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Research article

Optimization of density and durability of pellets using the response surface methodology in ultrasonic vibration-assisted pelleting of corn stover

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ABSTRACT: Top-quality pellets can significantly increase density and durability of agricultural residues, reducing logistic costs. However, these pellets depend on numerous parameters, including feedstock properties and production conditions. To ensure high-quality pellets, a single-factor experiment and the response surface methodology were used to investigate the effects of particle size, moisture content, molding pressure, pelleting time, ultrasonic power, and interaction effects between variables on density and durability of pellets for ultrasonic vibration-assisted pelleting of corn stover. The response surface models between variables and response were established. The results showed that all variables affect the density and durability of pellets. An optimal condition for density and durability was obtained, and a further experiment was conducted to validate the values. The results suggested that desirability (0.999) under optimal conditions confirmed the validation of models. The optimal combination of process parameters included particle size of 1.5 mm, moisture content of 10 %, molding pressure of 379 kPa, pelleting time of 80 s, ultrasonic power of 250 W, with values of 1,381.14 kg m^{-3} and 97.58 % for density and durability of pellets, respectively.

Keywords: pellet properties, process parameters, prediction models, crop residues

Introduction

Biomass is the only renewable resource in repository and logistics and has great energy potential. Approximately 10 % of the world energy consumption is supplied by biomass resources (Song et al., 2018). Crop residues are a type of biomass resource, and their production has increased to 5,280 Mt in 2020-2021 (Shinde et al., 2022). The high moisture content and transportation costs (Zhang et al., 2016) as well as low density (Kulokas et al., 2021), slagging and fouling (Nosek et al., 2020), and the non-uniform size (Frodeson et al., 2019) are the main reasons for the reduced use of crop residues. China has ruled several policies to increase the use of crop residues to relieve rural energy predicament and achieve carbon neutrality. Pellet, one clean, renewable energy source, has noticeably attracted attention (Zhang et al., 2017b).

Traditional methods to manufacture pellets require high temperature, high pressure, and proper moisture content. These methods are energy-consuming and lead to severe abrasion for molds. However, ultrasonic vibration-assisted (UV-A) pelleting can avoid external heat and high pressure (Li et al., 2016; Zhang et al., 2013). Pellets processed by UV-A pelleting have higher density and stability than those processed by other methods (Cong et al., 2011). Previous studies showed that density and durability of pellets made from wheat straw increased with high ultrasonic power, moisture content, and pressure and decreased particle size enlargement (Fan et al., 2013; Song et al., 2015). Pellets made from wheat straw and switchgrass achieved the highest density at a moisture content of 13 % (Fan et al., 2013). The response surface methodology (RSM) has been widely accepted to study the effects of multiple process variables on response variables (Cui et al., 2019; Kaveh et al., 2021). A predictive model was developed using RSM to predict the variables on pellet density using UV-A pelleting of wheat straw. The effects of ultrasonic power on pellet density were stronger at high pressure and low biomass moisture content (Fan et al., 2014). Predictive models for density and durability and optimal conditions were obtained using RSM in UV-A pelleting of sorghum stalk (Zhang et al., 2018).

However, agricultural residues have physical and chemical diversities, resulting in significant differences in their molding characteristics. Few studies used the RSM method to establish models and optimize parameters in UV-A pelleting of corn stover. In addition, the input parameters mainly included pelleting time, pressure, and ultrasonic power. The effect of particle size and moisture content to the model is unclear. The present study aimed (1) to establish a mathematical model; (2) to investigate the main effects of process parameters on density and durability; (3) to determine the interactions among the variables tested; (4) to optimize the process parameters for top-quality pellets.

Materials and Methods

Materials and experimental apparatus

The raw material used in this study was corn stover from southern Kansas (KS, USA). Experimental apparatus for UV-A pelleting was obtained from the Department of Industrial and Manufacturing Systems Engineering, Kansas State University. Corn stover was milled into particles using a cutting miller (model SM 2000; Retsch, GmbH, Haan). The moisture content of materials was sprayed with distilled water. The machine included a pneumatic loading system, ultrasonic generation system, and biomass holding system (Song et al., 2018), as shown in Figure 1. The ultrasonic power (500 W, 20,000 Hz, AP-1000; Sonic-Mill Inc.) was adjusted from 0 to 100 %, and the air compressor's pressure ranged from 0 to 689.5 kPa. The diameters of the pelleting tool and the aluminum mold were 17.4 and 18.6 mm, respectively.

Experimental design

A single-factor experiment was first carried out to determine the rational ranges and effects of particle size, moisture content, molding pressure, pelleting time, and ultrasonic power on preparation using RSM experiments. For this purpose, 1.5 g of corn stover particles were loaded into the mold and the titanium tool was fed down to compress the particles into pellets. Variables and levels used in the single-factor experiment are shown in Table 1. The experimental data were analyzed by OriginPro 2018 software.

Based on the results of the single-factor experiment, RSM was used to determine the interactions between the variables tested and their effective ranges. A central composite rotatable design with five factors and five levels (1/2 implementation) was carried out. The experimental variables and levels used in RSM are listed in Table 2.

The experimental data were analyzed by Design-Expert 8.0.6.1 software and RSM was used to obtain three-dimensional (3D) surface graphs between the variables and the response. The second-order polynomial of Eq. (1) was used to fit the experimental data (Askari et al., 2021; Cui et al., 2019; Le et al., 2019). The optimization function in Design-Expert was applied to investigate the optimal conditions for maximizing the density and durability of pellets.

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

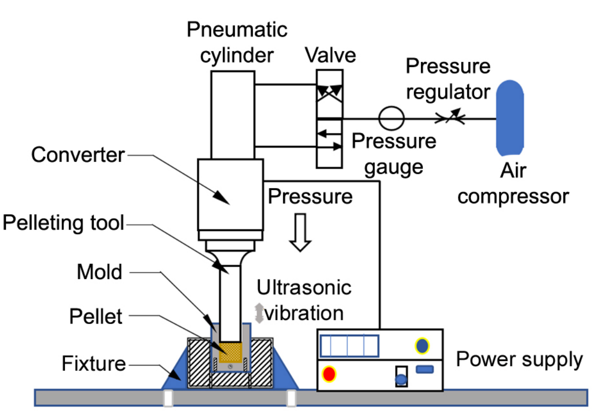

Figure 1 – Ultrasonic assisted molding device.

Table 1 – Variables and levels in the single factorial design.

Variables	Levels				
Particle size (mm)	0~1.0	1.0~1.5	1.5~2.0	2.0~2.5	2.5~3.0
Moisture content (%)	6	8	10	12	14
Molding Pressure (kPa)	138	207	276	345	414
Pelleting time (s)	30	50	70	90	110
Ultrasonic power (W)	0	100	200	300	400

Table 2 – The independent variables and levels for RSM design.

Variables	Symbols	Level				
		–2	–1	0	+1	+2
Particle size (mm)	x_1	0.5	1.0	1.5	2.0	2.5
Moisture content (%)	x_2	6	8	10	12	14
Molding Pressure (kPa)	x_3	276	310	345	379	414
Pelleting time (s)	x_4	30	50	70	90	110
Ultrasonic power (W)	x_5	100	150	200	250	300

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

Test method

Pellet density was calculated by the ratio of its mass to its volume (Zhang et al., 2018). An electronic scale measured pellet mass and the volume was calculated by pellet diameter and height. Three replications were carried out under each condition. The average value from the three replications was used as pellet density for the condition. Pellet density was calculated using Eq. (2):

$$\rho = \frac{4m}{\pi d^2 h} \times 10^6 \quad (2)$$

where: ρ is pellet density, kg m^{-3}; m represents pellet mass, g; d is pellet diameter, mm; h represents pellet height, mm.

Pellet durability is the capacity of the pellet to withstand impact and other forces during handling and transportation (Tabil and Sokhansanj, 1996). Five pieces of pellets were weigheded and then placed into a pellet durability tester (Seedburo Equipment Co.), which was kept tumbling under a rotation speed of 50 rpm (Tang et al., 2015). Pellets were next taken out from the tester and sieved through a No.6 U.S. sieve (Zhang et al., 2013). The weight of pellets retained by each sieve was recorded. Pellet durability was calculated using Eq. (3):

$$DU = \frac{m_2}{m_1} \quad (3)$$

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

Results and Discussion

Single-factor experiment

Effects of particle size on density and durability of pellets

Density and durability of pellets were obtained with different particle sizes under moisture content of 10 %, ultrasonic power of 200 W, pressure of 276 kPa, and pelleting time of 70 s. Pellet density gradually decreased with the enlargement of the particle size. The maximum value of density was 1,261.29 kg m^{-3} obtained at 1.0 ~ 1.5 mm and the minimum value was 1,167.52 kg m^{-3} at 2.5 ~ 3.0 mm. However, durability increased first and then gradually decreased (range, 93-95 %), as shown in Figure 2, mainly because particle size significantly influenced compression, contact between the adjacent particles, flowability, and friction (Siyal et al., 2021a). The chemical bonding was established when the maximum attractive force reached the minimum potential energy (Kaliyan and Morey, 2010). The effectiveness of these forces diminished dramatically as the size of the particles or inter-particle distance increased (Matsunaga et al., 2000; Yub Harun et al., 2018). The effect of particle size on pellet durability is that the coarse and large particles reduced the adhesion area, producing poorly structured pellets (Azargohar et al., 2018). As for small particle sizes, attraction forces were mainly associated to the larger rate of particle deformations. Internal resilience decreased compared to large particle sizes under the same condition, which increased of density and durability. Similar studies also showed that the smaller particle size increased pellet density by filling the voids and facilitating the interlocking of particles during the compression process (Hettiarachchi et al., 2019; Mani et al., 2004; Whittaker and Shield, 2017). However, particle size below a particular limit has a negligible effect on pellet quality (Pradhan et al., 2018; Stelte et al., 2012). The results confirmed that the particle size appropriate for RSM was less than 2.5 mm.

Effects of molding pressure on density and durability of pellets

Pressure significantly influenced pellet density, durability, and energy consumption (Stelte et al., 2012). Density and durability of pellets were obtained under different molding pressure with a particle size of 1.0 ~ 1.5 mm, moisture content of 10 %, ultrasonic power of 200 W, and pelleting time of 70 s. Density and durability of pellets increased with higher molding pressure. Higher pressure caused solid particles to be closer to each other with fewer voids and gaps (Siyal et al., 2021b), enhancing van der Waals forces and hydrogen bonding (Anukam et al., 2021). Density rose fast from 1,047.23 to 1,244.67 kg m^{-3} before 276 kPa, indicating an increase of 18.85 %, and increased slowly afterward. Density reached 1,280.47 kg m^{-3} at 414 kPa, which only increased by 0.18 % compared with 345 kPa. Durability also had a similar trend to that of density. The maximum value was 96.21 % at 414 kPa, while the minimum was 93.09 % at 138 kPa, as shown in Figure 3. The results showed that density and durability were challenging to improve effectively when pressure reached a certain limit, indicating that porosity was very small. Higher pressure did not significantly influence pellet density (Adapa et al., 2010; Cui et al., 2021). If a higher pressure had been applied, the wearing of the mold might have risen. In order to obtain high-quality pellets, the range of molding pressure appropriate for RSM was confirmed to be 276-414 kPa.

Effects of moisture content on density and durability of pellets

Density and durability of pellets were obtained with different moisture contents under particle size of 1.0 ~ 1.5 mm, molding pressure of 345 kPa, ultrasonic power of 200 W, and pelleting time of 70 s. Density and

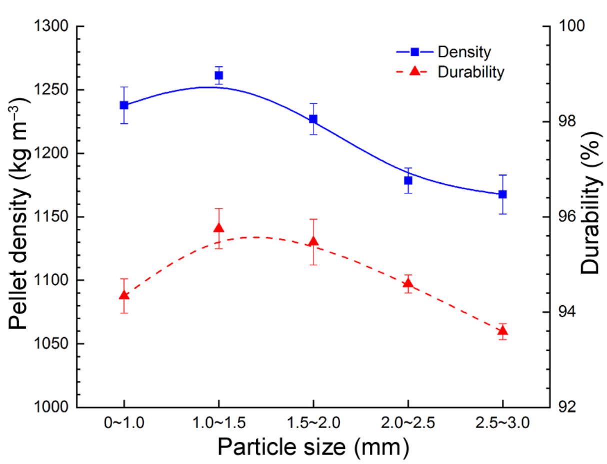

Figure 2 – Pellet density and durability with different particle sizes.

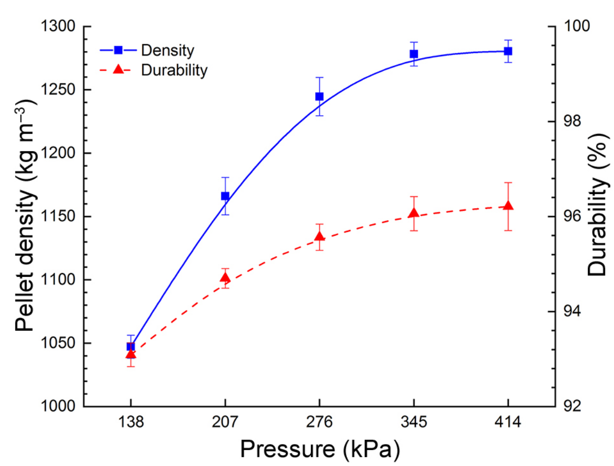

Figure 3 – Pellet density and durability with different molding pressures.

durability first increased and then slightly decreased with increased moisture content. The maximum density was 1,367.28 kg m^{-3} at a moisture content of 10 %, indicating an increase of 17.9 % compared to the density at a moisture content of 6 %. The final density was 1,353.00 kg m^{-3} at 14 %. Durability ranged from 92.56 % to 96.27 %, in which the highest and the lowest durability levels were obtained at 10 % and 6 %, respectively, as shown in Figure 4.

An appropriate moisture content not only plays a role in lubrication, reducing friction between particles of raw materials and increasing fluidity (Yılmaz et al., 2021), but it also mixes with sugar in raw materials to form colloids to help the bonding of particles together. A similar result has been reported in which a proper moisture content facilitated starch gelatinization, protein denaturation, and fiber solubilization during pelletization (Carone et al., 2011). However, excessive moisture contents weaken hydrogen bonds and the van der Waals forces due to the increased distance between particles (Hettiarachchi et al., 2019), reducing density and durability (Tumuluru, 2019). The range of moisture content appropriate for RSM was confirmed to be 6-14 %.

Effects of pelleting time on density and durability of pellets

Density and durability of pellets were obtained at different pelleting time points under particle size of 1.0 ~ 1.5 mm, molding pressure of 345 kPa, ultrasonic power of 200 W, and moisture content of 10 %. The extension of pelleting time increased density and durability of pellets before 90 s and then slightly decreased after reaching the maximum value. The maximum density and durability values were 1,340.84 kg m^{-3} and 98.07 % at 90 s, as shown in Figure 5.

Some factors contributed to this phenomenon. On the one hand, it might be because the holding time was not set in the process. With the extension of pelleting time, including the destruction of the structure of cell walls, holding time also had the function of dwell time. Previous research showed that holding time was beneficial to pellet density and durability due to the increase of pellet deformation resistance and it could offset the spring-back effect of biomass grinds (Tang et al., 2018). On the other hand, when the pelleting time was short, the ultrasonic effect was not clear enough to generate more heat and softened lignin, resulting in low density and durability. However, extended pelleting time could generate huge heat and cause charring inside the pellet, leading to low density and durability (Zhang et al., 2017b). The pelleting time appropriate for RSM was confirmed to be 30–110 s.

Effects of ultrasonic power on density and durability of pellets

Density and durability of pellets were obtained with different ultrasonic powers under particle size of 1.0 ~ 1.5 mm, molding pressure of 345 kPa, pelleting time of 90 s, and moisture content of 10 %. Pellet density and durability increased from 0 to 300 W. The maximum density was 1,345.44 kg m^{-3} at 300 W, while the lowest density was 978.69 kg m^{-3} without applying ultrasonic power. Similar results showed that pellet density processed with ultrasonic vibration was higher than without for switchgrass and wheat straw (Zhang et al., 2011; Zhang et al., 2012). Pellet durability ranged from 0 to 97.37 %. The blasting phenomenon occurred during the compression process when the ultrasonic power reached 400 W. Part of the center in a pellet was charred, which caused several cracks, as shown in Figure 7. During UV-A pelleting, the temperature was a key factor affecting pellet quality (Zhang et al., 2017a). The highest temperature always appeared at the center of a pellet with the increase in ultrasonic power (Li et al., 2016; Tang et al., 2015). Crack size became larger when ultrasonic power, pelleting pressure, pellet weight, and moisture content were higher and when pelleting time

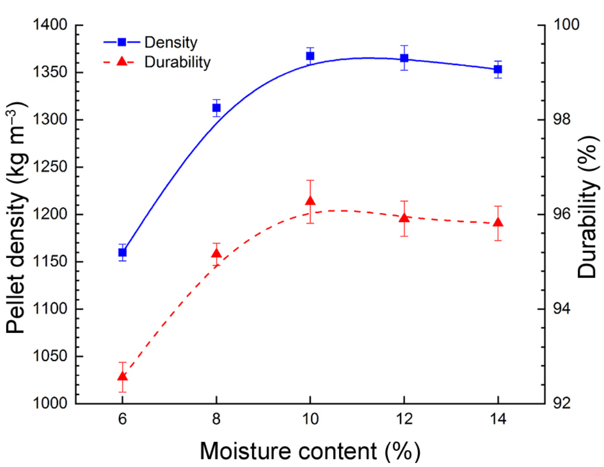

Figure 4 – Pellet density and durability with different moisture contents.

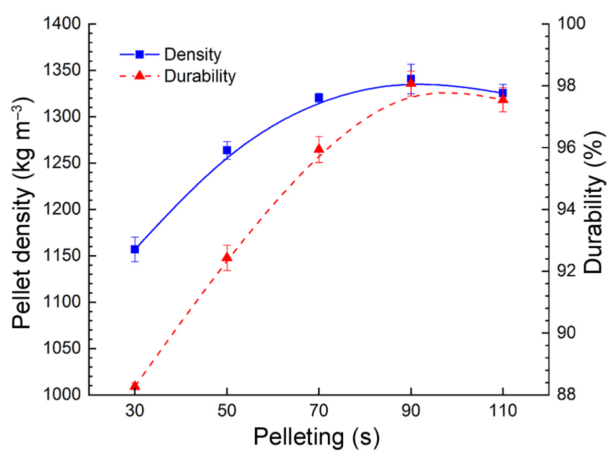

Figure 5 – Pellet density and durability with different pelleting times.

was longer (Tang et al., 2012). The range of ultrasonic power appropriate for RSM was confirmed to be 100-300 W, as shown in Figure 6.

RSM experiment

Response surface model of density and durability of pellets

Experimental results of RSM are listed in Table 3. The experimental data were fitted to second-order polynomial models by the analysis of variance (ANOVA), as shown in Tables 4 and 5.

The ANOVA from Table 4 indicated that the density model was highly significant (F = 28.42, p < 0.0001). At the same time, the lack-of-fit test was insignificant (F = 2.10, p = 0.1517), demonstrating that the model was well fitted to the experimental data. The p-value of particle size (x_1), moisture content (x_2), molding pressure (x_3), and ultrasonic power (x_5) were < 0.0001, while the pelleting time (x_4) was 0.0004. The p-value of x_1x_5, x_2x_3, and x_3x_4 were 0.0005, 0.0220, and 0.0008, respectively. The coefficient of determination (R^2) and Adj-R^2 were 0.97 and 0.94, respectively. The coefficient of variation (CV) and Adeq Precision (AP) obtained were 1.81 % and 22.86, respectively. This suggested that the density data were consistent with the second-order polynomial response surface model and that the response surface model could provide reasonable predictions for pellet density. Moreover, the ANOVA showed that pellet density was greatly affected by all parameters, as well as the interaction of particle size and ultrasonic power, the interaction of pressure, and pelleting time at the statistical level of p < 0.01. The interaction of moisture content and pressure had a prominent effect at the statistical level of p < 0.05. After removing the statistically insignificant items in which p-value exceeded 0.05, the response surface model for pellet density was formulated as Eq. (4):

$$y_1 = -1365.84 + 904.69x_1 + 38877x_2 - 0.10x_3 - 4.47x_4 - 0.74x_5 - 0.98x_1x_5 - 0.20x_2x_3 + 0.03x_3x_4 - 155.03x_1^2 - 13.52x_2^2 + 5.51\times10^{-3}x_5^2 \quad (4)$$

The ANOVA from Table 5 indicated that the durability model was highly significant (F = 20.75, p < 0.0001), while the lack-of-fit test was insignificant (F = 3.25, p = 0.0550). The p-value of particle size (x_1), moisture content (x_2), molding pressure (x_3), pelleting time (x_4), and ultrasonic power (x_5) were < 0.0001, 0.0018, 0.0025, < 0.0001, 0.0005, and < 0.0001, respectively. The p-value of x_1x_2 was 0.0336. The values of R^2 and Adj-R^2 were 0.97 and 0.92. The values of CV and AP obtained were 1.67 % and 16.61, which confirmed that this quadratic regression model for pellet durability was reliable and accurate. The p-values of variables suggested that ultrasonic power and molding pressure had the most remarkable effects on durability, followed by pelleting time, particle size, and moisture content, respectively. The effects of all the interaction terms on durability were negligible (p > 0.05) except for the interaction of particle size and moisture content at the statistical level of p < 0.05. After removing the statistically insignificant items in which p-value exceeded 0.05, the response surface model for pellet durability was formulated as Eq. (5):

$$y_2 = -171.49 + 30.55x_1 + 17.91x_2 + 0.48x_3 + 0.46x_4 + 0.37x_5 - 0.88x_1x_2 - 9.78x_1^2 - 0.81x_2^2 - 4.87\times10^{-4}x_3^2 - 1.64\times10^{-3}x_4^2 - 2.8\times10^{-4}x_5^2 \quad (5)$$

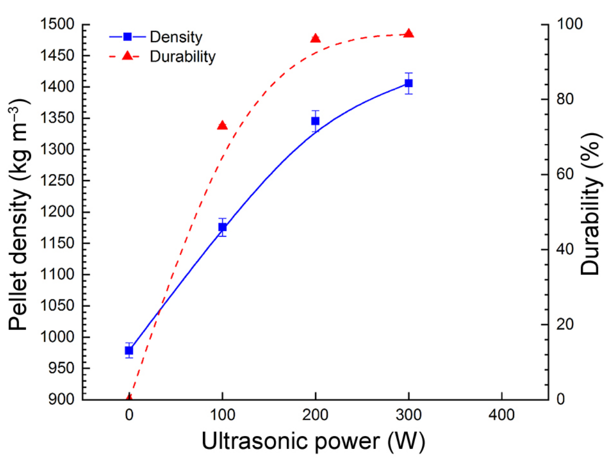

Figure 6 – Pellet density and durability with different ultrasonic powers.

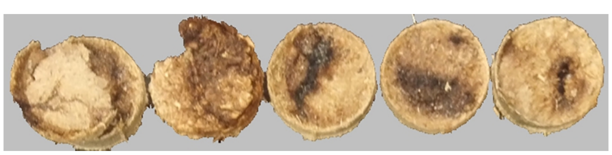

Figure 7 – Charring pellet.

Interaction effects on pellet density

The interaction effects between particle size and ultrasonic power, moisture content and molding pressure, and molding pressure and pelleting time on pellet density were investigated. The interaction effect of particle size and ultrasonic power on pellet density was obtained with moisture content, molding pressure, and pelleting time at 10 %, 345 kPa, and 70s, respectively, as shown in Figure 8A. Pellet density increased from 1,232.87 to 1,333.38 kg m^{-3} (increase of 8.15 %) at a smaller particle size (1.0 mm), and increased from 1,213.59 to 1,217.44 kg m^{-3} (an increase of 0.32 %) at a larger particle size (2.0 mm) due to the interaction effects between particle size and ultrasonic power when ultrasonic power increased from 150 to 250 W. It can be concluded that the effect of ultrasonic power on pellet density was more significant at the smaller particle size. Moreover, pellet density increased first before 1.5

Table 3 – The experimental results of pellet density and durability based on a central composite design.

Run	Particle size	Moisture content	Molding pressure	pelleting time	Ultrasonic power	Pellet density / y_1	Pellet durability / y_2
	mm	%	kPa	s	W	kg m^{-3}	%
1	2.0	12	310	50	250	1203.78	84.84
2	2.0	12	379	90	250	1212.33	90.85
3	1.0	12	379	90	150	1274.91	94.16
4	1.0	10	345	70	200	1290.16	96.40
5	2.0	12	310	90	150	1180.14	85.86
6	1.5	10	345	70	200	1269.39	96.79
7	1.5	10	414	70	200	1355.04	98.12
8	1.0	8	379	90	250	1335.88	90.10
9	1.5	6	345	70	200	1001.49	81.08
10	2.0	8	310	90	250	1095.39	88.81
11	1.0	12	379	50	250	1310.12	94.74
12	1.5	10	345	70	200	1303.60	96.42
13	2.0	12	379	50	150	1160.06	86.16
14	1.5	10	345	70	200	1303.60	95.71
15	2.5	10	345	70	200	1027.81	84.71
16	2.0	8	379	50	300	1133.67	90.17
17	2.0	8	379	50	150	1123.17	86.93
18	1.5	10	345	70	100	1256.84	87.58
19	1.0	12	310	50	150	1210.30	86.21
20	1.5	10	345	70	200	1257.36	93.85
21	1.5	10	345	110	200	1303.83	97.08
22	1.5	10	345	70	200	1267.60	94.02
23	2.0	8	379	90	150	1212.59	86.28
24	1.0	8	310	90	150	1099.11	83.29
25	1.0	8	310	50	250	1174.00	88.98
26	0.5	10	345	70	200	1198.92	86.23
27	2.0	8	310	50	150	1109.63	78.59
28	1.5	10	345	70	300	1390.10	97.33
29	1.5	14	345	70	200	1102.70	83.54
30	1.5	10	345	70	200	1280.44	94.29
31	1.5	10	276	70	200	1195.45	87.75
32	1.5	10	345	70	200	1261.10	95.97
33	1.5	10	345	70	200	1257.44	95.63
34	1.5	10	345	70	200	1259.87	96.40
35	1.0	12	310	90	250	1260.91	92.14
36	1.5	10	345	30	200	1186.00	88.18

mm and then decreased at the same ultrasonic power. The transfer of heat generated by ultrasonic power was retarded with the enlarged particle size could explain this phenomenon. The increase in biomass temperature was one of the densification mechanisms of UV-A pelleting, which may lead to the local melting of biomass and the subsequent formation of strong solid bridges (Fan et al., 2014; Kaliyan and Morey, 2009). In addition, a larger biomass size resulted in more voids and weakened the van der Waals forces during the compression process (Pradhan et al., 2021). In contrast, a smaller biomass size might reduce pelletization temperature and cause a low degree of lignin softening thereby inhibiting the natural binding tendency (Kirsten et al., 2016).

The interaction effect of moisture content and molding pressure on pellet density originated from the particle size, ultrasonic power, and pelleting time at 1.5 mm, 200 W, and 70 s, respectively, as shown in Figure 8B. Pellet density increased from 1,146.95 to 1,236.55 kg m^{-3} (increase of 7.81 %) at a lower moisture content (8 %), and increased from 1,234.96 to 1,269.29 kg m^{-3} (increase of 2.78 %) at a higher moisture content (12 %) due to the interaction effects between moisture content and molding pressure, when molding pressure increased from 310 to 379 kPa. It can be concluded that the effect of molding pressure on pellet density was more significant at the lower moisture content. There was a positive correlation between pellet density and molding pressure at all moisture contents because the higher pressure resulted in greater particle size change and less porosity. The highest density was obtained when the moisture content was 10 % - 11 %. Moisture lowered the glass transition temperature of lignin and increased the contact area of particles (Siyal et al., 2021a). When the moisture content

Table 4 – ANOVA results for pellet density in a full model.

Source	Sum of square	Degree of freedom	Mean square	*F*-value	*p*-value
Model	275300	20	13767.05	28.42	< 0.0001**
x_1- Particle sizes	28223.72	1	28223.72	58.26	< 0.0001**
x_2- Moisture content	22296.43	1	22296.43	46.03	< 0.0001**
x_3- Molding pressure	23352.55	1	23352.55	48.21	< 0.0001**
x_4- Pelleting time	9687.37	1	9687.37	20.00	0.0004**
x_5- Ultrasonic power	16155.99	1	16155.99	33.35	< 0.0001**
x_1x_2	885.58	1	885.58	1.83	0.1964ns
x_1x_3	1807.13	1	1807.13	3.73	0.0725ns
x_1x_4	224.25	1	224.25	0.46	0.5066ns
x_1x_5	9538.95	1	9538.95	19.69	0.0005**
x_2x_3	3160.95	1	3160.95	6.53	0.0220*
x_2x_4	1569.56	1	1569.56	3.24	0.0920ns
x_2x_5	66.91	1	66.91	0.14	0.7154ns
x_3x_4	8596.00	1	8596.00	17.74	0.0008**
x_3x_5	466.25	1	466.25	0.96	0.3421ns
x_4x_5	406.53	1	406.53	0.84	0.3741ns
x_1^2	48068.58	1	48068.58	99.23	< 0.0001**
x_2^2	93571.35	1	93571.35	193.16	< 0.0001**
x_3^2	93.84	1	93.84	0.19	0.6661ns
x_4^2	1102.62	1	1102.62	2.28	0.1522ns
x_5^2	6066.52	1	6066.52	12.52	0.0030**
Residual	7266.48	15	484.43	-	-
Lack of Fit	4242.81	6	707.14	2.10	0.1517
Pure error	3023.67	9	335.96	-	-
Total	282600	35	-	-	-

**High significance ($p \leq 0.01$); *Significant ($p < 0.05$); nsnot significant.

Table 5 – ANOVA results for pellet durability in a full model.

Source	Sum of square	Degree of freedom	Mean square	*F*-value	*p*-value
Model	944.51	20	47.23	20.75	< 0.0001**
x_1- Particle sizes	32.75	1	32.75	14.39	0.0018**
x_2- Moisture content	29.83	1	29.83	13.11	0.0025**
x_3- Molding pressure	110.17	1	110.17	48.41	< 0.0001**
x_4- Pelleting time	44.44	1	44.44	19.53	0.0005**
x_5- Ultrasonic power	115.49	1	115.49	50.75	< 0.0001**
x_1x_2	12.44	1	12.44	5.47	0.0336*
x_1x_3	0.00	1	0.00	0.00	0.9954ns
x_1x_4	5.32	1	5.32	2.34	0.1472ns
x_1x_5	0.36	1	0.36	0.16	0.6964ns
x_2x_3	0.58	1	0.58	0.25	0.6211ns
x_2x_4	3.29	1	3.29	1.45	0.2478ns
x_2x_5	10.20	1	10.20	4.48	0.0514ns
x_3x_4	4.08	1	4.08	1.79	0.2006ns
x_3x_5	4.50	1	4.50	1.98	0.1799ns
x_4x_5	4.54	1	4.54	1.99	0.1783ns
x_1^2	191.37	1	191.37	84.09	< 0.0001**
x_2^2	335.03	1	335.03	147.21	< 0.0001**
x_3^2	10.73	1	10.73	4.72	0.0463*
x_4^2	13.75	1	13.75	6.04	0.0266*
x_5^2	15.63	1	15.63	6.87	0.0193*
Residual	34.14	15	2.28	-	-
Lack of Fit	23.37	6	3.89	3.25	0.0550
Pure error	10.77	9	1.20	-	-
Total	978.64	35	-	-	-

**High significance ($p \leq 0.01$); *Significant ($p < 0.05$); nsnot significant.

was low, straw fiber was easy to break and caused poor fluidity (Carone et al., 2011). In addition, several silicide compounds and waxiness in the straw epidermis were challenging to connect under the low moisture content. However, excessive moisture prevented the complete release of natural binders from biomass (Carone et al., 2011; Siyal et al., 2021a), increased the spacing among particles, and relaxed degrees of pellets after ejection from the die (Adapa et al., 2011).

The interaction effect of pelleting time and molding pressure on pellet density was obtained with particle size, moisture content, and ultrasonic power at 1.5 mm, 10 %, and 200 W, respectively. Pellet density increased from 1,241.76 to 1,258.22 kg m^{-3} (increase of 1.33 %) at a shorter pelleting time (50 s), and increased from 1,235.73 to 1,344.10 kg m^{-3} (increase of 8.77 %) at a longer pelleting time (90 s) due to the interaction effects between pelleting time and molding pressure, when molding pressure increased from 310 to 379 kPa, as shown in Figure 8C. It can be concluded that the effect of molding pressure on pellet density was more significant at the longer pelleting time, which allowed more heat to be transferred to soften lignin to eliminate the elastic stress.

Interaction effects on pellet durability

The interaction effect of particle size and moisture content on pellet durability was analyzed under pelleting time, molding pressure, and ultrasonic power at 70 s, 345 kPa and 200 W, respectively. Pellet durability increased first and then decreased with the increase in particle size and moisture content. The lowest pellet durability was obtained at a larger particle size and lower moisture content, followed by a small particle size and high moisture content. The decline of pellet durability was rapid under larger particle sizes with the high moisture content condition compared with other conditions. Under the high moisture content, the moisture inside the particles may prevent the release of the perfectly smooth natural binder due to water incompressibility (Cui et al., 2021; Kaliyan and Morey, 2010). The highest pellet durability was found at a particle size of around 1.5 mm and moisture content of 9-11 %. The results showed that particle size and moisture content interaction greatly influenced pellet durability due to their effect on the contact surface and biomass fluidity, as shown in Figure 9.

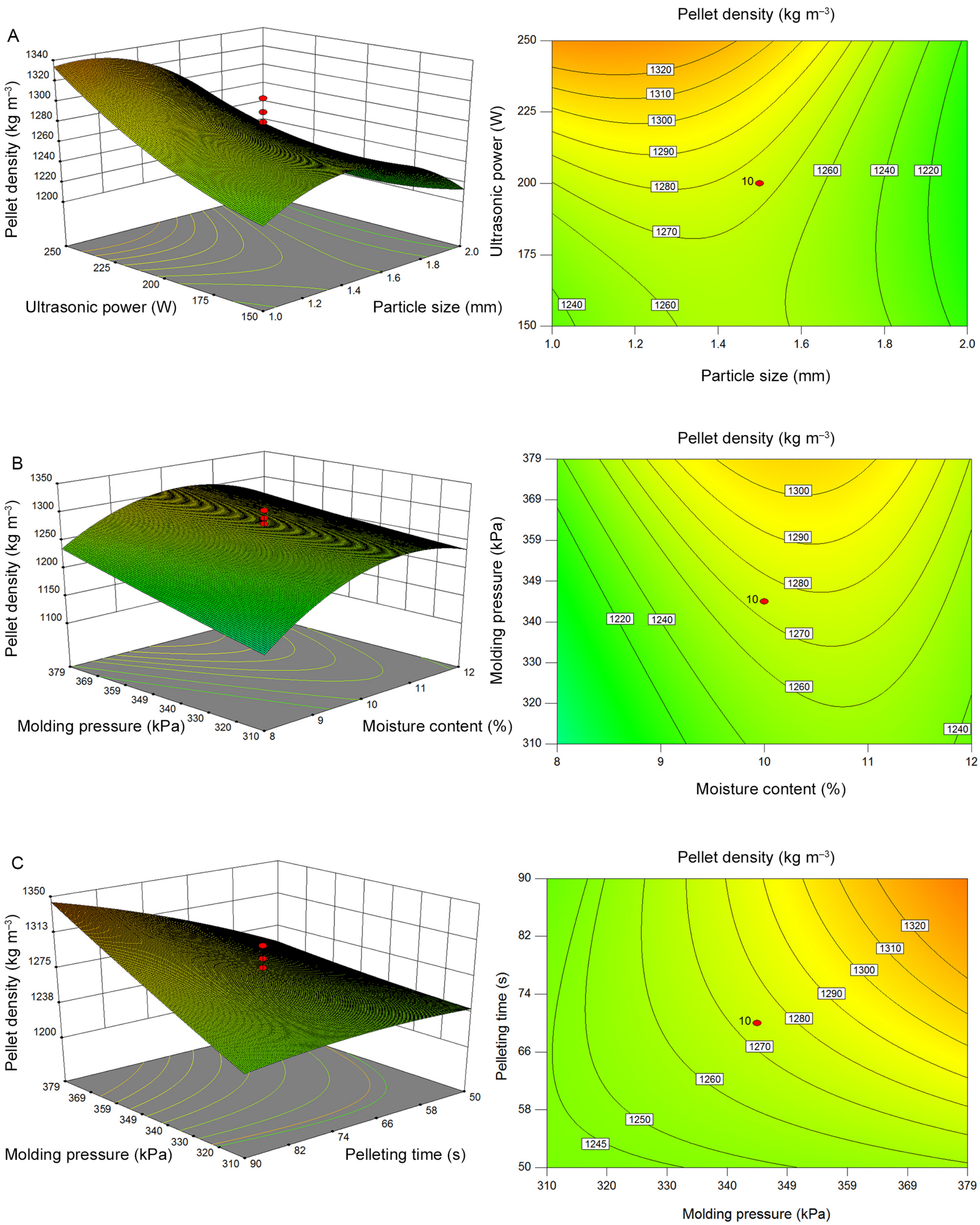

Figure 8 – Response surface plots of pellet density between variables. (A) Particle size and ultrasonic power; (B) Moisture content and molding pressure; (C) Molding pressure and pelleting time.

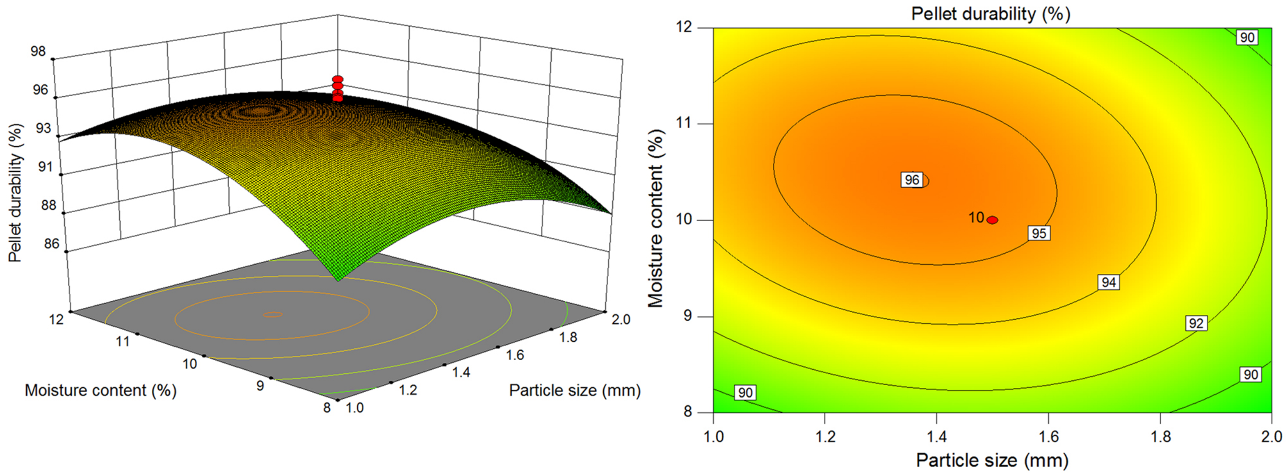

Figure 9 – Response surface of particle size and moisture content on pellet durability.

Table 6 – The results of the confirmation experiment.

Comparison	Process variables					Pellet density	Pellet durability
	Particle size	Moisture content	Molding pressure	Pelleting time	Ultrasonic power		
	mm	%	kPa	s	W	kg m^{-3}	%
Predicted value	1.33	10.30	379	80.26	250	1390.10	98.07
Experimental value	1.5	10	379	80	250	1381.14	97.58

Optimization of process variables for density and durability of pellets and verification

The effects of process variables and their interactions on the density and durability of pellets were assessed. The optimization function in Design-Expert was used to optimize the input variables for high density and durability simultaneously and object function and constraint conditions are shown in Eq. (6).

$$\begin{cases} Max(y_1, y_2) \\ s.t \begin{cases} 0.5 \le x_1 \le 2.5 \\ 6 \le x_2 \le 14 \\ 276 \le x_3 \le 414 \\ 30 \le x_4 \le 110 \\ 100 \le x_5 \le 300 \end{cases} \end{cases} \quad (6)$$

The optimal conditions were obtained with a particle size of 1.33 mm, moisture content of 10.30 %, molding pressure of 379 kPa, pelleting time of 80.26 s, and ultrasonic power of 250 W. To compare the results of the model, additional confirmation experiments were conducted to verify the simulation results at the particle size of 1.5 mm, moisture content of 10 %, molding pressure of 379 kPa, pelleting time of 80 s, and ultrasonic power of 250 W. Pellet density and durability were 1,381.14 kg m^{-3} and 97.58 %, respectively, which were very close to the predicted values, while desirability was 0.999, suggesting an outstanding agreement, as shown in Table 6.

Conclusions

In this study, single-factor experiments were carried out to determine the rational ranges for UV-A pelleting of corn stover. RSM was developed to predict thedensity and durability of pellets. The results showed that particle size, molding pressure, moisture content, pelleting time, and ultrasonic power significantly affected density and durability of pellets in UV-A pelleting. The mathematical regression models of factors for density and durability of pellets were established and verified by ANOVA through quadratic regression rotatable orthogonal test. According to *p*-values, the effects of variables on pellet density were summarized as follows: particle size, molding pressure, moisture content, ultrasonic power, and pelleting time. The effects of variables on pellet durability were presented as follows: ultrasonic power, molding pressure, pelleting time, particle size, and moisture content. The optimal combination of process parameters included particle size of 1.5 mm, moisture content of 10 %, molding pressure of 379 kPa, pelleting time of 80 s, and ultrasonic power of 250 W, with values of 1,381.14 kg m^{-3} and 97.58 % for density and durability of pellets, respectively.

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Authors' Contributions

Conceptualization: Li WT. **Data curation:** Li WT, Zhang B. **Formal analysis:** Li WT, Li CX, Zhou GX. **Funding acquisition:** Zhou GX, Liu D. **Investigation:** Zhang B, Li CX. **Methodology:** Li WT, Zhang B. **Resources:** Zhou GX, Liu D, Li CX. **Supervision:** Li WT. **Writing-original draft:** Li WT. **Writing-review & editing:** Zhou GX, Liu D, Li CX.

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