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The Effect of Urea Pretreatment Combined with Ultrasonic Vibration-Assisted Pelleting on Pellet Solid Density and Durability

Wentao Li [1,2,*], Jianning Sui [1], Liming Wang [1] and Jindong Song [3]

[1] College of Engineering, Heilongjiang Bayi Agricultural University, Daqing 163319, China
[2] Department of Biological and Agricultural Engineering, Kansas State University, Manhattan, KS 66506, USA
[3] Heilongjiang Boneng Ecological Environmental Protection Co., Ltd., Harbin 150028, China
* Correspondence: yuesetingquan@byau.edu.cn; Tel.: +86-0459-6819-216

Abstract: Pellets, as a clean and renewable energy source, can overcome the shortcomings of high moisture content, expensive transportation costs, and non-uniform sizes for agricultural residues. However, traditional pelleting methods are associated with high energy consumption and severe mold abrasion due to the application of high temperature and pressure. To address these issues while improving pellet solid density and durability, urea pretreatment combined with ultrasonic vibration-assisted (UV-A) pelleting is investigated in the present research. Comparative experiments were initially conducted to verify the feasibility of the approach, followed by a central composite rotatable design (CCRD) to investigate the relative contributions and interactions of tested variables on pellet solid density and durability during pretreatment. The results revealed that combining urea pretreatment with UV-A pelleting could enhance pellet solid density and durability. Urea content, temperature, waste soybean flour (WSF) content, and distilled water significantly impacted pellet solid density, and all variables except distilled water had a significant effect on pellet durability. The optimal conditions were determined and a subsequent experiment was conducted to verify the agreement between experimental data and predicted results. The optimal conditions consisted of 42% distilled water, temperature of 45 °C, 10% urea content, and 12% WSF content resulting in pellet solid density and durability values of 1438.28 kg m^{-3} and 98.67%, respectively.

Keywords: urea pretreatment; ultrasonic vibration-assisted pelleting; pellet solid density; pellet durability

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1. Introduction

Biomass resources are the sole renewable resource capable of participating in repository and logistics [1], with a total straw resource volume of 856 million tons and a collectible resource volume of 720 million tons in 2020 [2], indicating their significant potential for bioenergy utilization. It is projected that by 2050, approximately 77% of the global energy supply will be derived from renewable sources [3,4]. Under the context of carbon peak and carbon neutrality, the increasing demand for low-cost, clean, and renewable energy necessitates a comprehensive utilization of agricultural residues, which are associated with high costs in handling, transportation, and storage [5]. Pelleting, a process that compresses biomass into uniform pellets of consistent shape and size, has been shown to significantly increase the density of cellulosic biomass [6–8]. The solid density of biomass could reach 1200 kg/m^3 [9]. Traditional methods for pellet manufacturing include screw extrusion, piston press, and ring die techniques [10,11], which offer reduced costs for handling, transportation, and storage compared to raw biomass. However, pelleting requires high temperature and pressure as well as strict moisture content control [12].

To produce high-quality pellets, the temperature range used was 90–140 °C, with the corresponding moisture content of raw materials ranging from 6% to 14% and pressure levels between 90–110 MPa [13–15]. They were also highly energy-consuming processes

that led to severe abrasion of the mold. To reduce both abrasion and energy consumption, comminution and adhesives have traditionally been widely used. Comminution aims to increase the flow and filling characteristics of raw materials by crushing straw into fine particles, increasing specific surface area, and destroying crystalline structures [16,17]. With the addition of adhesives such as pea and soybean waste, pellet durability and combustion characteristics have been improved [18]. In addition to the aforementioned pretreatment methods, alkaline solution [19–22], acid solution [23,24], organic solvent [25], hydrothermal pretreatment [26], and steam explosion [27] are often used to separate lignin, hemicellulose, and cellulose. However, those methods mostly focus on the content of sugar or volatile fatty acids in the liquid phase for production of bioethanol or methane. Ultrasonic vibration-assisted (UV-A) pelleting employs a converter to transform the high-frequency oscillation signal from the ultrasonic generator into high-frequency mechanical vibration. The pelleting tool harnesses this energy to compress biomass, while avoiding the use of high temperature, pressure, and adhesives [28–30] that are typically associated with traditional pelleting methods.

The existing literature on UV-A pelleting has solely focused on the impact of various factors on pellet quality, while the potential effects of combining UV-A pelleting with chemical pretreatment for pellet production remain unclear. Urea pretreatment, as a mild alkaline treatment, can disrupt the chemical bonds between lignin and hemicellulose, promote carbohydrate liberation, increase contact area and crude protein content, as well as roughen and disintegrate cellulose fibers [3,31–33]. The aim of this study is to investigate the feasibility of combining urea pretreatment with ultrasonic vibration-assisted pelleting for producing pellets, and to determine the optimal conditions for achieving higher pellet solid density and durability.

2. Materials and Methods

2.1. Materials and Experimental Setup

The raw material used in this study was the whole corn stover from southern Kansas. Corn stover was milled using a cutting mill (model SM 2000, Retsch, Inc., Haan, Germany) into particles. Urea was purchased from Sigma Chemical Co. (St. Louis, MO, USA), and waste soybean flour (WSF) was from farmers market and milled into 0.5 mm flour.

Experimental setup for UV-A pelleting came from the Department of Industrial and Manufacturing Systems Engineering, Kansas State University. The machine (Figure 1) included a pneumatic loading system, ultrasonic generation system, and biomass holding system [8,29,34]. The ultrasonic power can be adjusted from 0 to 100%, and the pressure from air compressor was from 0 to 0.689 MPa. The diameter of pelleting tool and aluminum mold was 17.4 mm and 18.6 mm, respectively.

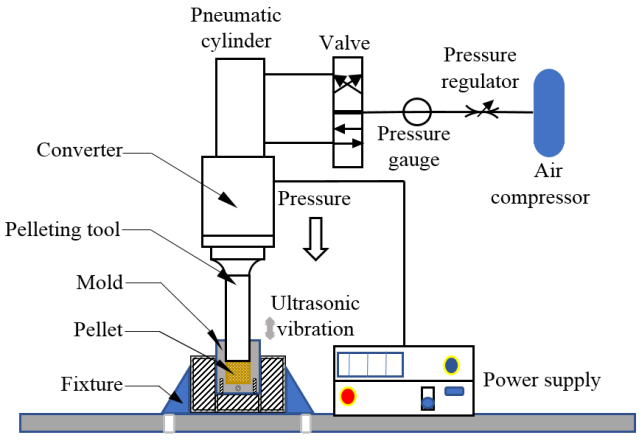

Figure 1. Ultrasonic-assisted molding device.

2.2. Experimental Design

First of all, corn stover was milled into five levels of particles (0~1.0 mm, 1.0~1.5 mm, 1.5~2.0 mm, 2.0~2.5 mm, and 2.5~3.0 mm). Then, all of the milled corn stover was divided into two parts: one was urea pretreatment and the other was without any pretreatment. Urea pretreatment was carried out in 500 mL Erlenmeyer flasks [35]. The milled corn stover, distilled water (40%, mass ratio to corn stover), urea (6%, mass ratio to corn stover), and WSF (6%, mass ratio to corn stover) were put into Erlenmeyer flask and mixed sufficiently by hand, which was then sealed by screw cap with polytetrafluoroethylene sealing tape. The sealed flasks were placed into biochemical incubator and maintained at 45 °C for 8 days. After pretreatment with urea, the materials were dried and adjusted to a moisture content of 10%.

To assess the efficacy of urea pretreatment on pellet quality, milled materials without urea pretreatment were subjected to UV-A pelleting alone (UV-A). Urea pretreatment materials combined with UV-A pelleting (UPUV-A) and urea pretreatment materials without UV-A pelleting (UP) at varying particle sizes were investigated. The resulting pellets were analyzed for density and durability as indicators. Each time, 1.5 g of particles were loaded into the mold and the titanium tool was used to compress the particles into pellets. All pellets were obtained at a moisture content of 10%, pressure of 0.276 MPa, pelleting time of 70 s, and ultrasonic power of 40 w if necessary. Based on the results from comparative experiments, a central composite rotatable design (CCRD) was carried out to investigate the relative contributions of tested variables and the interactions to pellet solid density and durability. The experimental variables and levels used in the study are listed in Table 1.

Table 1. The independent variables and levels for CCRD.

Level	Variables			
	Distilled Water (x_1) /%	Temperature (x_2) /°C	Urea Content (x_3) /%	WSF Content (x_4) /%
+2	60	65	12	12
+1	50	55	9	9
0	40	45	6	6
−1	30	35	3	3
−2	20	25	0	0

The experimental data were analyzed by Design-Expert 8.0.6.1, and three-dimensional (3D) surface graphs between variables and responses were obtained by RSM. The second-order polynomial of Equation (1) was used to fit the experimental data [36,37]. The optimization function in Design-Expert was applied to investigate the optimal conditions of maximizing density and durability of pellets.

$$y = \beta_0 + \sum_{i=1}^{k} \beta_i x_i + \sum_{i=1}^{k} \beta_{ii} {x_i}^2 + \sum \sum_{i<j=1}^{k} \beta_{ij} x_i x_j \quad (1)$$

where y values represent investigated responses (density and durability); β_0, β_i, β_{ii}, and β_{ij} indicate constant regression coefficients of intercept, linear, quadratic, and interaction terms, respectively; and x_i and x_j signify the independent coded variables.

2.3. Test Methods

The density of a pellet was calculated by the ratio of its mass over its volume. For pellet durability, five pieces of pellets were weighted and then put into a pellet durability tester (Seedburo Equipment Co., Des Plaines, IL, USA), which was kept tumbling under a rotation speed of 50 rpm [38]. Pellets were next taken out from the tester and sieved

through a No.6 U.S. sieve [29]. The weight of pellets retained by each sieve was recorded. Pellet durability was calculated using Equation (2):

$$DU = \frac{m_2}{m_1} \times 100 \quad (2)$$

where DU is the pellet durability, %; m_1 represents the initial weight, g; and m_2 is the weight of the remaining pellets, g.

3. Results and Discussion

3.1. Pellet Solid Density and Durability of Comparative Experiments

Pellet solid density and durability were compared among UV-A pelleting, UPUV-A pelleting, and UP pelleting with different particle sizes, as shown in Figure 2. The results showed that the pellet solid density of UPUV-A pelleting was significantly higher than that of other methods, ranging from 1385 kg/m^3 to 1443 kg/m^3. In contrast, the pellet solid density of UP pelleting was the lowest at approximately 1000 kg/m^3. Moreover, the pellet durability for UV-A pelleting ranged from 93% to 95%, which was lower than that for UPUV-A pelleting (>97%) but much higher than that for UP pelleting (<57%). The durability of the UP pellet failed to meet the standards of storage and transportation. The results indicated that ultrasonic assistance exerted a significant impact on the pelletization process. This was attributed to the high-frequency vertical vibration and thermal effect generated by the ultrasonic wave through the converter, which facilitated lignin softening via heat absorption in the corn stover. Additionally, urea pretreatment could effectively disrupt hydrogen bonds, leading to cleavage of both lignin and carbohydrate linkers [3,4] and destruction of the cellular structure. With the dissolution of lignin, hemicellulose, and cell soluble substances, the corn stover became more porous and less dense, which facilitated pelletization. In addition, the hydrophilic surface of pretreated corn stover enhanced Van der Waals forces between particles [39]. According to the experimental results, in order to further improve pellet quality and determine optimal process parameters, it was necessary to investigate the effect of urea pretreatment parameters on raw materials.

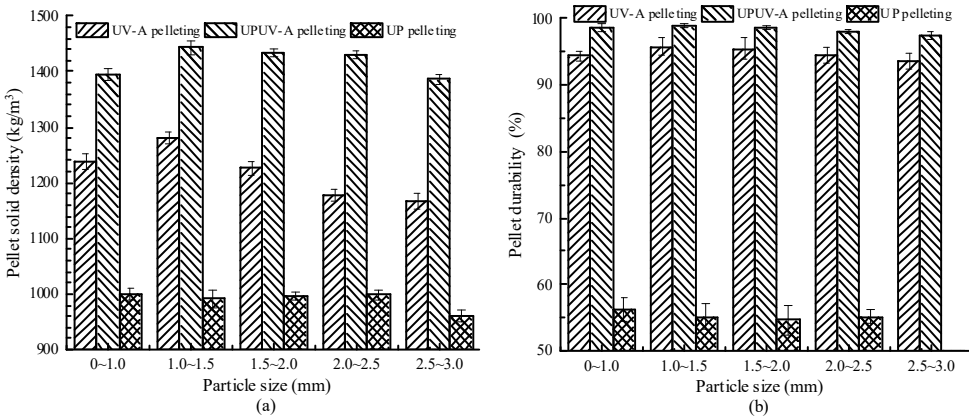

Figure 2. The pellet solid density and durability of comparative experiments: (**a**) pellet solid density; (**b**) pellet durability.

3.2. Response Surface Model of Density and Durability

Experimental results of the RSM are listed in Table 2. The experimental data were fitted to second-order polynomial models by conducting analysis of variance (ANOVA), as shown in Tables 3 and 4.

Table 2. The results of pellet solid density and durability based on CCRD.

Run	Distilled Water (x_1)/%	Temperature (x_2)/°C	Urea Content (x_3)/%	WSF Content (x_4)/%	Pellet solid Density y_1/(kg/m^3)	Pellet Durability y_2/%
1	50	55	3	9	1334.28	97.88
2	40	25	6	6	1320.44	96.38
3	30	35	3	9	1296.11	96.44
4	30	55	9	3	1393.78	96.04
5	50	55	9	3	1359.49	96.21
6	50	55	9	9	1389.60	97.95
7	30	35	9	3	1369.76	96.33
8	50	35	9	9	1413.62	97.90
9	30	35	9	9	1410.47	98.27
10	40	45	6	6	1398.90	97.21
11	30	55	3	3	1379.30	96.84
12	20	45	6	6	1364.18	96.94
13	30	55	9	9	1414.62	97.96
14	60	45	6	6	1352.23	97.68
15	40	45	6	0	1356.10	95.71
16	50	35	9	3	1349.32	96.49
17	30	35	3	3	1330.47	95.95
18	40	65	6	6	1395.15	97.88
19	40	45	12	6	1366.59	97.35
20	40	45	6	6	1425.19	97.75
21	40	45	6	6	1420.83	97.74
22	40	45	6	6	1402.25	97.59
23	40	45	6	6	1407.16	97.88
24	50	55	3	3	1365.15	96.73
25	50	35	3	9	1323.57	96.58
26	30	55	3	9	1362.23	97.99
27	40	45	6	12	1424.30	98.74
28	40	45	6	6	1428.60	97.85
29	50	35	3	3	1300.67	95.47
30	40	45	0	6	1268.19	95.58

Table 3. ANOVA results of the pellet solid density in a full model.

Source	Sum of Squares	df	Mean Square	F Value	p-Value
Model	49,079.22	14	3505.66	19.99	<0.0001 **
x_1—Distilled water	875.32	1	875.32	4.99	0.0411 *
x_2—Temperature	5217.96	1	5217.96	29.75	<0.0001 **
x_3—Urea content	15,285.34	1	15,285.34	87.15	<0.0001 **
x_4—WSF content	2261.27	1	2261.27	12.89	0.0027 *
x_1x_2	418.00	1	418.00	2.38	0.1435
x_1x_3	64.64	1	64.64	0.37	0.5529
x_1x_4	364.05	1	364.05	2.08	0.1702
x_2x_3	1932.04	1	1932.04	11.02	0.0047 **
x_2x_4	512.34	1	512.34	2.92	0.1080
x_3x_4	2898.75	1	2898.75	16.53	0.0010 **
x_1^2	4552.01	1	4552.01	25.95	0.0001 **
x_2^2	4624.74	1	4624.74	26.37	0.0001 **
x_3^2	14,618.74	1	14,618.74	83.35	<0.0001 **
x_4^2	654.20	1	654.20	3.73	0.0726
Residual	2630.87	15	175.39	--	--
Lack of Fit	1833.18	10	183.32	1.15	0.4664
Pure Error	797.69	5	159.54	--	--
Cor Total	51,710.09	29	--	--	--

Note: ** indicates high significance ($p \leq 0.01$); * indicates significance ($p < 0.05$).

Table 4. ANOVA results of the pellet durability in a full model.

Source	Sum of Squares	df	Mean Square	F Value	*p*-Value
Model	22.17	14	1.58	19.67	<0.0001 **
x_1—Distilled water	0.032	1	0.032	0.39	0.5408
x_2—Temperature	2.14	1	2.14	26.61	0.0001 **
x_3—Urea content	1.93	1	1.93	24.00	0.0002 **
x_4—WSF content	12.00	1	12.00	149.05	<0.0001 **
x_1x_2	0.015	1	0.015	0.19	0.6721
x_1x_3	0.016	1	0.016	0.20	0.6596
x_1x_4	5.063×10^{-4}	1	5.063×10^{-4}	6.288×10^{-3}	0.9378
x_2x_3	2.12	1	2.12	26.39	0.0001 **
x_2x_4	0.064	1	0.064	0.79	0.3876
x_3x_4	0.60	1	0.60	7.51	0.0152 *
x_1^2	0.31	1	0.31	3.79	<0.0704
x_2^2	0.62	1	0.62	7.72	0.0141 *
x_3^2	2.75	1	2.75	34.19	<0.0001 *
x_4^2	0.44	1	0.44	5.48	0.0335 *
Residual	1.21	15	0.081	--	--
Lack of Fit	0.90	10	0.090	1.47	0.3500
Pure Error	0.31	5	0.061	--	--
Cor Total	23.38	29	--	--	--

Note: ** indicates high significance ($p \leq 0.01$); * indicates significance ($p < 0.05$).

As listed in Table 3, the ANOVA indicated that the density model was highly significant ($p < 0.0001$) and the lack-of-fit test was insignificant. The coefficient of determination (R^2), Adj-R^2, and pred-R^2 were 0.9491, 0.9016, and 0.7736, respectively. The values of coefficient of variation (C.V.) and Adeq Precision (AP) obtained herein were 0.97% and 15.36, demonstrating that the model was well fitted with the experimental data. Comparison between the actual numerical values and the predicted values for the density (Figure 3) further confirmed that this quadratic regression model for pellet solid density was reliable and accurate. The *p*-values of variables suggested that x_2, x_3, x_4, x_2x_3, x_3x_4, x_1^2, x_2^2, and x_3^2 had the most significant effects on density, followed by x_1. After removing the statistically insignificant items in which the *p*-value exceeded 0.05, the response surface model for pellet solid density was formulated as Equation (3):

$$y_1 = 554.88 + 11.45x_1 + 18.54x_2 + 49.38x_3 + 2.90x_4 - 0.37x_2x_3 + 1.50x_3x_4 - 0.13x_1^2 - 0.13x_2^2 - 2.57x_3^2 \quad (3)$$

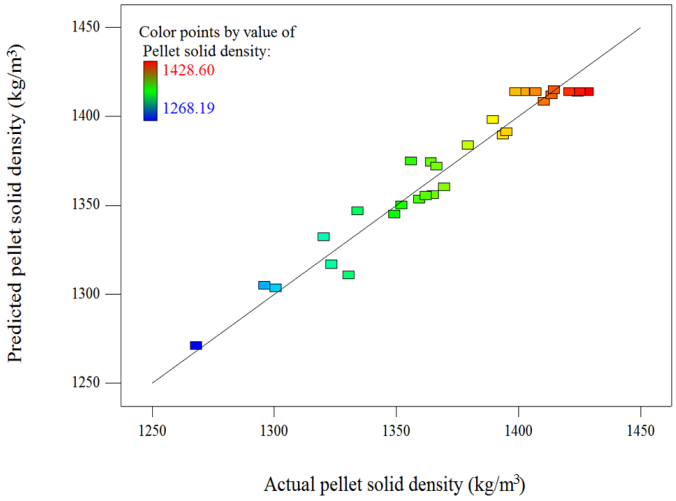

Figure 3. Comparison between the actual density and the predicted values.

The ANOVA from Table 4 indicated that the durability model was extremely significant ($p < 0.0001$) and the lack-of-fit test was insignificant ($p > 0.05$). The coefficient of determination (R^2), Adj-R^2, and pred-R^2 were 0.9483, 0.9001, and 0.7590, respectively. The values of the coefficient of variation (C.V.) and Adeq Precision (AP) obtained herein were 0.29% and 15.07, respectively; these suggest that the durability data were consistent with the second-order polynomial response surface model. To further validate the accuracy of the model, a comparison between the actual numerical values and the predicted values for the durability was carried out, shown in Figure 4. The points were uniformly distributed around the diagonal line. It can be considered that the response surface model could provide reasonable predictions for pellet durability.

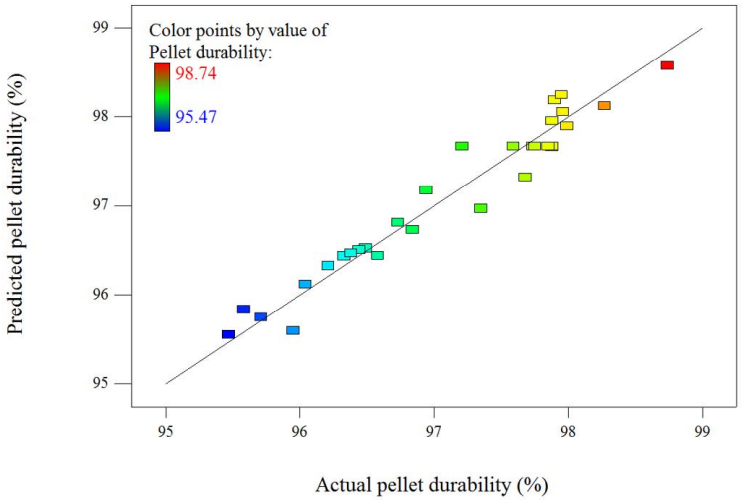

Figure 4. Comparison between the actual durability and the predicted values.

The p-values of x_2, x_3, x_4, x_2x_3, and x_3^2 were all less than 0.01, suggesting that they had extremely significant effects on pellet durability at the statistical level of $p < 0.01$. The p-values of x_3x_4, x_2^2, and x_4^2 were 0.0152, 0.0141, and 0.0335, respectively, suggesting that they had a significant effect at the significance level of $p < 0.05$. The p-values exceeding 0.05 had no statistically significant effect. After removing all statistically insignificant items, the response surface model for pellet durability was formulated as Equation (4):

$$y_2 = 86.52 + 0.21x_2 + 0.89x_3 + 0.19x_4 - 0.012x_2x_3 + 0.021x_3x_4 \\ -1.51 \times 10^{-3}x_2^2 - 0.035x_3^2 - 0.014x_4^2 \quad (4)$$

3.3. Predicted Effects of Variables on Pellet Solid Density and Durability

Equation (3) was utilized to predict the impact of distilled water on pellet solid density, and the results are illustrated in Figure 5. The pellet solid density increased with an increase in distilled water up to 40% under identical urea content but gradually decreased thereafter. Furthermore, an increase in urea content from 0% to 9% resulted in a corresponding increase in pellet solid density; however, at 12%, it was lower than that at 9%. It can be concluded that for urea with little distilled water, it was difficult to reach homogeneous mixing with raw corn stover, despite sufficient stirring. Minimal ammonia gas was converted from the urea solution, making it difficult to completely disrupt the chemical linkages between lignin and hemicellulose. This resulted in a reduced surface area of raw materials. However, excess distilled water diluted the concentration of urea solution and lowered the solid loading. Previous studies had shown that the low solid loading had a negative impact on ammonia gas conversion from urea [31].

The effects of temperature on pellet solid density and durability were predicted by Equations (3) and (4), and the results are depicted in Figure 6. The pellet solid density increased with the increase in temperature with a urea content of less than 6%. However, after initially increasing up to 55 °C, it subsequently decreased for urea contents of 9% and

12%. The pellet durability increased with temperature at urea contents of 0% and 3%, but only slightly at a urea content of 6%. However, the pellet durability decreased rapidly with increasing temperature for urea contents of 9% and 12%, especially at a urea content of 12%. This was due to the decrease in hemicellulose and water-soluble inorganic ions caused by swelling and hydrolysis at higher temperatures, while lignin content remained constant in corn stover pellets without added urea. The lignin, as a natural binder, could improve pellet solid density and durability. The delignification efficiency is improved by higher temperature and urea content, resulting in more removal of lignin. However, this process weakens the binding force between particles despite an increase in density.

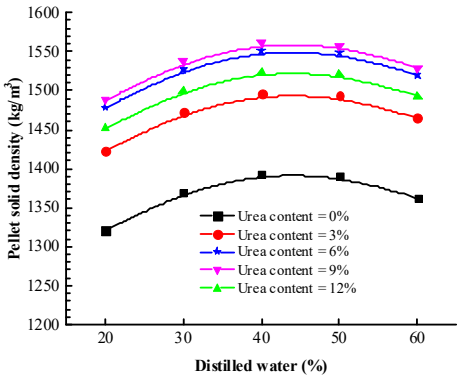

Figure 5. The effects of distilled water (mass ratio of distilled water added to corn stover) on pellet solid density (temperature = 45 °C; WSF content = 6%).

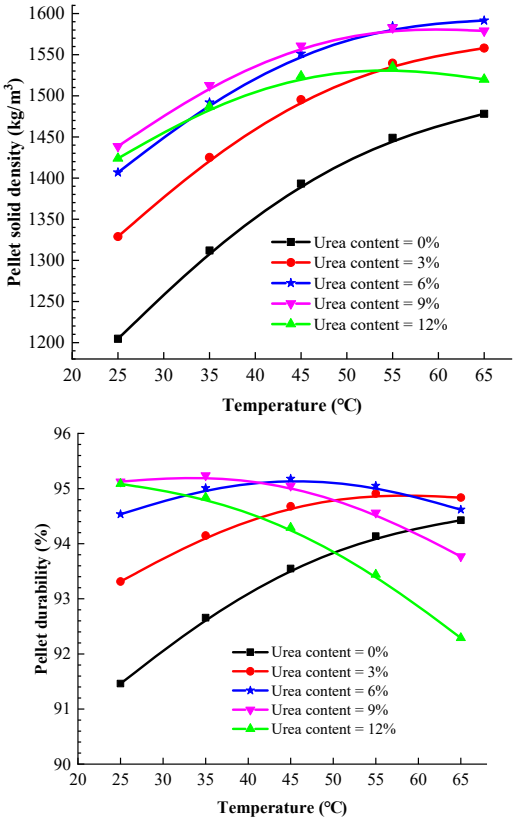

Figure 6. The effects of temperature on pellet solid density and durability (distilled water = 40%; WSF content = 6%).

The effects of urea content on pellet solid density and durability were predicted by Equations (3) and (4), with the results illustrated in Figure 7. Both pellet solid density

and durability exhibited a similar trend. With the increase in urea content, all increased to the peak value and subsequently declined. Within a specific range, the increase in urea concentration led to an elevation in the levels of aqueous ammonia and ammonium carbonate. However, excessive mass fractions of urea could impede the activity of urease, thereby hindering conversion [40]. To achieve optimal pellet solid density and durability, a high urea content combined with WSF was necessary. The WSF acted as both a source of urease and an additive.

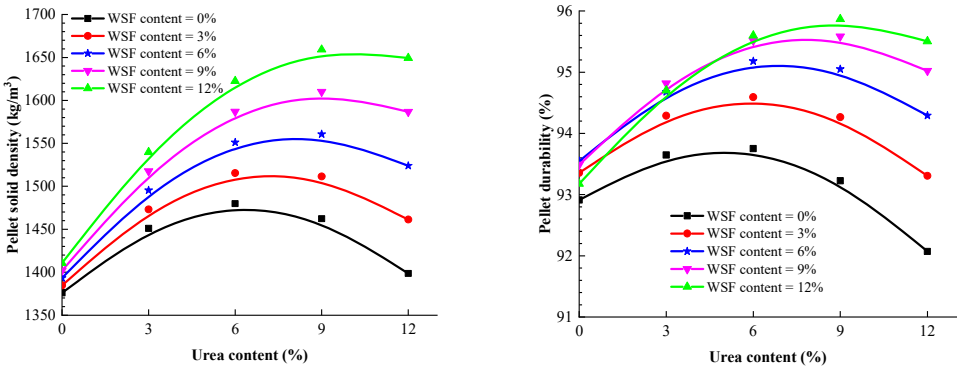

Figure 7. The effects of urea content on pellet solid density and durability (distilled water = 40%; temperature = 45 °C).

Equations (3) and (4) were used to predict the effects of WSF content on pellet solid density and durability, which are depicted in Figure 8. A linear relationship between pellet solid density and WSF content was observed, while the increase in pellet durability slowed down after reaching 9%. The effect of WSF content on pellet solid density was stronger than that on pellet durability. An increase in WSF content led to a rise in the amounts of additives present in corn stover. Kaliyan et al. [18] reported that highly polar components, such as lignin, starch, and protein, could enable hydrogen bonding and solid bridges. The high temperature increased the activation energy of the reaction system, intensified the activity of OH^-, and raised the possibility of breaking the cross-linking bond between lignin and hemicellulose.

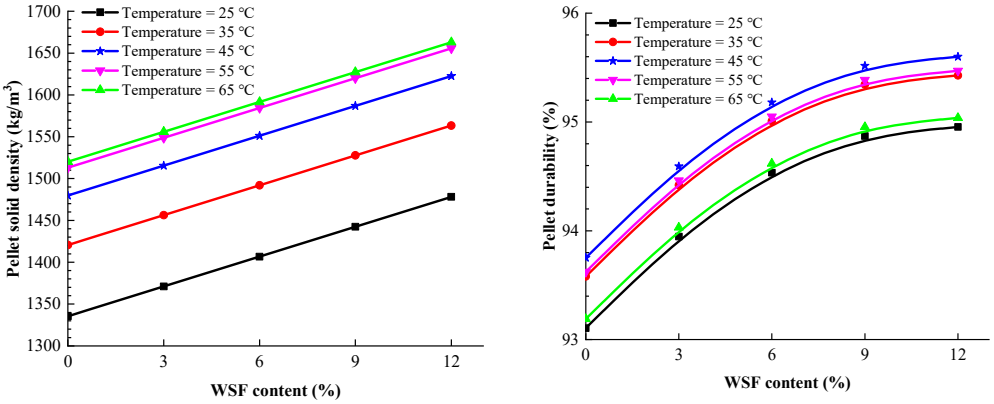

Figure 8. The effects of WSF content on pellet solid density and durability (distilled water = 40%; urea content = 6%).

3.4. Interaction Effects on Pellet Solid Density

Figure 9a illustrates the interaction effect of temperature and urea on pellet solid density. The minimum value of pellet solid density, which was 1327.05 kg/m^3, occurred at a urea content of 3% and a temperature of 35 °C, followed by an increase with the rise in variables. At a temperature of 55 °C, the density first increased and then decreased as the urea content increased. A higher density was achieved when the urea content exceeded 6% at temperatures ranging from 45 to 55 °C. It can be inferred that urease present in WSF

had the ability to convert urea into ammonia and carbamate [41]. The released ammonia further removed the fibers and lignin in the cell wall, released cellulose, and increased the contact area [40]. It was beneficial to the combination of particles, thus improving the pellet solid density. Figure 9b presents the interaction effect of urea content and WSF on pellet solid density. The pellet solid density initially increased with the increase in urea content but decreased at lower WSF level. With the increase in urea content at a higher WSF level, the density also increased, reaching a maximum value of 1434.25 kg/m^3 when both urea and WSF contents were 9%. The presence of sufficient amounts of WSF and urea facilitated greater conversion of ammonia gas from urea, thereby enhancing its effect on increasing the contact area between particles [31]. Although more lignin may be removed, the remaining WSF as adhesive could compensate for lignin's connection. The pellet solid density remained relatively stable at lower urea concentrations, primarily due to the limited availability of urea for releasing ammonia and removing lignin. Sufficient binders were present in the materials even at lower WSF levels.

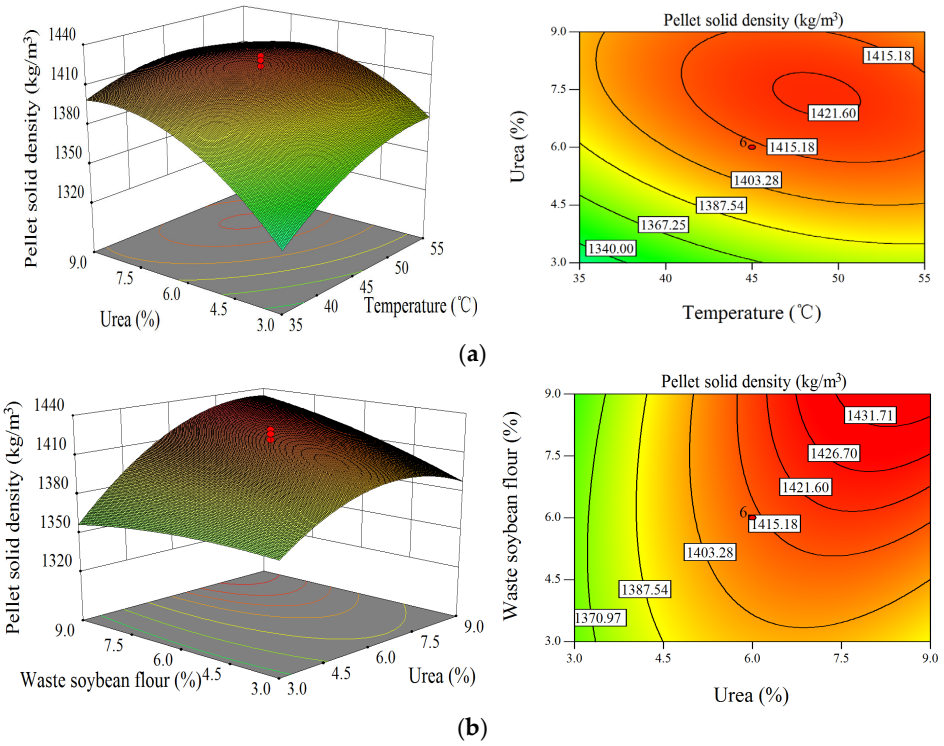

Figure 9. Response surface plots of pellet solid density between variables: (**a**) urea and temperature; (**b**) WSF and urea.

The interaction effects of variables on pellet durability were investigated. As depicted in Figure 10a, an increase in temperature led to a rise in pellet durability from 96.26% to 97.59% at a lower urea content (3%), while it decreased from 97.55% to 97.42% at a higher urea content (9%). It can be concluded that the effect of temperature on pellet durability was more significant with lower urea content. The minimum value of durability was observed when the urea content and temperature were set at 3% and 35 °C, respectively. However, higher temperatures resulted in greater removal of lignin from corn stover, leading to a decline in durability. Figure 10b presents the interaction effect of WSF and urea. The lowest pellet durability was observed at a WSF content of 3% and urea content of 3%, with a value of 96.44%. As the WSF content increased, so did the pellet durability. In addition, the durability increased faster at the higher level of urea than the lower one. The maximum pellet durability was achieved at 98.42% when the WSF and urea contents were both set to 9%. The increased amounts of WSF acted as an adhesive, enhancing attraction forces between solid particles and mechanical interlocking bonds, ultimately leading to

improved pellet durability [42]. Therefore, it can be concluded that additional additives were necessary for urea-pretreated corn stover to improve pellet durability.

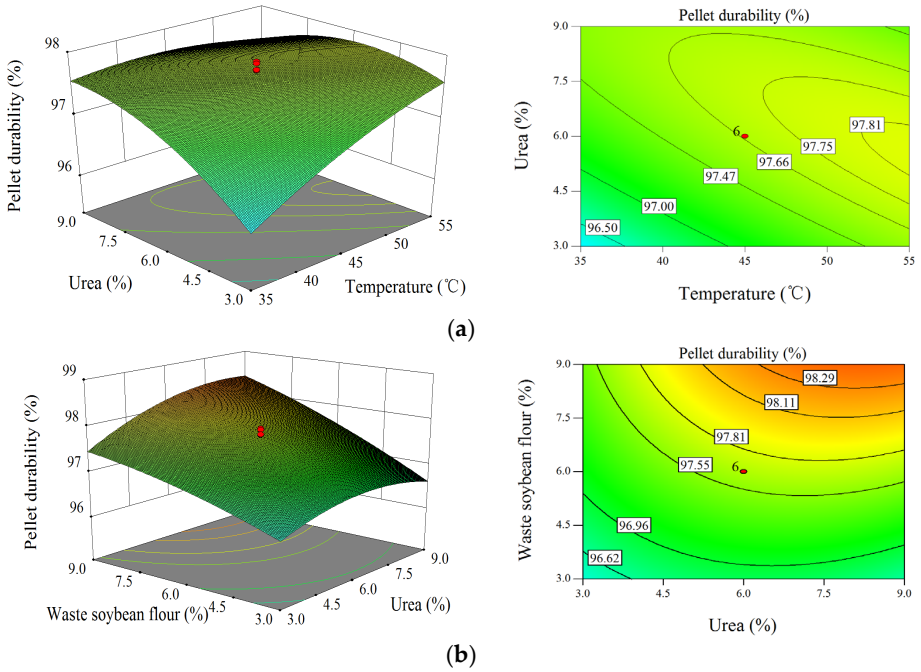

Figure 10. Response surface plots of pellet durability between variables: (**a**) urea and temperature; (**b**) WSF and urea.

3.5. Optimization and Model Verification

To optimize the pretreatment variables for high density and durability simultaneously, the optimization function in Design-Expert (v8.0.6.1) was employed. The optimal conditions were determined to be 42.06% distilled water, temperature of 44.69 °C, 10.18% urea content, and 11.96% WSF content. To compare the results of the model, additional confirmation experiments were conducted to verify the simulation results under the condition of 42% distilled water, temperature of 45 °C, 10% urea content, and 12% WSF content. The density and durability were 1438.28 kg m^{-3} and 98.67%, respectively, which were very close to the predicted values, and the desirability was 1.000, suggesting an outstanding agreement, as shown in Table 5.

Table 5. The results of the confirmation experiment.

Comparison	Process Variables				Pellet Solid Density (kg m^{-3})	Pellet Durability (%)
	Distilled Water /%	Temperature /°C	Urea Content /%	WSF Content /%		
Predicted value	42.06	44.69	10.18	11.96	1441.41	98.91
Experimental value	42	45	10	12	1438.28	98.67
Error (%)	1.43	0.42	1.8	0.33	0.22	0.24

4. Conclusions

In the present research, comparative experiments results demonstrated that combining urea pretreatment with UV-A pelleting could significantly improve the pellet solid density and durability. The CCRD was applied to determine the optimal conditions of urea pretreatment. Mathematical regression models were established to predict pellet solid density and durability based on various factors, which were then verified by ANOVA. The results indicated that the regression model with an R^2 value greater than 0.90 could accurately

predict pellet solid density and durability under the optimal conditions. According to the *p*-values, the effects of variables on pellet solid density were summarized in descending order of significance as follows: urea content, temperature, WSF content, and distilled water; meanwhile, the effects of variables on pellet durability were WSF content, temperature, and urea content. Interaction effects of urea content and temperature, as well as urea content and WSF content, were found to have statistically significant effects on pellet solid density and durability. The optimal conditions were obtained with 42% distilled water, temperature of 45 °C, 10% urea content, and 12% WSF content.

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