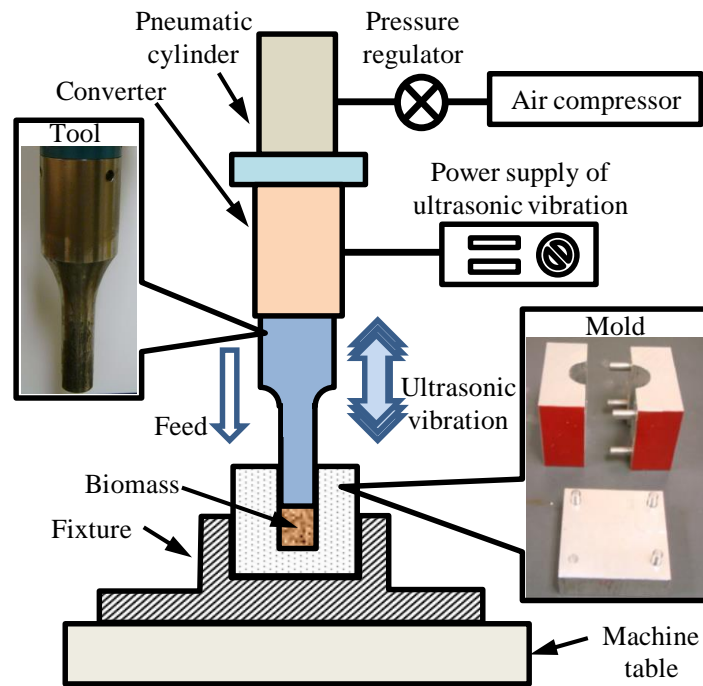
The pelleting pressure represented the air pressure in the pneumatic cylinder. A higher pressure indicated a higher compressive pressure applied on the biomass in the mold. Ultrasonic power, supplied by a power supply, controlled the vibration amplitude of the tool. A higher ultrasonic power indicated a greater vibration amplitude. The ultrasonic power could be adjusted from 0 (no ultrasonic power) to 100% (the maximum ultrasonic power).

One gram of biomass materials was used for each pellet. Three replications were conducted under each pelleting condition. After removal from the mold, the dimensions of the cylinder-shaped pellets were  $(4.57 \pm 0.4 \text{ mm}) \times (18.62 \pm 0.1 \text{ mm})$ , height  $\times$  diameter. When the range of ultrasonic power was 50%-100%, the range of pressure was 30-50 psi, and the frequency was fixed at 20 kHz, the pellet density was controlled to be  $780 \pm 50 \text{ kg/m}^3$  in order to compare power consumption.

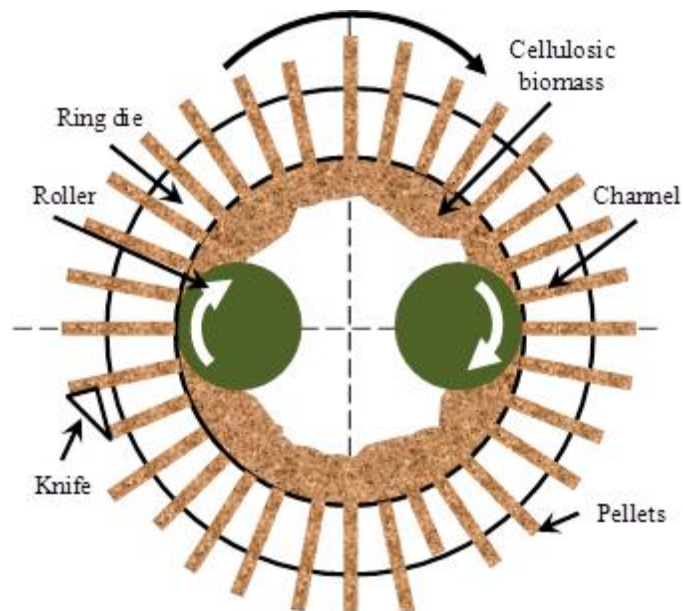
### ***7.2.3 Ring-die pelleting***

Ring-die pelleting experiments were conducted using a 22.1 kW (30 hp) ring-die pelleting mill (CPM Master Model Series 2000, California Pellet Mill Co., Crawfordsville, IN). The die size was 6.35 mm  $\times$  44.45 mm (0.25 inch  $\times$  1.75 inch), hole diameter  $\times$  effective thickness. Figure 7.4 illustrates the ring-die pelleting. The ring die was turning clockwise, causing two rollers to turn in the same direction. The roller turned as the die was rotated, forcing the biomass materials through the die holes. As the pellets were extruded, adjustable knives cut them to the desired length. Cylinder-shaped pellets,  $(18.9 \pm 1.6 \text{ mm}) \times (6.4 \pm 0.1 \text{ mm})$ , height  $\times$  diameter, were produced by extruding the biomass particles through the channels of the ring die. Each test run lasted for 10 -11 minutes and used 25 lbs of each type of biomass material. Three replications were conducted under each pelleting condition. Totally, 24 runs (2 particle size  $\times$  4 types of biomass  $\times$  3 replications) were conducted.

**Figure 7.3 Illustration of experimental set-up for uv-a pelleting**



**Figure 7.4 Illustration of ring-die pelleting**





### **7.3 Measurement procedures for power consumption**

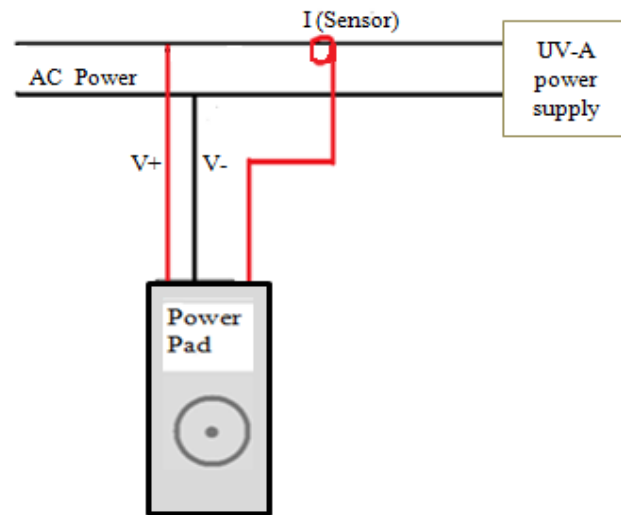
The term “power consumption” in this paper refers to the electrical energy consumed. A power analyzer (AEMC 2010.86 PowerPad Jr. Model 8230, AEMC-Instruments, Foxborough, MA) was used to measure the electricity consumption.

Figure 7.5 (a) shows the measuring method for UV-A pelleting where single phase electricity was used. A power cable was connected in parallel with the UV-A machine. Voltage probe leads were connected to the 120 Volt AC cable and a current sensor was clamped around the positive AC cable. The device took voltage/current readings. The power analyzer began recording data when the tool started dropping and stopped recording data when the tool started retreating.

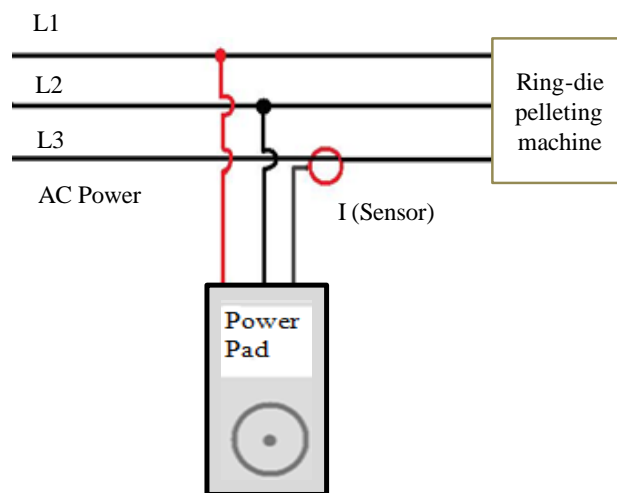
Figure 7.5 (b) shows the measuring method for ring-die pelleting where three phase electricity was used. Three cables (L1, L2, and L3) were connected in parallel with the ring-die pelleting mill. The current sensor was clamped on L3. A positive probe (red wire) was connected to L1 and a negative probe (black wire) was connected to L2. The power analyzer started recording data when the mill was turned on and stopped recording data when the mill was turned off.

For electricity consumption of both pelleting methods, the average value of measurements was calculated and plotted to the graphs in Figures 7.6 to 7.10. Results were obtained from the instruments as Wh/g and converted to kWh/ton.

**Figure 7.5 Measuring methods for power consumption**



**(a) For UV-A pelleting**



**(b) For ring-die pelleting**

## **7.4 Experimental results on power consumption in UV-A pelleting**

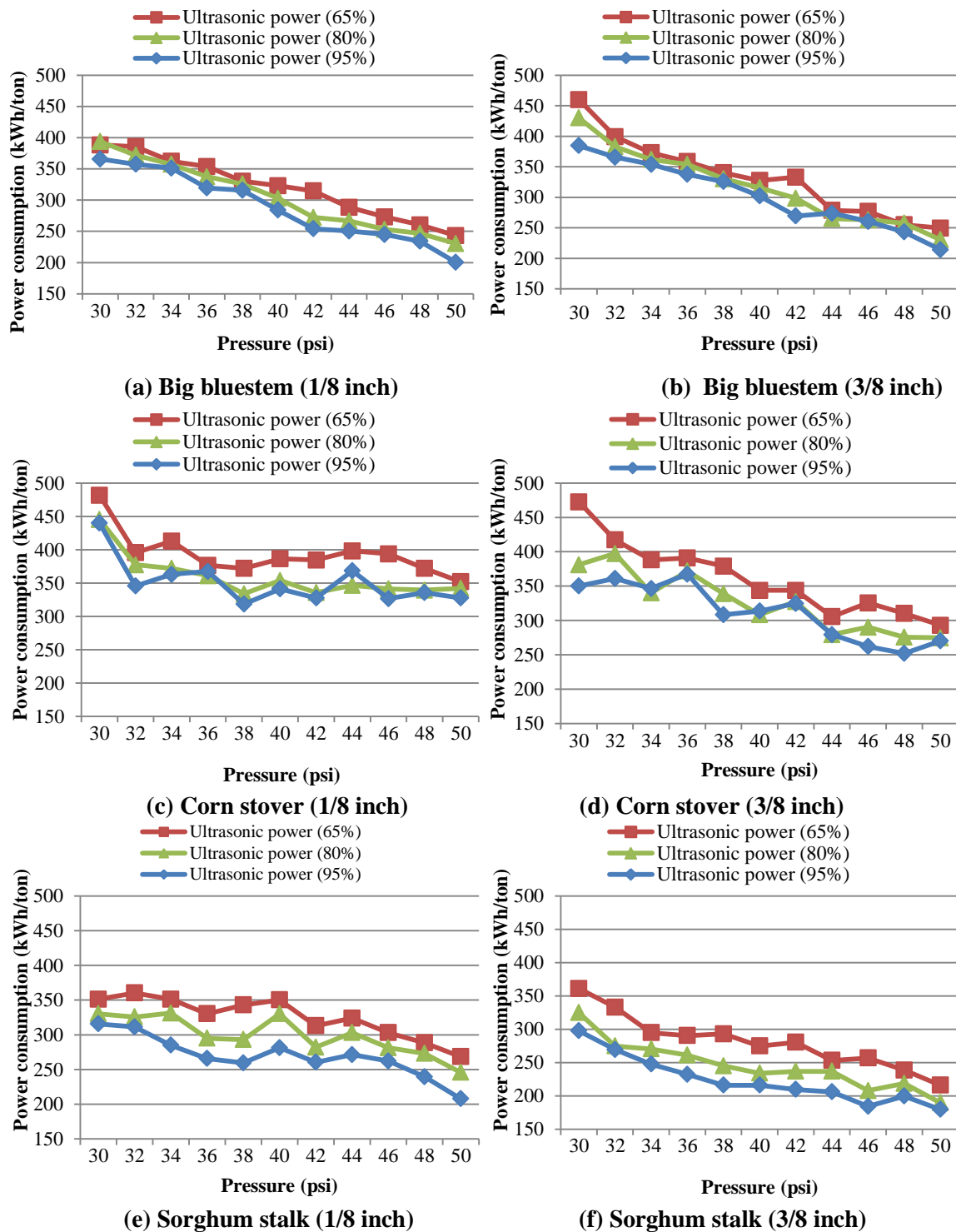
### ***7.4.1 Effects of pelleting pressure***

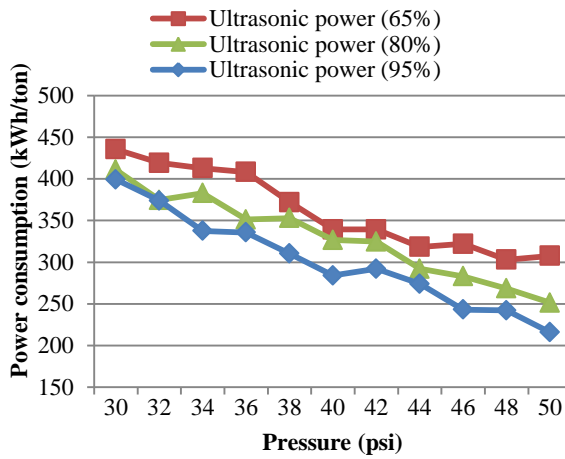
Figure 7.6 shows effects of pelleting pressure on power consumption. It can be observed that, as pressure increased, the power consumption decreased. A similar trend can be found with different ultrasonic power levels. Preliminary studies show that as the pressure increased from 20 to 30 and to 40 psi, both pellet density and pellet durability of sorghum stalk increased

dramatically [22]. The highest density for a UV-A pellet is 950 Kg/m<sup>3</sup>. However, the pellet density was almost the same for pressure levels of 40 psi and 50 psi [21].

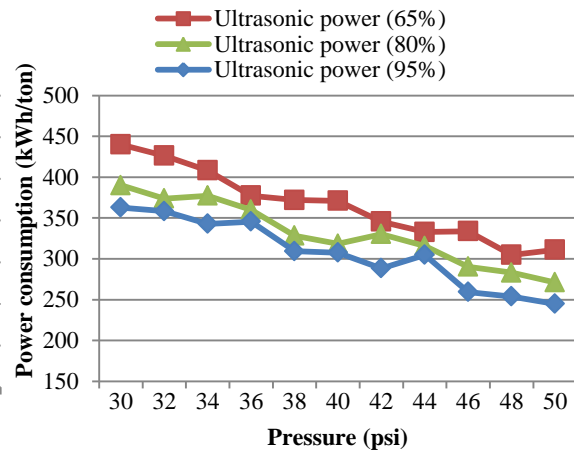
Biomass materials can be further processed to produce sugar after pretreatment and enzymatic hydrolysis. Sugar yield is a key factor affecting the ethanol yield. Zhang et.al [22] also reported that pressure had significant effects on sugar yield.

**Figure 7.6 Effects of pelleting pressure**





(g) Wheat straw (1/8 inch)

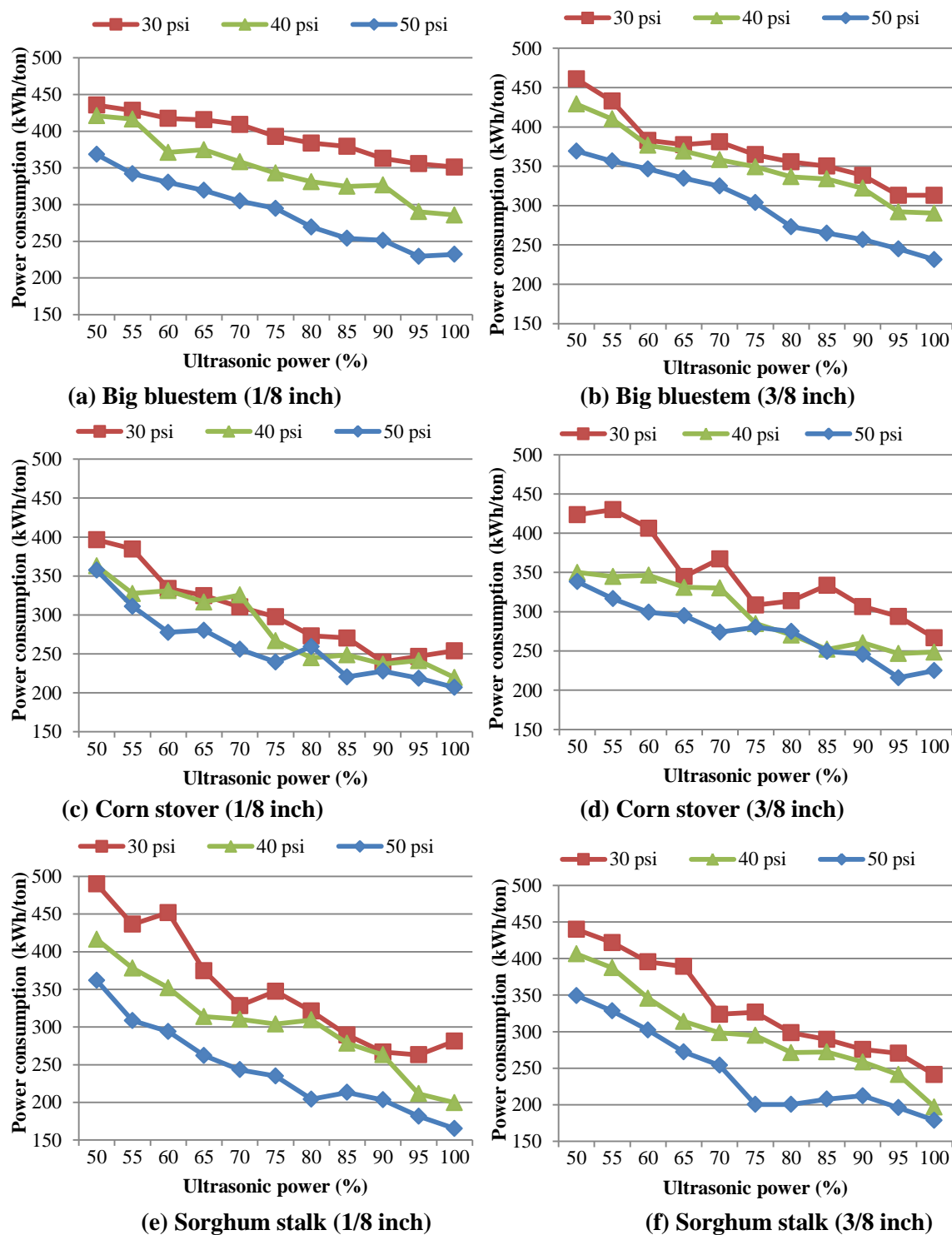


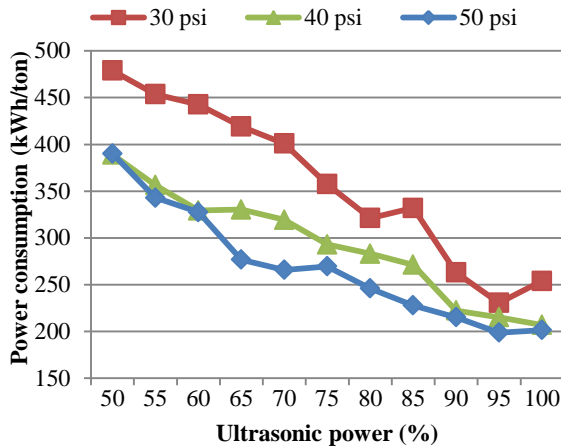
(h) Wheat Straw (3/8 inch)

#### 7.4.2 Effects of ultrasonic power

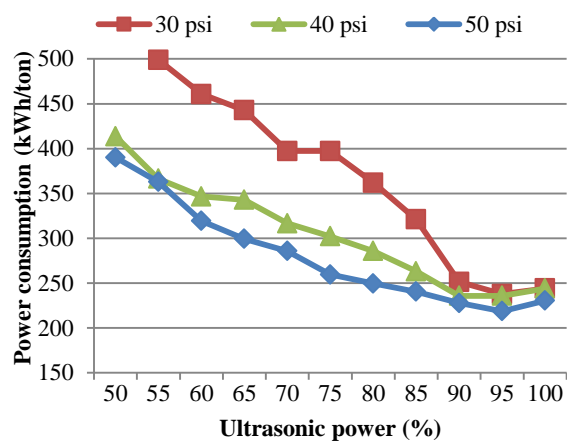
Figure 7.7 shows effects of ultrasonic power on power consumption. It can be seen that, as ultrasonic power increased, the power consumption decreased. Preliminary studies show that as the ultrasonic power was increased from 30% to 50%, both pellet density and pellet durability increased significantly [22]. The sugar yield increased about 20% as the ultrasonic power was increased from 30% to 55%. Considering the results above, a higher ultrasonic power is preferred because it could be beneficial for physical properties and sugar yield of UV-A pellets.

**Figure 7.7 Effects of ultrasonic power**





(g) Wheat straw (1/8 inch)



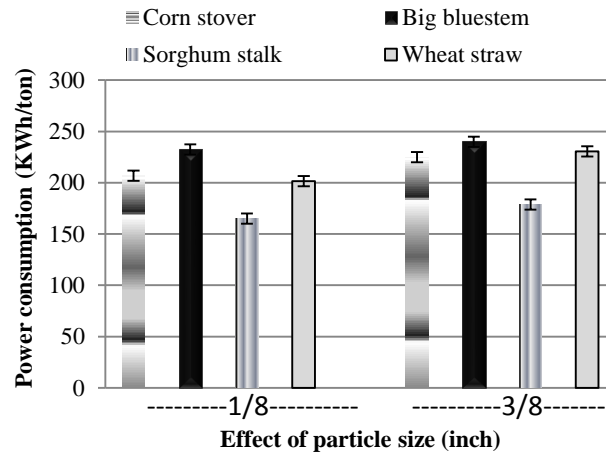
(h) Wheat Straw (3/8 inch)

### 7.4.3 Effects of particle size

Figure 7.8 shows effects of particle size on power consumption. The error bars represent standard deviation. Minitab 16 was used to perform paired T test for these two particle sizes on each type of material. Results showed that particle size had significant effects on power consumption ( $P$  value  $< 0.05$ ). It can also be seen that smaller particle sizes (1/8 inch) consumed less power than larger particle sizes (3/8 inch).

Preliminary studies showed that smaller particle sizes produced higher pellet density and smaller spring-back [21]. Spring-back measures the percentage of volume expansion after a pellet is formed. Hill and Pulkine [23] reported a 15% improvement in pellet durability from 6.4 mm to 2.8 mm. Meanwhile, smaller particle size increased the surface area of biomass [24]. The increase in surface area allowed easier access by enzymes and chemicals, resulting in higher sugar yield. However, other researchers reported smaller particle sizes required more power in size reduction [25,26]. In conclusion, smaller particle sizes were desirable in pellet quality, sugar yield, and pelleting power consumption but not preferred in size reduction. A comprehensive study was desirable to find an optimal particle size.

**Figure 7.8 Effects of particle size**



#### ***7.4.4 Effects of biomass type***

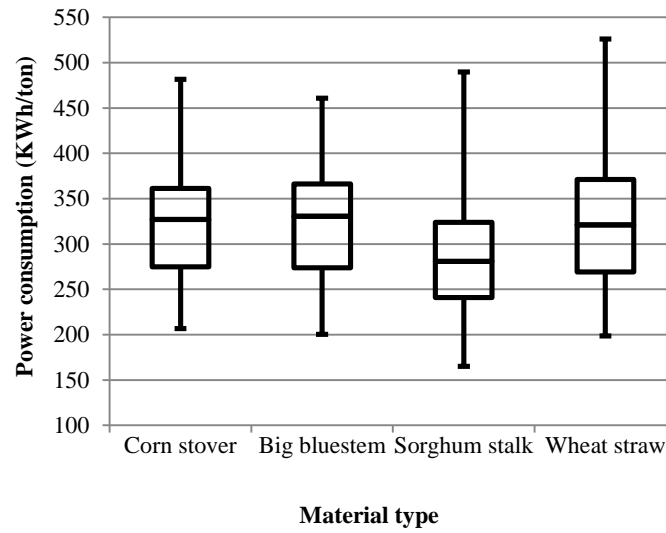
Figure 7.9 shows effects of material type on power consumption. Box-plot is used to display the data. Five values from a set of data are used: the minimum, the maximum, the lower and upper percentile, and the median [27]. The bottom and top of the box are the 25<sup>th</sup> and 75<sup>th</sup> percentile, respectively. The band near the middle of the box is always the 50<sup>th</sup> percentile (the median). It can be seen that different materials resulted in different power consumptions. Sorghum stalk consumed the least power. However, there was no significant difference between corn stover and big bluestem.

### **7.5 Comparison of power consumption between two pelleting methods**

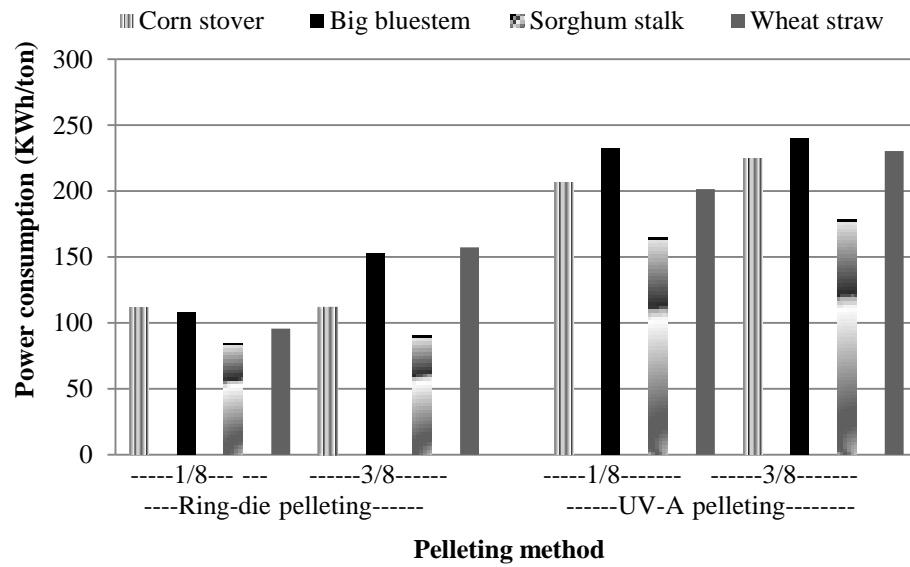
Figure 7.10 compares power consumption between two pelleting methods. The lowest power consumption of each type of material in both pelleting methods was displayed in the graph. UV-pelleting consumed more power than ring-die pelleting. It is noted that the ring-die pelleting was conducted using a manufacturing-scale machine while UV-A pelleting was conducted on a lab-scale set up. It is anticipated that the power consumption in UV-A pelleting will go down on a manufacturing-scale machine.



**Figure 7.9 Effects of biomass material type**



**Figure 7.10 Comparison between two pelleting methods**



## **7.6 Conclusions and remarks**

This paper presents the effects of pelleting parameters on power consumption of UV-A pelleting. In addition, it compares the power consumption of two pelleting methods: ring-die pelleting and UV-A pelleting. The following conclusions can be drawn from the study:

1. As pelleting pressure increased, the power consumption in UV-A pelleting decreased.
2. As ultrasonic power increased, the power consumption in UV-A pelleting decreased.
3. In UV-A pelleting, larger particles sizes (3/8 inch) required more power than smaller particle size (1/8 inch). The obtained trends are consistent with those in ring-die pelleting.
4. Material type had significant effects on power consumption in both UV-A pelleting and ring-die pelleting. Sorghum stalk consumed the least power in UV-A pelleting among the four types of cellulosic materials.
5. Ring-die pelleting consumed less power than UV-A pelleting.

Higher pressure and ultrasonic power were preferred because less power was consumed in pelleting, resulting in better pellet quality and sugar yield. For particle size, smaller particle sizes required less power in the pelleting process, resulting in better pellet quality but also required more power in size reduction. A comprehensive study is needed to find an optimal particle size. Furthermore, Comparison of these two pelleting methods on sugar yield is a future research direction.

## **Acknowledgements**

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## Chapter 8 - Investigation on power consumption

### Paper Title:

Ultrasonic-vibration assisted pelleting for cellulosic biofuel manufacturing: investigation on power consumption

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### **Abstract**

Cellulosic ethanol produced from cellulosic biomass is an alternative to petroleum-based transportation fuels. Raw cellulosic biomass has low density, causing high costs in their storage, transportation, and handling. Ultrasonic vibration-assisted (UV-A) pelleting can increase the density of cellulosic biomass. Effects of UV-A pelleting variables on pellet quality (density, durability, stability, and strength) and sugar yield have been reported. However, power

consumption in UV-A pelleting has not been fully investigated. This paper presents an experimental investigation on power consumption in UV-A pelleting of wheat straw. Effects of input variables (biomass moisture content, biomass particle size, pelleting pressure, and ultrasonic power) on power consumption are investigated. Results show that power consumption in UV-A pelleting increases as moisture content and particle size decrease, and as pelleting pressure and ultrasonic power increase.

## **8.1 Introduction**

Liquid transportation fuels (including gasoline, diesel, and jet fuels) account for 70% of the U.S. petroleum consumption [1]. In 2010, the U.S. transportation sector consumed about 19 million barrels of petroleum every day, and about half of them were imported [2]. Use of petroleum-based liquid transportation fuels contributes to the accumulation of GHG (greenhouse gas) in the atmosphere. These conditions plus other concerns (finite reserves, non-uniform distribution, and volatile price of petroleum) make it critically important to develop domestic sustainable alternatives to petroleum-based liquid transportation fuels [3,4].

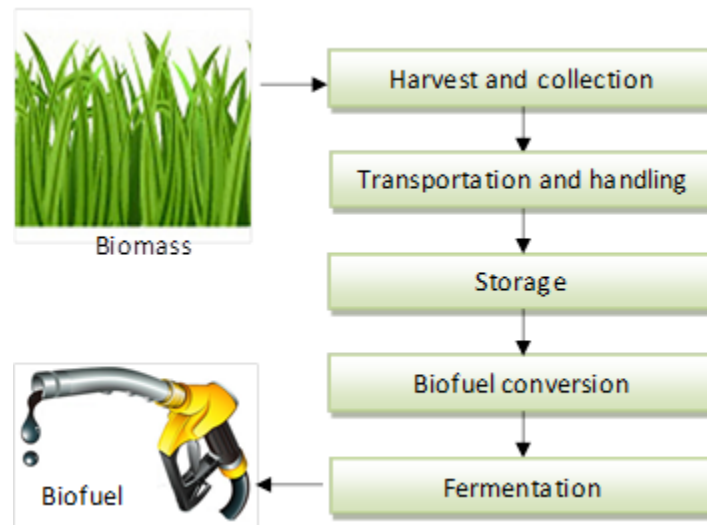
One such alternative is cellulosic ethanol made from cellulosic biomass (herbaceous, woody, and generally inedible portions of plant matter). Cellulosic biomass is abundant and relatively inexpensive. Land resources in the U.S. are sufficient to sustain production of enough cellulosic biomass (about 1.3 billion dry tons) annually to replace 30% or more of the nation's current consumption of liquid transportation fuels [5,6]. Cellulosic ethanol reduces GHG emissions by 85% over petroleum-based fuels [5,7]. In addition, a cellulosic ethanol industry would create jobs, increase farmers' income, and boost rural economy [6].

Figure 8.1 shows major steps in manufacturing of cellulosic ethanol. A major challenge to cellulosic biofuel manufacturing is the high costs in storage, transportation, and handling of low density biomass. Pelleting of cellulosic biomass can significantly increase the density of cellulosic biomass and reduce the costs in biomass storage, transportation, and handling [10].

Traditional pelleting methods (e.g., using a screw extruder, a briquetting press, or a rolling machine [11,12]) usually involve high-temperature steam, high pressure, and binder materials. It is difficult to realize cost-effective pelleting at or near the fields where cellulosic biomass is available by using traditional pelleting methods. Ultrasonic vibration-assisted (UV-A) pelleting, without using high-temperature steam and binder materials, can produce pellets whose density is comparable to those produced by using traditional pelleting methods [13].

The literature on UV-A pelleting is focused on experimental investigations on pellet quality (density, durability, and stability) and sugar yield. However, power consumption in UV-A pelleting has not been fully investigated. The objective of this paper is to investigate the effects of input variables on power consumption in UV-A pelleting. The input variables include biomass moisture content, biomass particle size, pelleting pressure, and ultrasonic power.

**Figure 8.1 Major steps in biofuel manufacturing (after [8,9])**



## 8.2 Materials and methods

### 8.2.1 Raw biomass material

The pelleting feedstock used in this study was wheat straw harvested in northwestern Kansas in July of 2010. The wheat straw had been run through a John Deere 9600 combine (that

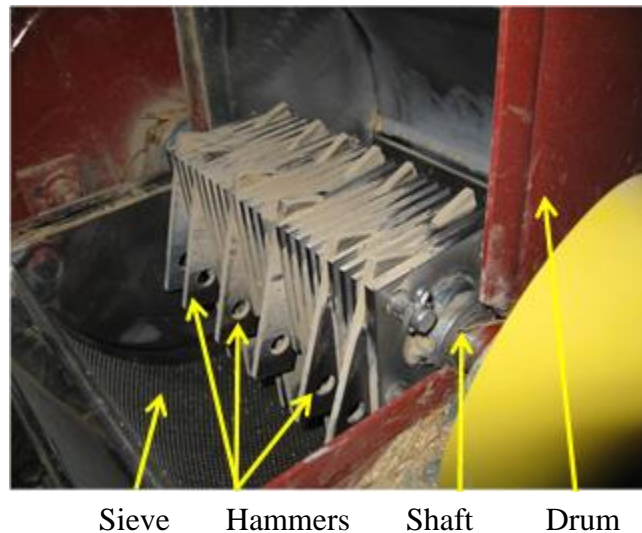


removed wheat grains from wheat straw and chaff) and collected. The collected wheat straw had an average length of 25 cm. After harvesting and collection, wheat straw was stored in bags before use.

### ***8.2.2 Size reduction***

The size of wheat straw was further reduced using a hammer mill (model 35, Meadows Mills, Inc., North Wilkesboro, NC, USA), as shown in Figure 8.2. The hammer mill used a 240-volt, 5-horsepower electric motor. The hammer mill had a steel drum containing a rotating shaft on which 24 hammers were mounted. The rotation speed of shaft was fixed at 3600 rpm and the hammers were free to swing. The size of hammers was 101.6 x 25.4 x 4.8 mm. The wheat straw was fed into the grinding drum from the top of the hammer mill. The rotating hammers impacted the wheat straw to reduce the size of wheat straw. The produced particles would pass through the sieve at the bottom of the grinding chamber when they were small enough [14]. The screen size of the sieve in the hammer mill was 2 mm.

**Figure 8.2 Size reduction by a hammer mill**

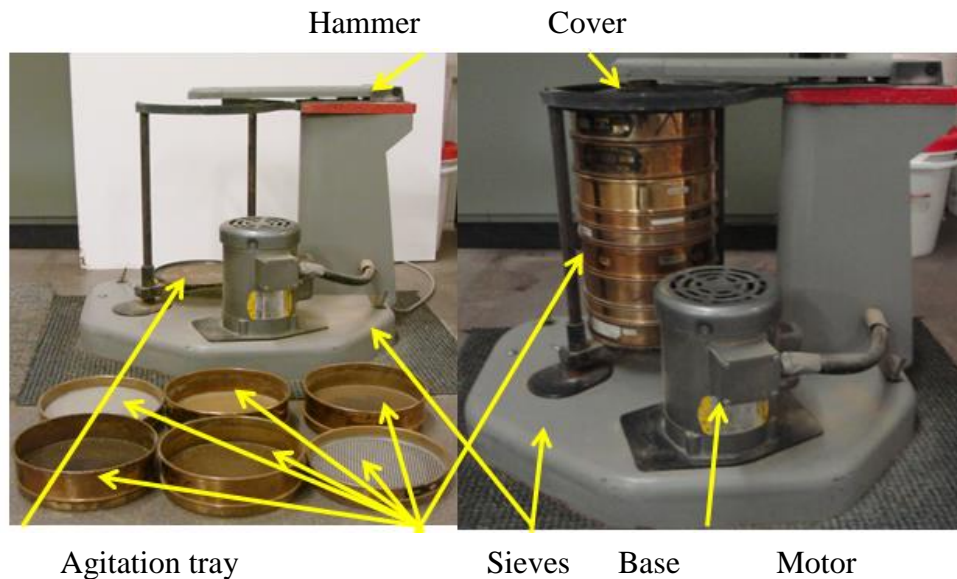


### 8.2.3 Separation of particle sizes

Wheat straw particles from hammer milling had a wide size distribution. The particles were then separated into different size ranges using a sieve shaker (model RX-29, W.S. Tyler, Inc., Mentor, OH, U.S.), as shown in Figure 8.3. A series of sieves with different screen sizes were loaded on an agitation tray. Particles were put on the top sieve that had the largest screen size. A hammer stroke a cover located above the sieves three times per second. Meanwhile, the agitation tray moved circularly at 200 rpm. The running time of the sieve shaker was 10 minutes.

Particle sizes were determined by the screen size of the sieves. Table 8.1 lists the screen sizes of the six sieves used to separate the wheat straw particles. Theoretically, particles should be separated into seven different size ranges with these six sieves. However, almost all particles fell through the 2.4 mm sieve, so the particle size range of  $> 2.4$  mm was excluded. Therefore, particles were separated into six different size ranges:  $< 0.2$ ,  $0.2 - 0.3$ ,  $0.3 - 0.4$ ,  $0.4 - 0.6$ ,  $0.6 - 1.2$ , and  $1.2 - 2.4$  mm. These six particle size ranges were investigated in this study.

**Figure 8.3 Sieve shaker**



**Table 8.1 Screen sizes of sieves**

Sieve #	Screen Size (mm)
1	2.4
2	1.2
3	0.6
4	0.4
5	0.3
6	0.2

#### ***8.2.4 Adjustment of biomass moisture content***

Biomass moisture content represents the amount of moisture (water) contained in a certain amount of biomass (wheat straw in this study). The initial moisture content was determined by drying about 25 g of wheat straw particles (after hammer milling) in an oven (Blue M Electric Co., Blue island, IL, USA) at 103 °C for 24 hours according to ASABE standard S358.2 [15]. After drying, the dried particles were weighed by using an electronic scale (Ohaus, Pine Brook, NJ, USA). The initial moisture content was calculated as the ratio of the loss in weight during drying to the weight of pre-dried sample. In this study, the initial moisture content was determined as 5%.

Another four levels of moisture content were also investigated in this study: 10%, 15%, 20%, and 25%. The initial moisture content was adjusted to the higher levels by adding distilled water based on the ASABE standard [15]. Then, the wheat straw particles were stored in zip-lock bags until being pelleted.

#### ***8.2.5 UV-A pelleting***

Pelleting was performed on a modified ultrasonic machine (model AP-1000, Sonic-Mill, Albuquerque, NM, U.S.). Figure 8.4 is a schematic illustration of the experimental set-up for UV-A pelleting. The machine included a power supply (which converts 60 Hz electrical power into 20,000 Hz electrical power), a converter (which converts high frequency electrical energy into vibration), and a titanium tool (which was connected to converter). The tip of the tool was a

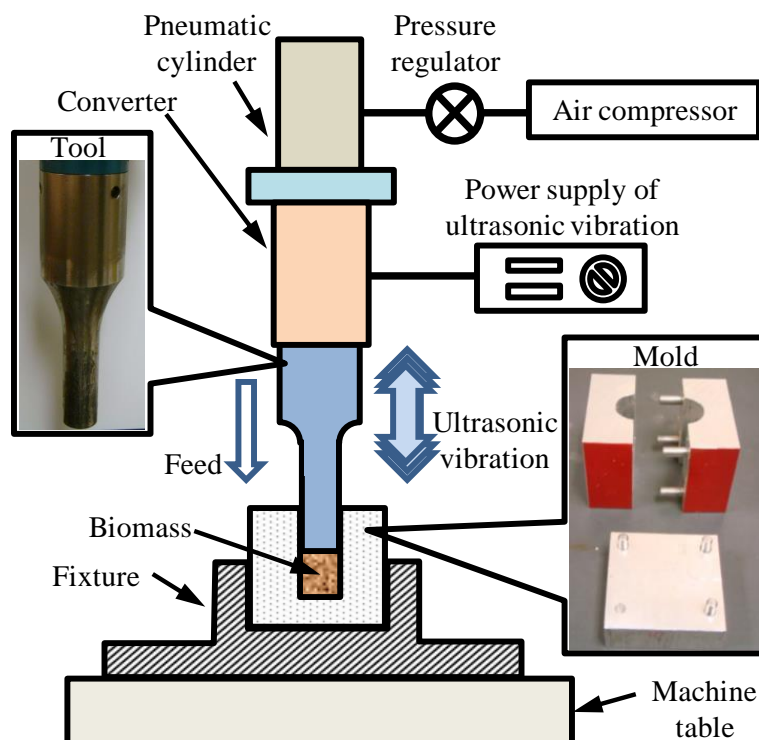
solid cylinder (17.4 mm in diameter) with a flat end. The vibration frequency of the tool was fixed at 20 kHz.

The pneumatic cylinder was driven by compressed air provided by a 1.6 HP, 33 gallon air compressor (Sears, Roebuck and Co., Hoffman Estates, IL, U.S.). The pelleting pressure represented air pressure in the pneumatic cylinder. The air pressure was controlled by a pressure regulator. A higher air pressure in the cylinder would cause a higher pressure applied on the wheat straw particles in the mold by the tool.

Ultrasonic power was referred to the power provided by the power supply. It controlled the amplitude of the tool vibration. A larger ultrasonic power would result in larger vibration amplitude. Ultrasonic power was expressed as a percentage of the maximum ultrasonic power for the power supply. It could be adjusted from 0 (no ultrasonic power) to 100% (the maximum ultrasonic power).

Before each pelleting test, one gram of wheat straw particles was loaded into the center cavity of the mold and the mold was clamped by a fixture. An aluminum mold was made in three parts that were assembled together with pins. The upper two parts formed a cylindrical cavity (18.6 mm in diameter). The bottom part served as a base. During pelleting, the tool was fed into the wheat straw particles in the mold. The pneumatic cylinder applied a pressure to the wheat straw particles through the tool, and converter provided ultrasonic vibration to the tool. After a period of time during which the tool was in contact with the wheat straw particles, the tool was retracted and the mold was disassembled to unload the cylinder-shaped pellet. Table 8.2 shows experimental parameters and their values. Ten replicates were made under each experimental condition.

**Figure 8.4 Illustration of UV-A pelleting**



**Table 8.2 Experimental parameters and values**

MC (%)	Particle size (mm)	Pressure (psi)	Ultrasonic power (%)
10	<0.2; 0.2-0.3; 0.3-0.4; 0.4-0.6; 0.6-1.2; 1.2-2.4	50	95
5; 10; 15; 20; 25	2*	50	95
10	2*	30; 35; 40; 45; 50	95
10	2*	50	50; 60; 70; 80; 90; 100

\*Particles were obtained from hammer milling with 2mm sieve size.

### **8.2.6 Measurement of power consumption**

The term “power consumption” in this paper refers to the electricity consumed by the ultrasonic power supply. It measured the power consumed to produce pellets with a specific density (around 930 kg/m<sup>3</sup>). Different pelleting time might be needed to produce the specific density under different conditions. In each measurement of power consumption, the pelleting time and pellet density were also recorded.

Power consumption was measured by a power analyzer (AEMC 2010.86 PowerPad Jr. Model 8230, AEMC-Instruments, Foxborough, MA). Voltage probe leads were connected to the 120 Volt AC cable and a current sensor was clamped around the AC cable. The power analyzer began recording voltage and current when the tool started dropping and stopped recording data when the tool started retreating.

### ***8.2.7 Measurement of pellet density***

Pellet density means the density of an individual pellet and was determined by ratio of its weight to its volume. Weight of the pellet was measured by an electronic scale (Ohaus, Pine Brook, NJ, U.S.). The volume of a pellet was determined by its diameter and height measured with a vernier caliper.

## **8.3 Results and discussion**

### ***8.3.1 Effects of moisture content***

The effects of moisture content on power consumption are shown in Table 8.3 and Figure 8.5. The error bars in Figure 5 represent the standard deviations presented in Table 3. The pellet densities at different levels of moisture content are presented in Figure 8.5 (a). Statistic tests were conducted to compare the densities. The results showed that there was no significant difference between the densities at the significance level of 0.05.

Different pelleting time was needed to produce the same pellet density at different levels of moisture content when other input variables were kept the same. As shown in Figure 8.5 (b), pelleting time increased slightly as moisture content increased from 5% to 15%. Much longer pelleting time was needed when moisture content was 15% and 25%. This indicates that higher moisture content would lead to lower pellet density if same pelleting time was used. This was consistent with the results of earlier studies. Song et al. [16] reported that, with the same pelleting time, lower moisture content (13%) produced wheat straw pellets with higher density

than higher moisture content (20% and 25%) in UV-A pelleting. Similar results were also reported by Zhang et al. [17].

As shown in Figure 8.5 (c), power consumption in UV-A pelleting increased slightly as moisture content increased from 5% to 15%. As moisture content increased from 15% to 25%, power consumption had a dramatic increase. This trend is very similar to that between moisture content and pelleting time. When moisture content was 5%, 10%, and 15%, both pelleting time and power consumption slightly increased. When moisture content was 20% and 25%, both pelleting time and power consumption increased dramatically. There was an obvious correlation between pelleting time and power consumption. This indicates that the higher power consumption for higher moisture content was at least partially caused by longer pelleting time.

Power consumption rate for different levels of moisture content is presented in Figure 5 (d). When moisture content increased to 20% or 25%, power consumption rate was much higher than those when moisture content was lower. This indicates that, excluding the effects of pelleting time, higher moisture content in itself would lead to higher power consumption. Therefore, from the viewpoint of power consumption, the moisture content of wheat straw particles in UV-A pelleting should be lower than 15%.

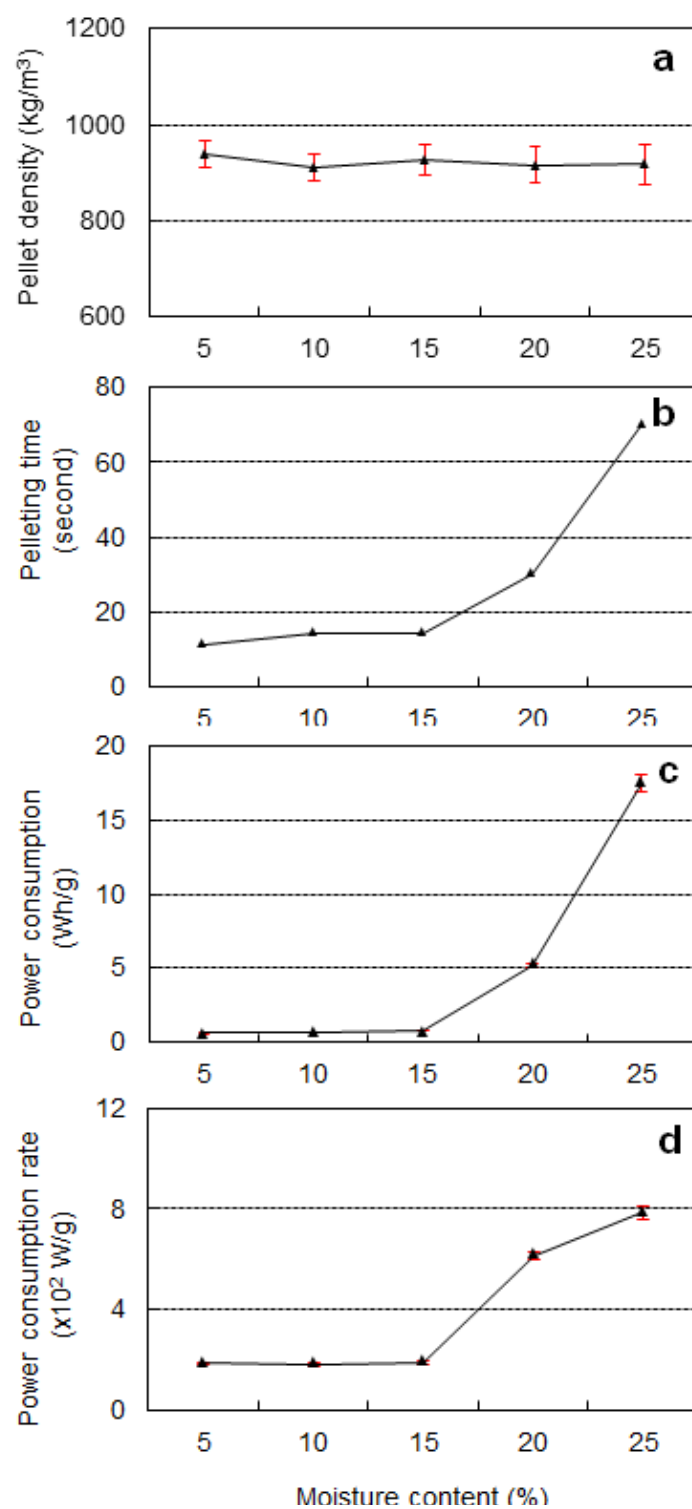
Earlier studies also showed that higher moisture content (higher than 15%) in UV-A pelleting led to lower pellet durability and stability [17]. Therefore, lower moisture content is preferable in UV-A pelleting.

Similar relations between moisture content and pellet quality (such as density, durability, and stability) were reported in the literature by using other pelleting methods and other biomass materials. Fasina and Sokhansanj [18] reported effects of moisture content on pellet durability. When pelleting of alfalfa, as moisture content increased, durability increased first before reaching a maximum value of about 86% when moisture content was 3% to 5%, and then decreased. Fasina [19] studied the effects of moisture content on durability of peanut hull pellets. As moisture content increased, pellet durability increased first and reached a maximum value of 90% when moisture content was 9%, and then decreased. An increase in moisture content also

resulted in decrease in pellet density. Colley et al. [20] studied the effects of moisture content on switchgrass pellets. As moisture content increased, pellet density decreased. Mani et al. [21] reported that moisture content significantly affected pellet density of barley straw, corn stover, and switchgrass using a single pellet unit (piston press pelleting).



**Figure 8.5 Results for different levels of moisture content**



**Table 8.3 Results for different levels of moisture content**

Moisture content (%)	Density* (kg/m <sup>3</sup> )	Pelleting time (second)	Power consumption* (Wh/g)	Power consumption rate* (x10 <sup>2</sup> W/g)
5	938 (28)	11	0.57 (0.01)	1.86 (0.04)
10	911 (29)	12	0.60 (0.02)	1.81 (0.06)
15	926 (33)	14	0.74 (0.03)	1.89 (0.07)
20	916 (39)	30	5.13 (0.13)	6.16 (0.16)
25	918 (42)	80	17.47 (0.57)	7.86 (0.26)

\* Numbers outside parentheses are mean values and numbers enclosed in parentheses are standard deviations for n = 10

### ***8.3.2 Effects of particle size***

The effects of particle size on power consumption are shown in Table 8.4 and Figure 8.6. The error bars in Figure 6 represent the standard deviations presented in Table 4. The pellet densities of different particle sizes are presented in Figure 8.6 (a). There was no significant difference between the densities at the significance level of 0.05.

As shown in Figure 8.6 (b), pelleting time increased as particle size increased. This indicates that larger particle size would lead to lower pellet density if same pelleting time was used. This trend was consistent with earlier results on relations between sieve size and pellet density. Zhang et al. [13] reported that, with the same pelleting time, smaller sieve size (0.25 mm) produced higher density for wheat straw pellets than larger sieve size (8 mm) in UV-A pelleting. Zhang et al. [22] studied pellet density in UV-A of wheat straw particles using a 24 full factorial design with two levels of sieve size (1 and 2 mm). They reported that wheat straw particles milled with the smaller sieve size produced much higher pellet density than those milled with the larger sieve size. Similar results were reported by Mani et al. [21] who studied effects of particle size on pellet density of barley straw, corn stover, and switchgrass using a single pellet unit. When particle size decreased from 3.2 mm to 0.8 mm, the pellet density increased linearly.

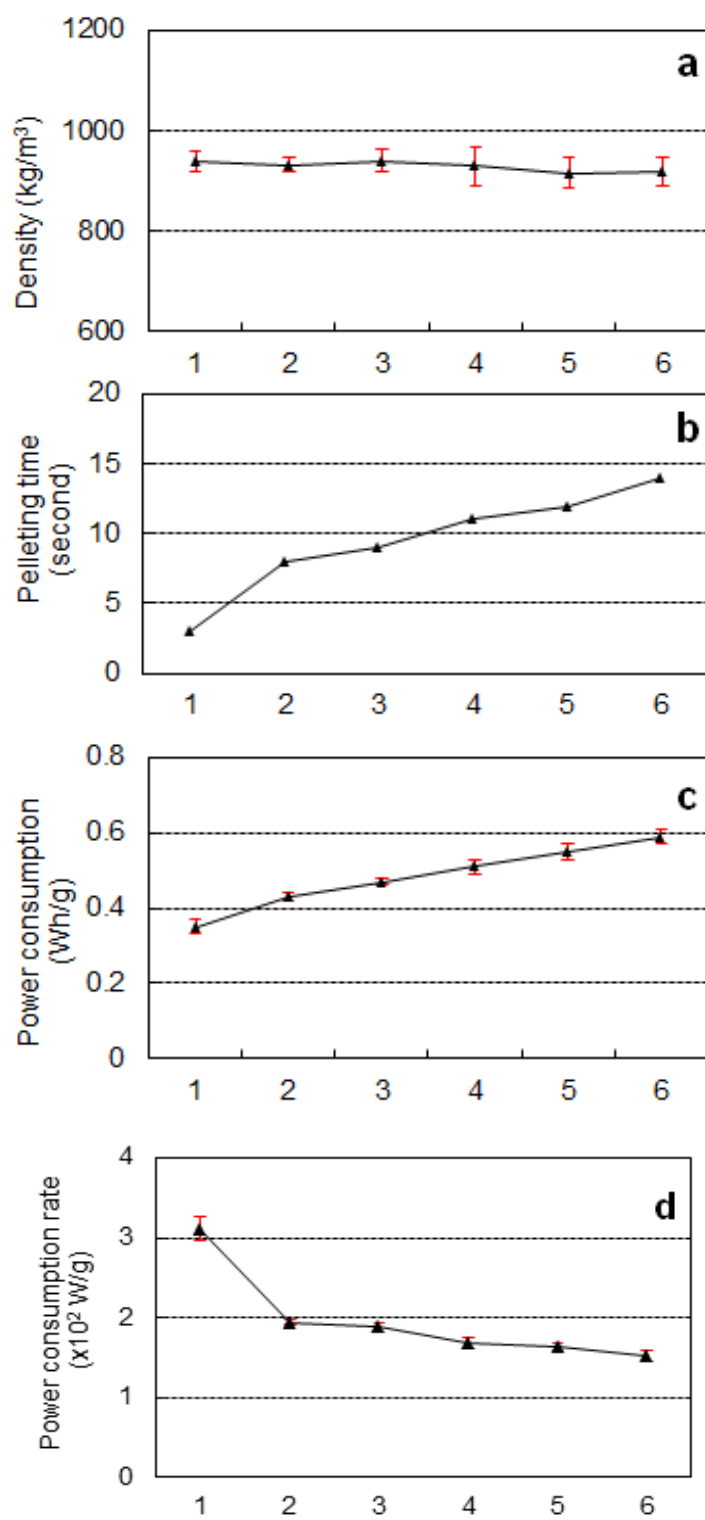
The effects of particle size on power consumption are shown in Figure 8.6 (c). It is obvious that power consumption increased as particle size increased. This result is consistent with a previous study in which effects of particle sizes (3.2 and 9.6 mm) on pelleting power

consumption of corn stover, sorghum stalk, big blue, and wheat straw were investigated by using two pelleting methods [23]. One was ring-die pelleting (a traditional pelleting method) and the other was UV-A pelleting. It is reported that 3.2 mm particles consumed less power than 9.6 mm particles in both pelleting methods. It can be seen from Figure 6 (c) that the trend between particle size and power consumption is similar to that between particle size and pelleting time. The correlation between pelleting time and power consumption indicates that the higher power consumption of larger particle size might be caused by longer pelleting time.

Power consumption rate for different particle size is presented in Figure 8.6 (d). As particle size increased, power consumption rate decreased. This indicates that the higher power consumption for larger particle size was totally caused by the longer pelleting time needed to produce the specific pellet density. Therefore, from the viewpoint of energy consumption in UV-A pelleting, smaller particle size was preferable to producing pellets with a specific density.

Earlier studies on UV-A pelleting showed that smaller particles were also preferable to produce pellets with high durability and stability [13,22]. However, inconsistent relations between particle size and pellet quality were reported by other studies in which other pelleting methods were used. Theerarattananoon et al. [24] reported that sieve size of hammer mill did not have significant effects on pellet density and durability in ring-die pelleting. Similar results were also reported by Tabil and Sokhansanj [25].

Figure 8.6 Results for different particle size



1-6 stands for particle size ranges of <0.2, 0.2-0.3, 0.3-0.4, 0.4-0.6, 0.6-1.2, and 1.2-2.4 mm, respectively.

**Table 8.4 Results for different particle size**

Particle size (mm)	Density* (kg/m <sup>3</sup> )	Pelleting time (second)	Power consumption* (Wh/g)	Power consumption rate* (×10 <sup>2</sup> W/g)
<0.2	942 (21)	4	0.35 (0.02)	3.11 (0.15)
0.2-0.3	931 (14)	8	0.43 (0.01)	1.94 (0.04)
0.3-0.4	940 (23)	9	0.47 (0.01)	1.88 (0.06)
0.4-0.6	929 (39)	11	0.51 (0.02)	1.68 (0.06)
0.6-1.2	916 (32)	12	0.55 (0.02)	1.64 (0.05)
1.2-2.4	918 (29)	14	0.59 (0.02)	1.53 (0.07)

\* Numbers outside parentheses are mean values and numbers enclosed in parentheses are standard deviations for n = 10

Some researchers believed that smaller particle size increased the surface area of biomass [26]. The increase in surface area of biomass allowed easier access by enzymes, resulting in higher sugar yield in hydrolysis. However, Zhang et al. [27] reported that particle size in the range of 0.2-2.4 mm of switchgrass did not have significant effects on sugar yield in hydrolysis after UV-A pelleting. This result might be due to the narrow ranges of particle sizes.

Smaller particle sizes required more power in size reduction [28]. Deines and Pei [29] reported that more power was consumed to produce smaller particle size in knife milling of switchgrass.

### ***8.3.3 Effects of pressure***

The effects of pelleting pressure on power consumption are shown in Table 8.5 and Figure 8.7. The error bars in Figure 8.7 represent the standard deviations presented in Table 8.5. In this study, the pelleting pressure has been set to five levels (30, 35, 40, 45, and 50 psi). The metric unit was converted. The pellet densities of different pressure are presented in Figure 8.7 (a). There was no significant difference between the densities at the significance level of 0.05.

As shown in Figure 8.7 (b), pelleting time decreased as pressure increased. This indicates that lower pressure would lead to lower pellet density if same pelleting time was used. This trend was consistent with earlier results. Zhang et al. [30] investigated four levels of pelleting pressure

(137, 206, 275, and 344 kPa) in UV-A pelleting. A significant increase in pellet density was found as pressure increased from 137 to 344 kPa. Similar trend was also reported by Zhang et al. [22].

The effects of pressure on power consumption are shown in Figure 8.7 (c). It is obvious that power consumption decreased as pressure increased. The relations between power consumption rate and pressure are presented in Figure 7 (d). As pressure increased, there was no significant change in power consumption rate. This indicates that the higher power consumption for lower pressure was totally caused by the longer pelleting time needed to produce the specific pellet density. Therefore, from the viewpoint of energy consumption and productivity in UV-A pelleting, higher pressure was preferable.

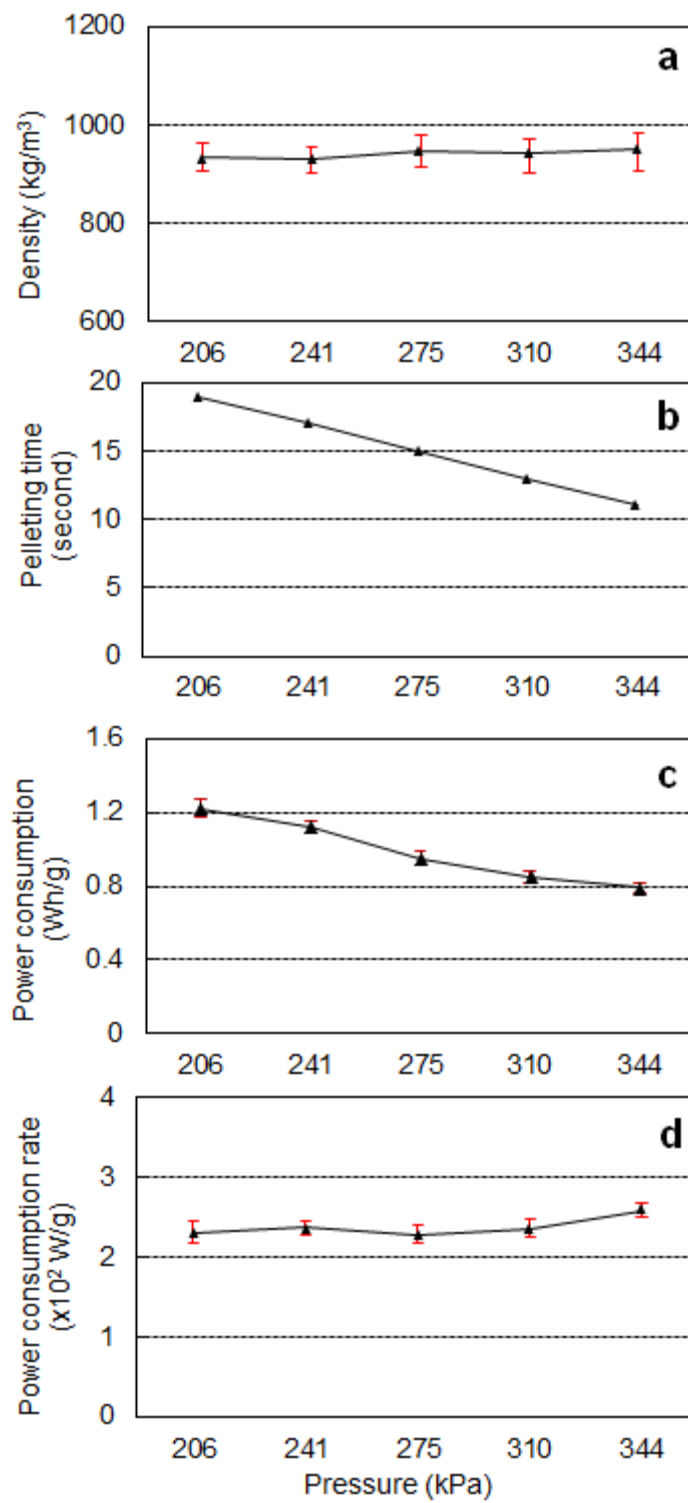
Previous studies also showed that as pressure increased from 137 to 275 kPa, pellet durability increased [30]. As pressure increased from 137 to 275 kPa, there was no significant change in pellet stability [30].

**Table 8.5 Results for different pelleting pressure**

Pressure (kPa)	Pressure (psi)	Density* (kg/m <sup>3</sup> )	Pelleting time (second)	Power consumption* (Wh/g)	Power consumption rate* (W/g)
206	30	933 (32)	19	1.22 (0.05)	2.31 (0.14)
241	35	932 (25)	17	1.12 (0.03)	2.37 (0.08)
275	40	946 (33)	15	0.95 (0.04)	2.29 (0.11)
310	45	942 (30)	13	0.85 (0.03)	2.36 (0.11)
344	50	950 (35)	11	0.79 (0.03)	2.59 (0.08)

\* Numbers outside parentheses are mean values and numbers enclosed in parentheses are

**Figure 8.7 Results for different pressure**



### ***8.3.4 Effects of ultrasonic power***

The effects of ultrasonic power on power consumption are shown in Table 8.6 and Figure 8. The error bars in Figure 8.8 represent the standard deviations presented in Table 8.6. The pellet densities for different levels of ultrasonic power are presented in Figure 8.8 (a). There was no significant difference between the densities at the significance level of 0.05.

As shown in Figure 8.8 (b), pelleting time decreased rapidly as ultrasonic power increased. This indicates that lower ultrasonic power would lead to lower pellet density if same pelleting time was used. This trend was consistent with earlier results. Zhang et al. [30] investigated four levels of ultrasonic power (30%, 40%, 50%, and 55%) in UV-A pelleting. A significant increase in pellet density was found as ultrasonic power increased from 30% to 55%. Similar trend was also reported in the results of a test with a 24 factorial design [22].

The effects of pressure on power consumption are shown in Figure 8.8 (c). Power consumption decreased rapidly as ultrasonic power increased from 50% to 100%. The effects of ultrasonic power on power consumption rate are presented in Figure 8.8 (d). As ultrasonic power increased, power consumption rate increased. This indicates that the higher power consumption for lower ultrasonic power was totally caused by the longer pelleting time needed to produce the specific pellet density. Therefore, from the viewpoint of energy consumption in UV-A pelleting, higher ultrasonic power was preferable.

Higher ultrasonic power could produce pellets with higher durability and stability [30]. Sugar yield in hydrolysis also increased as ultrasonic power in UV-A pelleting increased [30]. Overall, using higher ultrasonic power was beneficial to UV-A pelleting.

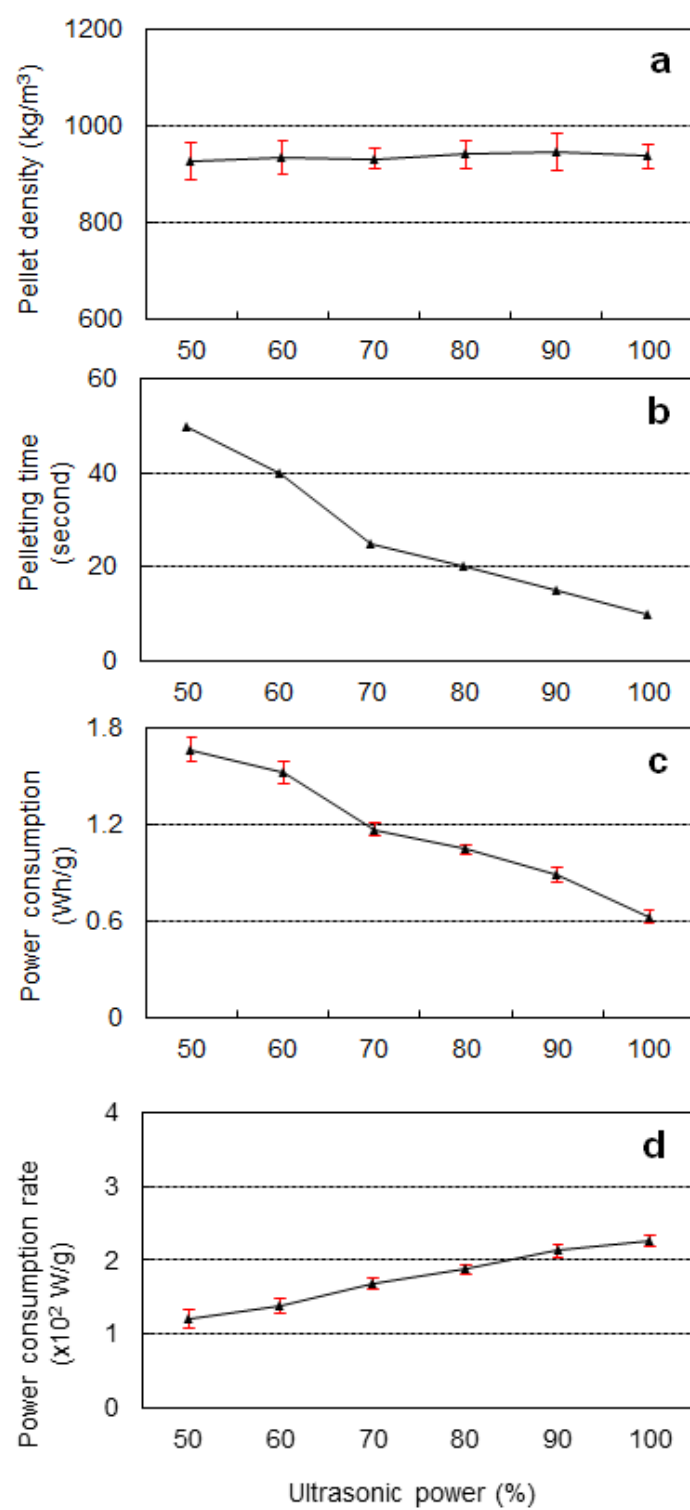


**Table 8.6 Results for different ultrasonic power**

Ultrasonic power (%)	Density* (kg/m <sup>3</sup> )	Pelleting time (second)	Power consumption* (Wh/g)	Power consumption rate* (W/g)
50	928 (38)	50	1.67 (0.08)	1.20 (0.13)
60	935 (35)	40	1.53 (0.07)	1.38 (0.11)
70	932 (21)	25	1.17 (0.04)	1.68 (0.07)
80	943 (29)	20	1.05 (0.03)	1.88 (0.06)
90	946 (38)	15	0.89 (0.05)	2.13 (0.09)
100	938 (26)	10	0.63 (0.04)	2.26 (0.08)

\* Numbers outside parentheses are mean values and numbers enclosed in parentheses are standard deviations for n = 10

**Figure 8.8 Results for different ultrasonic power**



### **8.3 Conclusions**

This paper presents the effects of moisture content, particle size, pressure, and ultrasonic power on power consumption in ultrasonic vibration-assisted (UV-A) pelleting. The following conclusions can be drawn from the study.

As moisture content increased from 5% to 15%, power consumption in UV-A pelleting did not change much. As moisture content increased from 15% to 25%, power consumption increased dramatically. Excluding the effects of pelleting time, higher moisture content in itself would lead to higher power consumption. Lower moisture content is preferable in UV-A pelleting.

As particle size increased, power consumption in UV-A pelleting increased but power consumption rate decreased. The higher power consumption for larger particle size was totally caused by the longer pelleting time needed to produce the specific pellet density. Smaller particle size is beneficial to UV-A pelleting in terms of high pellet quality and low power consumption. However, the power consumption in size reduction is increased dramatically to produce smaller particles.

As pressure increased from 206 to 344 kPa, power consumption in UV-A pelleting decreased but there was no significant change in power consumption rate. The higher power consumption for lower pressure was totally caused by the longer pelleting time needed to produce the specific pellet density.

As ultrasonic power increased from 50% to 100%, power consumption in UV-A pelleting decreased but power consumption rate increased. The higher power consumption for lower ultrasonic power was totally caused by the longer pelleting time needed to produce the specific pellet density.

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## **Chapter 9 - Investigation on power consumption with design of experiment**

### Paper Title:

Ultrasonic vibration-assisted pelleting for cellulosic biofuel manufacturing: investigation on power consumption with design of experiment

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## **Abstract**



Cellulosic biomass is abundantly available in the nature. It is an attractive feedstock to make alternative fuels to petroleum-based transportation fuels. Because of low bulk density and irregular shape, raw biomass materials are difficult to handle, transport, and store. Pelleting can increase the density of cellulosic biomass. Pellets can be easily handled, resulting in reduced transportation and storage costs. Ultrasonic vibration-assisted (UV-A) pelleting is a new pelleting method. Moisture content, particle size, pelleting pressure, and ultrasonic power are four important input parameters affecting pellet quality and sugar yield (proportional to biofuel yield). However, their effects on power consumption in UV-A pelleting have not been adequately investigated. Since power consumption directly affects ethanol manufacturing costs, it is desirable to understand how input parameters affect power consumption. This paper reports an experimental investigation of power consumption in UV-A pelleting. A  $2^4$  factorial design is employed to evaluate the effects of four input parameters (moisture content, particle size, pelleting pressure, and ultrasonic power) on power consumption in UV-A pelleting. Results show that three input parameters (moisture content, particle size, and ultrasonic power) significantly affect power consumption. Higher moisture content, lower ultrasonic power, and larger particle size result in higher power consumption. Only one interaction of two parameters is significant, i.e. with the increase of pelleting pressure, power consumption will increase at the high level of particle size while decrease at the low level of particle size.

## **KEYWORDS**

Biofuel; Cellulosic biomass; Moisture content; Particle size; Pellet; Power consumption; Pelleting pressure; Ultrasonic vibration

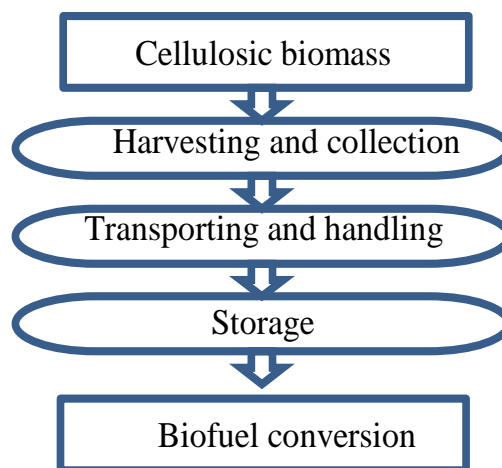
## **9.1 Introduction**

It is important to substitute petroleum-based transportation fuels with renewable energy. More than 50% of the petroleum consumed in the U.S is imported, making the U.S. energy depend on other countries. Cellulosic ethanol, produced from cellulosic biomass, is used as a

substitute for petroleum-based transportation fuels and is a renewable energy source. Cellulosic biomass (herbaceous, woody, and generally inedible portions of plant matter) is abundantly available in the nature. Land resources in the U.S. are sufficient to produce 1.3 billion dry tonne of cellulosic biomass annually, from which enough ethanol can be made to replace over 50% of gasoline used currently in the U.S. [1]. Comparing with corn-based biofuels, cellulosic biomass is outside the food chain, thus will not compete with food and feed production for limited agricultural land. Furthermore, use of cellulosic biomass in place of liquid transportation fuels would reduce greenhouse gas (GHG) emissions by 85% over petroleum-based fuels [2,3].

In order to use cellulosic materials for manufacturing liquid transportation fuels, some challenges must be resolved. Figure 9.1 shows major steps in manufacturing of biofuels. After harvesting, cellulosic biomass materials are transported from fields to warehouse and stored for future use. High moisture content, irregular shape and size, and low bulk density make cellulosic biomass very difficult to handle, transport, store in its original form, resulting in high transportation and storage costs [6,7].

**Figure 9.1 Major steps in manufacturing of cellulosic biofuels (after [4,5])**



One solution to these problems is densification of biomass materials into pellets. Pelletizing is the agglomeration of small particles into larger particles by the means of a mechanical process, and thermal processing in some applications [8]. The density of pelleted

cellulosic biomass can be as high as 1200 kg/m<sup>3</sup> [9], while the density of raw biomass materials is usually 40 to 250 kg/m<sup>3</sup> [10,11]. Furthermore, pellets with uniform size and shape are easier to handling and storage using existing equipment, resulting in reduced costs of transportation, storage, and handling.

Traditional pelleting methods (e.g., using a screw extruder, a briquetting press, or a rolling machine [11,12]) usually involve high-temperature steam, high pressure, and binder materials. It is difficult to realize cost-effective pelleting at or near the fields where cellulosic biomass is available by using traditional pelleting methods. Ultrasonic vibration-assisted (UV-A) pelleting is a new pelleting method without using high-temperature steam, high pressure, and binder materials. It can produce biomass pellets whose density is comparable to those produced by traditional pelleting methods [13].

Earlier studies on UV-A pelleting show that moisture content (MC), particle size, pelleting pressure, and ultrasonic power are four important parameters affecting pellet quality (in terms of density, durability, and spring back), and sugar yield (proportional to biofuel yield) [14-17]. However, their effects on power consumption in UV-A pelleting have not been adequately investigated. Since power consumption directly affects biofuel manufacturing costs, it is desirable to understand how these input parameters affect power consumption. The objective of this paper is to study main and interaction effects of input parameters (moisture content, particle size, pelleting pressure, and ultrasonic power) on power consumption in UV-A pelleting of wheat straw via employing a 2<sup>4</sup> full factorial design.

## **9.2 Experimental conditions and procedures**

### ***9.2.1 Collection of wheat straw***

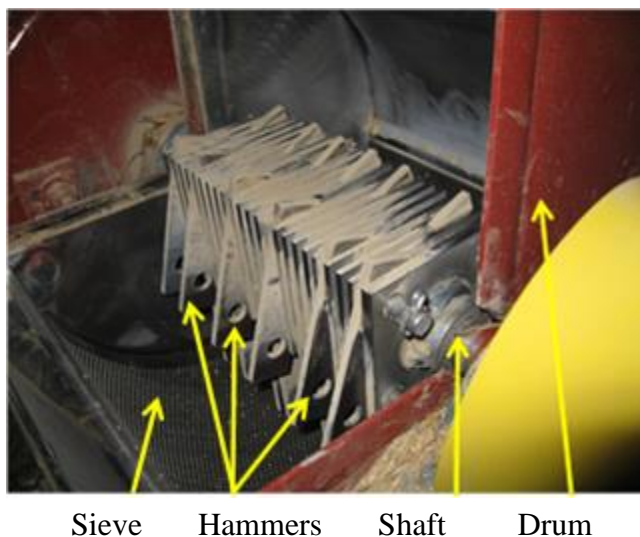
Wheat straw was harvested by Deines Farms in northwestern Kansas in July of 2010. The wheat straw had been run through a John Deere 9600 combine that removed wheat grains from wheat straw and chaff. Wheat straw, exited from the back of the combine, had an average length

of 25 cm. After harvesting, wheat straw was transported to a lab located in Durland Hall at Kansas State University.

### ***9.2.2 Further reduction of particle size***

The size of wheat straw was further reduced using a hammer mill (model 35, Meadows Mills, Inc., North Wilkesboro, NC, USA) as shown in Figure 9.2. The hammer mill had a steel drum containing a rotating shaft on which hammers were mounted. The shaft rotated at 3600 rpm and the hammers were free to swing. The wheat straw was fed into the grinding drum from the top of the hammer mill. The rotating hammers impacted the wheat straw to further reduce the particle size. If wheat straw particles were small enough, they would pass through the sieve at the bottom of the grinding chamber [19]. The size of the sieve on the hammer mill was 2 mm.

**Figure 9.2 Further size reduction by a hammer milling**



### ***9.2.3 Separation of particle sizes***

Particle sizes had a wide size distribution after milling. They were separated into different size ranges using a sieve shaker (model RX-29, W.S. Tyler, Inc., Mentor, OH, U.S.) as shown in Figure 9.3. A series of sieves with different screen sizes were loaded on an agitation tray. Particles were put on the top sieve that had the largest screen size. A hammer stroke the black

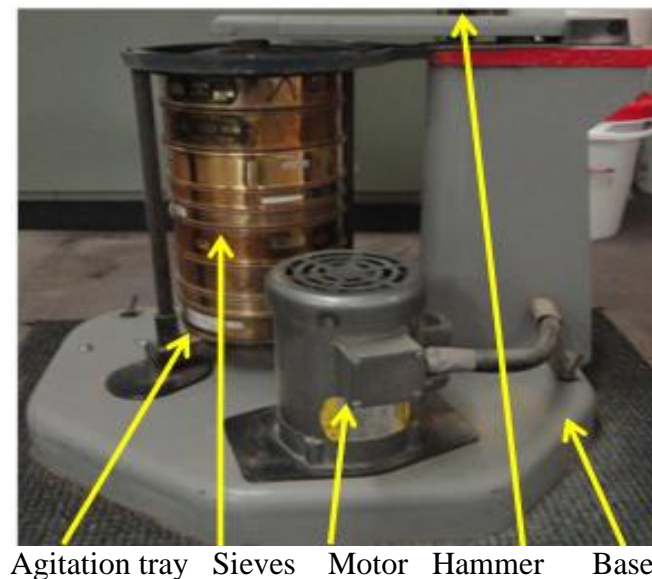
cover located above the sieves three times per second. Meanwhile, the agitation tray moved circularly at 200 rpm. The running time of the sieve shaker was ten minutes.

The particle size was determined by the screen size of the sieves. Table 9.1 lists screen sizes of the six sieves used to separate the wheat straw particle size ranges. Theoretically, particles should be separated into seven different size ranges with these six sieves. However, almost all particles fell through the 2.4 mm sieve, so the particle size range of  $> 2.4$  mm was excluded. Therefore, particles were separated into six different sizes:  $<0.2$ ,  $0.2 - 0.3$ ,  $0.3 - 0.4$ ,  $0.4 - 0.6$ ,  $0.6 - 1.2$ , and  $1.2 - 2.4$  mm. Only two particle size ranges were used in this study:  $<0.2$ , and  $0.6 - 1.2$  mm.

**Table 9.1 Screen sizes of sieves**

Sieve #	Screen size (mm)
1	2.4
2	1.2
3	0.6
4	0.4
5	0.3
6	0.2

**Figure 9.3 Sieve shaker**



#### ***9.2.4 Adjustment of moisture content***

Moisture content (MC) in this paper refers to the moisture content of the wheat straw particles right before UV-A pelleting. It represents the amount of moisture (water) contained in wheat straw particles. MC was calculated by the ratio of the weight of the water in the wheat straw particles to the total weight of the wheat straw particles.

MC of biomass particles after milling was measured and adjusted according to ASAE standard S 358.2 [19]. The initial MC of biomass was measured by the following procedure. 25 grams of biomass was weighed by an electronic scale (Ohaus, Pine Brook, NJ, USA) and then heated in an oven at 103 °C for 24 hours to evaporate the moisture. After heating, the weight of the dry sample was weighed again. The initial MC was calculated by Equation (1).

The initial MC of wheat straw was 5%. The MC was adjusted to desired level (wet basis) - 15% - by mixing tap water with wheat straw particles and stirring manually for 2 mins at room temperature. The water added was calculated by Equation (2). After MC was adjusted, the wheat straw particles were stored in zip-lock bags until being pelleted.

$$\text{Initial moisture content} = \frac{\text{Initial biomass weight} - \text{Weight after heating}}{\text{Initial biomass weight}} \times 100\% \quad (1)$$

$$\text{Moisture content (wet basis)} = \frac{\text{Weight of biomass} \times \text{Initial moisture content} + \text{Water added}}{\text{Weight of biomass} + \text{Water added}} \times 100\% \quad (2)$$

#### ***9.2.5 UV-A pelleting***

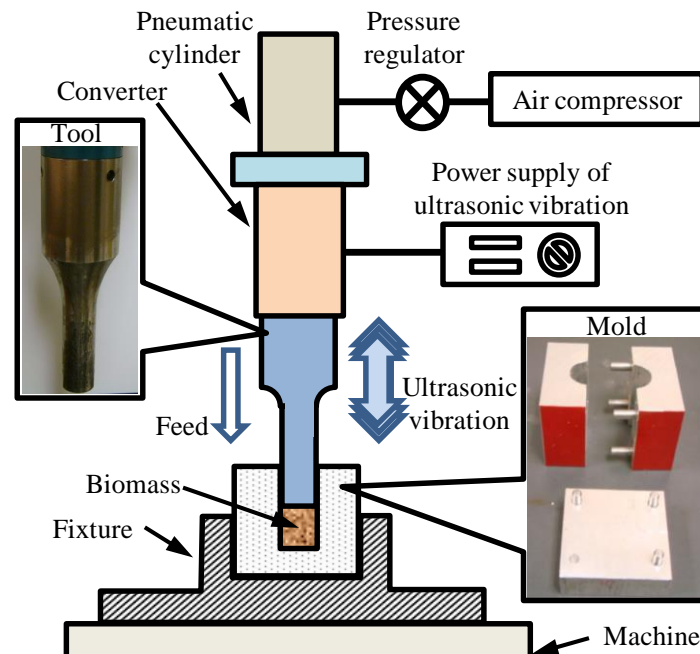
Pelleting is performed on a modified ultrasonic machine (model AP-1000, Sonic-Mill, Albuquerque, NM, U.S.). Figure 9.4 is a schematic illustration of the experimental set-up for UV-A pelleting. An aluminum mold was made in three parts that were assembled together with pins. The top two parts formed a cylindrical cavity (18.6 mm in diameter) and the bottom part served as a base. For each pellet, one gram of wheat straw particles was loaded into the center cavity of the mold and the mold was clamped by a fixture. The titanium tool was connected to an ultrasonic converter. The tip of the tool was a round solid (17.4 mm in diameter) with a flat end.

The tool was fed into the wheat straw particles in the mold. During pelleting, the pneumatic cylinder applied a pressure to the wheat straw particles through the tool, and the ultrasonic converter provided ultrasonic vibration to the tool. After a period of time during which the tool was in contact with the wheat straw particles, the tool was retracted and the mold was disassembled to unload the cylinder-shaped pellet. The pellet was weighed again by an electronic scale (Ohaus, Pine Brook, NJ, USA). The density of a pellet is determined by the ratio of its weight to its volume. The volume of a pellet was determined by its diameter and height measured with a vernier caliper. The height of pellets was  $4.57 \pm 0.4$  mm and diameter of pellets was  $18.62 \pm 0.1$  mm. Density of all pellets were kept in the range of 900 to 950 Kg/m<sup>3</sup>.

Pelleting pressure and ultrasonic power are two important pelleting parameters. The pelleting pressure represented the air pressure in the pneumatic cylinder. A higher air pressure would cause a higher pressure applied on the wheat straw particles by the tool.

Ultrasonic power was referred to the power provided by a power supply. It controlled the amplitude of the tool vibration. A higher ultrasonic power would result in a larger vibration amplitude. Ultrasonic power was expressed as a percentage of the maximum ultrasonic power for the power supply. It could be adjusted from 0 (no ultrasonic power) to 100% (the maximum ultrasonic power). The vibration frequency of the tool was fixed at 20 kHz.

**Figure 9.4 Illustration of experimental set-up for UV-A pelleting**



### 9.2.6 Design of experiments

A  $2^4$  (four parameters, two-levels) full factorial design was used for the experiments with four replications under each condition. Table 9.2 lists four parameters and their levels. For each parameter, there are two levels. The matrix of the experiments is shown in Table 9.3. These tests were conducted in a random order. The parameter levels were determined based on the results of a previous experimental study [29].

**Table 9.2 Variables and their levels**

Variable	Unit	Low level (-)	High Level (+)
Moisture content	%	5	15
Particle size	mm	<0.2	0.6-1.2
Pelleting pressure	psi	35	50
Ultrasonic power	%	65	95



### 9.3 Measurement procedure for power consumption

The term “power consumption” in this paper refers to the consumed electrical energy measured by a power analyzer (AEMC 2010.86 PowerPad Jr. Model 8230, AEMC-Instruments, Foxborough, MA). Voltage probe leads were connected to the 120 Volt AC cable and a current sensor was clamped around the AC cable. The power analyzer began recording voltage and current when the tool started dropping and stopped recording data when the tool started retreating. The power analyzer displayed power consumption in the unit of Wh.

After dividing the power consumption by the mass of biomass in the pellet, the unit of power consumption became Wh/g. The average value of measurements was calculated and used to plot the graphs in Figures 9.5 and 9.6.

**Table 9.3 Matrixes of the experiments**

Test number	Moisture content	Particle size	Pelleting pressure	Ultrasonic power
Test 12, 32, 49 ,52	-1	-1	-1	-1
Test 33, 45, 51, 58	-1	-1	-1	1
Test 4, 15, 26, 56	-1	-1	1	-1
Test 9, 19, 22, 34	-1	-1	1	1
Test 3, 25, 29, 50	-1	1	-1	-1
Test 16, 42, 47, 57	-1	1	-1	1
Test 7, 36, 41, 55	-1	1	1	-1
Test 10, 21, 23, 60	-1	1	1	1
Test 6, 14, 27, 44	1	-1	-1	-1
Test 31, 35, 37, 40	1	-1	-1	1
Test 17, 24, 39, 46	1	-1	1	-1
Test 2, 5, 8, 28	1	-1	1	1
Test 20, 43, 48, 54	1	1	-1	-1
Test 1, 11, 62, 63	1	1	-1	1
Test 18, 38, 53, 59	1	1	1	-1
Test 13. 30, 61, 64	1	1	1	1

## 9.4 Experimental results

Minitab (Version 16) was used to process the experimental data. To identify the significant effects, the analysis of variance (ANOVA) was performed.

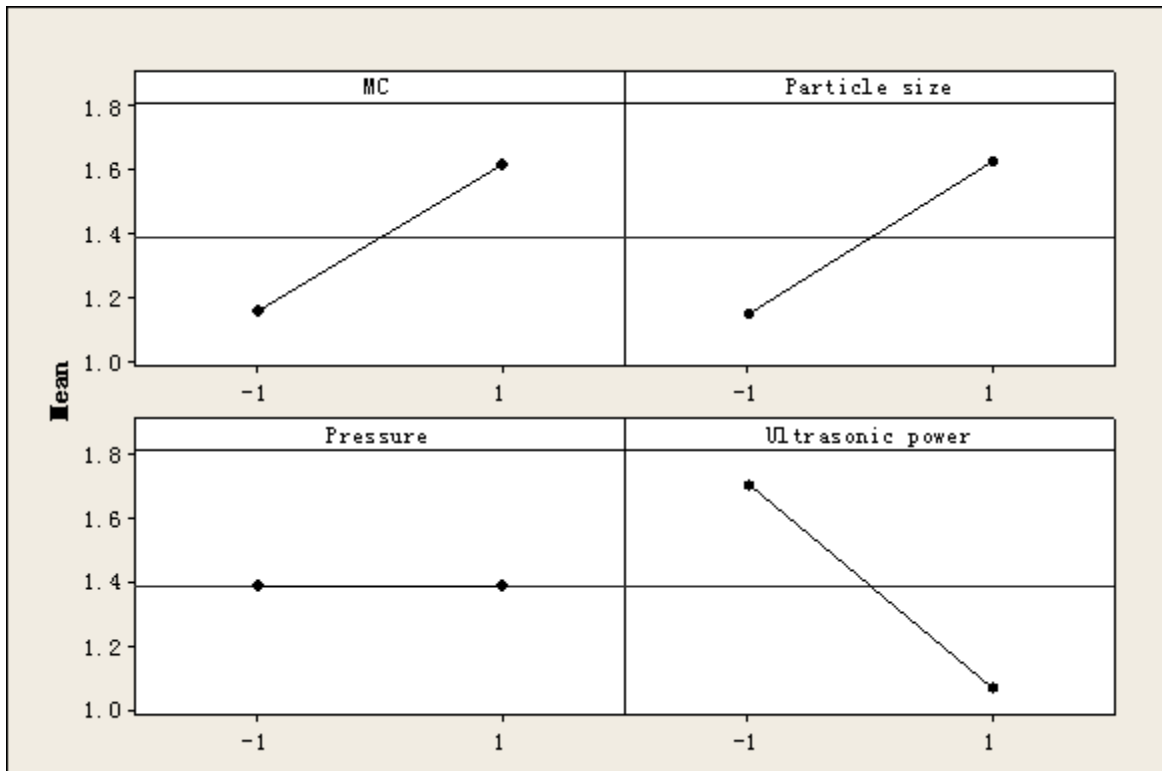
The results on power consumption are presented in Table 9.4. The significant main and interaction effects of parameters on power consumption are shown in Figures 9.5 and 9.6, respectively. All the main effects of MC, particle size, and ultrasonic power are significant at the significance level of  $\alpha = 0.05$ . The main effect of pelleting pressure is not significant because P value = 0.982 >  $\alpha$  (0.05). The null hypothesis (there is no significant difference in the power consumption among two levels of pelleting pressure) cannot be rejected. From Figure 9.5, it can be seen that power consumption increases with an increase of MC and particle size. Power consumption decreases as ultrasonic power increases.

Figure 9.6 shows interaction effects between particle size and pressure. The analysis results show the interaction (P-value = 0.008) is significant at the significance level of  $\alpha = 0.05$ . With the increase of pelleting pressure, power consumption will increase at the high level of particle size while decrease at the low level of particle size. The analysis results from ANOVA also indicate interaction effects of MC and particle size (P-value = 0.094), MC and pelleting pressure (P-value = 0.36), MC and ultrasonic power (P-value = 0.399), particle size and ultrasonic power (P-value = 0.071), and pelleting pressure and ultrasonic power (P-value = 0.638) are not significant the significance level of  $\alpha = 0.05$ .

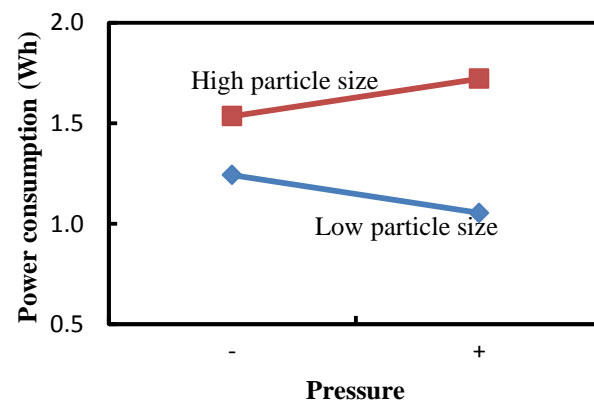
**Table 9.4 Results on power consumption**

Test number	Moisture content	Particle size	Pelleting pressure	Ultrasonic power	Power consumption			
					Replication 1	Replication 2	Replication 3	Replication 4
Test 12, 32, 49, 52	-1	-1	-1	-1	1.18	1.03	1.89	1.18
Test 33, 45, 51, 58	-1	-1	-1	1	0.45	0.57	0.65	0.63
Test 4, 15, 26, 56	-1	-1	1	-1	0.89	1.06	0.81	1.09
Test 9, 19, 22, 34	-1	-1	1	1	0.55	0.65	0.61	0.52
Test 3, 25, 29, 50	-1	1	-1	-1	1.35	1.22	1.37	1.34
Test 16, 42, 47, 57	-1	1	-1	1	1.43	1.15	1.2	1.4
Test 7, 36, 41, 55	-1	1	1	-1	2.12	2.1	2.03	2.49
Test 10, 21, 23, 60	-1	1	1	1	1.1	1.08	0.83	1.09
Test 6, 14, 27, 44	1	-1	-1	-1	1.86	1.94	1.77	1.93
Test 31, 35, 37, 40	1	-1	-1	1	1.01	1.32	1.28	1.19
Test 17, 24, 39, 46	1	-1	1	-1	1.63	1.49	1.28	1.42
Test 2, 5, 8, 28	1	-1	1	1	1.37	1.19	1.22	1.08
Test 20, 43, 48, 54	1	1	-1	-1	2.16	1.75	2.75	2.33
Test 1, 11, 62, 63	1	1	-1	1	1.15	1.15	1.81	1.01
Test 18, 38, 53, 59	1	1	1	-1	2.37	2.21	2.07	2.48
Test 13, 30, 61, 64	1	1	1	1	1.27	1.2	1.46	1.64

**Figure 9.5 Main effects on power consumption**



**Figure 9.6 Interaction effects of pressure and particle size**



## 9.5 Conclusions

This paper presents the main and interaction effects of moisture content (MC), particle size, pelleting pressure, and ultrasonic power on power consumption in UV-A pelleting of wheat straw. The following conclusions can be drawn from the study.

Main effects of MC, particle size, and ultrasonic power are significant at the significance level of  $\alpha = 0.05$ . Larger particle size, higher MC, and lower ultrasonic power result in higher power consumption.

Interaction effects of particle size and pelleting pressure are significant. With the increase of pressure, power consumption will increase at the high level of particle size while decrease at the low level of particle size.

### Acknowledgements

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## Chapter 10 - Investigation on temperature in UV-A pelleting

### Paper Title:

Ultrasonic vibration-assisted pelleting of cellulosic biomass for ethanol manufacturing:  
an investigation on pelleting temperature

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### **Abstract**

Ethanol made from cellulosic biomass is an alternative to petroleum-based liquid transportation fuels. However, large-scale manufacturing of cellulosic ethanol is hindered by the low density of cellulosic biomass. Ultrasonic vibration-assisted (UV-A) pelleting can effectively increase biomass density by compressing raw biomass into pellets. Temperature of biomass in pelleting (referred to as pelleting temperature) has been identified as a key factor influencing



pellet quality. This paper reports an investigation on pelleting temperature in UV-A pelleting of wheat straw. The precision of temperature measurement was first evaluated. The relationships between pelleting temperature and pelleting time were then investigated. The pattern of pelleting temperature distribution was evaluated by ranking the pelleting temperatures at six different locations in a pellet. Also, the main and interaction effects of three input variables (ultrasonic power, pelleting pressure, and pellet weight) on pelleting temperature were investigated.

## **10.1 Introduction**

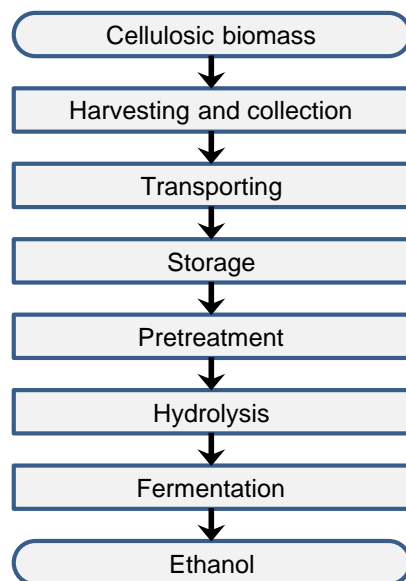
Ethanol produced from cellulosic biomass (herbaceous, woody, and generally inedible portions of plant matter) is an alternative to petroleum-based liquid transportation fuels. Land resources in the U.S. are sufficient to sustain production of enough biomass annually to displace 30% or more of the nation's current petroleum consumption [1]. Furthermore, manufacturing and using of cellulosic ethanol can mitigate the accumulation of greenhouse gas in the atmosphere and boom rural economy [2].

Major steps for manufacturing of cellulosic ethanol are shown in Figure 10.1. One major challenge to cost-effective manufacturing of cellulosic ethanol is high transportation and storage costs caused by low density of raw cellulosic biomass (ranging from 24 to 266 kg/m<sup>3</sup> [4]). Pelleting processes can effectively increase density of cellulosic biomass to higher than 600 kg/m<sup>3</sup> [5-7]. In turn, costs for transporting and storing of pelleted cellulosic biomass are less than 1/2 and 1/10 of those of raw cellulosic biomass respectively [8,9]. In addition, pellets with uniform size and shape are easier to handle with existing equipment for grain processing than raw cellulosic biomass [5].

Several traditional pelleting methods have been reported in the literature, including ring-die pelleting [5,10-13], flat-die pelleting [10,11], screw extrusion [12,14], piston press [15], and roll press [13]. For these pelleting methods, the temperature of biomass has been identified as a key factor affecting pellet quality in terms of density, durability, and stability. In general, biomass needs to be heated (by high-temperature steam or heated dies) to a high temperature to

achieve good pellet quality [13]. Faborode [16] reported that biomass should be heated to 100°C to produce high pellet density and durability. Reece [17] reported that heating biomass to 60 – 70°C was needed to produce stable pellets. A possible reason for this is that high temperature could melt lignin (a major component in biomass) which would exhibit thermosetting properties and act as the binder material for biomass particles to form pellets [18].

**Figure 10.1 Major steps for manufacturing of cellulosic ethanol (after [3]).**



Ultrasonic vibration-assisted (UV-A) pelleting is a new pelleting method which uses an ultrasonically vibrating tool to compress biomass and make pellets (more detailed information will be provided in Section 10.2). Unlike most traditional pelleting methods in which biomass is heated before being compressed, UV-A pelleting uses unheated biomass as feedstock and produces pellets whose density and durability are comparable to those produced by traditional pelleting methods [6,19]. In addition, a previous study reported that cellulosic biomass (switchgrass) processed by UV-A pelleting produced 20% higher sugar yield than that not processed by UV-A pelleting [6].

Table 10.1 summarizes the input and output variables in UV-A pelleting that have been investigated. There are few studies that investigated temperature in UV-A pelleting. Feng et al. [34] investigated the effects of input variables of UV-A pelleting on mold temperature (the mold was used to contain biomass in UV-A pelleting). However, the results cannot reflect the temperature of biomass in UV-A pelleting. Tang et al. [35] measured temperature of biomass in UV-A pelleting. However, their measurements were taken under one specific pelleting condition. So far, there is no systematic study in the literature on temperature of biomass in UV-A pelleting.

There are three objectives in this paper: (1) to investigate the relationships between pelleting temperature (the temperature of biomass during UV-A pelleting) and pelleting time (the period of time when biomass is treated by UV-A pelleting), (2) to investigate the distribution of pelleting temperature in a pellet in UV-A pelleting, and (3) to investigate effects of three input variables (ultrasonic power, pelleting pressure, and pellet weight) on pelleting temperature.

**Table 10.1 Investigated input and output variables in UV-A pelleting**

Output variable	Input variable						Reference
	Biomass type	Moisture content	Particle size	Pellet weight	Pressure	Ultrasonic power	
Charring	√	√	√			√	[20]
Density	√	√	√	√	√	√	[6,21-27]
Durability		√	√	√	√	√	[6,22,24-26,28]
Power consumption	√	√	√		√	√	[29-31]
Stability		√	√		√	√	[6,22,24-27]
Sugar yield		√	√		√	√	[6,25,26,32,33]
Temperature		√	√		√	√	[34,35]

## 10.2 Material and methods

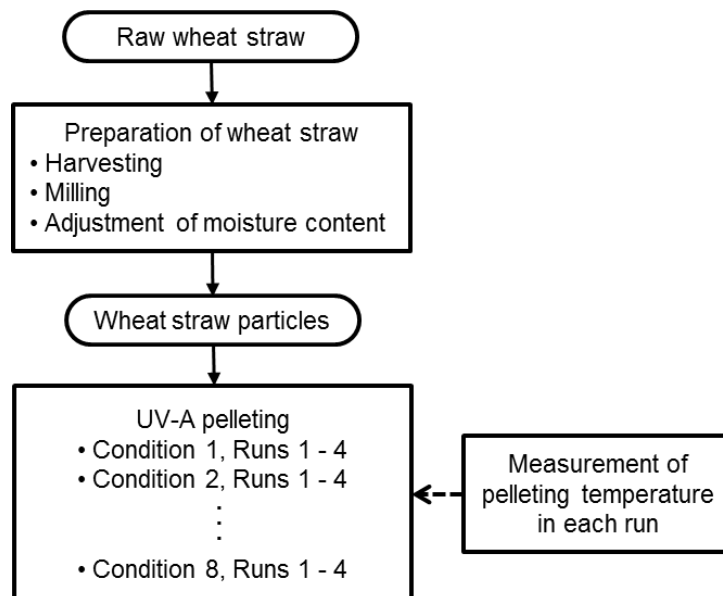
### 10.2.1 Experimental procedure

The experimental procedure is shown in Figure 10.2. Raw wheat straw was converted into particles and prepared for UV-A pelleting through three steps: harvesting, milling, and adjustment of moisture content. The prepared wheat straw particles were processed by UV-A pelleting under eight different pelleting conditions (with three input variables and two levels for each input variable) and four runs of UV-A pelleting were conducted under each pelleting condition. For each run, pelleting temperature was measured.

### 10.2.2 Preparation of wheat straw

The wheat straw used in this investigation was harvested and collected at Deines Farm in Northwest Kansas in late June of 2011. It had been run through a John Deere 9600 combine. The combine separated the grains from the straw and chaff. Wheat straw and chaff exited through the back of the combine. The straw chopper on the combine was disconnected to allow the straw to be baled. The average length of the wheat straw was 17 - 25 cm. After being collected, the wheat straw was transported to the lab in Durland Hall at Kansas State University.

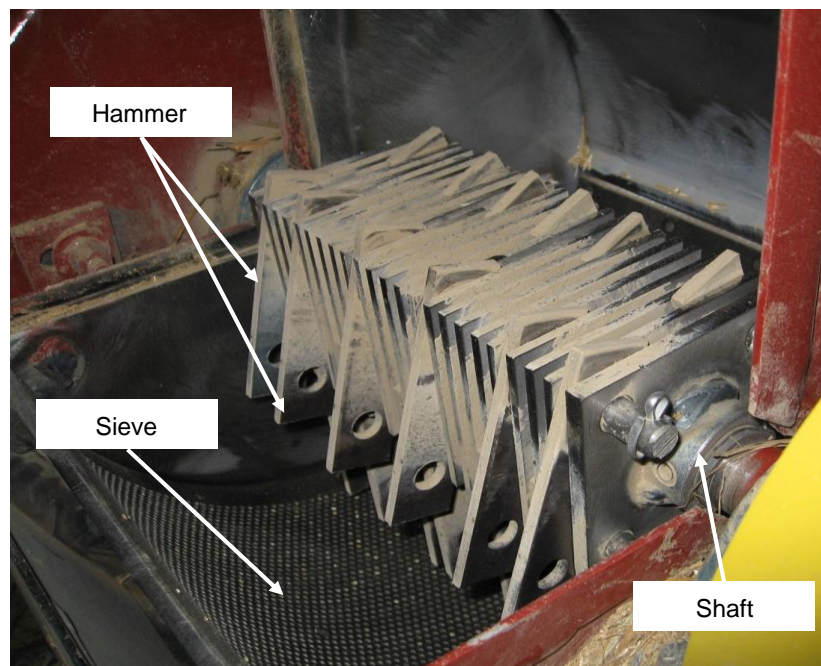
**Figure 10.2 Experimental procedure**



The wheat straw was milled into small particles using a hammer mill (Model 5 from Meadows Mill Inc., North Wickeabord, NC, USA) using a sieve with size of 2 mm. The hammer mill used a 240-volt, 5-horsepower electric motor with a fixed rotation speed (3600 rpm). As shown in Figure 10.3, 24 hammers were mounted on the rotating shaft and were free to swing. The size of these hammers was  $101.6 \times 25.4 \times 4.8$  mm. The wheat straw was fed into the grinding chamber from the top of the hammer mill. The rotating hammers impacted the wheat straw to reduce particle size. Particles would pass through the sieve at the bottom of the grinding chamber if they were smaller than 2 mm. The milled wheat straw was kept in sealed plastic bags at room temperature.

Moisture content of biomass represents the amount of moisture (water) contained in a certain amount of biomass. Moisture content of the wheat straw after milling was measured as 5% by following the ASABE Standard S358.2 [36] and adjusted to 10% by following the NREL LAP [37].

**Figure 10.3 Illustration of hammer milling**

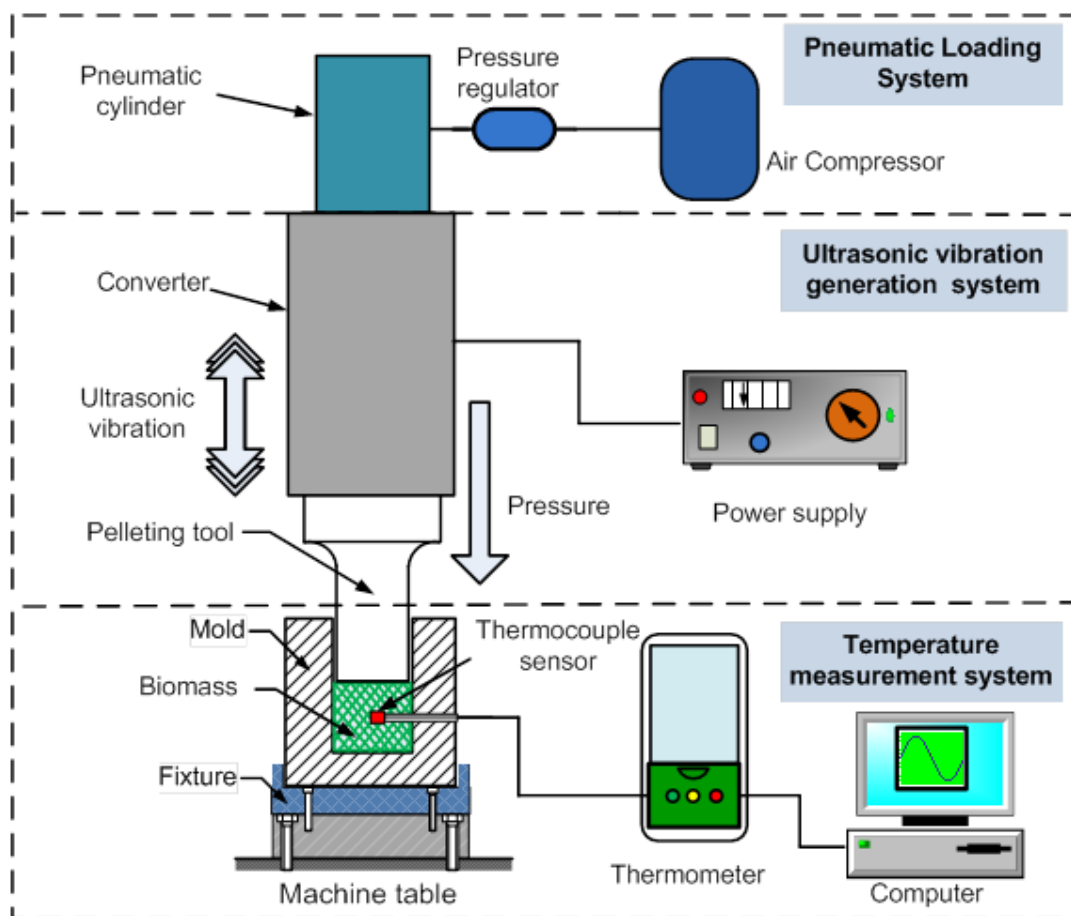


### 10.2.3 Experimental set-up for UV-A pelleting

Pelleting was performed on a modified ultrasonic machine (model AP-1000, Sonic-Mill, Albuquerque, NM, USA). Figure 10.4 is a schematic illustration of the experimental set-up for UV-A pelleting. The machine was composed of two systems: an ultrasonic-vibration generation system, and a pneumatic loading system.

The ultrasonic-vibration generation system was consisted of three major parts: a power supply, a converter, and a pelleting tool. The power supply converted 60-Hz electrical power into 20-kHz electrical power. The converter converted high-frequency electrical energy into mechanical motion. The pelleting tool was made from titanium. It was connected to the converter and used to compress biomass. The tip of the tool was a solid cylinder (17.4 mm in diameter) with a flat end.

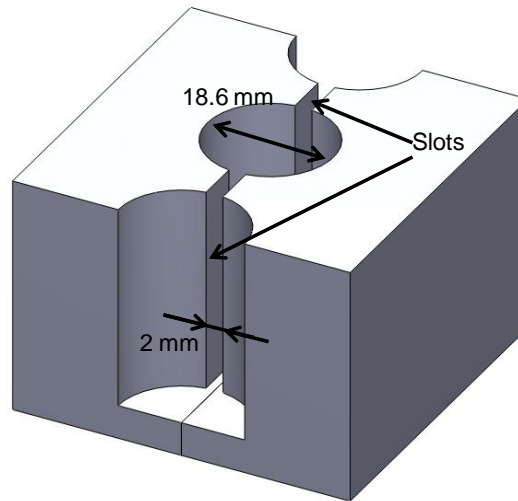
**Figure 10.4 Illustration of experimental set-up for UV-A pelleting**



The pneumatic loading system was consisted of three major parts: an air compressor (1.2 kw, 125 liter, Sears, Roebuck and Co., Hoffman Estates, IL, USA), a pressure regulator, and a pneumatic cylinder (104.4 mm in diameter). Air pressure in the air compressor was set at 0.8 MPa (120 psi). The pressure regulator controlled the air pressure in the pneumatic cylinder. The pneumatic cylinder was driven by the compressed air and pushed the pelleting tool against biomass in a mold. The mold, as illustrated in Figure 10.5, was consisted of two pieces. When the two pieces were assembled together, they formed two slots (the width of the slots was 2 mm) and a cylinder cavity (the diameter of the cavity was 18.6 mm). The mold was clamped by a fixture before pelleting.

The temperature measurement system was consisted of three major parts: thermocouples, a thermometer, and a computer with data acquisition software package. Four thermocouples of metal wire-type (K-type, Model SC-GG-K-30-36, OMEGA Engineering, Inc, Stamford, CT, USA) were inserted in pellets through the slots of the mold, as shown in Fig 10.4. The thermometer (Fluke, HH147U, OMEGA Engineering, Inc., Stamford, CT, USA) had four channels and each channel was connected with one thermocouple. The measured data were recorded by the data acquisition software package (Temp Monitor\_S2, OMEGA Engineering, Inc., Stamford, CT, USA) with a frequency of one measurement per second. The recorded temperature data were saved in an Excel file.

**Figure 10.5 Illustration of the mold used for temperature measurement**



#### ***10.2.4 Input variables in UV-A pelleting and their values***

The eight pelleting conditions used in this study were selected by changing three input variables in UV-A pelleting: ultrasonic power, pelleting pressure, and pellet weight. Ultrasonic power was the power provided by the power supply. It controlled the amplitude of tool vibration in UV-A pelleting. A larger ultrasonic power would result in a larger vibration amplitude. Ultrasonic power was expressed as a percentage of the maximum ultrasonic power for the power supply. It could be adjusted from 0 (no ultrasonic power) to 100% (the maximum ultrasonic power). Pelleting pressure was the pressure in the pneumatic cylinder. A higher pelleting pressure means that a higher air pressure in the pneumatic cylinder applied on the tool. The air pressure in the pneumatic cylinder can be adjusted from 0 to 0.34 MPa (50 psi) by the regulator. Pellet weight was the weight of the wheat straw particles used to make a pellet.

Table 10.2 lists the values of the input variables. Ultrasonic power was from 20% to 60%. 20% ultrasonic power was the lowest value applicable for making good-quality pellets, and 60% was the highest value which would not generate charring of biomass (a phenomenon of biomass burning due to extremely high pelleting temperature, more information about charring of



biomass in UV-A pelleting can be found in [20]). Pelleting pressure was from 0.14 to 0.34 MPa (20 to 50 psi). Any pressure below 0.14 MPa was too low to make pellets and 0.34 MPa was the highest value available for the machine. Pellet weight was from 1.4 to 2.6 g. On the one hand, if pellet weight was less than 1.4 g, it was difficult to put three thermocouples in biomass. On the other hand, it was difficult to put more than 2.6 g of biomass in the mold. Table 10.3 lists the combination of these three input variables for each pelleting condition.

**Table 10.2 Input variables and their values**

Variable	Unit	Value
Pressure	MPa	0.14; 0.34
Ultrasonic Power	%	20; 60
Weight	g	1.5; 2.5

**Table 10.3 Values of input variables for each pelleting condition**

Condition #	Ultrasonic power (%)	Pelleting pressure (MPa)	Pellet weight (g)
1	20	0.14	1.4
2	60	0.14	1.4
3	20	0.34	1.4
4	60	0.34	1.4
5	20	0.14	2.6
6	60	0.14	2.6
7	20	0.34	2.6
8	60	0.34	2.6

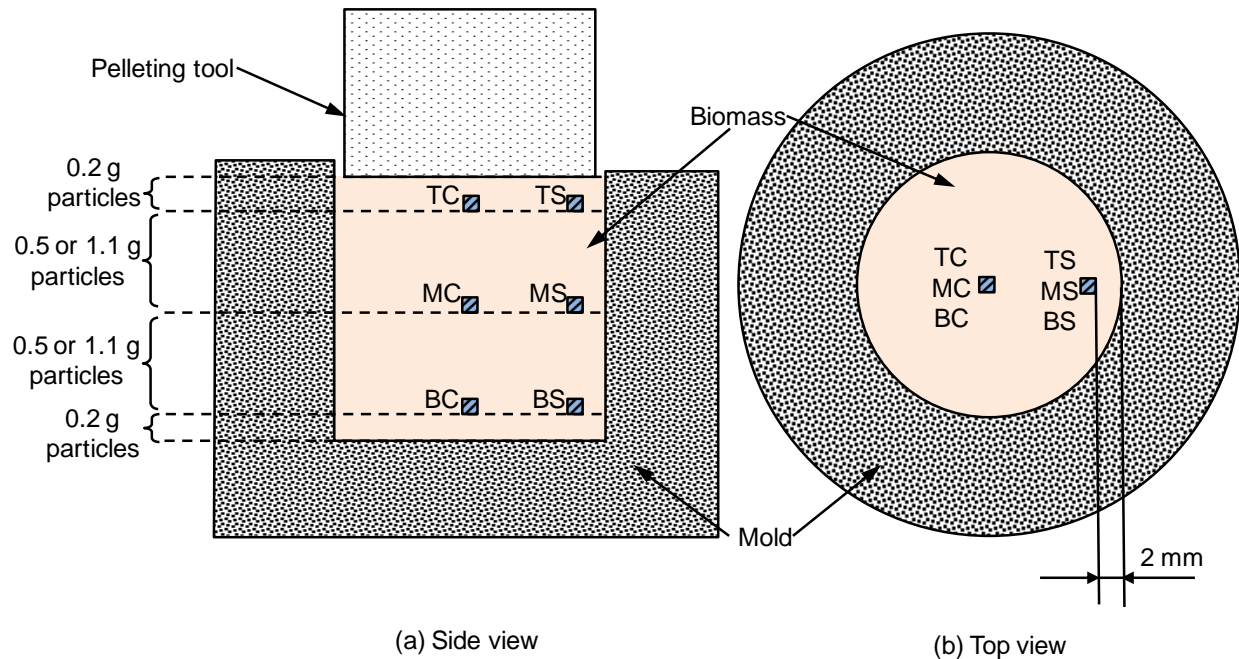
### ***10.2.5 Temperature measurement***

Temperature data were recorded when ultrasonic power was turned on, and the data recording lasted for 180 seconds (which was the longest pelleting duration used in one run of UV-A pelleting in previous studies). In each run, pelleting temperature was measured at six

locations in a pellet based on an assumption that temperature distribution is symmetric in a pellet. As shown in Figure 10.6, top center (TC), middle center (MC), and bottom center (BC) were located at the center axis of a pellet. Top side (TS), middle side (MS), and bottom side (BS) were at the side (2 mm away from the mold) of a pellet. It was ensured that thermocouples were not in contact with the mold or the pelleting tool because such contact could adversely affect the accuracy of measurements.

In each run, 0.2 gram of wheat straw particles was first loaded in the mold. Then, two thermocouples were placed at BC and BS. When pellet weight was 1.4 gram, 0.5 gram of wheat straw particles was put between MC (MS) and BC (BS), and another 0.5 gram between MC (MS) and TC (TS). When pellet weight was 2.6 gram, 1.1 gram of wheat straw particles was put between MC (MS) and BC (BS), and another 1.1 gram between MC (MS) and TC (TS). Finally, 0.2 gram of wheat straw particles was loaded above TC and TS.

**Figure 10.6 Illustration of the six locations in a pellet where temperatures were measured**



## 10.3 Results and discussion

### 10.3.1 Precision of the measurements

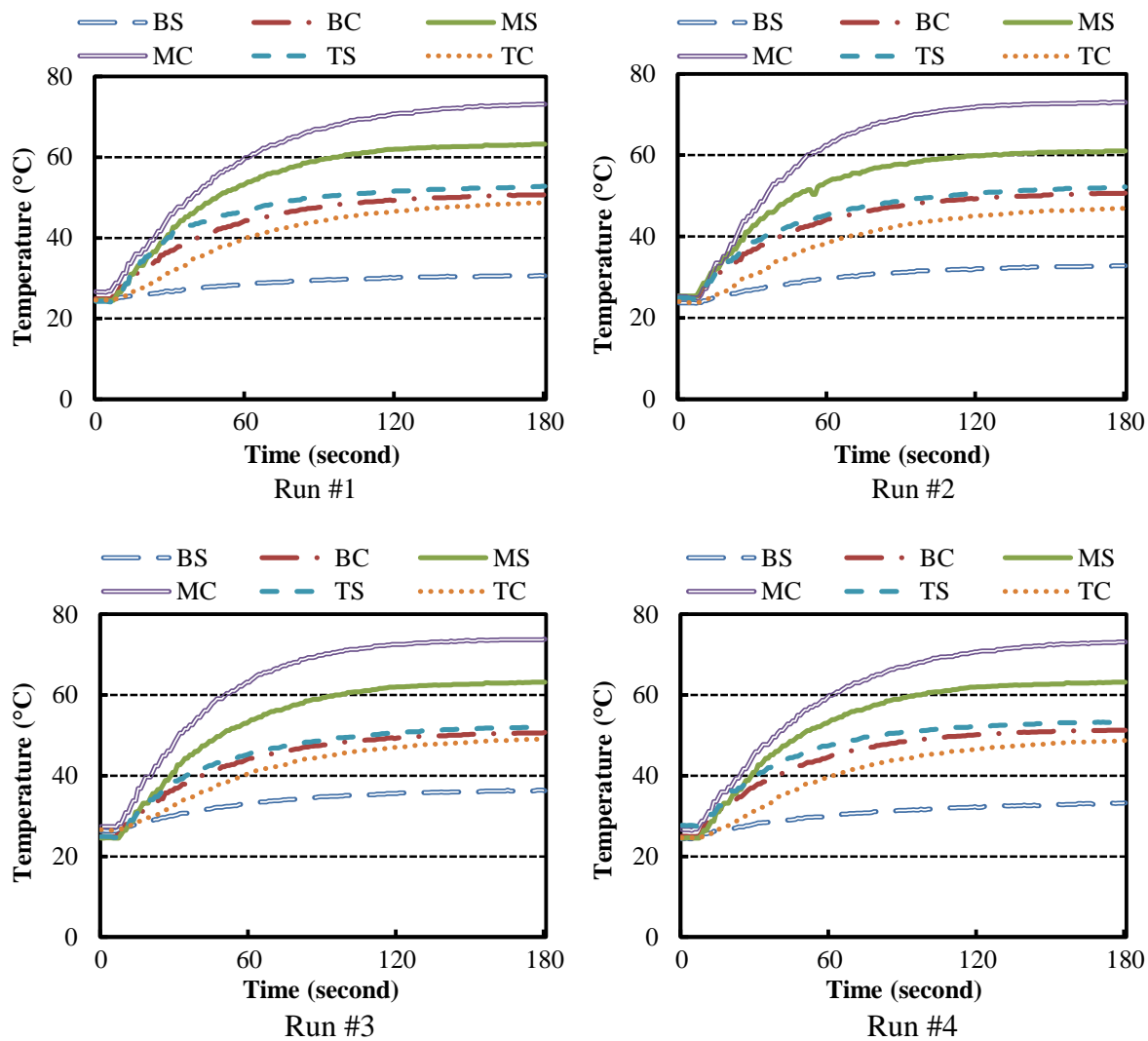
Under each pelleting condition, similar temperature-time curves were obtained for all runs. This indicates that high precision (or good repeatability) in temperature measurements was obtained. To illustrate the precision of the measurements, the temperature-time curves under two pelleting conditions are shown in Figures. 10.7 and 10.8. One condition (pressure = 0.14 MPa, ultrasonic power = 20%, and pellet weight = 1.4 g) had the best precision, and the other condition (pressure = 0.34 MPa, ultrasonic power = 60%, and pellet weight = 1.4 g) had the worst precision.

In Figure 10.7, the same shape of temperature-time curves at all locations was obtained for all four runs. It can be seen that there was a rapid increase in temperature at five locations ( $T_{mc}$ ,  $T_{ms}$ ,  $T_{ts}$ ,  $T_{bc}$ , and  $T_{tc}$ ) during the first 60 seconds and a gradual increase afterwards. In contrast, for all four runs,  $T_{bs}$  showed a consistently gradual increase over the entire pelleting duration. In addition, the overall rate of temperature increase at the six locations has the same order ( $T_{mc} > T_{ms} > T_{ts} > T_{bc} > T_{ts} > T_{bs}$ ). The average and standard deviation of the measurement data at the end of pelleting time are shown in Table 10.4. The standard deviations at all locations were smaller than 3°C.

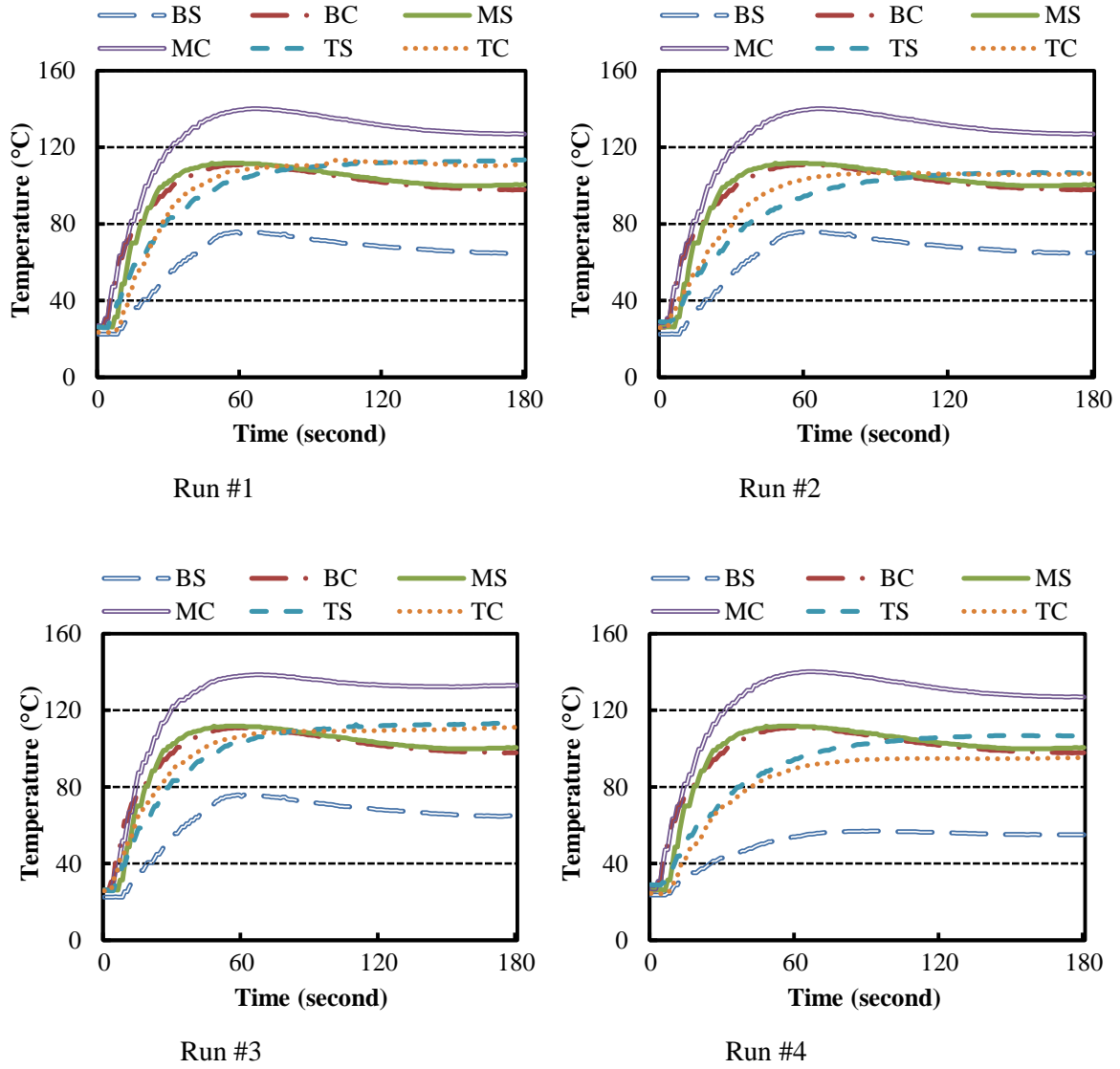
In Figure 10.8, the same shape of temperature-time curves at all locations was obtained for all four runs. The temperatures at four locations ( $T_{mc}$ ,  $T_{ms}$ ,  $T_{bc}$ , and  $T_{bs}$ ) underwent an initial increase followed by a decrease. In contrast, the temperatures at the other two locations ( $T_{tc}$  and  $T_{ts}$ ) underwent a consistent increase. However, these four run did not share the same order of the temperatures at different locations at the end of pelleting duration. The order in the fourth run ( $T_{mc} > T_{ts} > T_{ms} > T_{bc} > T_{tc} > T_{bs}$ ) was slightly different from those in the other three runs ( $T_{mc} > T_{ts} > T_{tc} > T_{ms} > T_{bc} > T_{bs}$ ). The average and standard deviation of the measurement data at the end of pelleting duration are shown in Table 10.5. The standard deviations at locations BS, TS,

and TC in Table 10.5 were 3.9, 3.2, and 5.1°C higher than those in Table 10.4, respectively. The standard deviations were larger than those in Table 10.4 (P-value < 0.05).

**Figure 10.7 Temperature-time curves under the best-precision condition**



**Figure 10.8 Temperature-time curves under the worst-precision condition**



**Table 10.4 Temperature measurements (°C) under the best-precision condition**

Run #	Bottom side	Bottom center	Middle side	Middle center	Top side	Top center
1	33.8	50.7	63.4	75.1	53.0	48.8
2	32.8	54.6	64.5	73.0	56.8	47.1
3	36.4	50.2	61.0	73.8	52.3	49.2
4	33.3	55.3	59.7	73.1	53.4	53.7
Mean	34.1	52.7	62.2	73.8	53.9	49.7
Standard deviation	1.6	2.6	2.2	1.0	2.0	2.8

**Table 10.5 Temperature measurements (°C) under the worst-precision condition**

Run #	Bottom side	Bottom center	Middle side	Middle center	Top side	Top center
1	65.9	87.8	101.8	127.5	113.5	113.4
2	68.0	91.8	103.0	126.9	106.2	106.7
3	66.3	86.3	106.5	133.4	116.9	111.4
4	55.8	88.2	104.6	128.0	107.0	95.3
Mean	64.0	88.5	104.0	129.0	110.9	106.7
Standard deviation	5.5	2.3	2.0	3.0	5.2	8.1

The precision of the temperature measurements for other conditions was between those for the two conditions described above. Overall, a good precision in temperature measurement was achieved using the experimental setup. Under each pelleting condition, the temperature-time curves obtained from different runs had the same shape. Therefore, one measurement from each pelleting condition was conducted and used in the following discussions.

### ***10.3.2 Relationships between pelleting temperature and pelleting time***

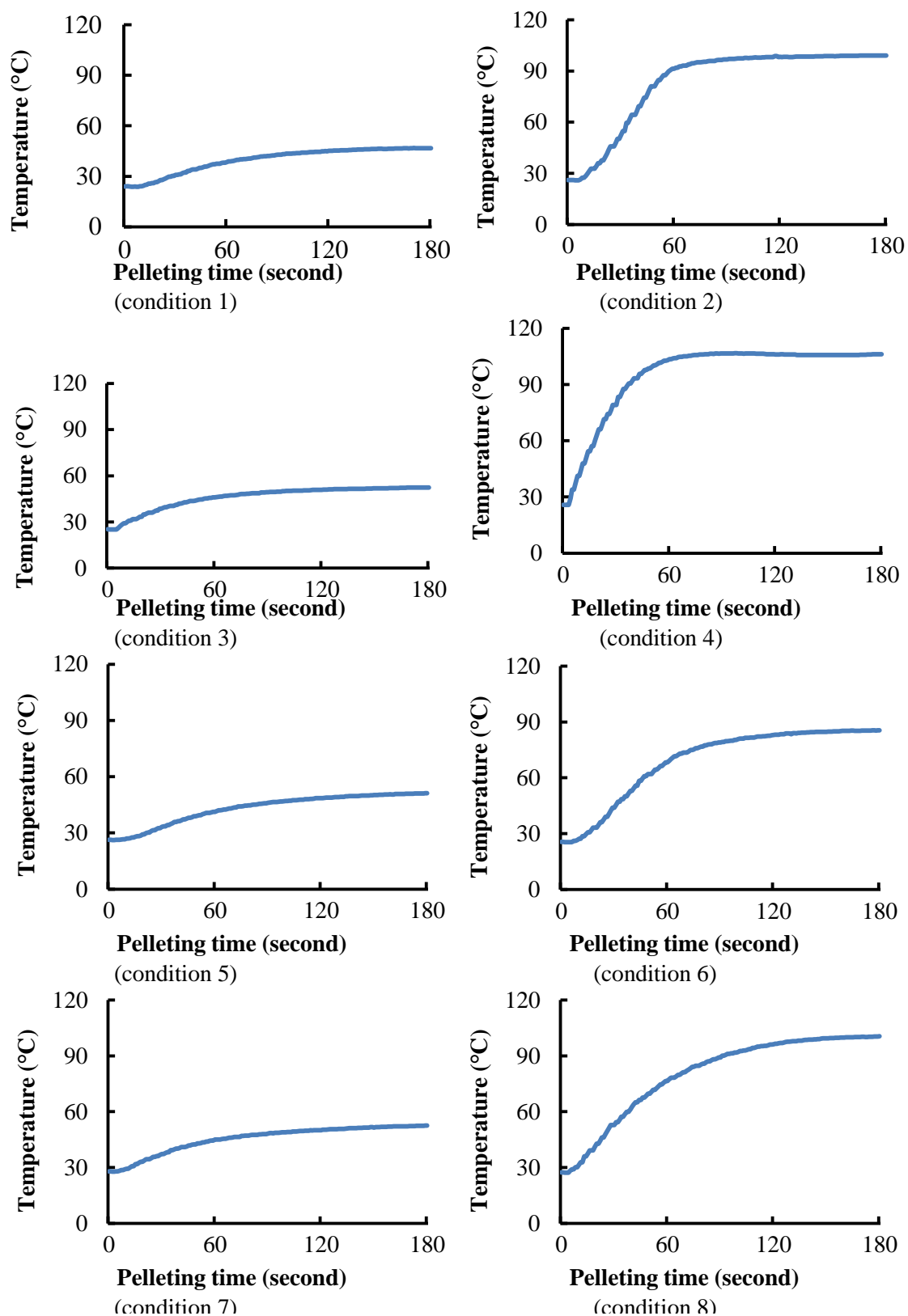
The relationships between pelleting temperature and pelleting time are presented as temperature-time curves shown in Figures 10.9–10.14. These curves can be classified into three basic groups based on two attributes of their shapes (increasing rate of temperature and existence of decline in pelleting temperature): (a) pelleting temperature increases rapidly during the first 60 seconds, then increases slowly, and becomes constant eventually; (b) pelleting temperature

increases consistently slowly for almost the entire duration, and becomes constant eventually; and (c) pelleting temperature increases rapidly during the first 60 seconds, then decreases slightly, and remain constant eventually.

The majority of the curves in Figures 10.9–10.14 belong to group (a), including conditions 1–8 in Figures 10.9 and 10.10, conditions 1-3, 6, and 8 in Figures 10.11 and 10.12; conditions 1, 3, and 8 in Figure 10.13; and conditions 1 and 3 in Figure 10.14. Group (b) includes conditions 5 and 7 in Figures 10.11–10.13, and conditions 5, 7, and 8 in Figure 10.14. Group (c) includes condition 4 in Figures 10.11 and 10.12, and conditions 2, 4, and 6 in Figures 10.13 and 10.14.

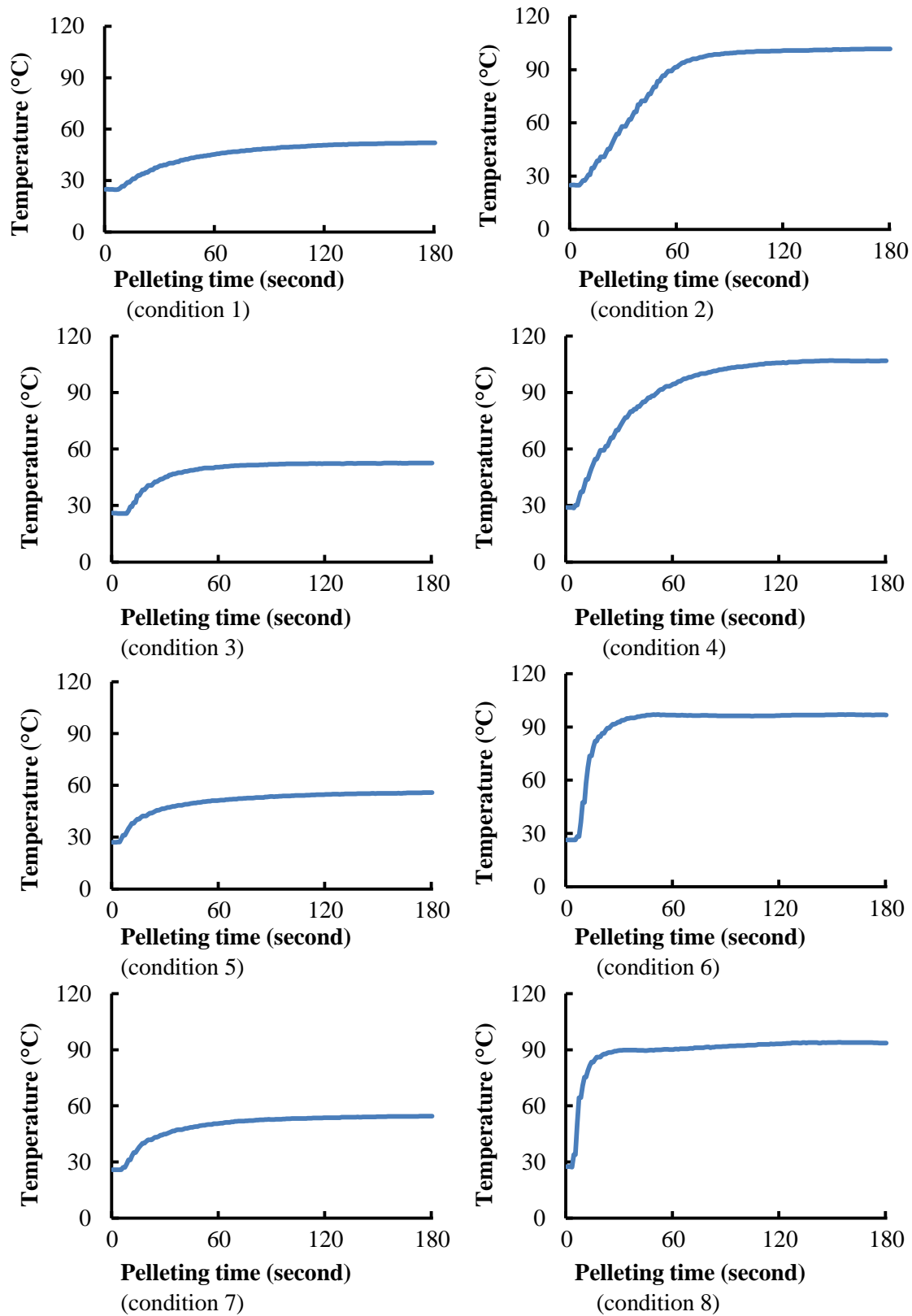
Tang et al. [35] reported the temperature-time curves for four positions (TC, MC, BC, and BS) under one pelleting condition (pelleting pressure = 0.28 MPa, ultrasonic power = 40%, and pellet weight = 2 g). The shape of three curves reported (MC, BC, and BS) was consistent with that obtained in this study. However, the shape of their TC curve (belonging to group (c)) was different from that of TC curve obtained in this study (belonging to group (a)). It is noted that the thermocouple at TC contacted the pelleting tool in their study but not (there was 0.2 g of biomass

**Figure 10.9 Temperature-time curves at top center**

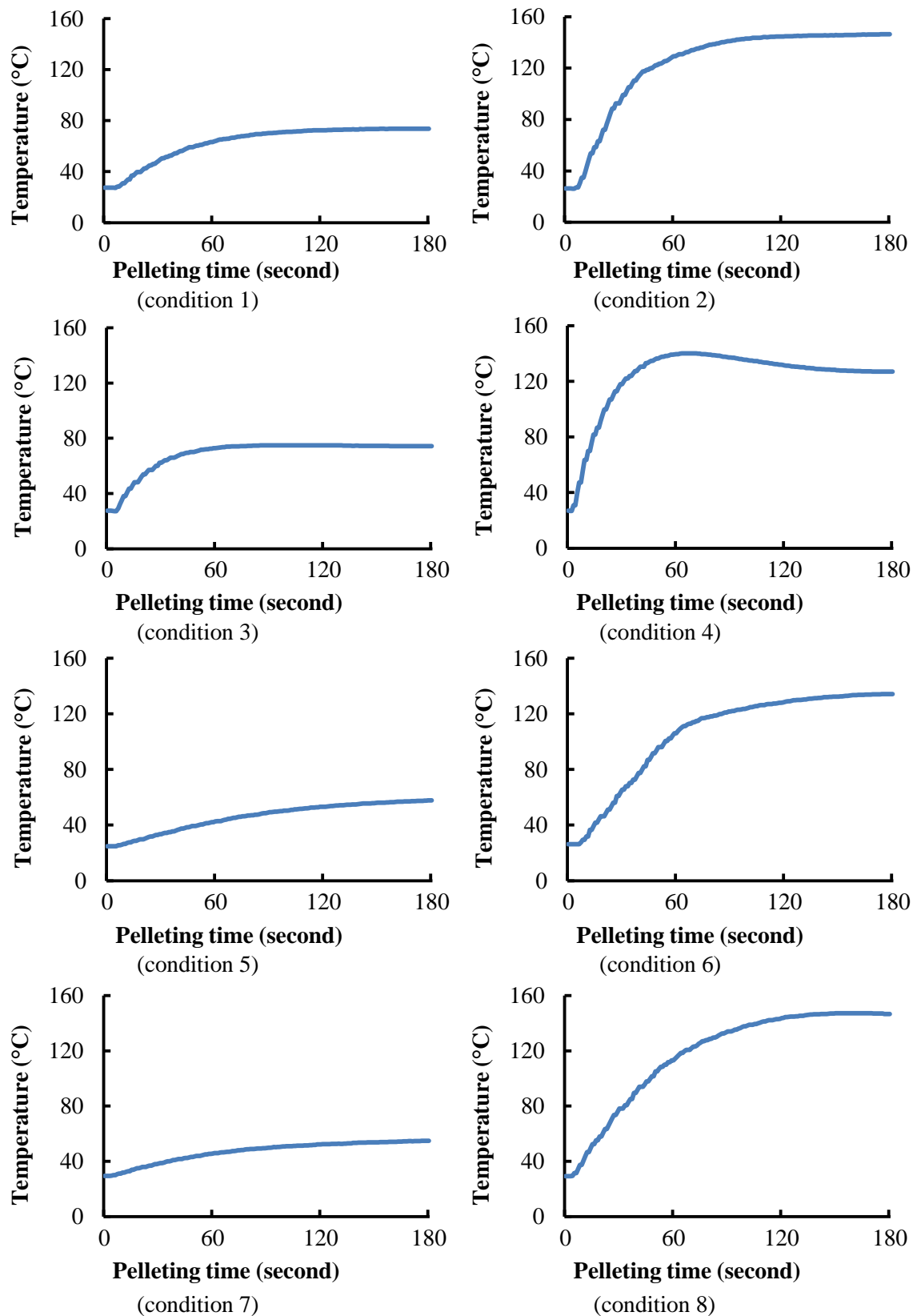




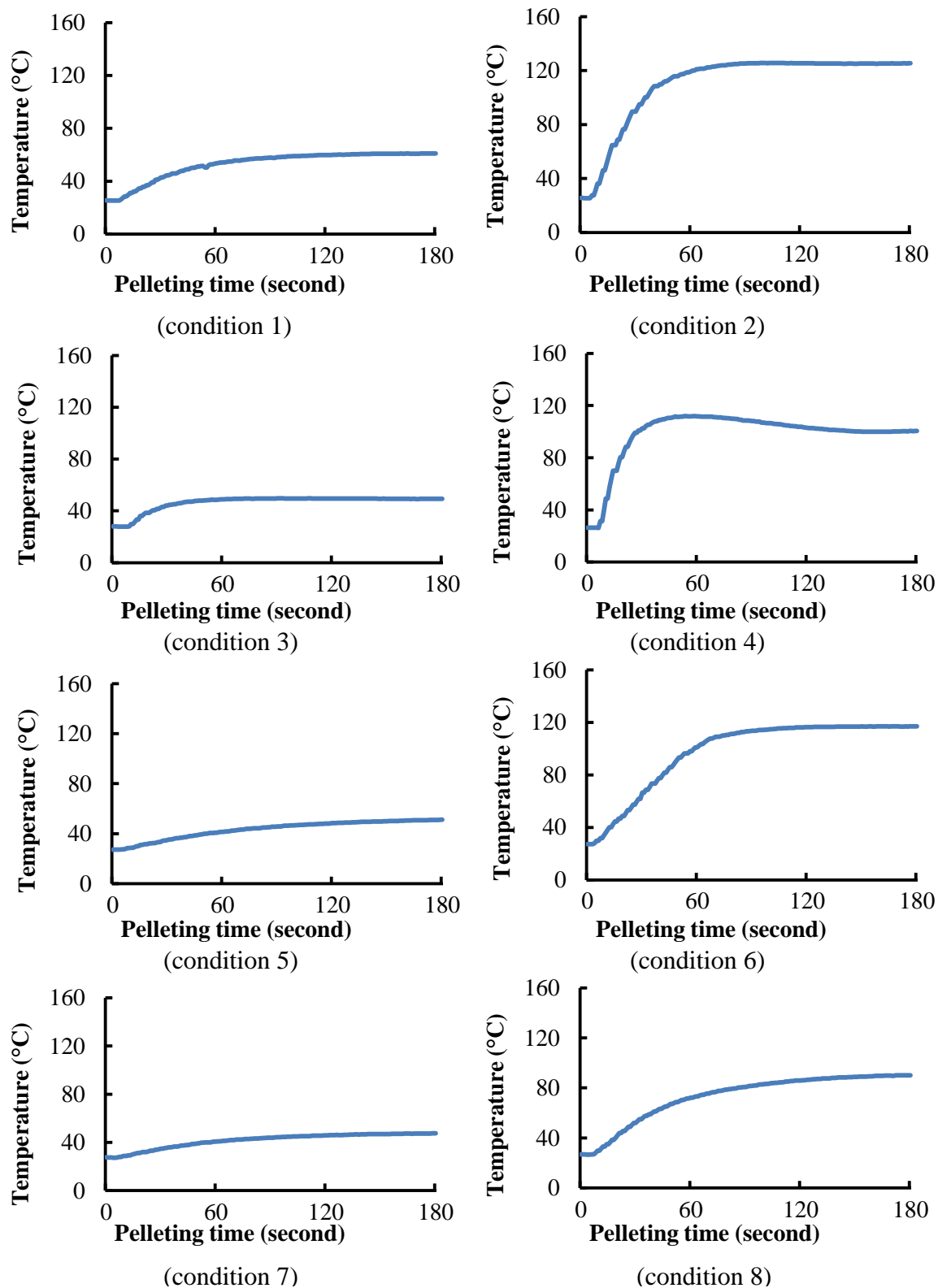
**Figure 10.10 Temperature-time curves at top side**



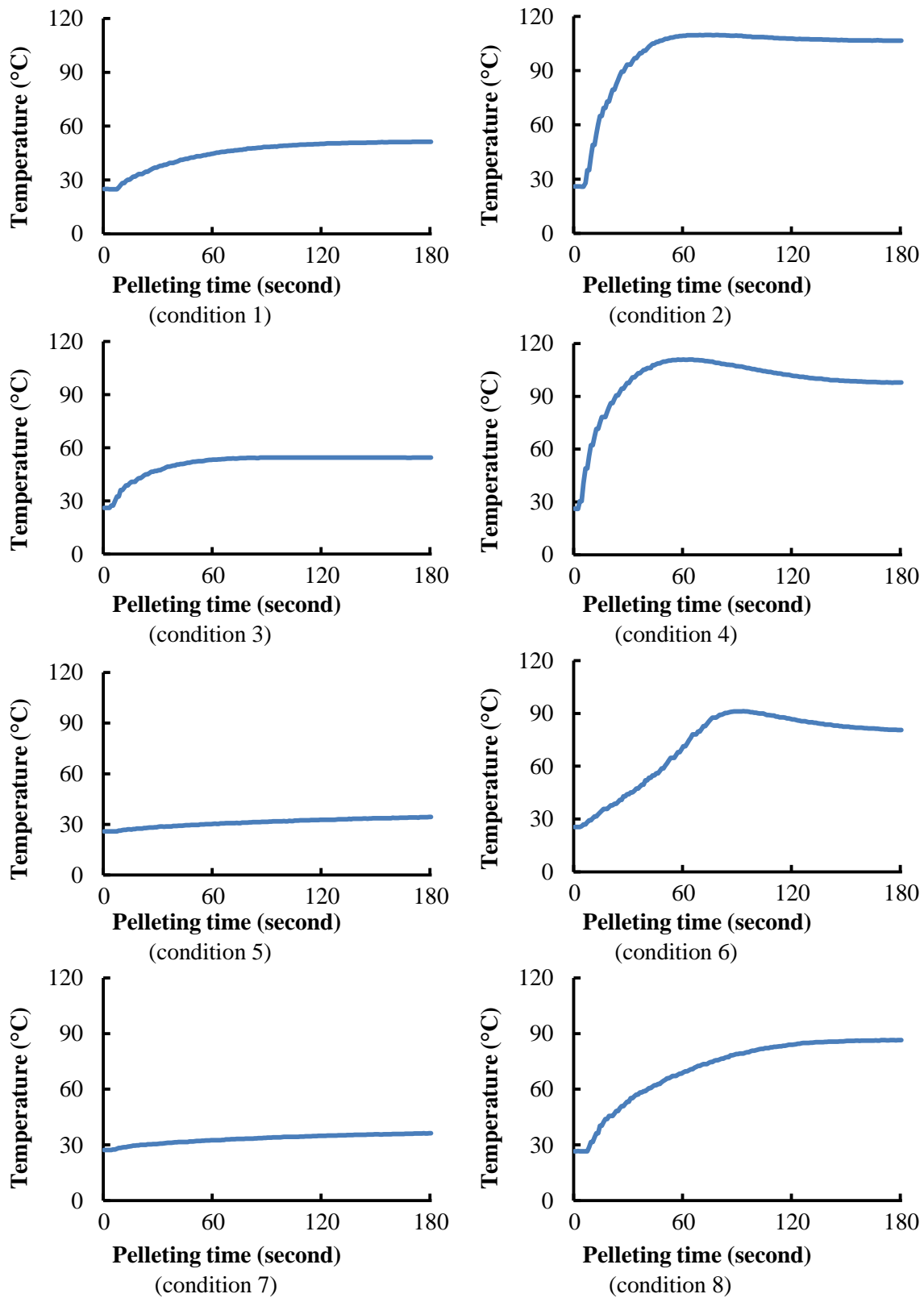
**Figure 10.11 Temperature-time curves at middle center**



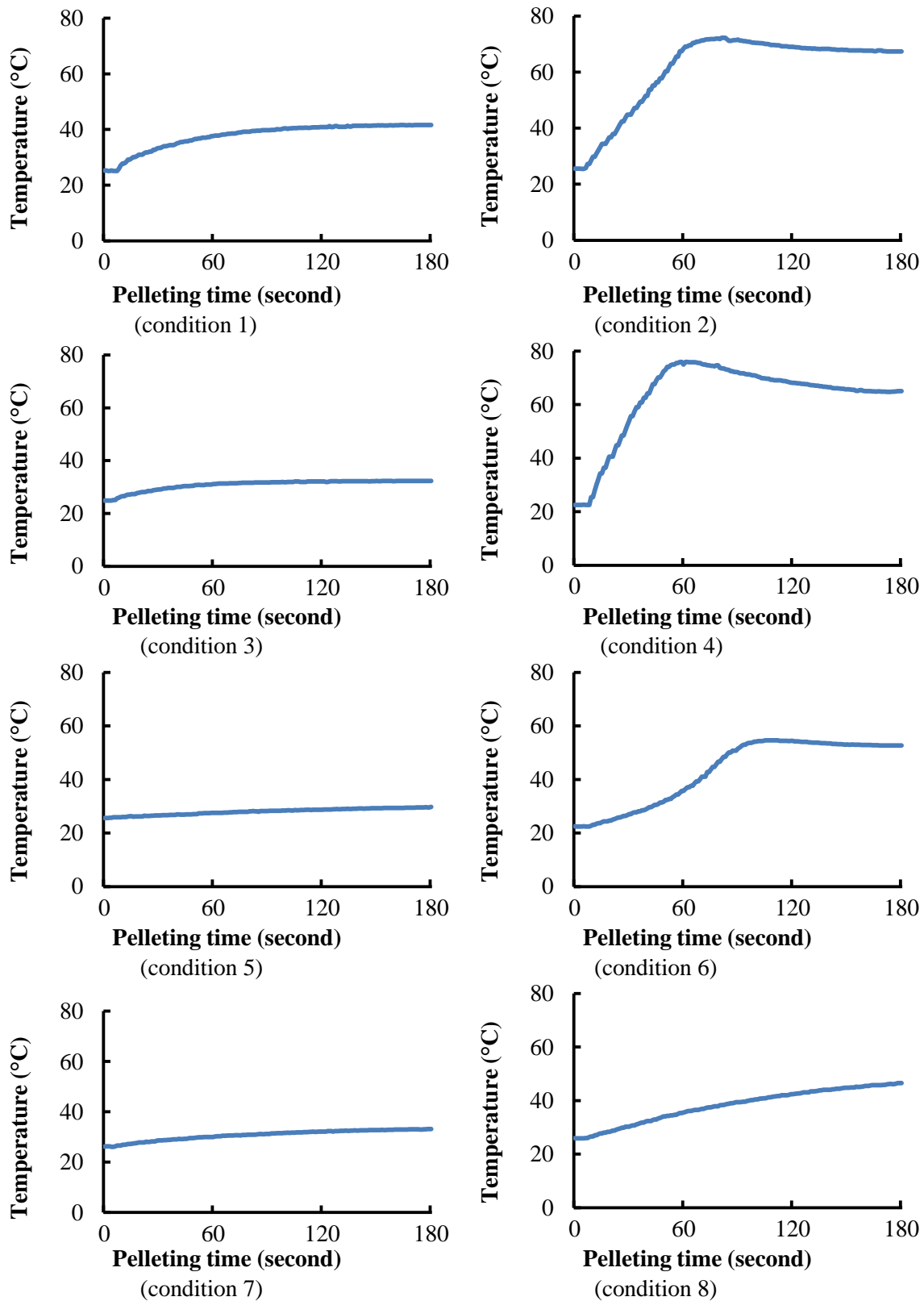
**Figure 10.12 Temperature-time curves at middle side**



**Figure 10.13 Temperature-time curves at bottom center**



**Figure 10.14 Temperature-time curves at bottom side**



between the thermocouple and the pelleting tool) in this study. This could lead to two possible explanations for the inconsistent curve shape at TC. First,  $T_{tc}$  in these two studies refers to the temperature at different locations—at very top of pellet in Tang et al.' study and at a certain distance (the height of 0.2-gram biomass) away from the very top of pellet. Second, the direct contact between the thermocouple and the metal pelleting tool could probably affect the temperature measurement, resulting in changed curve shape.

It can be seen, from Figures 10.9–10.14, that, for each location and each pelleting condition, the temperature at the end of pelleting duration was higher than the initial temperature, indicating that UV-A pelleting increased biomass temperature. Also, pelleting temperature became constant sooner or later regardless of the position and pelleting condition, indicating that a thermal balance (the heat generated equaled the heat dissipated) in a pellet was achieved during UV-A pelleting. Therefore, the highest pelleting temperature occurred during the first 180 seconds in UV-A pelleting. These findings are consistent with the results reported by Tang et al. [35].

Increasing rate of pelleting temperature during the first 60 seconds was greatly affected by ultrasonic power. For each location, the increasing rate when the higher ultrasonic power (60%) was used was much higher than that when the lower ultrasonic power (20%) was used. There was no obvious effect of pelleting pressure on the increasing rate. Effects of pellet weight on the increasing rate depended on the ultrasonic power used. When the lower ultrasonic power (20%) was used, there was no obvious effect of pellet weight on the increasing rate. When the higher ultrasonic power (60%) was used, the increasing rate for smaller pellet weight (1.4 g) was higher than that for larger pellet weight (2.6 g) for most locations (TC, MC, MS, BC, and BS). An opposite trend was obtained for TS. Under the same pelleting condition, the increasing rate was different at different locations. For example, under condition 4 (ultrasonic power = 60%,

pelleting pressure = 0.34 MPa, and pellet weight = 1.4 g), the increasing rate at MC was the highest, followed by MS, BC, TC, TS, and BS.

Under some pelleting conditions, a slight decline in pelleting temperature occurred in the middle stage of pelleting duration was observed at some locations. Such decline was not observed for any condition at TC and TS. It was observed under condition 4 at MC and MS, and under conditions 2, 4, and 6 at BC and BS. The decline in pelleting temperature occurred more frequently at the bottom of a pellet. This is possibly due to the fact that the heat dissipation at the bottom of a pellet was faster than that at the middle or top of a pellet (because the contact surface between the bottom of a pellet and the mold was much larger than that between the middle or top of a pellet and the mold). It was also observed that the decline in pelleting temperature occurred only when the higher ultrasonic power (60%) was used. Under condition 4, the decline in pelleting temperature was found at four locations (MC, MS, BC, and BS); under conditions 2 and 6, the decline in pelleting temperature was found only at BC and BS.

### ***10.3.3 Rank of pelleting temperature at different locations***

Pelleting temperatures at different locations were ranked to describe a basic pattern of biomass temperature distribution in UV-A pelleting. Given that the highest pelleting temperature generated was of high interest in UV-A pelleting (the highest pelleting temperature was closely related to pellet charring [20]), the temperatures at different locations were ranked based on the highest pelleting temperature and time at 180s obtained during the entire pelleting time.

Under each pelleting condition, ANOVA was performed to compare pelleting temperature at each location using Tukey's method at significance level of 0.05. The temperatures were then ranked based on the comparison results. Table 10.6 shows the ranked temperatures at different locations when value of temperature was based on the highest pelleting temperature point in pelleting process. Under each condition, MC was the highest or one of the highest among the six temperatures, and BS was the lowest or one of the lowest. This indicates that the highest temperature always occurred in the pellet's core, primarily because the

increasing rate of  $T_{mc}$  was much higher than those of temperatures at other locations. This result can explain the finding in our previous study [20] that charring in UV-A pelleting always started in a pellet's core. The ranking of temperatures at 180s was the same as that of the highest temperature.

**Table 10.6 Rank of temperatures at different locations**

Condition	Rank of temperature*
1	MC > MS > (BC,TC,TS) > BS
2	MC > MS > (BC,TC,TS) > BS
3	MC > (MS,BC,TC,TS) > BS
4	MC > (MS,BC,TC,TS) > BS
5	(TC,TS, MC, MS) > (BC,BS)
6	MC > MS > (BC,TC,TS) > BS
7	(TC,TS, MC, MS) > (BC,BS)
8	MC > (MS,BC,TC,TS) > BS

\* The symbol ">" means "significantly higher than (at the significance level of 0.05)", and the temperatures encapsulated in one pair of parentheses are not significantly different from each other.

In general, the temperatures at the other four locations ( $T_{tc}$ ,  $T_{ts}$ ,  $T_{ms}$ , and  $T_{bc}$ ) were between  $T_{mc}$  and  $T_{bs}$ . When the lower pelleting pressure (0.14 MPa) was used (under conditions 1, 2, and 6),  $T_{ms}$  was significantly higher than  $T_{bc}$ ,  $T_{tc}$ , and  $T_{ts}$ . When the higher pelleting pressure (0.34 MPa) was used (under conditions 3, 4, and 8), there was no significant difference between  $T_{ms}$ ,  $T_{bc}$ ,  $T_{tc}$ , and  $T_{ts}$ . This indicates that a higher pelleting pressure could make pelleting temperature more evenly distributed in a pellet. When the lower ultrasonic power (20%) and larger pellet weight (2.6 g) was used (under conditions 5 and 7), there was no significant difference between  $T_{tc}$ ,  $T_{ts}$ ,  $T_{mc}$ , and  $T_{ms}$ ; but they were significantly higher than  $T_{bc}$  and  $T_{bs}$ .

#### ***10.3.4 Effects of input variables on pelleting temperature***

Pelleting temperature plays a major role in pellet quality [4]. In UV-A pelleting, pellet quality is related to two pelleting temperature values: the highest and the lowest pelleting



temperatures. The highest pelleting temperature (HPT) is related to charring of biomass in UV-A pelleting. Biomass was charred when the pelleting temperature was too high [20,35]. The lowest pelleting temperature (LPT) is related to pellet density and durability. There would be remarkable deterioration in pellet density and durability if pelleting temperature is not sufficiently high [4,13,38]. Therefore, effects of the three input variables (ultrasonic power, pelleting temperature, and pellet weight) on the highest and lowest pelleting temperature were investigated using a  $2^3$  (three variables, two levels) full factorial design with one center point with four replications. Levels of the three input variables are: 60% (+) and 20% (-) for ultrasonic power, 0.34 MPa (+) and 0.14 MPa (-) for pelleting pressure, and 2.6 g (+) and 1.4 g (-) for pellet weight. Four temperature measurements were obtained for each pelleting condition. A total of 36 measurements were obtained for HPT and LPT respectively, and they were obtained in a random order. The experimental matrix and temperature measurements are shown in Table 10.7.

The temperature data were analyzed with Design Expert (Stat-Ease, Inc., Minneapolis, MN, USA). ANOVA was conducted to determine the significance levels of main and interaction effects of input variables on pelleting temperature. ANOVA results on HPT are shown in Table 10.8. The P-value of curvature is 0.23, indicating that curvature is not significant at the significance level of  $\alpha = 0.05$  and HPT can be predicted based on the temperature data at the factorial points. At the significance level of  $\alpha = 0.05$ , two of the three main effects (ultrasonic power and pellet weight), one of the three two-factor interaction effects (interaction effect between ultrasonic power and pellet weight), and the three-factor interaction effect on HPT were

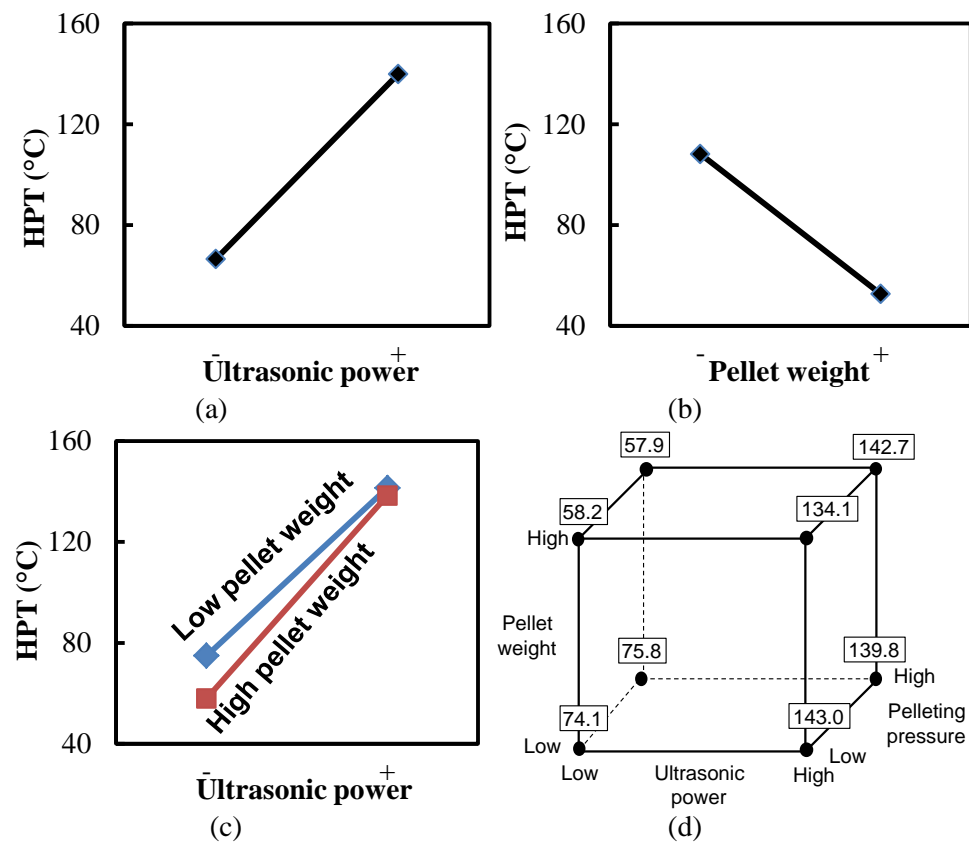
**Table 10.7 Experimental matrix and temperature measurements**

Run	Ultrasonic power	Pelleting pressure	Pellet weight	HPT	LPT
1	+	+	+	138.0	43.9
2	0	0	0	99.2	50.0
3	-	-	+	56.0	30.6
4	+	-	-	139.7	67.8
5	+	-	+	136.1	49.3
6	-	-	-	75.5	33.9
7	0	0	0	99.0	51.1
8	-	+	-	77.8	29.4
9	+	-	-	142.2	76.1
10	+	+	-	141.7	73.8
11	-	+	-	78.5	36.7
12	-	+	+	55.9	36.0
13	-	+	+	58.7	33.9
14	-	+	+	57.7	33.7
15	+	-	-	145.0	67.3
16	+	-	+	141.3	52.2
17	-	-	-	73.1	33.5
18	0	0	0	104.1	44.1
19	+	-	+	133.0	54.8
20	+	+	+	150.2	42.5
21	+	+	+	139.3	45.8
22	-	-	+	57.6	30.0
23	+	-	-	145.2	66.3
24	+	-	+	125.9	58.4
25	+	+	+	143.4	49.1
26	-	-	+	60.9	29.3
27	+	+	-	130.2	79.3
28	-	-	-	73.4	32.1
29	-	+	-	75.3	32.0
30	-	-	+	58.3	30.5
31	0	0	0	100.1	49.7
32	-	+	-	71.5	30.8
33	+	+	-	144.5	81.3
34	-	+	+	59.2	33.6
35	+	+	-	142.7	70.8
36	-	-	-	74.3	32.1

**Table 10.8 ANOVA results on HPT**

Source	Sum of squares	DF	Mean square	F-value	P-value
A-Ultrasonic power	43122.5	1	43122.5	2656.2	< 0.0001
B-Pelleting pressure	23.0	1	23.0	1.4	0.2444
C-Pellet weight	791.0	1	791.0	48.7	< 0.0001
AB	8.1	1	8.1	0.5	0.4858
AC	385.7	1	385.7	23.8	< 0.0001
BC	48.8	1	48.8	3.0	0.0941
ABC	97.0	1	97.0	6.0	0.0211
Lack of Fit	23.9	1	23.9	1.5	0.2319
Error	430.7	27	16.0		
Total	44930.6	35			

**Figure 10.15 Significant main and interaction effects on HPT**



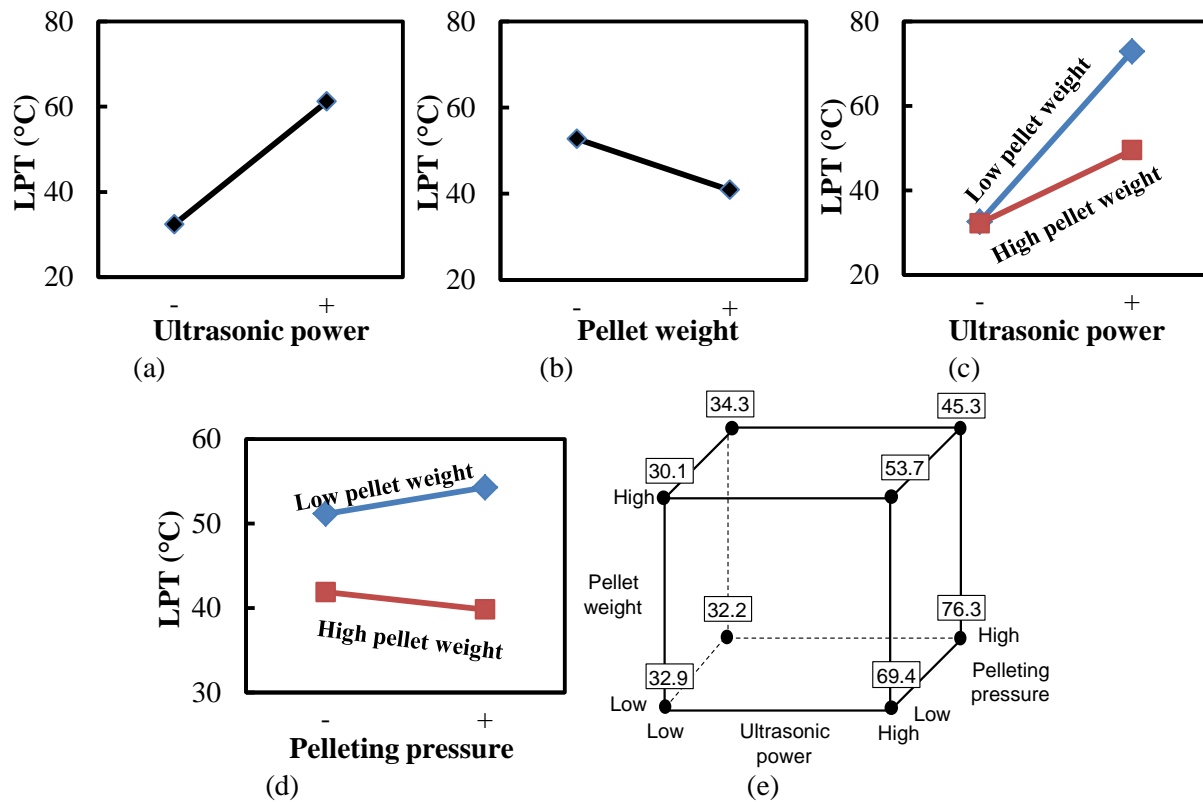
significant. The significant main and interaction effects of input variables on HPT are shown in Figure 10.15. HPT increased as ultrasonic power increased or pellet weight decreased. The interaction effects between ultrasonic power and pellet weight show that effects of ultrasonic power on HPT were stronger at the high level of pellet weight. From Figure 10.15(d), it can be seen that the highest HPT was obtained at the combination of high level of ultrasonic power and low level of pelleting pressure and pellet weight.

ANOVA results on LPT are shown in Table 10.9. The P-value of curvature is 0.25, indicating that curvature is not significant at the significance level of  $\alpha = 0.05$  and LPT can be predicted based on the temperature data at the factorial points. At the significance level of  $\alpha = 0.05$ , two of the three main effects (ultrasonic power and pellet weight), two of the three two-factor interaction effects (interaction effects between ultrasonic power and pellet weight and between pelleting pressure and pellet weight), and the three-factor interaction effect on LPT were significant. The significant main and interaction effects of input variables on LPT are shown in Figure 10.16. LPT increased as ultrasonic power increased or pellet weight decreased. From the interaction effects between ultrasonic power and pelleting weight, it can be seen that effects of ultrasonic power on LPT were stronger at the low level of pellet weight. The interaction effects between pelleting pressure and pellet weight show that, with the increase in pelleting pressure, LPT would increase at the low level of pellet weight while decrease at the high level of pellet weight. From Figure 10.16(e), it can be seen that the lowest LPT was obtained at the combination of low level of ultrasonic power and pressure and high level of pellet weight.

**Table 10.9 ANOVA results on LPT**

Source	Sum of squares	DF	Mean square	F-value	P-value
A-Ultrasonic power	6629.8	1	6629.8	657.9	< 0.0001
B-Pelleting pressure	2.2	1	2.2	0.2	0.6436
C-Pellet weight	1123.4	1	1123.4	111.5	< 0.0001
AB	12.3	1	12.3	1.2	0.2796
AC	1055.7	1	1055.7	104.8	< 0.0001
BC	54.1	1	54.1	5.4	0.0281
ABC	203.0	1	203.0	20.1	0.0001
Lack of Fit	13.5	1	13.5	1.4	0.2539
Error	268.7	27	10.0		
Total	9362.6	35			

**Figure 10.16 Significant main and interaction effects on LPT**



## 10.4 Conclusions

This paper reports an investigation on biomass temperature in ultrasonic vibration-assisted (UV-A) pelleting of cellulosic biomass. The following conclusions can be drawn:

- (1) A good precision in temperature measurement could be achieved using the experimental setup adopted in this study.
- (2) The obtained temperature-time curves could be classified into three groups based on their shapes. The curve shape depended on both measurement location and pelleting condition. For each combination of measurement location and pelleting condition, the speed of temperature increase and highest temperature point in each locations during the pelleting time (180 seconds) are different in this study.
- (3) In UV-A pelleting, the highest pelleting temperature always occurred at the middle center (the core) of a pellet. The lowest pelleting temperature always occurred at the bottom side of a pellet. The distribution of pelleting temperature (presented by the rank of pelleting temperatures at the six locations) was affected by pelleting condition. Pelleting temperature was more evenly distributed in a pellet (there was no significant difference in pelleting temperature between the four locations except middle center and bottom side) when a high level of pelleting pressure was used.
- (4) The highest and lowest pelleting temperatures in UV-A pelleting were significantly affected by two main effects (ultrasonic power and pellet weight) and the three-factor interaction effect. One two-factor interaction effect (between ultrasonic power and pellet weight) was significant on the highest pelleting temperature while two two-factor interaction effects (between ultrasonic power and pellet weight and between pelleting pressure and pellet weight) were significant on the lowest pelleting temperature.

## Acknowledgements

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## Chapter 11 - **Investigation on composition of pelleted biomass**

### Paper Title:

Effects of ultrasonic vibration-assisted pelleting on chemical composition and sugar yield  
of corn stover and sorghum stalk

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## **Abstract**

Ultrasonic vibration-assisted (UV-A) pelleting of cellulosic biomass can increase its density and reduce the costs of biomass transportation and handling. The objective of this paper is to investigate effects of UV-A pelleting on chemical composition and sugar yield of cellulosic biomass. The effects were investigated with and without dilute acid pretreatment using corn stover and sorghum stalk. It was found that there was no significant difference in chemical composition between pelleted and unpelleted biomass whether they went through dilute acid pretreatment or not. After dilute acid pretreatment, cellulose recovery of pelleted biomass was significantly higher than that of unpelleted biomass. UV-A pelleting could significantly increase the sugar yield in enzymatic hydrolysis for both corn stover and sorghum stalk.

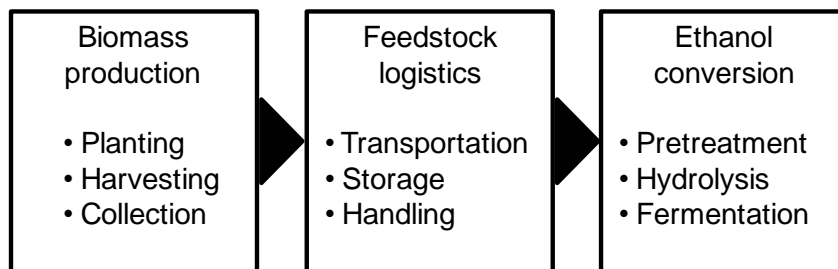
## **11.1 Introduction**

In 2011, about 140 billion gallons of liquid transportation fuels were consumed in the United States and more than half of these fuels were derived from foreign petroleum [1]. Increasing demands and concerns for reliable supply of liquid transportation fuels make it important to develop domestic sustainable alternatives to petroleum-based liquid transportation fuels. Cellulosic ethanol is one such alternative. Land resources in the United States are sufficient to sustain production of enough cellulosic biomass annually to displace 30% or more of the nation's current petroleum consumption [2]. In addition, using cellulosic biomass as feedstocks for ethanol production can mitigate the accumulation of greenhouse gas in the atmosphere and promote economic growth in rural areas [3].

Major steps in cellulosic ethanol production are shown in Figure 11.1. The cost-effectiveness of cellulosic ethanol manufacturing has been challenged by several technical barriers. One such barrier is related to the low density of raw cellulosic biomass (ranging from 24 to 250 kg/m<sup>3</sup> [5]), causing high costs of biomass transportation, handling, and storage. Another barrier is the difficulty in converting cellulose (a major sugar source in cellulosic

biomass) into fermentable sugars, leading to low sugar yield in enzymatic hydrolysis and making enzymatic hydrolysis an expensive and slow step.

**Figure 11.1 Major steps in cellulosic ethanol manufacturing (after [4])**



Pelleting of cellulosic biomass can significantly increase its density to higher than 600 kg/m<sup>3</sup> [6-8]. In turn, the costs of transporting and storing of pelleted cellulosic biomass could be reduced to less than 50% and 10% of those of raw cellulosic biomass, respectively [9,10]. Furthermore, pellets can be handled with existing grain-handling equipment at biorefinery plants. Unlike traditional pelleting methods (e.g., screw extrusion, press briquetting, or ring-die pelleting), UV-A pelleting does not use high-temperature steam, high pressure, and binder material. Pellet density and durability produced by UV-A pelleting are comparative to those produced by traditional pelleting methods [11].

It has been reported that traditional pelleting methods have positive effects on the sugar yield of cellulosic biomass in enzymatic hydrolysis. Theerarattananoon et al. [12] compared the chemical composition and sugar yield of pelleted (by ring-die pelleting) biomass with those of unpelleted biomass (wheat straw, corn stover, big bluestem, and sorghum stalk). They found that ring-die pelleting changed the chemical composition of tested biomass. In addition, sugar yields of biomass processed by ring-die pelleting were significantly higher than those of biomass not processed by ring-die pelleting. Lamsal et al. [13] compared the sugar yield of wheat bran processed by screw extrusion with that of unprocessed wheat bran. They reported that sugar yield of processed wheat bran was about 30% higher than that of unprocessed wheat bran. The same trend was reported by Yoo et al. [14,15] with soybean hulls. They reported that screw

extrusion could increase the sugar yield to 94% for soybean hulls. The sugar yield of soybean hulls not processed by screw extrusion was only 41%.

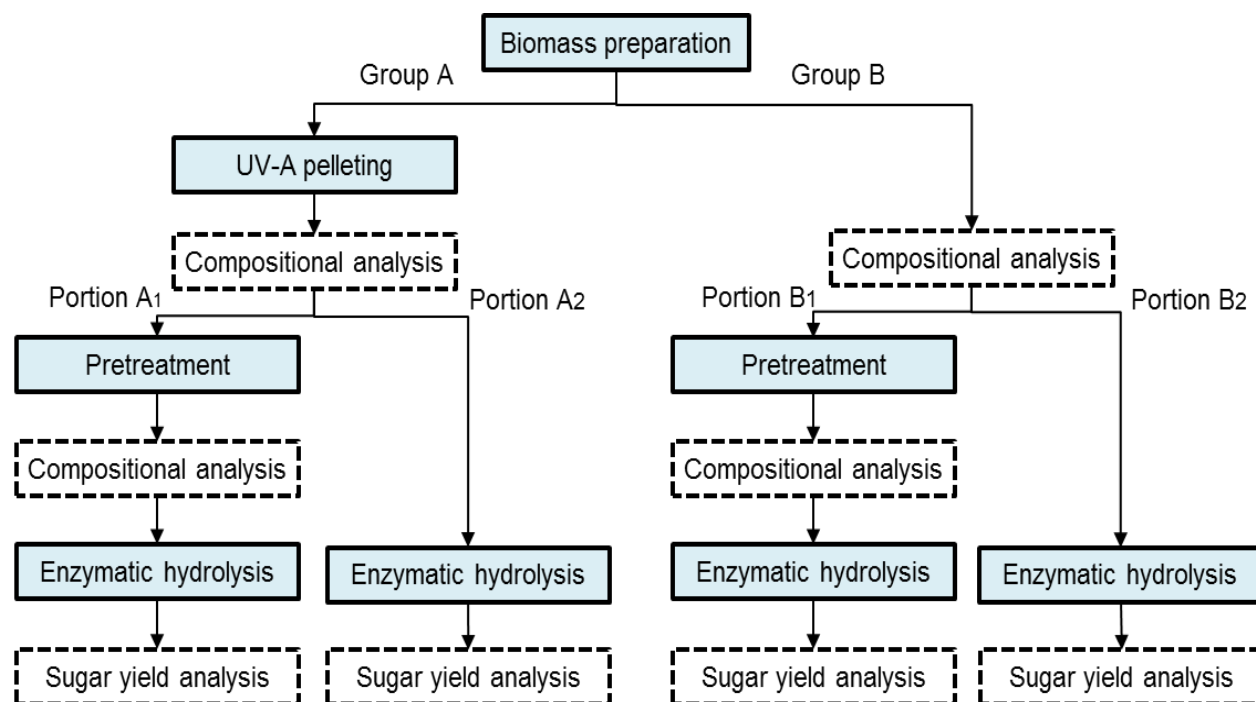
The abovementioned results cannot be directly applied to evaluate effects of UV-A pelleting on sugar yield of cellulosic biomass. The pelleting mechanism of UV-A pelleting is different from those of traditional pelleting methods. By now, effects of UV-A pelleting on chemical composition and sugar yield of cellulosic biomass have not been fully investigated. In this paper, chemical composition and sugar yield of biomass processed by UV-A pelleting were compared with those not processed UV-A pelleting. The comparisons were made with and without dilute acid pretreatment based on two types of cellulosic biomass (corn stover and sorghum stalk).

## **11.2 Materials and methods**

### ***11.2.1 Experimental procedure***

The experimental procedure is shown in Figure 11.2. Biomass preparation included steps of converting raw biomass into biomass particles suitable for pelleting. The prepared biomass particles were separated into two groups. Group A was processed by UV-A pelleting and group B was not. Each group of biomass was further separated into two portions: portions A<sub>1</sub> and B<sub>1</sub> went through dilute acid pretreatment and enzymatic hydrolysis, and portions A<sub>2</sub> and B<sub>2</sub> went through enzymatic hydrolysis without dilute acid pretreatment. Compositional analyses and sugar yield analyses were conducted to determine the chemical composition and sugar yield of different biomass samples.

**Figure 11.2 Experimental procedure of this study**



### ***11.2.2 Biomass preparation***

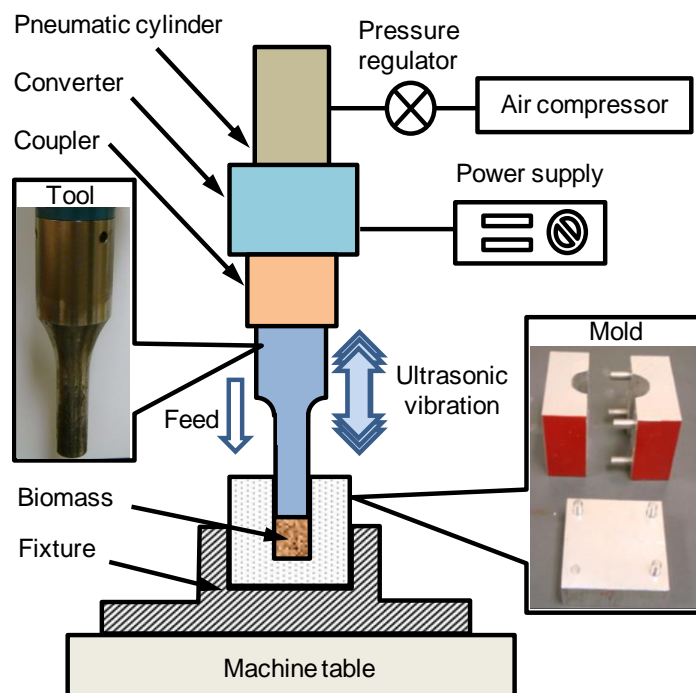
The corn stover and sorghum stalk used in this study were harvested on the Kansas State University Agronomy Farm in November, 2008. After harvesting, corn stover and sorghum stalk were chopped to the size of 18 – 23 cm using a tub grinder (Haybuster H-1150 series, DuraTech Industries International Inc., Jamestown, ND, USA). The chopped biomass (corn stover and sorghum stalk) was milled into small particles using a cutting mill (SM 2000, Retsch Inc., Newtown, PA, USA). Particle size was controlled by using a sieve with 1-mm screen size. After milling, the moisture content of biomass particles was measured and adjusted to 10% by following a NREL laboratory analytical procedure [16]. Previous studies showed that biomass particles with about 10% moisture content would produce pellets with high density and durability [17,18].



### 11.2.3 Experimental setup for UV-A pelleting

UV-A pelleting was conducted on a modified ultrasonic machine (model AP-1000, Sonic-Mill, Albuquerque, NM, USA). Figure 11.3 is a schematic illustration of the experimental setup for UV-A pelleting. The power supply converted 60-Hz electrical supply into 20-kHz electrical power. The high-frequency electrical power was applied to the piezoelectric converter and converted into high-frequency mechanical motion. The motion was amplified by the coupler and transmitted to the titanium pelleting tool. In this study, the pelleting tool vibrated at the frequency of 20 kHz.

**Figure 11.3 Illustration of experimental setup for UV-A pelleting**



The air compressor (1.2 kw, 125 liter, Sears, Roebuck and Co., Hoffman Estates, IL, USA) produced compressed air which was fed into the pneumatic cylinder (ARO Equipment Corporation, Bryan, OH, USA). The air pressure in the pneumatic cylinder was controlled by the pressure regulator and was 0.34 MPa in this study.

A three-piece aluminum mold was used to hold biomass particles in UV-A pelleting. The top two parts of the mold formed a central cylindrical cavity and the bottom part served as a base. The diameter of the mold cavity (18.6 mm) was slightly larger than that of the tip of the pelleting tool (17.4 mm). In this study, one gram of biomass was put in the mold and compressed for 3 minutes to make a pellet.

#### ***11.2.4 Pretreatment***

Two different pretreatment conditions were adopted in this study. Portions A<sub>1</sub> and B<sub>1</sub> (Figure 11.2) went through dilute acid pretreatment before enzymatic hydrolysis. Portions A<sub>2</sub> and B<sub>2</sub> were directly processed by enzymatic hydrolysis without pretreatment.

Dilute sulfuric acid pretreatment was carried out in a 600-mL reaction vessel of a pressure reactor (Parr Instrument Company, Moline, IL). A slurry of 20 grams of biomass (milled particles or pellets) and 200-mL diluted sulfuric acid solution (2% w/v) was loaded in the reaction vessel. The slurry was heated by a heater and temperature of the heater was maintained at 140°C for 30 minutes. Two four-blade impellers were used to stir the slurry. For pretreatment of biomass pellets, the pellets were soaked in the diluted sulfuric acid solution at room temperature until they were dissolved before they were heated. After pretreatment, the slurry was centrifuged and separated into two fractions: a liquid fraction (an acid solution) and a solid one. The solid fraction was then washed with 100°C distilled water three times to remove sulfuric acid. After each time of washing, the water was separated from biomass and added to the acid solution. The final solution (which consisted of the acid solution and all of the water added into it) was referred to as filtrate of biomass. A part of washed biomass was used for moisture content and chemical composition analyses. The other part was used for subsequent enzymatic hydrolysis.

### ***11.2.5 Enzymatic hydrolysis***

Enzymatic hydrolysis was conducted in a 100-mM sodium acetate buffer solution (pH 4.8) with the addition of 0.02% (w/v) sodium azide to prevent the microbial growth during hydrolysis. Biomass (1 gram dry weight) was mixed with 50-mL buffer solution in 125-mL flasks in a 50°C water bath shaker (Model C76, New Brunswick Scientific, Edison, NJ, USA) agitating at 180 rpm for 72 hours. The enzyme Accellerase 1500™ (Danisco, Inc., Genencor Division, Rochester, NY) was used. This enzyme complex contained multiple enzyme activities, including exoglucanase, endoglucanase (2200-2800 CMC U/g), hemicellulose, and  $\beta$ -glucosidase (525-778 pNPG U/g). The enzyme loading was 1mL/g of loaded cellulose. When hydrolyzing unpretreated pellets, the pellets were soaked in the buffer solution at room temperature. After they were dissolved, enzyme was loaded.

During enzymatic hydrolysis, the hydrolysis slurries were sampled every 24 hours by withdrawing 1 mL of slurry from each flask. Sample slurries (in 1.5-mL vials) were placed in boiling water for 15 minutes to deactivate the enzyme. Then sample slurries were centrifuged at 13,500 rpm for 15 minutes. 0.1 mL of supernatant was mixed with 0.9 mL double-distilled water and filtered into 1.5-mL autosampler vials through 0.2  $\mu$ m hydrophilic PTFE syringe filters (Millipore, Billerica, MA). Filtered samples were kept at 4°C before HPLC analysis.

The sugar yield was expressed as the percentage of cellulose enzymatically converted to glucose in hydrolysis. It was calculated by Equation (1)

$$\text{Sugar yield} = \frac{C \times V \times 10}{1.11 \times M} \times 100\% \quad (1)$$

where C is the concentration (g/L) of glucose in the diluted samples in the autosampler vials and determined by HPLC analysis, V is the total volume (L) of solutions in the flasks for hydrolysis, M is the weight (g) of cellulose in the biomass loaded for hydrolysis, and the number 1.11 is the cellulose-to-glucose conversion factor.

### ***11.2.6 Analytical methods***

Extractives in untreated biomass and chemical composition of different biomass samples were determined by following NREL laboratory analytical procedures [19,20]. Structural carbohydrates in biomass were reported as the percentage of glucan and xylan. Lignin, the major noncarbohydrate component, was reported as acid-soluble lignin (ASL), acid-insoluble lignin (AIL), and the sum of them (ASL + AIL). All chemical composition analyses and sugar yield analyses were repeated twice on same sample.

## **11.3 Results and discussion**

### ***11.3.1 Effects of UV-A pelleting on chemical composition of biomass***

Chemical composition of raw corn stover and sorghum stalk (which were not processed by pelleting and pretreatment) are shown in Table 11.1 [12,21-24]. In general, glucan is referred to as cellulose, and xylan is referred to as hemicellulose [12]. It can be seen from Table 11.1 that the composition determined in this study was comparable to those reported by other researchers. Figure 11.4 compares extractives of unpelleted and pelleted biomass. The extractives of sorghum stalk (about 30% dry basis) were higher than those of corn stover (about 20% dry basis). There was no significant difference in extractives between unpelleted and pelleted biomass (P-value = 0.56 for corn stover and P-value = 0.62 for sorghum stalk). A similar trend was reported by Theerarattananoon et al. [12] who compared the extractives of biomass (corn stover and sorghum stalk) processed and not processed by ring-die pelleting [12].

**Table 11.1 Chemical composition of raw corn stover and sorghum stalk**

Type of biomass	Chemical component	Chemical composition (% dry basis)		Reference
		This study*	Reported value	
Corn stover	Glucan	39.4 (0.5)	26.9 - 49.4	[18-20]
	Xylan	23.0 (0.5)	13.4 - 26.2	
	Lignin	16.6 (0.5)	14.3 - 26.0	
Sorghum stalk	Glucan	39.6 (0.8)	32.5 - 41.4	[12,21]
	Xylan	22.7 (0.8)	19.8 - 22.9	
	Lignin	19.5 (0.6)	11.7 - 18.7	

\* Expressed as “mean (standard deviation)”

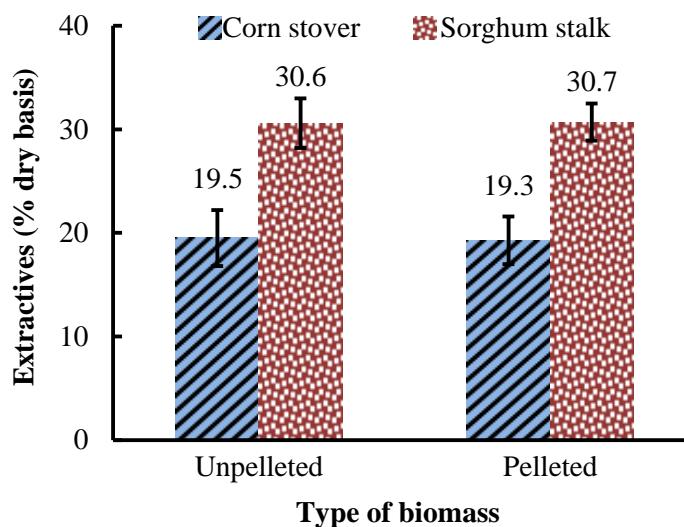
**Figure 11.4 Comparison of extractives between pellets and particles**

Table 11.2 shows composition of unpelleted and pelleted biomass before pretreatment. Statistical analysis (two-sample T-test) of these data revealed that there was no significant difference in the composition between unpelleted and pelleted biomass, indicating that UV-A pelleting did not change the chemical composition of cellulosic biomass. A similar trend was

reported for corn stover, sorghum stalk, wheat straw, and big bluestem processed by ring-die pelleting [12]. However, Yoo et al. [14] reported that chemical composition of soybean hulls was changed after screw extrusion—extruded soybean hulls had higher cellulose and hemicellulose contents and lower lignin content than unextruded soybean hulls. The inconsistency between the reported results might be attributed to the different pelleting methods, revealing that different pelleting methods had different effects on chemical composition of cellulosic biomass. Another possible reason for the inconsistent results is the type of biomass used in different studies. Consistent results were reported in the studies using corn stover and sorghum stalk, which were inconsistent with the results obtained using soybean hulls, implying that effects of pelleting on chemical composition of cellulosic biomass without pretreatment might depend on biomass type.

**Table 11.2 Chemical compositions of pellets and particles before pretreatment**

Type of biomass	Chemical composition (% dry basis)*					
	Glucan	Xylan	ASL	AIL	Total lignin	Ash
Unpelleted corn stover	40.9 (0.5)	23.0 (0.5)	1.1 (0.1)	15.5 (0.4)	16.6 (0.5)	2.6 (0.2)
Pelleted corn stover	41.6 (0.6)	23.4 (0.2)	1.1 (0.2)	16.1 (0.5)	17.2 (0.5)	2.4 (0.2)
Unpelleted sorghum stalk	40.5 (0.8)	22.7 (0.8)	0.9 (0.1)	18.1 (0.5)	19.5 (0.6)	2.8 (0.4)
Pelleted sorghum stalk	41.1 (0.9)	24.1 (0.6)	0.9 (0.1)	18.3 (0.9)	19.7 (0.7)	2.5 (0.2)

\* Expressed as “mean (standard deviation)”

Dilute acid pretreatment removed hemicellulose from biomass. As a result, xylan content was greatly decreased and the contents of other components (glucan, lignin, and ash) were greatly increased after dilute acid pretreatment. Table 11.3 shows the chemical composition of pretreated biomass. The mass recovery (the ratio of weight of biomass before pretreatment to that after pretreatment expressed as a percentage) of unpelleted biomass (60.1% for corn stover

and 56.9% for sorghum stalk) in the current study was close to that (60.6% for both corn stover and sorghum stalk) reported by Theerarattananoon et al. [12]. As shown in Table 11.3, for both corn stover and sorghum stalk, the mass recovery of pelleted biomass was slightly higher than that of unpelleted biomass. A similar trend was reported for corn stover processed by ring-die pelleting; however, an inverse trend was reported for sorghum stalk processed by ring-die pelleting [12].

**Table 11.3 Chemical compositions of pellets and particles after dilute-acid pretreatment**

Type of biomass	Chemical composition (% , dry basis)*						Mass recovery
	Glucan	Xylan	ASL	AIL	Total lignin	Ash	(%)*
Unpelleted corn stover	53.1 (0.7)	1.6 (0.2)	0.80 (0.05)	31.8 (1.0)	32.6 (0.9)	5.6 (0.4)	60.1 (1.1)
Pelleted corn stover	54.4 (0.4)	1.9 (0.4)	0.97 (0.03)	31.3 (0.6)	32.3 (0.5)	6.2 (0.6)	62.1 (1.2)
Unpelleted sorghum stalk	53.4 (0.5)	2.3 (0.1)	0.78 (0.04)	31.1 (0.9)	31.9 (1.0)	3.7 (0.2)	56.9 (1.0)
Pelleted sorghum stalk	54.3 (0.3)	2.5 (0.3)	1.01 (0.03)	32.2 (0.6)	33.2 (0.6)	3.8 (0.2)	58.1 (1.1)

\* Expressed as “mean (standard deviation)”

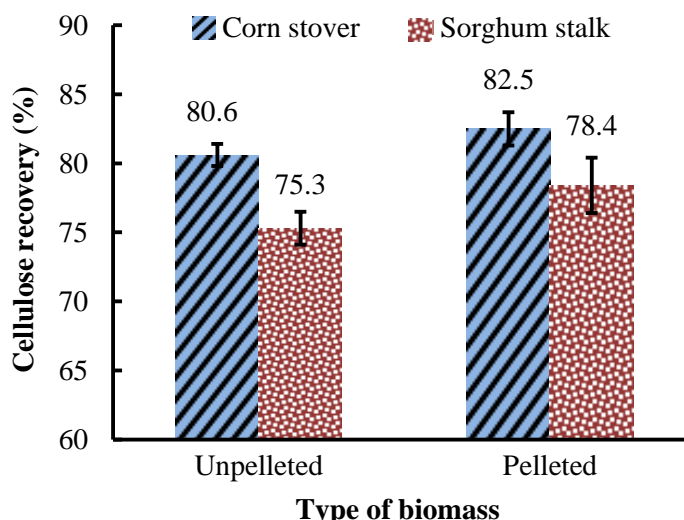
The composition data in Table 11.3 were comparable to those reported in the literature [12]. In the current study, ASL content in pelleted biomass was significantly higher than that in unpelleted biomass (P-value = 0.015 for corn stover and P-value = 0.004 for sorghum stalk), indicating that UV-A pelleting may have positive effects on removal of lignin from cellulosic biomass in dilute acid pretreatment. There was no significant difference in the content of other components between unpelleted and pelleted biomass. Different trends were reported by Theerarattananoon et al. using ring-die pelleting [12]. They reported that, after dilute acid pretreatment, the contents of glucan and ash in pelleted biomass (corn stover, sorghum stalk, wheat straw, and big bluestem) were significantly higher than those in unpelleted biomass. A

comparison between the results of this study and those by Theerarattananoon et al. implies that different pelleting methods might have different effects on the chemical composition of cellulosic biomass treated by dilute acid.

During dilute acid pretreatment, cellulose and hemicellulose were solubilized and converted to fermentable sugars to different extents. Cellulose recovery measured the degree to which cellulose was solubilized during the pretreatment; it was calculated as a ratio between cellulose contents (by weight) in pretreated and unpretreated biomass. A higher cellulose recovery means more cellulose remains in biomass after pretreatment. Figure 11.5 shows the cellulose recovery of different types of biomass. The cellulose recovery of unpelleted biomass in this study (80.6% for corn stover and 75.3% for sorghum stalk) was close to that reported by other researchers (80.6% for corn stover and 77.6% for sorghum stalk) [12]. For both corn stover and sorghum stalk, UV-A pelleting significantly increased their cellulose recovery after dilute acid pretreatment, indicating that less cellulose was solubilized during pretreatment when pelleted biomass was used. A similar trend was reported for corn stover and big bluestem by Theerarattananoon et al. using ring die pelleting [12]. However, they also reported that pelleted wheat straw had lower cellulose recovery than unpelleted wheat straw, and there was nearly no difference in cellulose recovery between pelleted and unpelleted sorghum stalk. Both pelleting methods had positive effects on cellulose recovery of corn stover, implying that pelleting would probably help retain more cellulose in dilute acid pretreatment for corn stover. UV-A pelleting had positive effects on cellulose recovery of both tested biomass types, whereas ring-die pelleting had different effects on cellulose recovery of different biomass types. For sorghum stalk, different pelleting methods had different effects on its cellulose recovery. These results indicate that effects of pelleting on cellulose recovery of biomass after dilute acid pretreatment depend on both pelleting method and biomass type.



**Figure 11.5 Comparison of cellulose recovery of pellets and particles after dilute-acid pretreatment**



The main components in filtrate of biomass after dilute acid pretreatment included glucose, xylose, and arabinose, and their contents are shown in Table 11.4. Xylose and arabinose were solubilized from hemicellulose in dilute acid pretreatment. More xylose was found in filtrate of unpelleted biomass than in that of pelleted biomass, indicating that more hemicellulose remained in pelleted biomass after dilute acid pretreatment than in unpelleted biomass. This is consistent with the results in Table 11.3. A similar trend was reported for sorghum stalk using ring-die pelleting; however, an inverse trend was found for corn stover, wheat straw, and big bluestem [12]. Both cellulose and hemicellulose contain glucose. Therefore, the glucose in the filtrate was considered to be from both cellulose and hemicellulose. More glucose was found in filtrate of unpelleted biomass than in that of pelleted biomass, possibly because more hemicellulose was solubilized in pretreatment for unpelleted biomass than for pelleted biomass. This is consistent with the results for corn stover and big bluestem using ring-die pelleting but inconsistent with the results for wheat straw and sorghum stalk using ring-die pelleting [12].

**Table 11.4 Sugar yield in filtrate of biomass after dilute acid pretreatment**

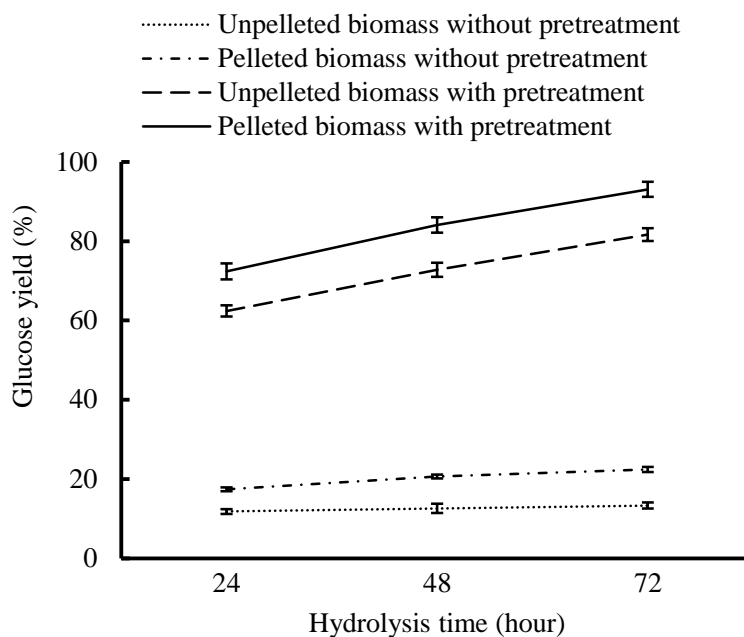
Type of biomass	Component in filtrate* (g/100g of dry, unpretreated biomass)		
	Glucose	Xylose	Arabinose
Unpelleted corn stover	3.7 (0.1)	19.6 (0.1)	3.1 (0.5)
Pelleted corn stover	3.6 (0.1)	18.7 (0.1)	3.6 (0.1)
Unpelleted sorghum stalk	5.2 (0.2)	18.6 (0.1)	3.2 (0.1)
Pelleted sorghum stalk	4.5 (0.1)	18.2 (0.1)	2.9 (0.1)

\* Expressed as “mean (standard deviation)”

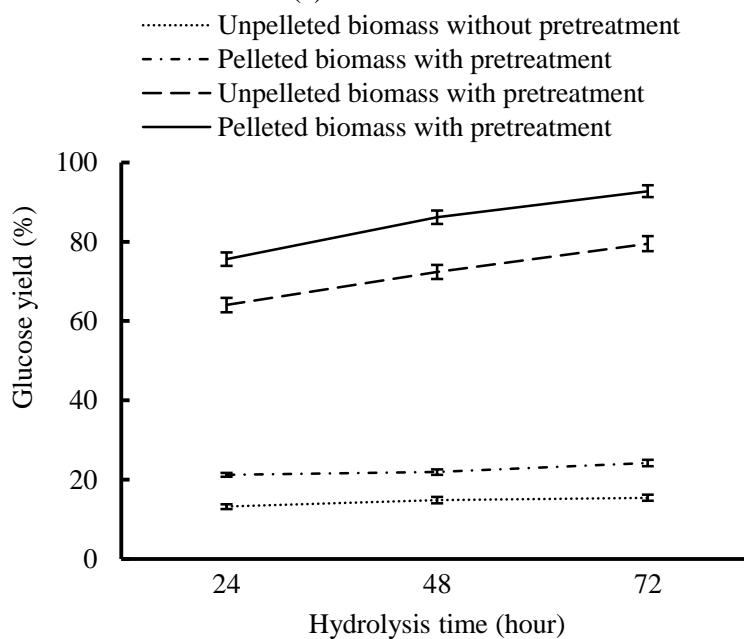
### ***11.3.2 Effects of UV-A pelleting on sugar yield in enzymatic hydrolysis***

Figure 11.6 shows the sugar yield of different biomass samples in enzymatic hydrolysis. It can be seen that UV-A pelleting significantly increased sugar yield for both corn stover and sorghum stalk. Without dilute acid pretreatment, sugar yield of pelleted corn stover (22.4%) was 70% higher than that of unpelleted corn stover (13.3%) after 72-hour hydrolysis (Figure 11.6(a)). With pretreatment, sugar yield (after 72-hour hydrolysis) of pelleted corn stover (93.1%) was 14% higher than that of unpelleted corn stover (81.7%). Similarly, without pretreatment, the sugar yield (after 72-hour hydrolysis) of pelleted sorghum stalk (24.2%) was 57% higher than that of unpelleted sorghum stalk (15.4%) (Figure 11.6(b)). With pretreatment, sugar yield (after 72-hour hydrolysis) of pelleted sorghum stalk (92.8%) was 17% higher than that of unpelleted sorghum stalk (79.5%). These results are consistent with those reported by other researchers with different pelleting methods. It was reported that ring-die pelleting could significantly increase sugar yield of corn stover, sorghum stalk, wheat straw, and big bluestem [12]; extrusion could significantly increase sugar yield of big bluestem [25], corn stover [25,26], prairie cordgrass [27], sorghum [28], soybean hulls [14], switchgrass [27,29], and wheat bran [13].

**Figure 11.6 Sugar yield for different types of biomass**



(a) Corn stover



(b) Sorghum stalk

Without pretreatment, the sugar yields of unpelleted biomass in this study (13.3% for corn stover and 15.4% for sorghum) are comparable to the literature data (17% for wheat bran [13]). The increased sugar yields caused by UV-A pelleting (22.4% for corn stover and 24.2%

for sorghum stalk) are comparable to that of extruded wheat bran (21.5-31% [13]) but slightly lower than that of extruded switchgrass (26.8-40.6% [29]). This indicates that, without pretreatment, effects of UV-A pelleting on sugar yield are comparable to or smaller than those of extrusion depending on biomass type and pelleting condition.

With pretreatment, the sugar yield of unpelleted biomass with pretreatment obtained in this study (81.7% for corn stover and 79.5% for sorghum stalk) is lower than the literature data (84.6% for corn stover and 87.1% for sorghum stalk [12]). The difference might be due to the lower glucan content and higher lignin content in the biomass used in this study. The sugar yield of pelleted biomass with pretreatment obtained in this study (93.1% for corn stover and 92.8% for sorghum stalk) is close to that of biomass processed by ring-die pelleting (93.1% for corn stover and 92.2% for sorghum stalk [12]). This indicates that UV-A pelleting might have greater effects on sugar yield of pretreated biomass than ring-die pelleting. The results on sugar yield indicate that UV-A pelleting of biomass could be considered as a preliminary pretreatment step to further increase the sugar yield of cellulosic biomass.

In the literature, two major mechanisms have been proposed to explain the increased sugar yield in enzymatic hydrolysis caused by pelleting of biomass. One mechanism is the shear developed during pelleting of biomass, which opens the biomass structure and thus increases enzyme access to cellulose [12,25,28]. The other mechanism is the heat generated during pelleting of biomass, which causes thermal-softening of biomass and increases sugar release from the biomass in enzymatic hydrolysis [27]. The shear mechanism may not be responsible for the increased sugar yield caused by UV-A pelleting for two reasons. First, instead of pushing biomass through one or more die openings like in ring-die pelleting, biomass is compressed in a closed mold to form pellets in UV-A pelleting. Therefore, biomass is mainly subject to compressive pressure rather than shear in UV-A pelleting. Second, the pressure applied on biomass in UV-A pelleting (less than 12 MPa) is much lower than those in other pelleting methods (100-200 MPa for ring-die pelleting or extrusion [30,31]). According to the reported

shear strength of biomass (approximately 9 MPa for sorghum stalk [32] and 8.6-13 MPa for wheat straw [33]), the low pressure in UV-A pelleting is probably not enough to open the biomass structure. However, the temperature of biomass in UV-A pelleting (160-200°C [34]) is higher than or comparable to those in other pelleting methods (74-82°C for ring-die pelleting [6] and 80-225°C for extrusion [14,15]), indicates that biomass is subject to greater or similar thermal-softening effects in UV-A pelleting compared with in ring-die pelleting or extrusion. Therefore, the high temperature in UV-A pelleting may be the major contributor to the increased sugar yield.

## **11.4 Conclusions**

This paper studies effects of ultrasonic vibration-assisted (UV-A) pelleting on chemical composition and sugar yield for corn stover and sorghum stalk. The following conclusions can be drawn from the study:

Without dilute acid pretreatment, there was no significant difference in the chemical composition between pelleted and unpelleted biomass (by UV-A pelleting). With dilute acid pretreatment, the content of acid soluble lignin (ASL) in pelleted biomass was significantly higher than those in unpelleted biomass. There was no significant difference in the other components and extractives between unpelleted and pelleted biomass. For both corn stover and sorghum stalk, the cellulose recovery of pelleted biomass was significantly higher than that of unpelleted biomass after dilute acid pretreatment.

UV-A pelleting could significantly increase the sugar yield in enzymatic hydrolysis for both corn stover and sorghum stalk. Without dilute acid pretreatment, the sugar yield of pelleted biomass was more than 50% higher than that of unpelleted biomass. With dilute acid pretreatment, the sugar yield of pelleted biomass was more than 10% higher than that of unpelleted biomass. The combination of UV-A pelleting and dilute acid pretreatment could achieve the sugar yield of 93.1% for corn stover and 92.8% for sorghum stalk.

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## Chapter 12 - **Investigation on characteristics of cellulosic biomass processed by ultrasonic-vibration assisted pelleting**

### Paper Title:

Investigation on characteristics of corn stover and sorghum stalk processed by ultrasonic-vibration assisted pelleting for sugar production

### To be submitted to:

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### **Abstract**

Ethanol produced from cellulosic biomass is an alternative to petroleum-based liquid transportation fuels. Two major challenges hindering cost-effective cellulosic ethanol

manufacturing are low density of raw cellulosic biomass and low sugar yield of cellulosic biomass in enzymatic hydrolysis. Ultrasonic vibration-assisted (UV-A) pelleting can remarkably increase biomass density. In addition, a preliminary study revealed that sugar yield of switchgrass processed by UV-A pelleting was higher than that of unprocessed switchgrass. The objective of this study was to confirm the preliminary results regarding sugar yield using two other types of biomass (corn stover and sorghum stalk) and to investigate the effects of UV-A pelleting on biomass characteristics (such as chemical composition, crystallinity index, thermal properties, and morphological structure). The results showed that pellets processed by UV-A pelleting had 13% higher sugar yield than biomass particles. There was no significant difference in chemical composition between pellets and particles. However, crystallinity of pellets was higher than that of particles. In addition, pellets had higher decomposition temperature than particles, indicating that pellets were more thermally stable than particles. Examinations on morphological structure of biomass showed that, after UV-A pelleting, softened surface regions of biomass were removed and cellulose microfibrils were revealed.

## **12.1 Introduction**

Ethanol produced from cellulosic biomass (herbaceous, woody, and generally inedible portions of plant matter) is an attractive alternative to petroleum-based liquid transportation fuels. Land resources in the U.S. are sufficient to sustain production of enough biomass annually to displace 30% or more of the nation's current petroleum consumption [1]. Furthermore, cellulosic ethanol has great environmental advantages over grain-based ethanol [2,3].

Cost-effective manufacturing of cellulosic ethanol has been facing several technical challenges. One challenge is related to low density of raw cellulosic biomass (ranging from 24 to 266 kg/m<sup>3</sup> [4]), causing high costs in biomass transportation and storage. Another challenge is low sugar yield in enzymatic hydrolysis, leading to low biomass-to-ethanol conversion rate and high costs in converting biomass into ethanol.

Pelleting (agglomeration of small particles into larger particles by means of mechanical or thermal processing [5]) can significantly increase density of cellulosic biomass. The density of biomass pellets could reach 1200 kg/m<sup>3</sup> [6]. In turn, costs for transporting and storing of pelleted cellulosic biomass are less than 1/2 and 1/10 of those of raw cellulosic biomass, respectively [7,8]. In addition, pellets with uniform size and shape are easier for handling with existing grain processing equipment. A variety of traditional pelleting methods have been reported in the literature, including ring-die pelleting [9-11], flat-die pelleting [9,11], screw extrusion [12], and piston press [13]. For these pelleting methods, cellulosic biomass usually needs to be preheated (by high-temperature steam or heated dies) and binders are often needed [4]. By contrast, ultrasonic vibration-assisted (UV-A) pelleting does not require preheated biomass as feedstock and binders. However, UV-A pelleting can produce pellets whose density and durability are comparable to those produced by traditional pelleting methods [6].

Pelleting of biomass has also been considered as an effective pretreatment method to increase biomass sugar yield in enzymatic hydrolysis. Theerarattananoon et al. [14] reported that sugar yield of biomass (big bluestem, corn stover, sorghum stalk, and wheat straw) processed by ring-die pelleting was 3-11% higher than that of unpelleted biomass. Lamsal et al. [15] reported that sugar yield of wheat bran processed by screw extrusion was about 30% higher than that of unpelleted wheat bran. A similar trend was reported by Yoo et al. [16,17] in screw extruding of soybean hulls. Our previous studies regarding UV-A pelleting of biomass revealed that UV-A pelleting of biomass was beneficial for increasing sugar yield of biomass in enzymatic hydrolysis. Zhang et al. [18] reported that, with dilute acid pretreatment, switchgrass processed by UV-A pelleting had 20% higher sugar yield than unpelleted switchgrass. Without pretreatment, pellets had 75% higher sugar yield than unpelleted switchgrass. However, effects of UV-A pelleting on sugar yield of other types of biomass are still unknown. Furthermore, there is no study in the literature regarding effects of UV-A pelleting and diluted acid pretreatment on

biomass characteristics. It is essential to know such effects to reveal the mechanisms through which UV-A pelleting increases biomass sugar yield.

In this paper, effects of UV-A pelleting on sugar yield and characteristics (such as crystallinity index, chemical structure, and thermal properties) of corn stover and sorghum stalk were investigated. To determine the characteristics, a variety of measurement methods were used, such as X-ray diffraction, Fourier transform infrared spectroscopy (FTIR), thermogravimetric analysis (TGA), and solid-state cross polarization/magic angle spinning (CP/MAS)  $^{13}\text{C}$  nuclear magnetic resonance (NMR) spectroscopy. Morphological structure of biomass was observed using scanning electron microscope (SEM).

## **12.2 Materials and methods**

### ***12.2.1 Materials***

Corn stover and sorghum stalk used in this study were harvested at the Kansas State Agronomy Farm in November, 2010. After harvesting, they were chopped to approximately 180-230 mm using a large tub grinder (Haybuster H-1150 series, DuraTech Industries International Inc., Jamestown, ND, USA). Chopped biomass was then transported to a lab located at Kansas State University in paper bags. Before UV-A pelleting, the chopped biomass was milled into particles using a cutting mill (model SM 2000, Retsch, Inc., Haan, Germany) with a sieve whose mesh size was 1 mm. The milled biomass was referred to as biomass particles.

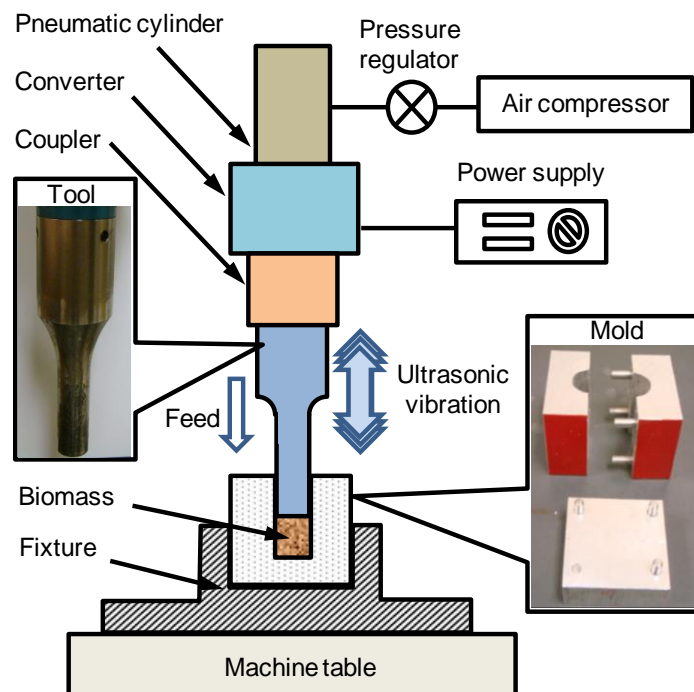
### ***12.2.2 Experimental setup for UV-A pelleting***

UV-A pelleting was conducted on a modified ultrasonic machine (model AP-1000, Sonic-Mill, Albuquerque, NM, USA). The schematic illustration of the experimental setup is shown in Figure 12.1. The power supply converted 60-Hz electrical supply into 20-kHz AC output which was fed to the piezoelectric converter and converted into high-frequency (20-kHz) mechanical motion. The mechanical motion was amplified by the coupler and transmitted to the pelleting tool. The ultrasonically vibrating tool was used to compress biomass particles into

pellets. Ultrasonic power (percentage of power from power supply) can be adjusted from 0 to 100% and controls amplitude of the tool vibration. The higher the ultrasonic power, the larger the tool vibration amplitude. The air compressor (1.2 kw, 125 liter, Sears, Roebuck and Co., Hoffman Estates, IL, USA) produced compressed air which was fed into the pneumatic cylinder (ARO Equipment Corporation, Bryan, OH, USA). The air pressure in the pneumatic cylinder was controlled by the pressure regulator and referred to as pelleting pressure. In UV-A pelleting, biomass particles were held by a three-piece aluminum mold. The top two pieces of the mold formed a central cylindrical cavity and the bottom part served as a base. The diameter of the mold cavity (18.6 mm) was slightly larger than that of the tip of the pelleting tool (17.4 mm).

There were five steps to make a pellet in this study: (1) assemble the mold and put 1 g of biomass particles in the central cavity of the mold, (2) adjust pelleting pressure to 50 psi and feed the pelleting tool down to compress particles, (3) turn on ultrasonic power at 50% and count 3 minutes using a stop watch, (4) turn off ultrasonic power and lift up the pelleting tool, and (5) disassemble the mold and take out the pellet.

**Figure 12.1 Illustration of experimental setup for UV-A pelleting**



### ***12.2.3 Pretreatment***

Pretreatment was carried out on a pressure reactor (Parr Instrument Company, Moline, IL, USA). A mixture of 20-g biomass and 200-mL 2% diluted sulfuric acid (solid content about 10%) were loaded into a 600-mL reaction vessel. The biomass slurry (biomass with diluted sulfuric acid) was treated at 140 °C for 30 minutes. Meanwhile, the slurry was stirred by two impeller mixers. In pellet pretreatment, pellets were dissolved in the 2% (w/v) sulfuric acid solution in the vessel at room temperature before being loaded in the reactor. Pretreated biomass was washed with distilled water and centrifuged three times to remove dissolved sugars and sulfuric acid. The supernatant was removed after centrifuging at 8000 rpm for 20 min for 3 times. Washed biomass samples were weighed and split into two portions. One portion was used for moisture content measurement and chemical compositional analysis, and the other portion was used for subsequent enzymatic hydrolysis.

### ***12.2.4 Enzymatic hydrolysis***

Pretreated biomass was enzymatically hydrolyzed in solution with 1.36% (w/v) sodium acetate and 0.02% (w/v) sodium azide to prevent microbial growth during hydrolysis. The pH value of the solution was adjusted to 4.8. Biomass (5 grams dry weight) was mixed with 100-mL buffer solution in 125-mL flasks in a 50°C water bath shaker (Model C76, New Brunswick Scientific, Edison, NJ, USA). The flasks were being agitated at 180 rpm for 72 hours. The enzyme loading (Accellerase 1500TM, Genencor Inc., Rochester, NY, USA) was 1 mL/g cellulose. After every 24-hour enzymatic hydrolysis, the hydrolysis slurry was sampled by withdrawing 1 mL of slurry from each flask into vials. The vials were placed in boiling water for 15 minutes to deactivate the enzyme. Then samples were centrifuged at 13500 rpm for 15 minutes. 0.1 mL supernatant was withdrawn and mixed with 0.9 mL distilled water in 1.5-mL vials. The supernatant was then filtered into 1.5-mL autosampler vials through 0.2 µm hydrophilic PTFE syringe filters. Filtered samples were stored at 0°C until HPLC analysis.



In this study, sugar yield means the percentage of cellulose enzymatically converted to glucose in hydrolysis. It was calculated by the following formula:

$$\text{Sugar yield} = \frac{c * v * 10}{1.11 * m} * 100\% \quad (2)$$

where c is the concentration (g/L) of glucose in the sampled hydrolysate determined by HPLC analysis, v is the total volume (L) of the slurry in the flasks, m is the weight (g) of cellulose before enzymatic hydrolysis (g), and the number 1.11 is the cellulose-to-glucose conversion factor.

### ***12.2.5 Analytical methods***

Moisture content (MC) of particles and pellets was measured following a NREL laboratory analytical procedure [19]. Composition of biomass before and after pretreatment was determined by following NREL laboratory analytical procedures [20,21]. The composition of biomass was reported as percentages of glucan (cellulose), xylan (major hemicellulose constituent), and lignin including acid-insoluble (AIL) and acid-soluble lignin (ASL). Glucose, xylose, mannose, and arabinose were determined via an HPLC instrument (Shi-madzu, Kyoto, Japan) equipped with an RCM monosaccharide column (300 x 7.8 mm; Phenomenex, Torrence, CA, USA), and a refractive index detector (RID-10A, Shimadzu, Kyoto, Japan). The mobile phase was 0.6 mL/min of double-distilled water, and oven temperature was 80 °C.

### ***12.2.6 X-ray diffraction analysis***

Crystallinity index of the biomass samples before and after pretreatment was analyzed by wide-angle X-ray diffraction (XRD) with a desktop X-ray diffractometer (Rigaku Instrument Co., Woodlands, TX, USA). Samples were irradiated with Cu K $\alpha$  ( $\lambda=1.5 \text{ \AA}$ ) rays at 15 KV and grade range was between 10° to 30° with a step size of 0.02° at room temperature. The scan speed was set at 2°/min.

Crystallinity index (CrI) was calculated using Equation (3) reported by Segal et al. [22]:

$$CrI = \frac{I_{002} - I_{amorphous}}{I_{002}} \times 100 \quad (3)$$

where  $I_{002}$  is the intensity of crystallinity portion of biomass at  $22.5^\circ$  and  $I_{amorphous}$  is the intensity of the background scatter (amorphous region) at  $18.7^\circ$ .

### ***12.2.7 Fourier transform infrared spectroscopy (FTIR)***

FTIR was used to investigate the structure of constituents and chemical changes in cellulosic biomass. FTIR analyses were performed using a PerkinElmer Spectrum 400 FT-IR spectrometer (PerkinElmer Inc., Waltham, MA, USA). All spectra were recorded in the absorbance mode in the wave number range of  $400\text{--}4000\text{ cm}^{-1}$  with a detection resolution of  $4\text{ cm}^{-1}$  in the transmission mode and 16 scans per sample.

### ***12.2.8 Thermogravimetric analysis (TGA)***

TGA was performed to measure the thermal properties of biomass samples. It was carried out on a Pyris 1 TGA Thermogravimetric Analyzer (The PerkinElmer Co., Norwalk, CT, USA). Dynamic thermogravimetric scans were conducted in a temperature range from  $30$  to  $600^\circ\text{C}$  at a heating rate of  $20^\circ\text{C}/\text{min}$ . In each TGA,  $5\text{--}6\text{ mg}$  biomass was used.

### ***12.2.9 Solid-State $^{13}\text{C}$ nuclear magnetic resonance (NMR) spectroscopy***

Solid-State  $^{13}\text{C}$  NMR spectroscopy was used to determine the changes in chemical structure of biomass. Solid state NMR spectra were acquired on a Bruker Avance III 400 spectrometer (Bruker Biospin, Billerica, MA, USA) operating at  $400.1\text{ MHz}$  for  $^1\text{H}$  and  $100.6\text{ MHz}$  for  $^{13}\text{C}$ . A  $7\text{-mm}$  spin module in a 4-module multiple sample solids (MSS) probe (Revolution NMR, Ft. Collins, CO) was used. Spectrometer setup used 3-methylglutaric acid (MGA) as a secondary external chemical shift reference via the methyl peak at  $18.84\text{ ppm}$  relative to tetramethylsilane (TMS). Each sample was packed into a  $7.0\text{-mm}$  zirconia rotor (Revolution NMR, Ft. Collins, CO, USA). Cross polarization was used for all measurements

with a contact time of 1 ms. The spectral width was 40 kHz and the acquisition time was 30 ms. Proton decoupling was performed with SPINAL-64 and a proton decoupling field of 64 kHz.

#### 12.2.10 Scanning electron microscopy (SEM)

SEM was used to observe the microstructure of biomass. An EVO MA10 SEM (Carl Zeiss Microscopy, Thornwood, NY, USA) was used to exam biomass samples. The samples were mounted on conductive adhesive carbon tapes, and observed using a voltage of 5 kV.

### 12.3 Results and discussion

#### 12.3.1 Sugar yield and compositional analysis

Figure 12.2 shows the sugar yield results of particles and pellets in enzymatic hydrolysis. It can be seen that pellets had 13 % higher sugar yield than particles after 72-hour hydrolysis. This result is consistent with that of our previous study with switchgrass [18]. This implies that UV-A pelleting might be considered as a pretreatment method to increase sugar yield in enzymatic hydrolysis.

**Figure 12.2 Sugar yield results**

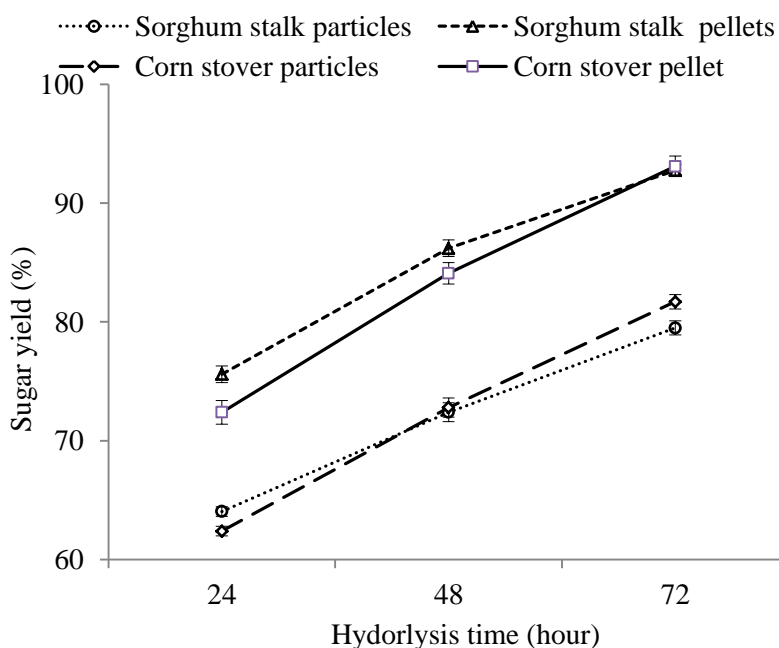


Table 12.1 shows the chemical compositions of various types of biomass samples. It can be seen that, without pretreatment, there was no significant difference between particles and pellets, indicating that UV-A pelleting did not alter the chemical composition of biomass. After pretreatment, the percentage of xylan was significantly reduced from 24% to 2%, indicating that most hemicellulose was removed during pretreatment. Glucan content increased from 40% to 52% and lignin content increased from 16% to 33%. Glucan content in pellets was slightly (but not significantly at the significance level of 0.05) higher than that in particles.

**Table 12.1 Chemical composition of particles and pellets before and after pretreatment**

Type of biomass			Chemical composition (% , dry basis)*						Mass recovery (%)*
			Glucan	Xylan	ASL	AIL	Total lignin	Ash	
Corn stover	Without pretreatment	Particles	40.9 (0.5)	23.0 (0.5)	1.1 (0.1)	15.5 (0.4)	16.6 (0.5)	2.6 (0.2)	
		Pellets	41.6 (0.6)	23.4 (0.2)	1.1 (0.2)	16.1 (0.5)	17.2 (0.5)	2.4 (0.2)	
	With pretreatment	Particles	53.1 (0.7)	1.6 (0.2)	0.80 (0.05)	31.8 (1.0)	32.6 (0.9)	5.6 (0.4)	60.1 (1.1)
		Pellets	54.4 (0.4)	1.9 (0.4)	0.97 (0.03)	31.3 (0.6)	32.3 (0.5)	6.2 (0.6)	62.1 (1.2)
Sorghum stalk	Without pretreatment	Particles	40.5 (0.8)	22.7 (0.8)	0.9 (0.1)	18.1 (0.5)	19.5 (0.6)	2.8 (0.4)	
		Pellets	41.1 (0.9)	24.1 (0.6)	0.9 (0.1)	18.3 (0.9)	19.7 (0.7)	2.5 (0.2)	
	With pretreatment	Particles	53.4 (0.5)	2.3 (0.1)	0.78 (0.04)	31.1 (0.9)	31.9 (1.0)	3.7 (0.2)	56.9 (1.0)
		Pellets	54.3 (0.3)	2.5 (0.3)	1.01 (0.03)	32.2 (0.6)	33.2 (0.6)	3.8 (0.2)	58.1 (1.1)

\* Expressed as “mean (standard deviation)”

### 12.3.2 Results on FTIR analysis

In FTIR analysis, the major structural components in cellulosic biomass (cellulose, hemicellulose, and lignin) are identified by absorption bands. Table 12.2 summarizes the assignments of FTIR absorption bands reported in the literature. The FRIT spectra of various biomass samples obtained in this study are shown in Figures 12.3-12.5.

**Table 12.2 Assignment of FTIR absorption bands for biomass**

Wave number (cm <sup>-1</sup> )	Assignment
---------------------------------	------------

1599	Aromatic skeletal vibration plus C=O stretching (related to lignin removal)
1509	C=C stretching from guaiacyl ring of lignin
1462	Aromatic C-H deformation; asymmetric in -CH <sub>3</sub> and -CH <sub>2</sub> -
1425	C-H in-plane deformation with aromatic skeletal stretching
1370	Weak C-O stretching in cellulose
1270	C-O of guaiacyl ring and C-O stretching
1260	Ester absorbance (related to removal of uronic acid)
1164	C-O-C asymmetric vibrations at $\beta$ -glucosidic linkages in cellulose and hemicellulose
1106	Antisymmetric in-phase ring stretch of cellulose
1060	C-O stretching in cellulose
1043	C-O stretching in C-O-C linkages of hemicellulose
1033	C-O deformation in primary alcohol or C-O, C=C and C-C-O vibration stretching in lignin
1030	C-O stretching in cellulose
991	$\beta$ -glucan in hemicellulose
900	Antisymmetric out-of-plane ring stretch of amorphous cellulose; C-O stretching

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Figure 12.3 FTIR spectra of pure hemicellulose and various biomass samples

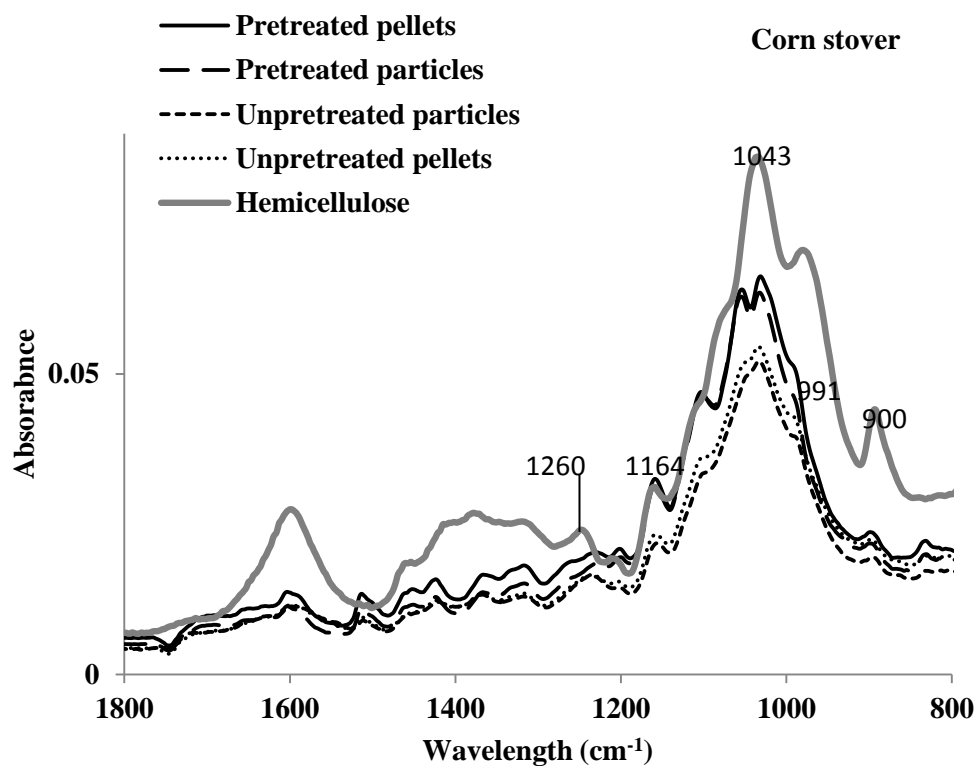
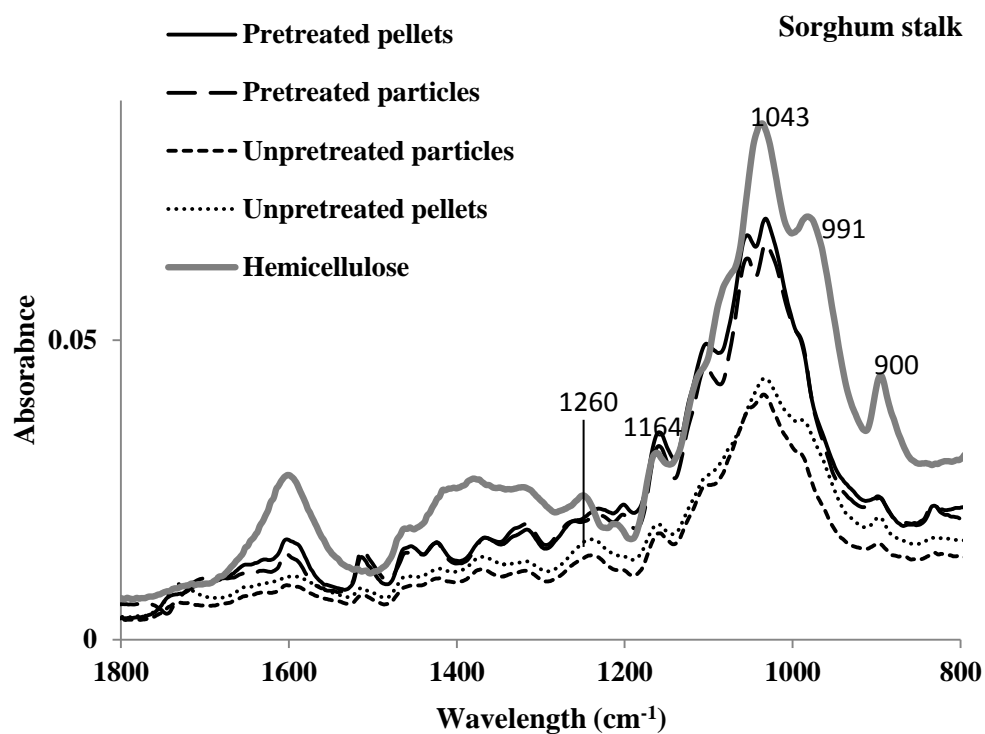


Figure 12.4 FTIR spectra of pure cellulose and various biomass samples

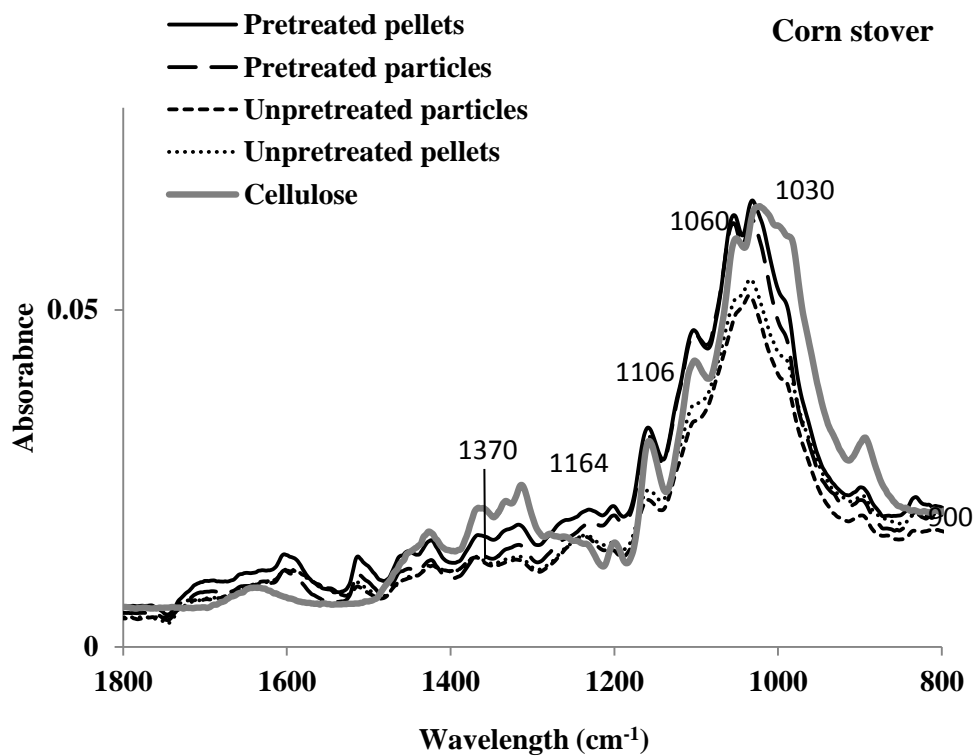
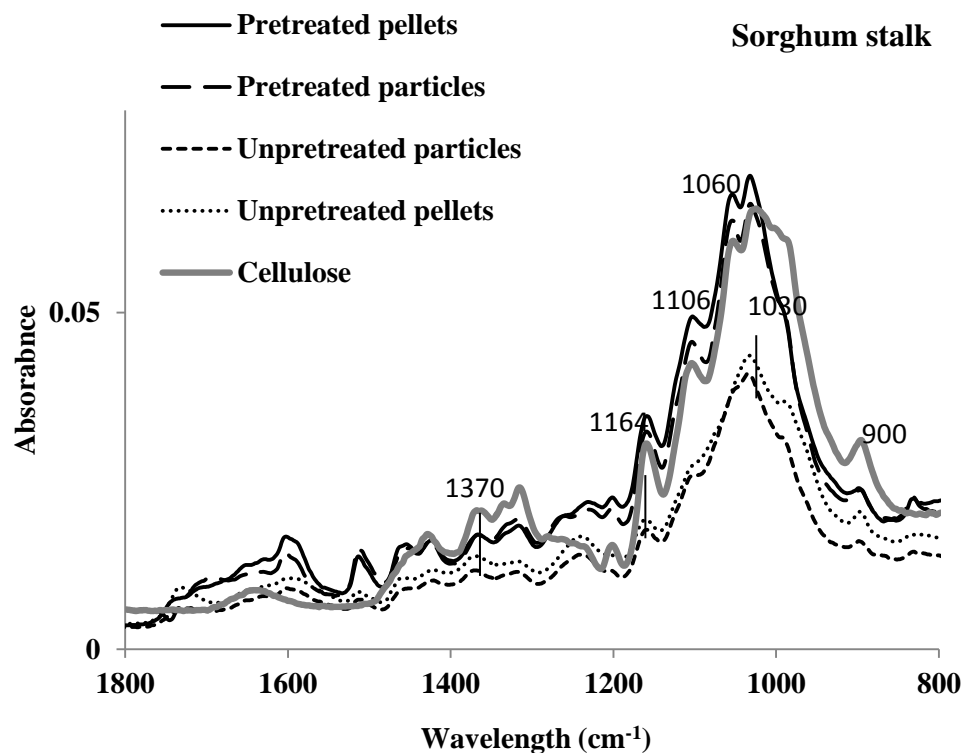
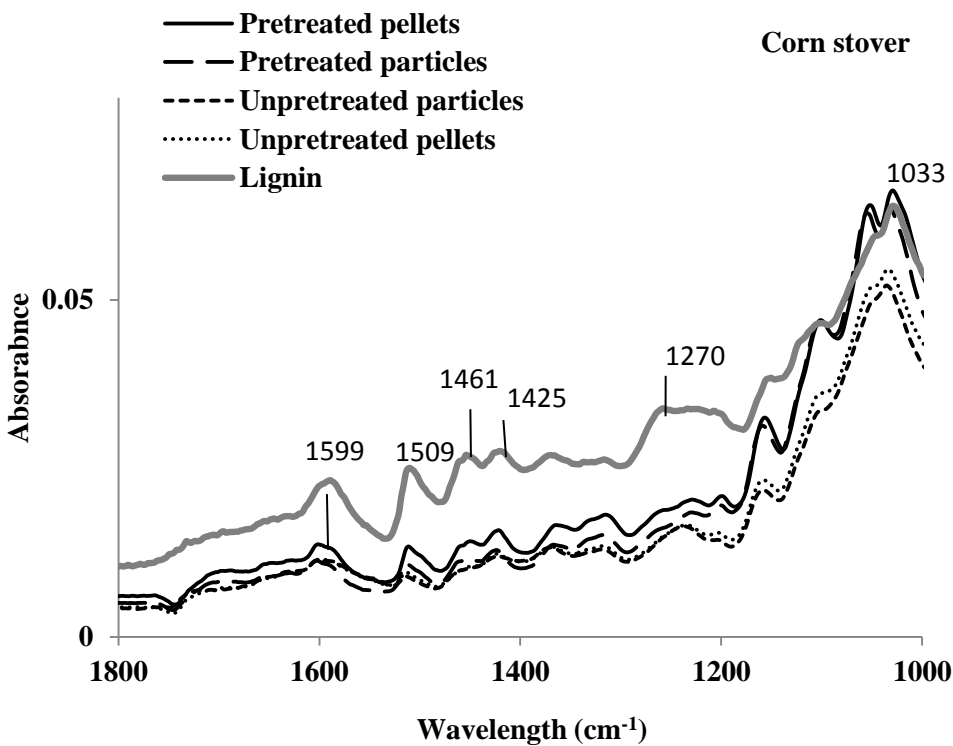
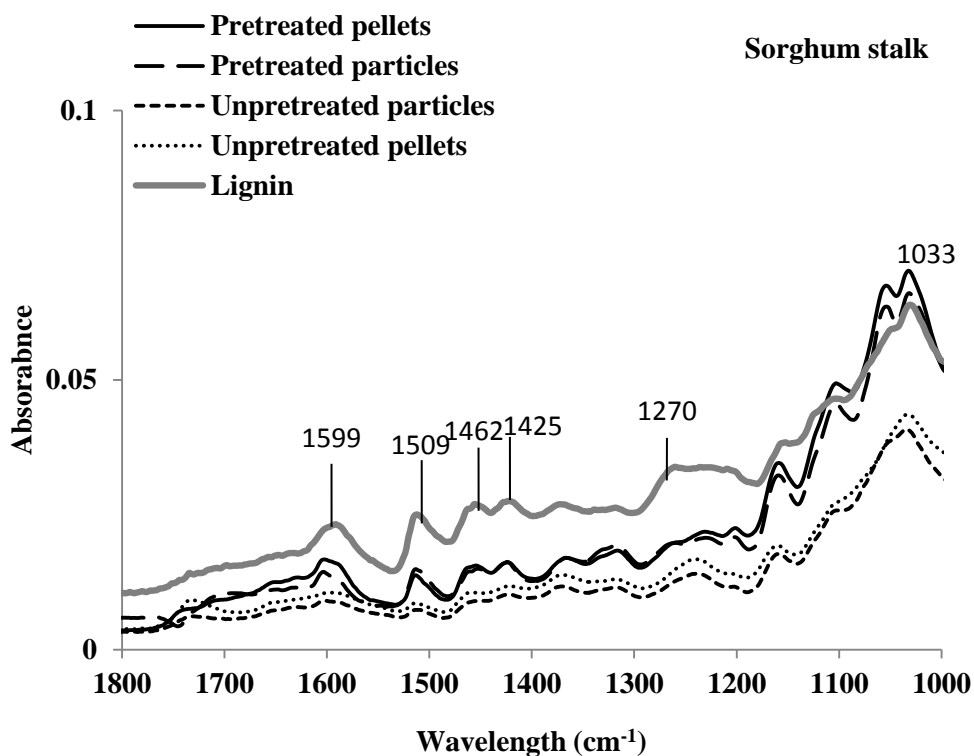


Figure 12.5 FTIR spectra of pure lignin and various biomass samples





FIRT spectra of pure hemicellulose, particles, and pellets are shown in Figure 12.3. The hemicellulose-related absorption bands can be identified from the characteristic peaks with wave numbers of 900, 991, 1043, 1164, and 1260  $\text{cm}^{-1}$  [23-25]. The location of these peaks is consistent with the observations of Chen et al. [23] and Ren et al. [25]. Before pretreatment, the five peaks could be observed in both biomass particles and pellets. There was no significant difference in the pattern of FRIT spectra of hemicellulose between particles and pellets, implying that UV-A pelleting did not alter hemicellulose content in biomass. This is consistent with the results in Table 12.1. However, after pretreatment, the peaks of at 1043 and 1260  $\text{cm}^{-1}$  were withered obviously, indicating a significant reduction in hemicellulose content in pretreatment biomass [23].

FTIR spectra of pure cellulose, particles, and pellets are shown in Figure 12.4. The cellulose-related absorption bands can be identified from the characteristic peaks with wave numbers of 900, 1033, 1060, 1106, 1164, and 1370  $\text{cm}^{-1}$  [23,24,26]. The location of these peaks is similar to those obtained by Chen et al. [23], Liu et al. [27], and Adapa et al. [24]. Before pretreatment, the peaks at 1033, 1060, 1106, and 1370  $\text{cm}^{-1}$  are not clear for both particles and pellets. But there is no clear difference in the spectra between particles and pellets, implying that UV-A pelleting did not change cellulose content in biomass. After pretreatment, the aforementioned peaks are notable for both particles and pellets. This indicates that removal of hemicellulose during pretreatment could increase the cellulose content in the biomass [23]. This result is consistent with the composition data in Table 12.1.

FTIR spectra of pure lignin, particles, and pellets are shown in Figure 12.5. The lignin-related bands can be identified from the characteristic peaks with wave numbers of 1033, 1270, 1425, 1462, 1509, and 1599  $\text{cm}^{-1}$  [24,26,28]. The location of these peaks is similar to those obtained by Chen et al. [23], Apada et al. [24], Xiao et al. [28], and Sun et al. [29]. The six peaks were clearly observed for both pretreated and untreated biomass samples, revealing that lignin content is not affected by dilute acid pretreatment. The peaks around 1599, 1509, 1462, and 1425

cm-1 were enhanced after pretreatment, indicating that pretreated samples had higher lignin content [23,28].

### ***12.3.3 Results on biomass crystallinity index***

In cellulosic biomass, cellulose consists of crystalline regions (with highly ordered molecule arrangement) and amorphous regions (with less ordered molecule arrangement) [30]. Crystallinity index (CrI) is defined as the percentage of the amount (weight) of crystalline material in biomass [22,31]. CrI has been correlated with the accessibility of cellulose to cellulase enzyme and considered as an influencing factor on sugar yield in enzymatic hydrolysis.

The XRD patterns of corn stover particles and pellets before and after pretreatment are shown in Figure 12.6. A major diffraction peak of cellulose crystallographic plane can be identified in the range of 22-23° [23,28]. Before pretreatment, the crystalline structure (around peak 22.5°) is not obvious due to the coverage of cellulose by hemicellulose and lignin [23]. After pretreatment, the peak around 22.5° is significantly increased, indicating that the crystalline cellulose of pretreated corn stover is clearly exhibited due to the removal of hemicellulose (amorphous region of biomass) and disruption of hydrogen bonding of cellulose chain by pretreatment [31,32]. The increased percentage of crystalline portion in biomass resulted in a higher CrI.

Figure 12.6 XRD patterns of corn stover particles and pellets before and after pretreatment

**Figure 12.6 XRD patterns of particles and pellets before and after pretreatment**

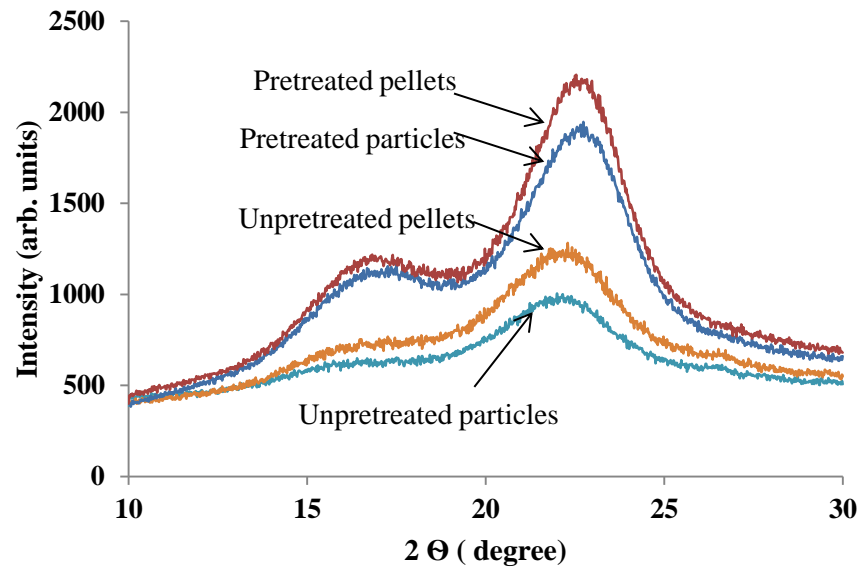
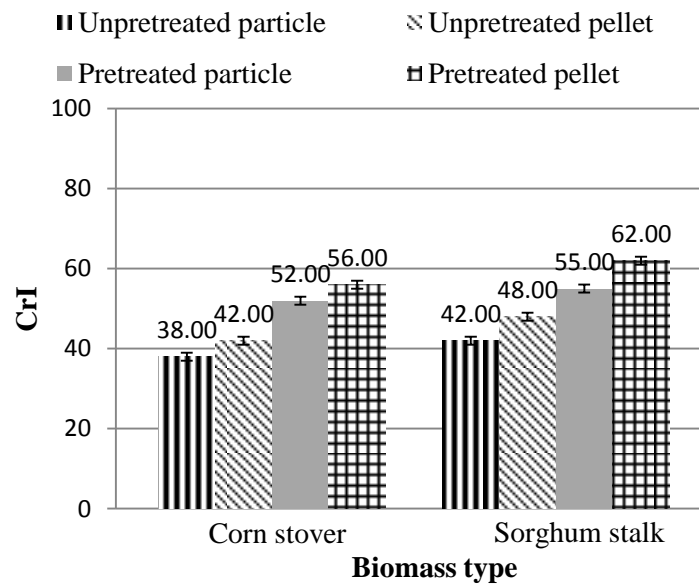


Figure 12.7 shows the effects of UV-A pelleting on the CrI of corn stover and sorghum stalk. It can be seen that pellets had higher CrIs than particles whether pretreatment was applied or not. Similar results were reported by other researchers with ring-die pelleting [33] and screw extrusion [16].

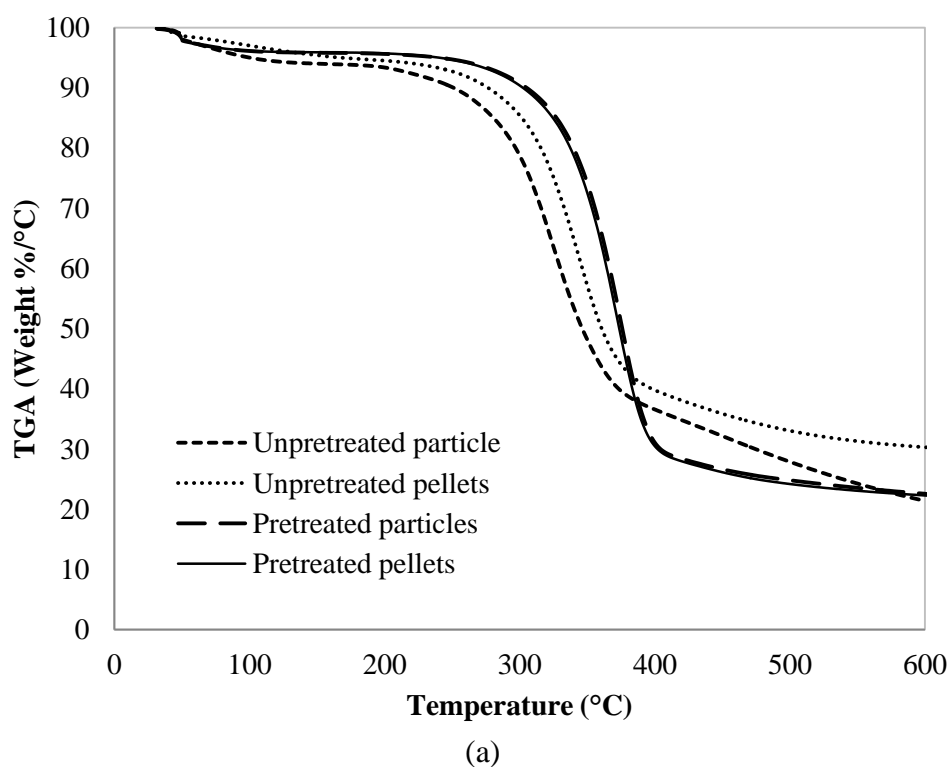
**Figure 12.7 Results on CrI values**



### 12.3.4 Results on thermogravimetric analysis

Results on TGA and derivative thermogravimetric (DTG) analysis for corn stover and sorghum stalk are displayed in Figures 12.8 and 12.9. It can be seen from Figures 12.8(a) and 12.9(a) that weight loss of samples that occurred at 30-280°C was less than 10% of the total sample weight. As heating temperature increased from 280 to 400°C, the samples had a remarkable weight loss of up to 70%. The remains of the samples were ashes. There was no evident difference in TGA results between particles and pellets after pretreatment. However, before pretreatment, there was obvious difference between them especially for corn stover.

**Figure 12.8 Distribution of (a) TGA and (b) DTG of corn stover samples**



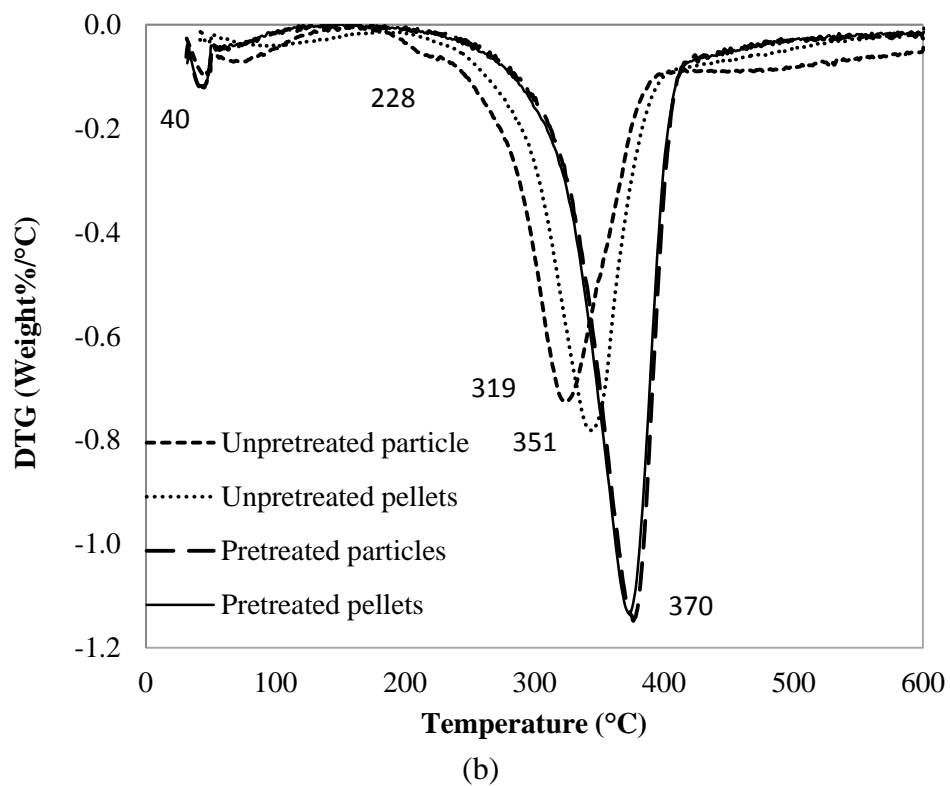
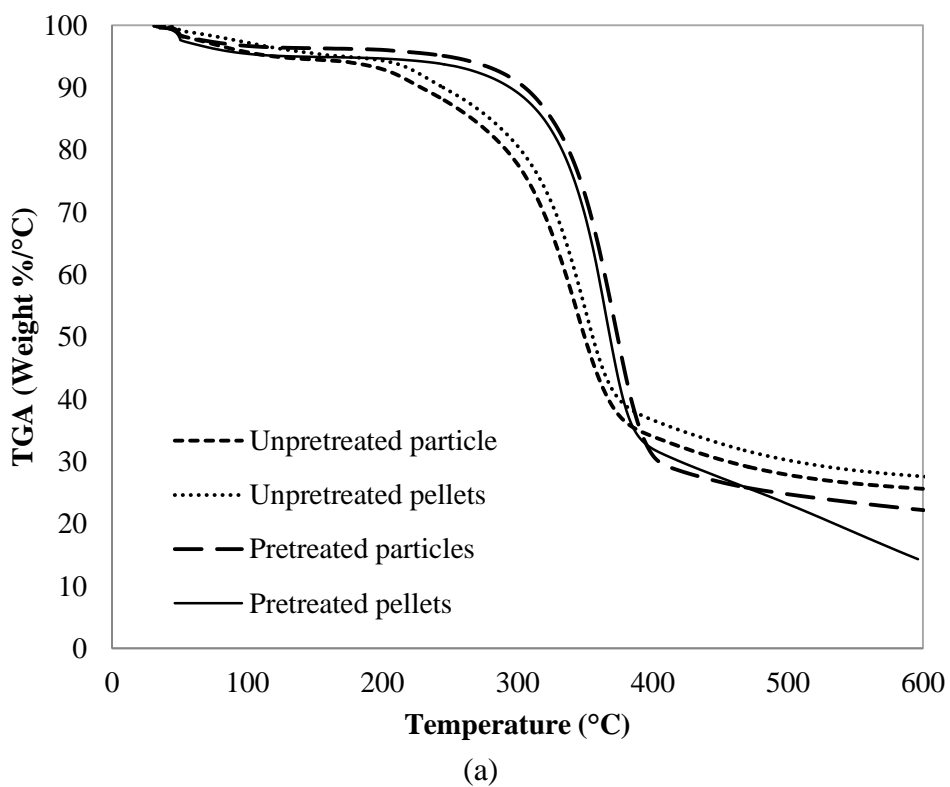
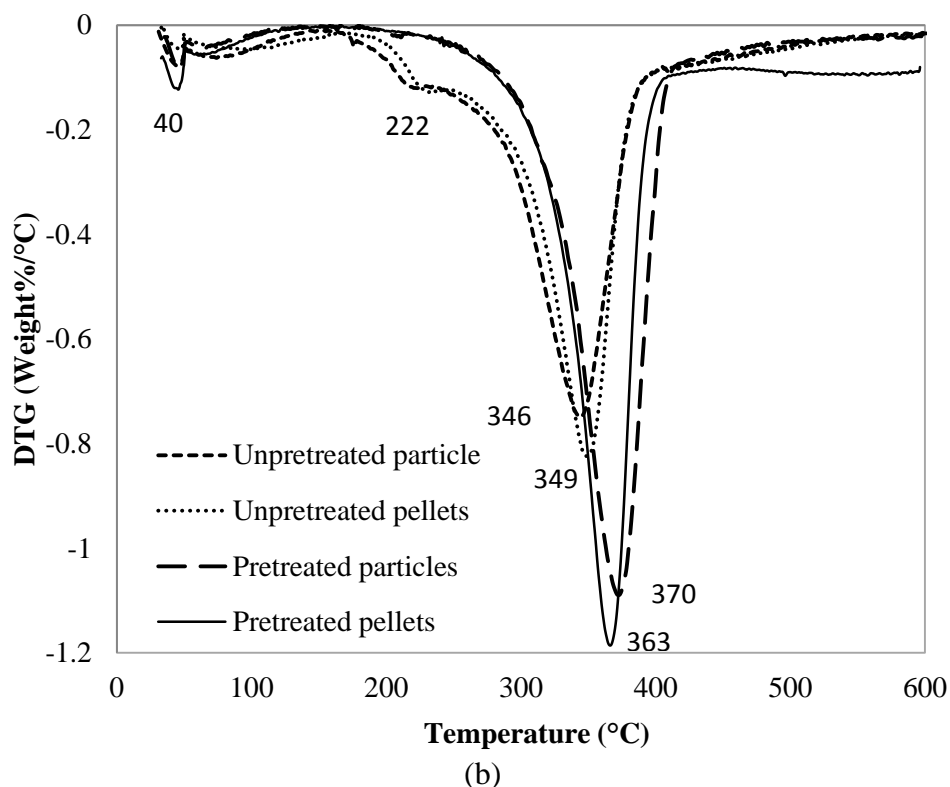


Figure 12.9 Distribution of (a) TGA and (b) DTG of sorghum stalk samples





The DTG (first derivative) profiles for the rate of weight loss of samples are displayed in Figures 12.8(b) and 12.9(b). The DTG curves of unpretreated biomass exhibit three peaks. The first peak appeared at the temperature around 30-85°C. It represented elimination of absorbed or combined water in the sample [34]. The second peak appeared at 222-228°C, which was attributed to decomposition of hemicellulose [35]. The third peak appeared at 319-351 °C, which was attributed to decomposition of cellulose [23]. Lignin decomposed at over 200-500 °C and decomposition of lignin did not form a peak because the various oxygen functional groups generated during lignin decomposition had different thermal stabilities [36]. For pretreated biomass, only two peaks were observed. The peak at 222-228°C disappeared, indicating that hemicellulose was removed during pretreatment.

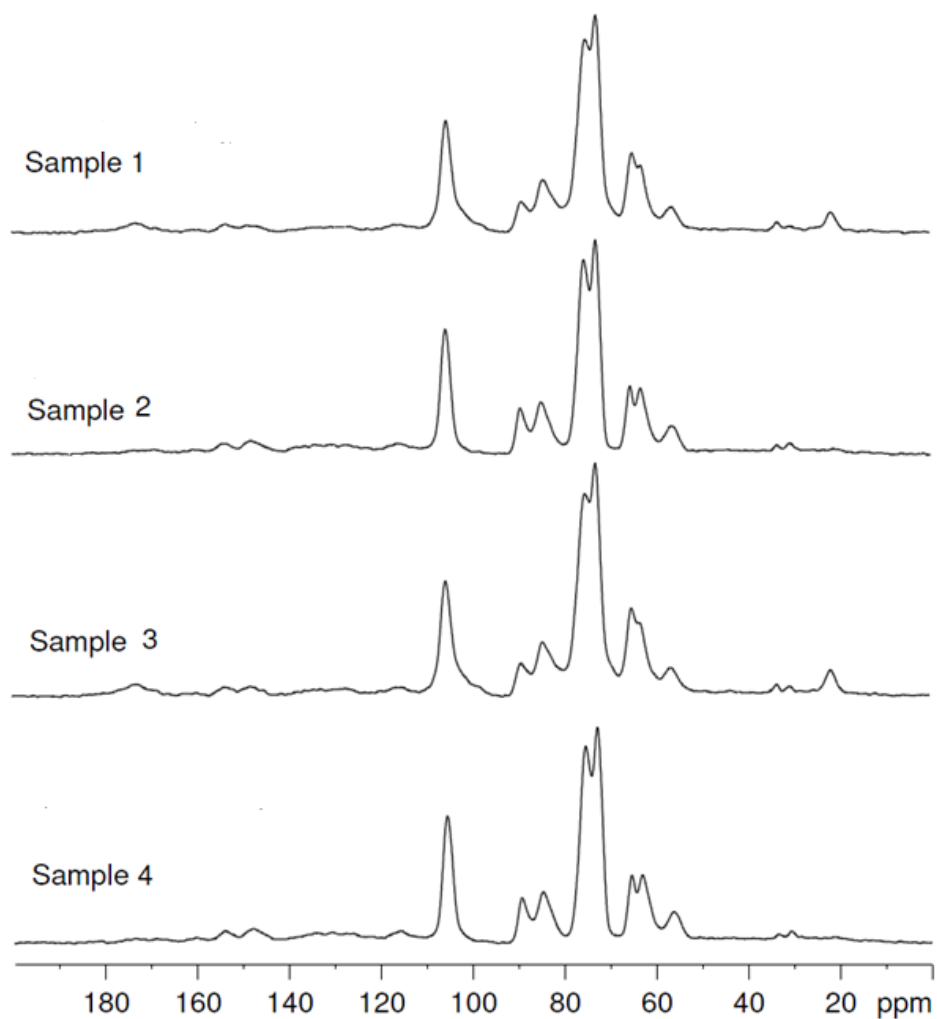
### 12.3.5 Solid-State $^{13}\text{C}$ NMR Analysis

Similar to FTIR analysis, the solid-state  $^{13}\text{C}$  NMR analysis is a qualitative technique used to determine the chemical structure and composition of biomass samples. Figures 12.10 and

12.11 show the  $^{13}\text{C}$  NMR spectra of biomass particles and pellets before and after pretreatment. Table 12.3 summarized chemical shift assignments of the biomass spectra reported in the literature [28,37-39].

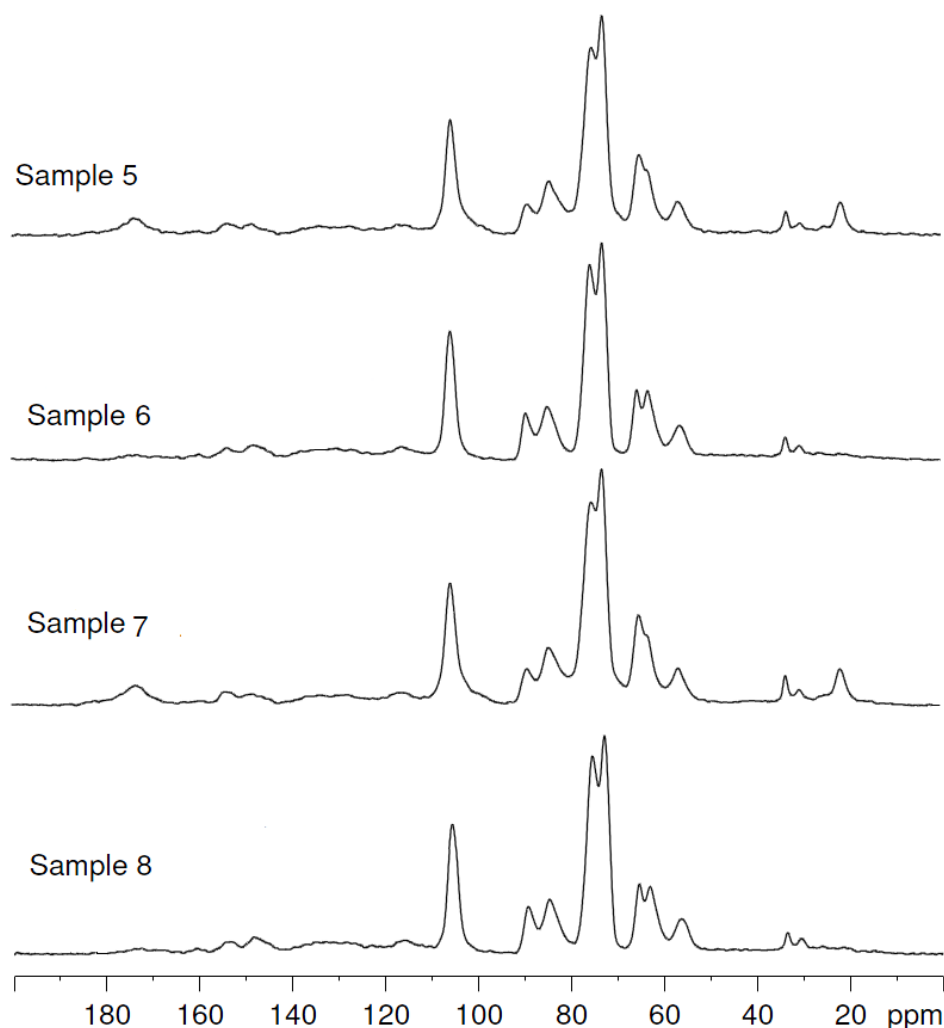
Signals appeared in the region of 50-105 ppm were assigned to various cellulosic carbons [28]. The sharp signals at 65.5, 89.2-93, and 105.4 ppm were assigned to C-6, C-4, and C-1 carbons of crystalline cellulose, respectively. The shape signals at 63 and 84 ppm were assigned to C-6, and C-4 carbons of amorphous cellulose, respectively. The signals at 72.9-75.2 ppm were assigned to C-2, C-3, and C-5 carbons of cellulose. Base on Figure 12.10, a comparison between samples 1 (corn stover particles) and 3 (corn stover pellets) reveals that there was no significant difference in the aforementioned cellulose-related signals. This indicates that UV-A pelleting does not have strong effects on either amorphous or crystalline cellulose. The same trend was found for sorghum stalk (Figure 12.11). However, signals at 56, 63, 75, and 89 ppm for both corn stover and sorghum stalk increased after pretreatment, indicating that pretreatment altered the cellulose structure of biomass.

**Figure 12.10 NMR spectra of corn stover (sample 1 – particles, sample 2 – pretreated particle, sample 3 – pellets, and sample 4 – pretreated pellets)**





**Figure 12.11 NMR spectra of sorghum stalk (sample 1 – particles, sample 2 – pretreated particles, sample 3 – pellets, and sample 4 – pretreated pellets)**



Signals at 23-25 and 172 ppm were assigned to methyl and carboxylic groups of the acetyl function of hemicellulose [28]. For both corn stover and sorghum stalk, there was no apparent difference in the intensity of these two signals between particles and pellets (e.g., by comparing samples 1 and 3 in Figure 12.10), indicating that UV-A pelleting did not remove hemicellulose. This result is consistent with the composition data in Table 12.1. By comparing samples 1 and 3 with samples 2 and 4 in Figures 12.10 and 12.11, it can be seen that the intensity of the two signals disappeared after pretreatment. This evidence was the removal of

hemicellulose during pretreatment. The results were consistent with the compositional analysis data in Table 12.1 and FTIR spectra in Figure 12.3.

**Table 12.3 Signal assignments for  $^{13}\text{C}$  CPMAS spectra of biomass**

Chemical Shift (ppm)	Types of carbons*
172.5-173.8	Carboxyl groups(COOH) in acetyl groups of hemicellulose
153.1-153.9	S3 (e), S5 (e), G4 (e) in lignin
147.8-148.1	S3 (ne),S5 (ne), G3 in lignin
136.5-137.1	S1 (e), S4 (e), G1 (e) in lignin
133.8-134.1	S1 (ne), S4 (ne), G1 (ne) in lignin
122.1-123.2	G6 in lignin
115.6-116.7	G5 in lignin
113.8	G2 in lignin
105.4	C-1 of cellulose
89.2-93	C-4 of crystalline cellulose
84.2-84.6	C-4 of amorphous cellulose
72.9-75.2	C-2 ,C-3, and C-5 of cellulose
65.5	C-6 of crystalline cellulose
63-63.3	C-6 of amorphous cellulose
56.1-56.7	Methoxyl groups(-OCH <sub>3</sub> ) in lignin
23.2-25.2	Methyl (CH <sub>3</sub> ) in acetyl groups of hemicellulose

\* S – syringyl, G – Guaiacyl, ne – nonetherified, e – etherified

Signals for aromatic carbons of lignin in the  $^{13}\text{C}$  NMR spectra were shown in a region of 110-155 ppm [28]. Guaiacyl (G) and Syringyl (S) are main components in lignin. Base on Table 12.3, G was identified by signals at 153 (C-4, G etherified), 147-148 (C-3, G etherified), 136-137 (C-1, G etherified), 133-134 (C-1, G nonetherified), 122-123 (C-6, G), 115-117 (C5, G), and 113.8 ppm (C-2, G) [28,29,37]. S was assigned by signals at 153-154 (C-3/C-5, S etherified),

147-148 (C-3/C-5, S nonetherified), 136-137 (C-1/C-4, S etherified), and 133-134 ppm (C-1/C-4, S nonetherified) [28,37,38]. Besides G and S, the signal at 56 ppm was assigned to methoxyl carbons in lignin [39]. The intensity of signals at 56, 147, and 153 ppm increased after pretreatment, as can be seen by comparing samples 2 and 4 in Figures 12.10 and 12.11. It indicates that lignin content in biomass increased after pretreatment. However, there was no significant difference in intensities of lignin signals before and after UV-A pelleting.

### ***12.3.6 Morphological structure analysis***

In the SEM images of corn stover particles before pretreatment (Figure 12.12(a)), samples exhibited regular and compact surface structure. After pelleting, soften surface region of biomass was removed, revealing cellulose microfibrils with 2.3-3.4  $\mu\text{m}$  in width, as shown in Figure 12.12(b).

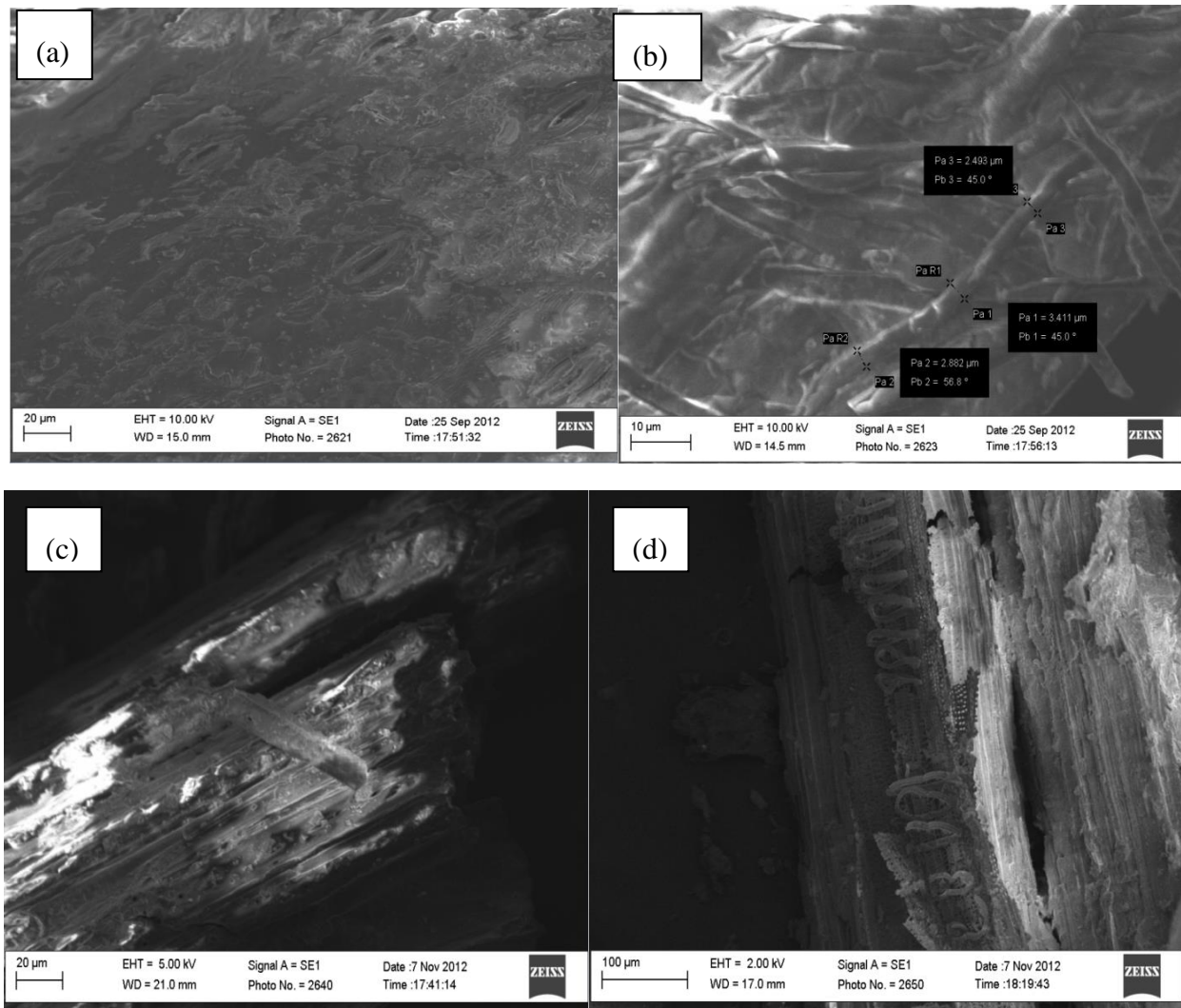
After pretreatment, microfibrils in corn stover were observed and its surface was clean and smooth, as shown in Figure 12.12(c). Although dilute-acid pretreatment disrupted the biomass network and removed hemicellulose, major microfibrinous cellulose structures were still preserved. Surface of cellulose fibers was covered by lignin or lignin carbohydrate complexes [28,40]. Furthermore, annular rings, which are parts of the biomass internal structure, were revealed in pelleted biomass after pretreatment, as shown in Figure 12.12(d).

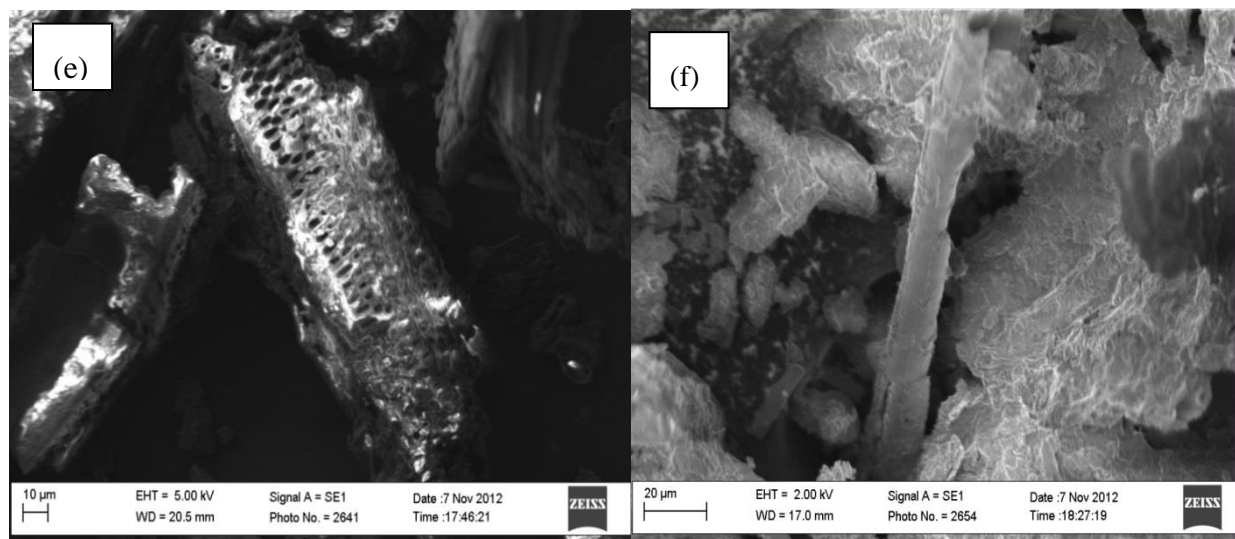
The SEM images of residues of corn stover after enzymatic hydrolysis were shown in Figures 12.12(e) and 12.12(f). Empty holes appeared on the surface of biomass residue (Figure 12.12(e)), indicating that cellulose was digested by enzyme. In addition, cellulose fibrils appeared after enzymatic hydrolysis, as shown in Figure 12.12(f).

Other than removal of soften surface region of biomass, there was no significant difference in the morphological structure between particles and pellets after pretreatment and hydrolysis. Lamsal et al. [15] reported that the increased sugar yield caused by screw extrusion was attributed to continuous removal of soften biomass surface and exposure of the interior of biomass to chemical and thermal actions. From the SEM images obtained in this study, removal

of soften biomass surface was also found in UV-A pelleting. Therefore, this might be the major reason responsible to the increased sugar yield caused by UV-A pelleting.

**Figure 12.12 SEM images of corn stover: (a) outer surface of untreated particles, (b) outer surface of untreated pellets, (c) cellulose bundles of pretreated pellets, (d) annular rings of pretreated pellets, (e) pellets after hydrolysis, and (f) microfibrils of pellet after hydrolysis**





## 12.4 Conclusions

This paper reports investigations on characteristics of corn stover and sorghum stalk processed by ultrasonic vibration-assisted (UV-A) pelleting. Effects of UV-A pelleting on sugar yield, chemical composition, crystallinity index, chemical structure, and thermal properties of corn stover and sorghum stalk were evaluated. Major conclusions are:

1. UV-A pelleting can increase sugar yield of cellulosic biomass in enzymatic hydrolysis by more than 10%. However, it does not change chemical composition of cellulosic biomass regardless of application of dilute acid pretreatment.
2. Crystallinity index of biomass processed by UV-A pelleting is significantly higher than that of unpelleted biomass. The increased biomass crystallinity index was probably due to the crystallization of amorphous cellulose in biomass and changes in lignin structure when the biomass was subjected to mechanical shear and heat generated in UV-A pelleting. In addition, dilute acid pretreatment significantly increases the crystallinity index of biomass by removing amorphous hemicellulose.
3. Results on FTIR and solid-state  $^{13}\text{C}$  NMR analyses indicate that there is no significant difference in biomass structure between particles and pellets. However, dilute acid pretreatment could alter the chemical structure of biomass due to degradation of hemicellulose.

4. The decomposition temperature of pellets is slightly higher than that of particles. In other words, pellets produced by UV-A pelleting are more thermally stable than particles.
5. Morphological structure of biomass is affected by UV-A pelleting. During UV-A pelleting, softened surface region of biomass is removed, revealing cellulose microfibrils. This could be a reason responsible to increased sugar yield caused by UV-A pelleting.

### **Acknowledgements**

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## Chapter 13 - Conclusions

### 13.1 Summaries and conclusions of this dissertation

In this dissertation, ultrasonic vibration-assisted (UV-A) pelleting of cellulosic biomass materials for ethanol manufacturing is investigated. Effects of input variables (such as biomass moisture content (MC), biomass particle size, biomass type, pelleting pressure, and ultrasonic vibration power) on pellet quality (such as density, durability, and stability), power consumption, and sugar yield are studied. Furthermore, pellets produced by UV-A pelleting are compared with those produced by ring-die pelleting in terms of pellet quality (pellet density and durability), power consumption, and sugar yield under different combination of pretreatment conditions (such as particle size, pretreatment acid concentration, solid content, pretreatment temperature, and pretreatment time). Temperature of biomass in UV-A pelleting is investigated. Effects of UV-A pelleting on biomass characteristics (such as chemical composition, crystallinity index, thermal properties, and morphological structure) are investigated. Table 13.2 presents areas in UV-A pelleting studied in this dissertation.

**Table 13.1 Areas of UV-A pelleting covered in this dissertation**

	Particle size	Biomass type	MC	Pressure	Ultrasonic power	Pellet weight
Temperature	✓		✓	✓	✓	✓
Power consumption	✓	✓	✓	✓	✓	
Sugar yield				✓	✓	
Pellet stability	✓			✓	✓	
Pellet durability	✓			✓	✓	
Pellet density	✓			✓	✓	

Below are the main conclusions drawn from this dissertation:

1. Particle size, pelleting pressure and ultrasonic power have significant effects on pellet properties (such as density, durability, and stability) and sugar yield. Smaller particle size, higher pressure, and higher ultrasonic power tend to produce higher density and durability. In some conditions, mixed-size particles can produce higher density than one-size particles. Smaller particle size and higher ultrasonic power result in higher stability while pressure have little effects on pellet stability. The stability of the pellets with mixed-size particles are between those of the pellets with one-size particles. Under certain experimental conditions, sugar yield increases with increasing pressure and ultrasonic power.
2. The five input variables (biomass type, biomass moisture content, biomass particle size, pelleting pressure, and ultrasonic power) investigated have significant effects on power consumption in UV-A pelleting. Sorghum stalk requires less power than corn stover, big bluestem, and wheat straw. As moisture content increases from 15% to 25%, the power consumption increases dramatically. As particle size increases, power consumption increases but power consumption rate decreases. As pressure increases, power consumption decreases but there is no significant change on power consumption rate. As ultrasonic power increases, power consumption decreases but power consumption rate increases. Only one two-factor interaction is significant, i.e., with the increase of pelleting pressure, power consumption will increase at the high level of particle size while decreases at the low level of particle size.
3. Comparisons are made between ring-die pelleting and UV-A pelleting in terms of pellet quality (such as density and durability), power consumption, and sugar yield under different combinations of pretreatment variables (such as particle size, acid concentration, solid content, pretreatment temperature, pretreatment time). Both pelleting methods could significantly increase pellet durability and density. Pellets

produced by UV-A pelleting have higher durability than those produced by ring-die pelleting. To produce same amount of pellets, ring-die pelleting consumed less power than UV-A pelleting. Wheat straw processed by UV-A pelleting has higher sugar yield than that processed by ring-die pelleting when the higher temperature and longer time are applied in pretreatment. The sugar yield of UV-A pellets are lower than those of ring-die pellets at same levels of acid concentration and solid content. There is no significant difference in sugar yield between two pelleting methods when small particles (3.2 mm) are used to make pellets. However, sugar yield of UV-A pellets is lower than that of ring-die pellets at large particle sizes (9.5 mm).

4. A good precision in temperature measurement could be achieved using the experimental setup adopted in this dissertation. The obtained temperature-time curves in UV-A pelleting could be classified into three groups based on their shapes. The curve shape depended on both measurement location and pelleting condition. For each combination of measurement location and pelleting condition, pelleting temperature became stable during the pelleting time (180 seconds) in this study. In UV-A pelleting, the highest pelleting temperature always occurred at the middle center (the core) of a pellet. The lowest pelleting temperature always occurred at the bottom side of a pellet. The distribution of pelleting temperature (presented by the rank of pelleting temperatures at six locations) was affected by pelleting condition. Pelleting temperature was more evenly distributed in a pellet (there was no significant difference in pelleting temperature between top center, bottom center, top side, and middle side) when a high level of pelleting pressure was used. The highest and lowest pelleting temperatures in UV-A pelleting were significantly affected by two main effects (ultrasonic power and pellet weight) and one three-factor interaction effect. One two-factor interaction effect (between ultrasonic power and pellet weight) was significant on the highest pelleting temperature while two two-factor interaction

- effects (between ultrasonic power and pellet weight, and between pelleting pressure and pellet weight) were significant on the lowest pelleting temperature.
5. Without dilute acid pretreatment, there was no significant difference in the chemical composition between pellets processed by UV-A pelleting and particles. With dilute acid pretreatment, the content of acid soluble lignin (ASL) in pelleted biomass was significantly higher than that in particles. There was no significant difference in the other components and extractives between particles and pellets. For both corn stover and sorghum stalk, the cellulose recovery of pellets was significantly higher than that of particles after dilute acid pretreatment. UV-A pelleting could significantly increase the sugar yield in enzymatic hydrolysis for both corn stover and sorghum stalk. Without dilute acid pretreatment, the sugar yield of pellets was more than 50% higher than that of particles. With dilute acid pretreatment, the sugar yield of pellets was more than 10% higher than that of particles. The combination of UV-A pelleting and dilute acid pretreatment could achieve the sugar yield of 93.1% for corn stover and 92.8% for sorghum stalk.
  6. Crystallinity index of pellets processed by UV-A pelleting is significantly higher than that of particles. The increased biomass crystallinity index was probably due to the crystallization of amorphous cellulose in biomass and changes in lignin structure when the biomass was subjected to mechanical shear and heat generated in UV-A pelleting. In addition, dilute acid pretreatment significantly increases the crystallinity index of biomass by removing amorphous hemicellulose. Results on FTIR and solid-state  $^{13}\text{C}$  NMR analyses indicate that there is no significant difference in biomass structure between particles and pellets. However, dilute acid pretreatment could alter the chemical structure of biomass due to degradation of hemicellulose. The decomposition temperature of pellets is slightly higher than that of particles. In other words, pellets produced by UV-A pelleting are more thermally stable than particles.

Morphological structure of biomass is affected by UV-A pelleting. During UV-A pelleting, softened surface region of biomass is removed, revealing cellulose microfibrils. This could be a reason responsible to increased sugar yield caused by UV-A pelleting.

### **13.2 Contributions of this dissertation**

The major contributions of this dissertation are:

1. This research is the first to systematic investigate input parameters (particle size, pelleting pressure, and ultrasonic power) on pellet properties (such as density, durability, and stability). This result will add to the literature of pelleting. In addition, this result is of practical use in pelleting industry for improving pellet quality.
2. This dissertation, for the first time, presents a systematic investigation on power consumption in ultrasonic vibration-assisted (UV-A) pelleting of cellulosic biomass. This research will fill gaps in the literature on cellulosic ethanol manufacturing.
3. This research is the first to make comparisons of pellets produced by UV-A pelleting and those produced by ring-die pelleting (a traditional pelleting method) in terms of pellet quality, power consumption, and sugar yield. This research will add to the literature of both pelleting methods.
4. This research is the first to systematically study temperature of biomass in UV-A pelleting. The result will add to the literature of temperature study in pelleting cellulosic biomass.
5. This research is the first to study effects of UV-A pelleting on biomass characteristics (such as chemical composition, crystallinity index, and thermal properties). Such investigations are essential to explain the mechanisms through which UV-A pelleting increases biomass sugar yield. The results will add to the literature of cellulosic ethanol manufacturing.

## **Appendix A – Publications during Ph.D. study**

### **Publications**

1. Zhang, P.F., Zhang, Q., Pei, Z.J., and Wang, D.H., 2013, “Cost estimates of cellulosic ethanol production: a review,” accepted to appear in Journal of Manufacturing Science and Engineering.
2. Zhang, Q., Zhang, P.F., Pei, Z.J., and Wang, D.H., 2013, “Ultrasonic vibration-assisted pelleting for cellulosic biofuel manufacturing: investigation on power consumption,” Renewable Energy, Vol. 55, pp. 775-781.
3. Zhang, P.F., Zhang, Q., Deines, T.W., Pei, Z.J., and Wang, D.H., 2012, “Ultrasonic vibration-assisted pelleting of wheat straw: a designed experimental investigation on pellet quality and sugar yield,” Journal of Manufacturing Science and Engineering, Vol. 134, No. 6, pp. 061013-1 – 061013-10.
4. Zhang, Q., Zhang, P.F., Chang, S., Pei, Z.J., and Wang, D.H., 2013, “Application of multiple response surface methodology for optimization of cellulosic ethanol manufacturing by ultrasonic vibration-assisted pelleting,” accepted to appear in Proceedings of the ASME 2013 International Manufacturing Science and Engineering Conference (MSEC), Madison, WI, June 10-14, MSEC 2013-1044.
5. Kennedy, E., Zhang, P.F., Zhang, Q., Pei, Z.J., and Wang, D.H., 2013, “Effects of ultrasonic vibration-assisted pelleting of biomass on sugar yield in biofuel manufacturing with different pretreatment methods,” accepted to appear in Proceedings of the ASME 2013 International Manufacturing Science and Engineering Conference (MSEC), Madison, WI, June 10-14, MSEC 2013-1044.
6. Zhang, Q., Zhang, P.F., Pritchett, G., Pei, Z.J., Zhang, M., Song, X., and Deines, T.W., 2012, “Ultrasonic-vibration assisted pelleting for cellulosic ethanol manufacturing: effects of particle size and moisture content on power consumption,” Proceedings of the



- ASME 2012 International Manufacturing Science and Engineering Conference (MSEC), Notre Dame, IN, June 4-8, MSEC 2012-7211.
7. Zhang, Q., Zhang, P.F., Pritchett, G., Pei, Z.J., Zhang, M., Song, X., and Deines, T.W., 2012, "Ultrasonic-vibration assisted pelleting for cellulosic biofuel manufacturing: investigation on power consumption with design of experiment," Proceedings of the ASME 2012 International Manufacturing Science and Engineering Conference (MSEC), Notre Dame, IN, June 4-8, 2012, MSEC 2012-7212.
  8. Wu, H., Zhang, P.F., Zhang, Q., and Pei, Z.J., 2012, "Effects of water soaking on biomass particle size in cellulosic biofuel manufacturing," Proceedings of the ASME 2012 International Manufacturing Science and Engineering Conference (MSEC), Notre Dame, IN, June 4-8, MSEC 2012-7227.
  9. Zhang, Q., Zhang, P.F., Zhang, Song, X., Pei, Z.J., and Siddiqui, O., 2012, "Sugar yield comparison of wheat straw processed by two pelleting methods for cellulosic biofuel manufacturing," Proceedings of the ASME 2012 International Manufacturing Science and Engineering Conference (MSEC), Notre Dame, IN, June 4-8, MSEC 2012-7228.
  10. Zhang, Q., Zhang, P.F., Pei, Z.J., Wilson, J., McKinney, L., and Prichett, G., 2011, "An experimental comparison of two pelleting methods for cellulosic ethanol manufacturing," Proceedings of the 2011 International Manufacturing Science and Engineering (MSEC) Conference, Corvallis, OR, US, June 13-17, MSEC 2011-50215.
  11. Zhang, Q., Zhang, P.F., Song, X.X., Zhang, M., Pei, Z.J., and Deines, T., 2011, "A study on amount of biomass pellets used in durability testing," Proceedings of the 2011 International Manufacturing Science and Engineering (MSEC) Conference, Corvallis, OR, US, June 13-17, MSEC 2011-50025.
  12. Zhang, Q., Zhang, P.F., Pei, Z.J., and Pei, L., 2011, "Effects of treatments on cellulosic biomass structure in ethanol manufacturing: a literature review," Proceedings of the 2011

- International Mechanical Engineering Congress and Exposition, Denver, CO, US, November 11-17, IMECE 2011-64304.
13. Zhang, Q., Zhang, P.F., Pei, Z.J., Pritchett, G., Wilson, J., and McKinney, L., 2011, "Ultrasonic vibration assisted pelleting for cellulosic ethanol manufacturing: an experimental investigation of power consumption," Proceedings of the 2011 International Mechanical Engineering Congress and Exposition, Denver, CO, US, November 11-17, IMECE 2011-64307.
  14. Zhang, Q., Zhang, P.F., Deines, T., Zhang, M., Song, X.X., and Pei, Z.J., 2010, "Ultrasonic vibration assisted pelleting of wheat straw: effects of particle size," Proceedings of the 2010 Flexible Automation and Intelligent Manufacturing International Conference, Hayward, CA, July 12-14.
  15. Zhang, Q., Zhang, P.F., and Pei, Z.J., 2010, "Effects of particle size on cellulosic biomass pellets: a literature review," Proceedings of the ASME International Conference on Manufacturing Science and Engineering (MSEC), Erie, PA, October 12-15.
  16. Zhang, Q., Zhang, P.F., Deines, T., Pei, Z.J., Wang, D.H., Wu, X.R., and Pritchett, G., 2010, "Ultrasonic vibration assisted pelleting of sorghum: effects of pressure and ultrasonic power level," Proceedings of the ASME International Conference on Manufacturing Science and Engineering (MSEC), Erie, PA, October 12-15.
  17. Zhang, P.F., Zhang, Q., Pei, Z.J., and Pei, L., 2011, "An experimental investigation on cellulosic biofuel manufacturing: effects of biomass particle size on sugar yield," Proceedings of the 2011 International Mechanical Engineering Congress and Exposition, Denver, CO, US, November 11-17.
  18. Zhang, P.F., Zhang, Q., Pei, Z.J., and Pei, L., 2011, "Ultrasonic vibration-assisted pelleting in manufacturing of cellulosic biofuels: an investigation on biomass particle size," Proceedings of the 2011 International Mechanical Engineering Congress and Exposition, Denver, CO, US, November 11-17.

19. Zhang, P.F., Zhang, M., Song, X.X., Zhang, Q., Cong, W.L., Qin, N., Pei, Z.J., Deines, T., and Wang, D.H., 2011, "Ultrasonic vibration-assisted pelleting of cellulosic biomass for biofuel manufacturing," CD-ROM Proceedings of 2011 NSF Civil, Mechanical and Manufacturing Innovation Grantees and Research Conference, Atlanta, GA, US, January 4-7.
20. Song, X.X., Zhang, M., Pei, Z.J., Deines, T., Zhang, Q., Zhang, P.F., and Wang, D.H., 2011, "Size reduction of poplar wood using a lathe for biofuel manufacturing: a preliminary experiment," Proceedings of the 2011 International Mechanical Engineering Congress and Exposition, Denver, CO, US, November 11-17.
21. Song, X.X., Zhang, M., Deines, T., Zhang, P.F., Zhang, Q., and Pei, Z.J., 2010, "Ultrasonic-vibration-assisted pelleting of wheat straw: effects of moisture content," Proceedings of the Industrial Engineering Research Conference, Cancun, Mexico, June 5-9.
22. Zhang, M., Song, X.X., Zhang, P.F., Zhang, Q., Pei, Z.J., Deines, T., and Wang, D.H., 2011, "Size reduction of cellulosic biomass in biofuel manufacturing: effects of milling orientation on sugar yield," Proceedings of the 2011 International Manufacturing Science and Engineering (MSEC) Conference, Corvallis, OR, US, June 13-17.
23. Zhang, M., Song, X.X., Deines, T.W., Zhang, P.F., Zhang, Q., Cong, W.L., Qin, N., and Pei, Z.J., 2010, "Ultrasonic-vibration-assisted pelleting of switchgrass: effects of binder material," Proceedings of the IIE Annual Conference and Expo 2010 – Cancun, Mexico, June 5 – 9, 2010.
24. Zhang, M., Song, X.X., Deines, T., Zhang, P.F., Zhang, Q., Cong, W.L., Qin, N., and Pei, Z.J., 2010, "Ultrasonic-vibration-assisted pelleting of switchgrass: effects of binder material," Proceedings of the Industrial Engineering Research Conference, Cancun, Mexico, June 5-9. 37.

### **Submitted papers**

1. Zhang, Q., Zhang, P.F., Pei, Z.J., and Wang, D.H., “Effects of cellulosic biomass particle size on enzymatic hydrolysis sugar yield: analysis of inconsistent results in the literature, accepted by Journal of Renewable Energy.
2. Zhang, P.F., Zhang, Q., Pei, Z.J., and Wang, D.H., Comparison of two pelleting methods for cellulosic ethanol manufacturing: ultrasonic vibration-assisted pelleting vs. ring-die pelleting, submitted to Journal of Renewable Energy.

### **Working papers**

1. Zhang, Q., Zhang, P.F., Pei, Z.J., and Wang, D.H., “Ultrasonic vibration-assisted pelleting of cellulosic biomass for ethanol manufacturing: an investigation on pelleting temperature,” to be submitted to Journal of Renewable Energy.
2. Zhang, Q., Zhang, P.F., Pei, Z.J., Praveen, V., Xu, F., and Wang, D.H., “Effects of ultrasonic vibration-assisted pelleting on chemical composition and sugar yield of corn stover and sorghum stalk,” to be submitted to Journal of Renewable Energy.
3. Zhang, Q., Zhang, P.F., Pei, Z.J., and Wang, D.H., “Investigation on characteristics of corn stover and sorghum stalk processed by ultrasonic-vibration assisted pelleting for sugar production,” to be submitted to Journal of Renewable Energy.

### **Posters**

1. Zhang, P.F., Feng, Q., Zinke, E., Cong, W.L., Zhang, M., Song, X.X., Zhang, Q., Pei, Z.J., and Wang, D.H., 2010, “Ultrasonic vibration-assisted pelleting of cellulosic biomass

- for bioethanol manufacturing,” Poster section of the ASME International Conference on Manufacturing Science and Engineering (MSEC), Erie, PA, October 12-15.
2. Zhang, M., Song, X.X., Zhang, P.F., Zhang, Q., Jones, E., Deines, T.W., Pei, Z.J., and Wang, D.H., 2010, “Fundamental study on size reduction of poplar for biofuel manufacturing,” Poster section at the ASME 2010 International Manufacturing Science and Engineering Conference (MSEC), Erie, PA, October 12-15, 2010, MSEC 2010-34352.
  3. Cong, W.L., Zhang, P.F., Qin, N., Zhang, M., Song, X.X., Zhang, Q., Nottingham, D., Clark, R., Denies, T., Pei, Z.J., and Wang, D.H., 2009, “Ultrasonic-vibration-assisted pelleting of cellulosic biomass for biofuel manufacturing,” Poster section of the ASME International Conference on Manufacturing Science and Engineering (MSEC), West Lafayette, IN, October 4-7.