Enhancing Density and Durability of Biomass Pellets through Optimized Pressurized Binder Spraying and Process Parameters

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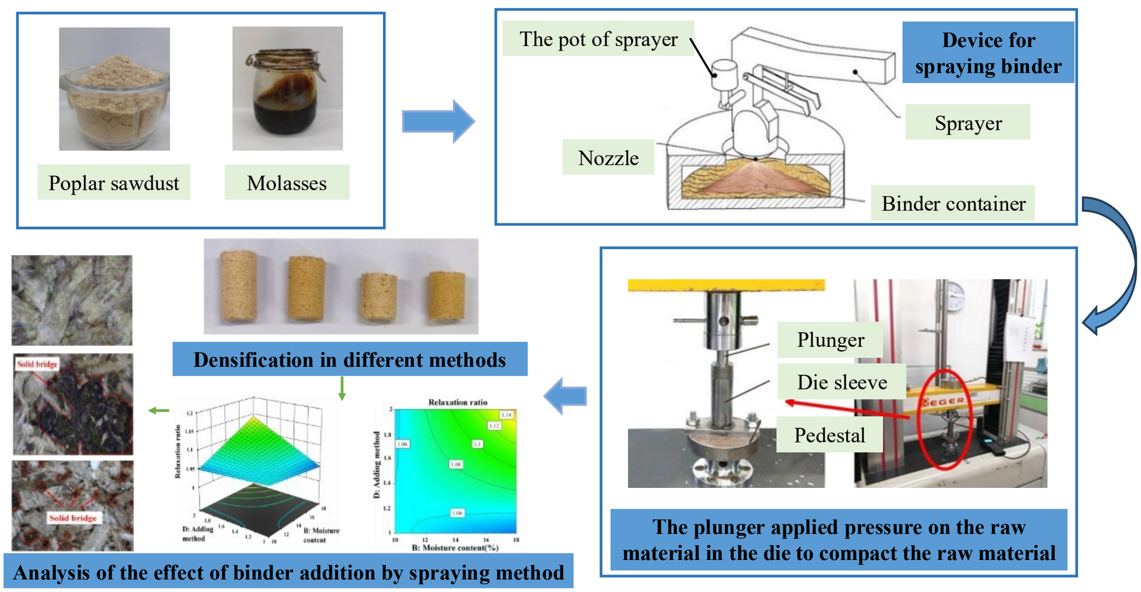
GRAPHICAL ABSTRACT



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To reduce the energy consumption of biomass densification, this study proposes a method of constructing solid bridges through pressurized binder spraying. The feasibility of this method for producing high-quality biomass molding was studied under ambient temperatures and lower pressures. Four-factor mixed-level orthogonal tests were designed to evaluate the relaxation ratio and durability of density pellets, in which the molasses served as the binder. Pressurized spraying of the binder resulted in a 27.0% increase of relaxation density, 8.21% decrease in relaxation ratio, and significantly enhanced durability compared to stirring method at pressure 40 MPa, which was determined in preliminary testing to conform to at least 95% durability. A multivariate quadratic regression equation through response surface analysis was established by selecting a 2FI model for the 100% importance in binder addition method. The relaxation ratio was normalized to the weights of the influencing factors obtained from model of multi-layer perceptron neural network. The test factors had a significant impact of on the relaxation ratio, and thus, the optimal combination condition for test was determined as 50 (MPa) densification pressure, 14% moisture content, 4% binder ratio, and pressurized spraying at 2 (MPa). These conditions reduced the minimum densification pressure required for biomass densification.

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Keywords: Biomass densification; Binder; Solid bridge; Pressurized spraying method

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INTRODUCTION

As the primary energy source in the world, fossil fuels face the problem of depletion and environmental pollution (Ates *et al.* 2020). Biomass is a green, non-polluting, and abundantly renewable energy source (Li and Hu 2003), and it has both practical and long-term significance for environmental protection, economic development, and the sustainable development of human society. Biomass raw materials have the disadvantages of low energy density, low bulk density, irregular shape and size, and high particle emission (Silva *et al.* 1998). In the course of the densification process, the biomass pellets become granular, rod-shaped, or blocky. Simultaneously, the energy density increases, which reduces the cost of transportation and storage (Duque *et al.*2023). Compared with biomass raw materials, biomass pellets are uniform in size and generally have higher compressive strength (Nunes *et al.* 2016). The dynamic changes of the raw material particles in the compression process and the performance of the pellets obtained after densification are fundamentally related to its microscopic densification mechanisms (Stelte *et al.* 2011). Therefore, studying forming mechanisms is the key to promoting the development of biomass compression densification technology.

The forming mechanisms include both physical changes and chemical reactions, and the transformation process behind it is quite sophisticated. Solid bridges are an important way of bonding internal particles during the densification of biomass (Kaliyan and Morey 2010). Two kinds of solid bridges can be formed during the densification process. One is to form solid bridges from the original material through the diffusion of the molecules, while the other is to form such bridges from added binders. Without the binder, components of the biomass such as protein, starch, and lignin become temporarily softened at high temperature and high pressure and then cool (sometimes crystallizing) to form the solid bridges (Zhen *et al.*2019). When binders were added during the densification process, they became the primary source of solid bridges (Chin and Siddiqui 2000). There are two types of solid bridges formed by the binders. The first one comes from the liquid bridge attached to the surface of the particles. When liquid binders are used during the densification process, liquid bridges were formed and they subsequently cool and dry to become solid bridges. The second type of bridging is formed by the inter-particle chemical reaction or viscosity on the surface of solid particles caused by increased temperature (Li *et al.* 2012). For instance, adding fibers to the raw materials can lead to forming the second type of solid bridge.

There exists a diverse range of binders, some of which form solid bridges that connect biomass particles, enhance their densification at room temperature, and ultimately improve pellet quality. The addition of waste paper fiber from a packaging box into sawdust as a binder and compression at room temperature enables low-pressure densification with low degree of rebounding, thereby improving the density, mechanical properties, and quality of biomass pellets (Kong *et al.* 2012). The performance of pellets, including porosity and density, depends strongly on the amount of binder and its dispersion within the pellets (Ileleji *et al.* 2016). Molasses, a by-product of sugarcane production, serves as a binder for biomass pellet densification and significantly enhances the mechanical properties and burning characteristics of biomass pellets (Zhai *et al.* 2018). Extensive research exists on molasses as a binder in pellet production. Molasses, as a high-viscosity binder, can adhere to the surface of solid particles and form a solid bridge (Kaliyan and Morey 2010). The essential role of molasses in producing high-quality biomass pellets has been confirmed (Manyuchi *et al.* 2018). The use of molasses and fructose results in better performance in the durability index of biomass pellets compared to other binders, regardless of raw material type (Soleimani *et al.* 2017).

Besides the binder, the densification process of biomass pellets is also affected by compression pressure, raw material moisture content, and particle size (Sharma *et al.*2021). Pressure represents the most critical condition for compressing biomass materials. Increasing pressure significantly enhances the durability and relaxation density of pellets, as demonstrated in a densification test using palm kernel cake (Razuan *et al.* 2011). The optimal moisture content for adding sawdust at room temperature falls within the range of 12 to 16%, due to the high sensitivity of wood materials to temperature and humidity (Hui 2006). Furthermore, particle size greatly affects the densification of biomass at room temperature (Yan 2013). The particle size of sawdust should not exceed 6 mm (Grover *et al.* 1994). Reducing particle size can improve the quality of pellet densification, as revealed by an electron microscope study on the micro-mechanism of the pellets (Huo *et al.* 2011).

Since most of the existing studies have used the stirring method to add binders, this paper aims to explore the effect of adding the binder using a pressurized spraying method. In the densification process, smaller particles encounter a larger surface area, resulting in stronger molecular attraction between particles, thereby increasing the relaxation density (Zhang *et al.* 2014). Therefore, by the pressurized spraying method, binder in microdroplets is added to the biomass raw materials, building solid bridges, and thus promoting the connection between the particles. The pellets are expected to be densified at room temperature under low pressure, with a low relaxation ratio, thereby improving the mechanical properties.

EXPERIMENTAL

Materials

Poplar sawdust used in this work had particle sizes ranging from 0 to 3 mm. These materials were sealed after collection because the physical and chemical properties of wood materials are influenced by temperature and humidity (Kumar *et al.* 2015). When an aqueous brown sugar solution with a concentration of 6% was added to poplar fibers, the density as well as the physical characteristics of biomass pellets increased (Liu *et al.* 2023). The environmentally friendly, relatively inexpensive industrial residue molasses was used as the binder. The molasses used in this experiment is a by-product of the sugar industry. Its main components are sugars, containing approximately 24%~36% sucrose, 12%~24% other sugars, and 8%~10% minerals, along with a small amount of crude protein. The experimental environment temperature was 15 to 30 °C, with a relative humidity of ≤80%. The particle size distribution of the binder droplets was as follows: the proportion of droplets with a particle size below 75 μm reached 85%, and the proportion between 0 and 50 μm was 69%.

According to ISO 13061-1 (2014), the initial moisture content of poplar sawdust was measured. The equation for calculating the moisture content was as follows,

$$W=\frac{m_1-m_0}{m_0}\times 100\% \quad (1)$$

where W is moisture content of raw material (%), m_1 is mass of sawdust before drying (g), and m_0 is sawdust mass (g) after drying. The initial moisture content of raw material was calculated as 8.5%, and the moisture content of raw material was adjusted to 10%, 12%, 14%, 16%, and 18% after binder addition, respectively. After the experiment, all materials were sealed.

Equipment

The pressurized spraying device had the following properties: barometer, nozzle diameter, 1.3 mm; spraying width, 165 mm; and outlet pressure, 0.4 MPa. The pressurized spraying device is shown in Fig. 1(a). The sprayer connected the air compressor with inlet 1 to obtain the compressed air, and the binder was filled into the sprayer pot 4. The liquid binder was sprayed out as a mist under high pressure when pressing the handle 3. The spraying method is shown in Fig. 1(b).

The plunger and die (inner diameter 16 mm) are shown in Fig. 1(c) and 1(d). During the densification, the plunger applied pressure on the raw material in the die to compact the raw material.

Other equipment used were a REGER microcomputer controlled electronic universal testing machine (model: 4050, Regel, Shenzhen, China), electronic balance (model: SF-400A, China) (precision: 0.01g), moisture meter (model: FK-50, Xiamen, China), air compressor (rated power: 750W), vernier caliper (precision: 0.01mm), and Leica S8 APO Body Vision Microscope (40×), *etc*.

Material Preparation

The biomass raw material and binder were mixed by stirring and pressurized spraying. Pressurized spraying: Poplar sawdust and molasses were put into container and sprayer pot, respectively. The air compressor was turned on to keep the air pressure at 0.4 MPa. Then, the molasses was evenly sprayed on the surface of sawdust particles by pressing the sprayer's handle. The mixture was stirred with an electric mixer for 2 minutes to ensure thorough homogenization of the raw materials and the binder. Stirring: After the binder was added, a mixer was used for blending. The mixing impeller was set to rotate clockwise at a constant speed of 200 r/min for 2 minutes to ensure thorough integration of the binder with the material.

In each test, 7 g of test material was filled into the die and then compressed by the universal testing machine at a speed of 5 mm/min. After reaching the set pressure value, the pressure was held for 30 seconds. Then the pellet was removed from the die and stored in a sealing bag.

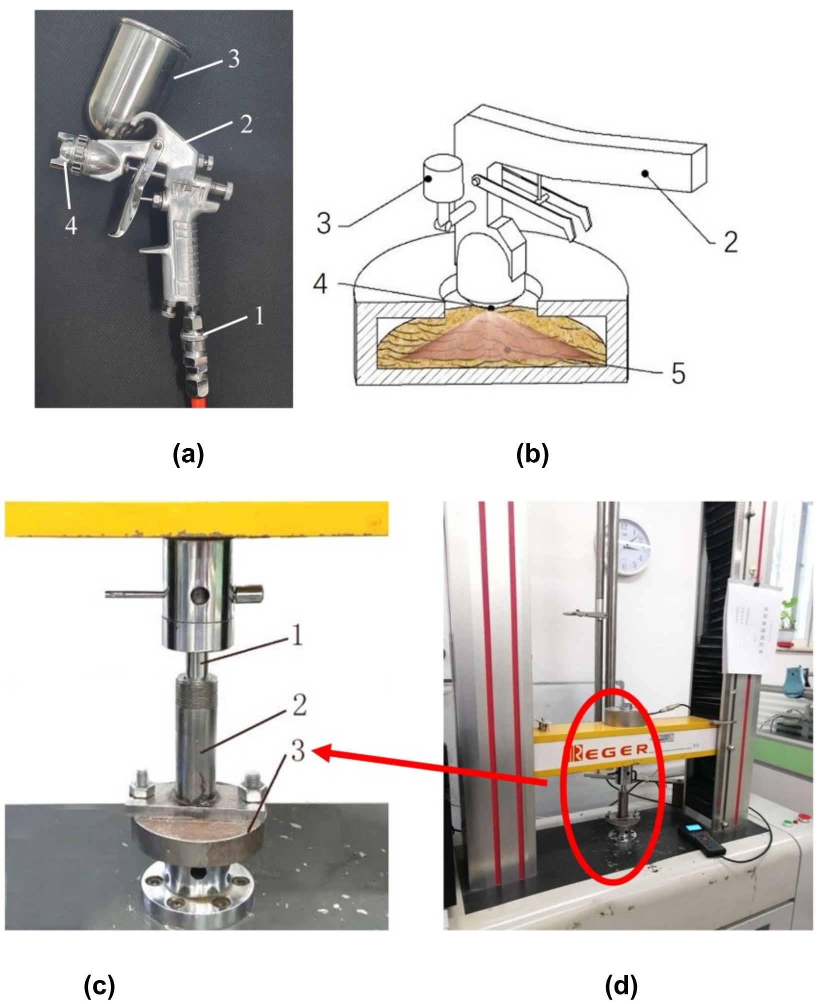


Fig. 1. (a)(b) Spraying device: 1-air inlet; 2-sprayer; 3-the pot of sprayer; 4-nozzle; 5-material

container. (c) Compaction apparatus: 1-plunger; 2-die sleeve; 3-pedestal. (d) Microcomputer-controlled electronic universal testing machine

Test Method and Index Determination

Apart from combustion characteristics, the physical characteristics of densified pellets are the most important ones. These characteristics directly influence the use requirements, transportation requirements, and storage conditions of pellets. This experiment selected the relaxation ratio, the durability, and the minimum densification pressure of densified pellets as the evaluation indices.

Relaxation ratio measures the pellet physical quality and combustion performance. It is calculated by dividing the maximal compression density by the relaxation density after biomass densification (Li *et al.* 2019). A smaller relaxation ratio indicates greater relaxation density. The length of three densified pellets in each group was measured, and the average value was calculated. To ensure the accuracy and reliability of the test data, the diameters at the top, middle, and bottom of each densified pellet were measured and averaged as the diameter values of each densified pellet. An electronic balance with the accuracy of 0.01 g (model: SF-400 A) was used to weigh the densification fuel quality. The relaxation density was calculated by Eq. 2,

$$\rho = \frac{4m}{\pi d^2 h} \tag{2}$$

where ρ is relaxation density (g/cm^3), m is mass of densification pellets (g), d is pellet diameter (cm), and h is pellet length (cm).

The relaxation ratio was calculated as follows,

$$\lambda = \frac{\rho_m}{\rho} \tag{3}$$

where λ is relaxation ratio, ρ_m is maximum compression density (density when pellets are removed from the die) (g/cm^3), and ρ is relaxation density (g/cm^3).

Durability reflects the ability of pellets to resist deformation after multiple falling and rolling collisions (Tu *et al.* 2015). To test their durability, densified pellets were dropped freely from height of 2 m, with 5 replicates. The percentage of pellet mass after five drops was greater than 95%, indicating that the pellets were of good quality. The sample quality was measured before and after the test, and the durability was calculated. The average value of each group's test results is chosen to determine the final durability to ensure the accuracy of the test. The durability was calculated as follows,

$$I = \frac{M_f}{M_b} \times 100\% \tag{4}$$

where I is durability (%), M_b is pellet mass before falling test (g), and M_f is mass of pellets after falling test (g).

The term "minimum densification pressure" denotes the pressure applied to biomass raw particles within the die to compress them into the desired shape. This pressure ensures that biomass pellets maintain their shape without becoming loose or breaking after squeezing out of the die. The specific value of the minimum pressure varies based on factors such as the characteristics of the raw materials, moisture content, particle size, and other parameters.

Design of Test Parameters

Test the effect of the spraying method to promote the densification of biomass

A four-factor mixed-level orthogonal test was designed to determine the influence of various test factors on the test evaluation index while reducing the number of tests and shortening the test cycle. Densification pressure (factor A), raw material moisture content (factor B), binder addition ratio (factor C), and binder addition method (factor D) were chosen to be the test factors. The L25 (2×5^3) orthogonal test was used. According to the pre-test results (Table 1), durability of at least 95% in the pre-test was measured at 40 MPa. Under this pressure, compared with that without binder, the relaxation density of the pellets obtained by the spraying method increased by 16.9%, the relaxation ratio decreased by 6.3%, and the durability increased by 6.8%. Compared with the stirring method, the relaxation density of the densified pellets obtained by the spraying method is increased by 27.0%, the relaxation ratio was reduced by 8.2%, and the durability was significantly improved. At the same time, the durability of densified pellets without binder and with a binder added by the stirring method cannot reach 95% of the industry standard value, and the durability of densified pellets with a binder was higher. During the densification process, the moisture content is best controlled with the range of 5 to 15%, and the maximum cannot exceed 20% (Li 2005).

To investigate the effect of binder forming a solid bridge by pressurized spraying on the densification of biomass at room temperature and low pressure, a series of tests were conducted. The densification pressure (factor A) was set to 30, 40, 50, 60, and 70 MPa, and the moisture content of raw materials (factor B) was set to 10, 12, 14, 16 and 18. Considering that adding too much binder in industrial applications will increase production costs, the binder addition ratio (factor C) was set to 1, 2, 3, 4, and 5%, and the binder addition method (factor D) was set as spraying method and the stirring method respectively.

Table 1. Average Value of each Dependent Variable at Different Densification Pressures

Densification pressure (MPa)	Relaxation density (g/cm³)	Relaxation ratio	Durability(%)
75 (spraying)	0.781 ± 0.009	1.047 ± 0.010	97.97 ± 0.070
62 (spraying)	0.751 ± 0.005	1.041 ± 0.007	98.04 ± 0.040
50 (spraying)	0.675 ± 0.007	1.066 ± 0.003	97.06 ± 0.980
45 (spraying)	0.723 ± 0.002	1.031 ± 0.005	97.19 ± 0.960
40 (spraying)	0.705 ± 0.005	1.028 ± 0.004	95.62 ± 0.759
40 (stirring)	0.555 ± 0.011	1.120 ± 0.010	92.75 ± 0.435
40 (no binder)	0.603 ± 0.001	1.097 ± 0.009	89.49 ± 1.055
37 (spraying)	0.642 ± 0.002	1.068 ± 0.004	91.15 ± 0.960

Note: moisture content is 10%, and binder proportion is 3%

Investigating the efficacy of the pressurized spraying method in forming solid bridges to enhance bonding of biomass pellets

The study assessed the impact of incorporating a binder through a pressurized spraying method in the formation of solid bridges to enhance the densification of biomass pellets. Based on the preliminary findings presented in Table 1, the densification pressure (Factor A) was selected for the subsequent tests (Table 2) by gradually reducing the pressure from 20 MPa in increments of one unit. The methods of binder addition (Factor B) were established as no binder addition, spraying method, and stirring method.

Table 2. Minimum Densification Pressure Test Chart

Level	Densification pressure A (MPa)	Adding method B
1	20	No Binder
2	10	Stirring
3	p	Spraying

Note: moisture content is 10%, binder proportion is 5%, p is the densification pressure, and P<20MPa, with no consideration of durability

Statistics Software

Two types of software were used based on the four-factor mixed-level orthogonal test. Firstly, SPSS (Statistical Product and Service Solutions) was used to improve the test design further, and the Muti-Layer Perception (MLP) model was used to calculate the influence of each factor. Secondly, the Design-Expert 12.0 data analysis software was used for variance and response surface analysis of test indices.

RESULTS AND DISCUSSION

Table 3 shows 25 groups of tests. Each group was repeated three times, using the orthogonal table L25 ($2{\times}5^3$).

Table 3. Orthogonal Test Results in L25 ($2{\times}5^3$)

Number	A(MPa)	B (%)	C (%)	D	Y1	Y2(%)
1	40	10	5	2	1.064 ± 0.015	96.36 ± 0.069
2	30	12	5	1	1.032 ± 0.005	94.82 ± 0.377
3	60	12	2	2	1.043 ± 0.008	97.27 ± 0.370
4	70	16	4	2	1.175 ± 0.075	96.67 ± 0.702
5	40	16	2	1	1.050 ± 0.006	99.22 ± 0.050
6	60	16	5	1	1.061 ± 0.015	99.91 ± 0.464
7	70	18	3	1	1.039 ± 0.002	99.57 ± 0.424
8	30	16	3	2	1.085 ± 0.048	93.22 ± 0.687
9	70	10	2	1	1.085 ± 0.010	99.17 ± 0.125
10	40	14	3	1	1.022 ± 0.002	98.39 ± 0.099
11	40	18	1	2	1.118 ± 0.028	95.06 ± 0.216
12	50	18	5	1	1.064 ± 0.009	99.36 ± 0.262
13	70	12	1	1	1.033 ± 0.002	99.10 ± 0.338
14	30	14	4	1	1.043 ± 0.008	94.33 ± 0.537
15	50	12	3	2	1.047 ± 0.010	95.00 ± 0.092
16	50	16	1	1	1.072 ± 0.015	99.12 ± 0.374
17	60	10	3	1	1.062 ± 0.010	99.64 ± 0.249
18	60	14	1	2	1.113 ± 0.010	98.39 ± 0.216
19	50	14	2	1	1.023 ± 0.010	98.18 ± 0.054
20	30	10	1	1	1.094 ± 0.004	88.28 ± 0.499
21	50	10	4	2	1.073 ± 0.004	96.61 ± 0.134
22	60	18	4	1	1.053 ± 0.002	99.54 ± 0.047
23	70	14	5	2	1.180 ± 0.020	98.25 ± 0.249
24	30	18	2	2	1.091 ± 0.015	97.22 ± 0.262
25	40	12	4	1	1.042 ± 0.005	99.64 ± 0.099

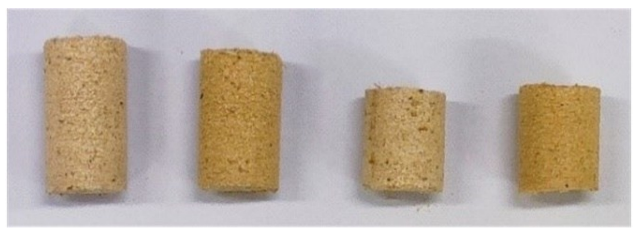
Notes: "A" represents densification pressure, "B" represents moisture content, "C" represents binder ratio, "D" represents addition methods, "Y1" represents relaxation ratio, "Y2" represents durability, "D=1" represents the binder addition method by pressurized spraying, and "D=2" represents the binder addition method by stirring.

In Table 3, parameters A, B, C, and D represent the densification pressure, moisture content, binder ratio, and addition methods, respectively. Y_1 and Y_2 represent the relaxation ratio and durability (%). Using the MLP neural network in SPSS, the influence of each factor was analyzed. The variance and response surface analysis of the test indices were carried out using Design-Expert data analysis, and the multivariate quadratic regression equation fitting between the test factors and the test evaluation indices was obtained. The response surface diagram and contour map were drawn to analyze the influence of the test factors and their interaction on the test evaluation indices. Finally, the regression model was solved using the optimized module, deriving the optimal densification conditions of biomass pellets.

The investigation focused on determining the minimum densification pressure required for the densification of biomass pellets with the inclusion of a binder through pressurized spraying, comparing it with pellets without any binder, with a binder added through stirring method, and with a binder added through spraying method. Morphological analysis of the biomass pellets was conducted using a microscope to evaluate the effect of binder incorporation in forming solid bridges on the microstructure.

The partly compressed biomass pellets are shown in Table 4.

Table 4. The Compressed Biomass Pellets

Biomass pellets / Test factors				
Densification pressure (MPa)	10	10	20	30
Moisture content	10%	10%	12%	12%
Binder ratio	4%	4%	5%	5%
Adding method	stirring	spraying	stirring	spraying
Calibre (mm)	15	15	15	15

The Influence of Test Factors on Relaxation Ratio

Weight calculation (normalization)

This paper used SPSS to design the model of the MLP neural network. The analysis process was divided into three steps: dividing the original data into a partitioned data set, model training, and results prediction. The data were divided into three blocks in the partitioned data set: “training sample”, “supporting sample”, and “test sample”. Among all the data, 70% was used as the training sample to construct the self-learning neural network model, and 30% was used as the test sample to evaluate the performance of the model. The supporting sample was temporarily not distributed in the meantime. In the training model, a hidden layer using a hyperbolic tangent activation function was constructed in the architecture of the neural network, and batch training was used to minimize the total error. The optimized algorithm selects the corresponding conjugate gradient, and the model parameters are shown in Fig. 2. The sum of squares error is 4.418%, and the relative error was 0.491%.

Through the training process, the neural network model was obtained, and the influence degree of each factor on the relaxation ratio of biomass pellets was calculated (shown in Fig. 3). The influence degree of the binder addition method, densification pressure, raw material moisture content, and binder addition ratio was 100%, 63.9%, 43.1%, and 40.9%, respectively. The tests showed that the binder addition method had the most significant influence on the relaxation ratio of biomass pellets, while other factors were less significant in comparison.

Train	sum of squares error	4.418
	Relative error	.491
	Abort rule used	The error does not decrease in one successive step a
	training time	0: 00: 00.00
Test	sum of squares error	.272
	Relative error	.184

dependent variable: relaxation ratio

a.Error calculation based on test sample

Fig. 2. Summary of relaxation ratio prediction models

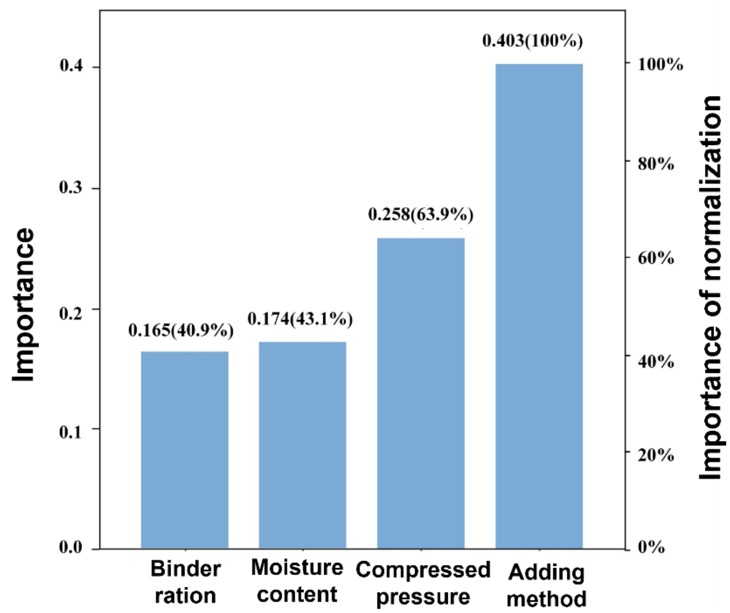


Fig. 3. Influence ratio of factors A, B, C, and D on relaxation ratio

Variance and response surface analysis

To analyze the degree of influence by densification pressure (factor A), raw material moisture content (factor B), binder addition ratio (factor C), and binder addition method (factor D) on each independent variable, the Design-Expert software was used to calculate the variance. The results showed that the smaller the significant coefficient, the greater its influence on variables, indicating that the test factor was the main influencing factor. The larger the significant coefficient, the smaller its influence on the variables, which is a secondary factor (Kang *et al.* 2020). According to the analysis of the test results, it is recommended to use the 2FI model for fitting, and the regression model was:

$$Y= k+k_1A+k_2B+k_3C+k_4D+k_5AB+k_6AC+k_7AD+k_8BC+k_9BD+k_{10}CD \quad (5)$$

The variance analysis results of the relaxation ratio are shown in Table 5. The regression equation between the relaxation ratio and test factors was:

$$Y=1.08+0.0204A+0.0182B+0.0121C+0.0223D+0.0029AB +0.0183AC+0.0143AD+0.0013BC+0.0288BD+0.0134CD \quad (6)$$

The R-squared value of the regression equation was 0.8189, indicating that fitting the relaxation ratio with the 2FI model was better. Table 5 shows that the P values of A, D, and BD were all less than 0.05, which suggests these related factors each had a significant influence on the relaxation ratio. According to the F value of each factor, the influences on the densified pellets relaxation ratio, in descending order, were the binder addition method, densification pressure, raw material moisture content, and binder ratio. The importance of each factor shown from the variance analysis is consistent with the results of the neural network analysis.

Table 5. Variance Analysis of Relaxation Ratio

Source of Variance	Sum of Squares	Degrees of Freedom	Mean Square	F-value	P-value	Significant
Model	0.0333	10	0.0033	6.33	0.0011	**
A- densification pressure	0.0037	1	0.0037	6.97	0.0194	*
B- moisture content	0.0021	1	0.0021	3.91	0.0681	
C- Binder ratio	0.0013	1	0.0013	2.44	0.1407	
D- Adding method	0.0077	1	0.0077	14.66	0.0018	**
AB	0.0000	1	0.0000	0.0560	0.8164	
AC	0.0011	1	0.0011	2.16	0.1641	
AD	0.0017	1	0.0017	3.31	0.0905	
BC	6.852E-06	1	6.852E-06	0.0130	0.9108	
BD	0.0060	1	0.0060	11.35	0.0046	**
CD	0.0013	1	0.0013	2.51	0.1357	
residual	0.0074	14	0.0005			
total	0.0407	24				

Note: P < 0.01 (highly significant**), 0.01≤P < 0.05 (significant*), P > 0.05 (not significant)

Figure 4(a) shows the response surface of the influence of the other two factors on the relaxation ratio when the two test factors were at the central level. Table 5 shows that BD had a very significant influence on the relaxation ratio. These two factors were selected to draw the response surface diagram. When the compaction pressure and binder ratio varied little, by increasing the moisture content or adding binder by stirring, the relaxation ratio significantly increased. To improve the quality of densified pellets, the moisture content can be appropriately reduced, or the spraying method can be used. Fig. 4(b) is the contour map corresponding to the response surface, which reflects the significance of the interaction between factors. The graph shows that the interaction between raw material moisture content and the binder addition method had a pronounced effect on the relaxation ratio. When the moisture content of raw material was high, adding the binder by spraying can significantly reduce the relaxation ratio of the densified pellets. The purpose of adding binder by spraying is to make smaller binder particles more evenly distributed among bio pellets, thus producing stronger adhesion and improving the quality of biomass pellets. Therefore, the relaxation ratio of densified biomass pellets obtained by the spraying method is smaller than that by the stirring method.

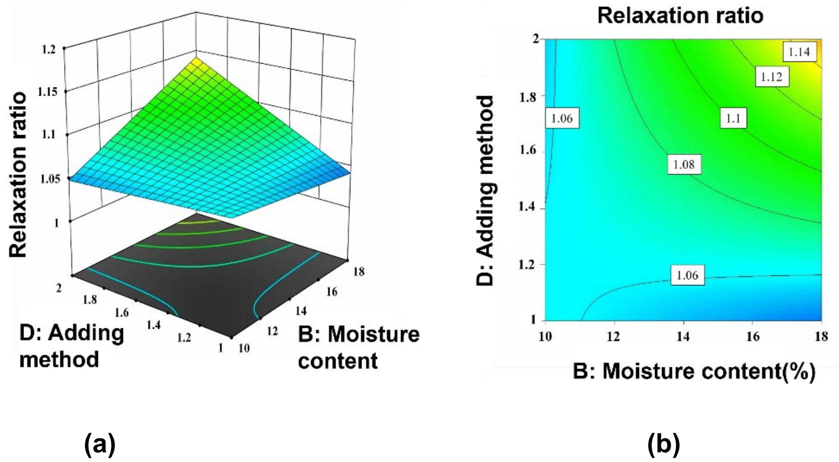


Fig. 4. (a) Response surface plot for effects of moisture content and binder adding method on relaxation ratio. (b) Contour plot for effects of moisture content and binder adding method on relaxation ratio.

Minimum Densification Pressure Test Analysis

According to the pre-test results in Table 1, the densification pressure was gradually reduced to 20 MPa, the binder addition ratio was fixed at 5%, and the raw material's moisture content was maintained at 10%. Compression tests were conducted without binder, with binder added through stirring method, and with binder added through spraying method to assess the densification effect and evaluate the surface quality of the densified pellets as the key performance indicator. Three samples were selected from each test group, and the average values were compared to ascertain the densification efficacy. The primary objective of this study is to validate the applicability of the pressurized spraying method and ascertain its influence on the minimum densification pressure required for the densification of biomass.

As shown in Table 6, when the densification pressure was 2 MPa, the biomass raw material without binder and the biomass raw material with binder added by the stirring method could not be densified. The biomass raw material with binder added by spraying method could be densified, but the sample pellets had cracks on the surface and were easy to break. After the densification pressure was 4 MPa, biomass pellets without binder, stirring method, and spraying method with binder were densified. Compared with the stirring method, the densified pellets obtained by the spraying method had fewer surface cracks and a better densification effect. When the densification pressure was lower than 2 MPa, no binder, stirring method, or spraying method with binder could not make the raw material densified. The 2 MPa densification effect is shown in Fig. 5.

Compared with no binder and binder added by stirring method, the method of pressurized spraying of binder reduced the minimum densification pressure of the biomass raw material and promoted the densification of biomass.

Table 6. Minimum Densification Pressure Analysis

Densification pressure (MPa)	Adding method	Can it be densified?
20	Spraying	Yes
20	Stirring	Yes
20	No binder	Yes
10	Spraying	Yes
10	Stirring	Yes
10	No binder	Yes
8	Spraying	Yes
8	Stirring	Yes
8	No binder	Yes
6	Spraying	Yes
6	Stirring	Yes
6	No binder	Yes
4	Spraying	Yes
4	Stirring	Yes
4	No binder	Yes
2	Spraying	Yes
2	Stirring	No
2	No binder	No
1	Spraying	No
1	Stirring	No
1	No binder	No

Note: moisture content is 10%, binder proportion is 5%, no consideration of durability

(a) (b) (c)

Fig. 5. (a) No binder densification effect; (b) stirring method densification effect; (c) spraying method densification effect.

Microscopic Observation of the Solid Bridge Structure Formed by the Binder

The room temperature densification test of poplar sawdust involved adding a binder to form solid bridges through pressurized spraying. This test allowed for the analysis of the relationship between each factor and the densification effect of biomass raw materials from a macroscopic perspective. However, it did not provide a way to verify the effect of different binder addition methods on the microscopic morphology of materials. Therefore, in this study, a Leica S8 APO body microscope was used to observe the distribution of solid bridges formed after binder curing on the cross-section of densification pellets at 40 times magnification. This allowed for a more comprehensive analysis of the influence of solid bridges formed by different binder addition methods on the densification effect.

Initially, biomass pellets without any binder addition were examined under a microscope to establish a baseline comparison. Figure 6(a) illustrates tightly bonded materials intertwined and embedded with each other. Subsequently, pellets produced with a binder added at a mass ratio of 4% through stirring and spraying methods were analyzed.

Microscopic observation revealed the presence of solid bridges formed by binder curing, identifiable by dark-colored regions. Figures 6(b) and (c) show that when the binder was added through stirring, the solid bridges appeared unevenly distributed or in clumps (b), whereas the spraying method yielded a more uniform distribution of smaller solid bridges evenly mixed within the poplar sawdust pellets (c).

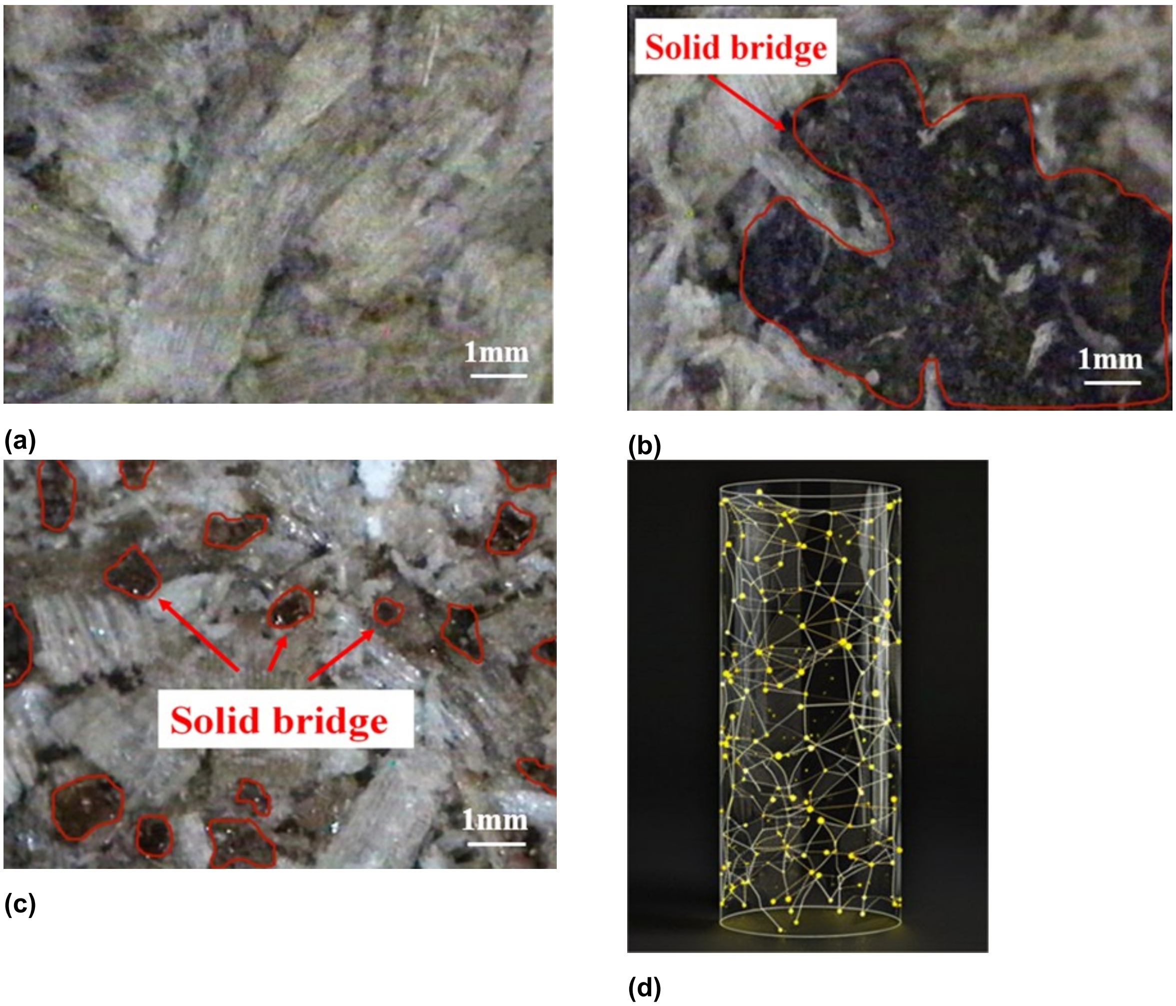


Fig. 6. (a) Micrograph of the section of the pellets without adding binder; (b) Micrograph of the section of the pellets with binder added by stirring method; (c) Micrograph of the section of the pellets with binder added by spraying method; (d)Solid bridge connection structure within the pellets

The transition of the binder from a liquid to a solid state resulted in the formation of solid bridges that enhanced particle adhesion. This solidification process tightened the material, facilitating densification and improving overall quality. Comparative analysis of cross-sectional micrographs of pellets produced through different addition methods highlighted that the pressurized spraying method enabled a more uniform distribution of the liquid binder among the materials during room-temperature densification. This uniform distribution led to the formation of a distinct and uniform solid bridge structure, reinforcing particle bonding and ensuring a tighter amalgamation. Consequently, the pressurized spraying method was shown to be more effective in promoting pellet densification.

It is supposed that the solid bridge structure inside the biomass pellets may form a mesh structure, as shown in Fig. 6(d), and its solid bonding is expected to enhance the durability, relaxation density, and relaxation ratio of the biomass pellets, thus promoting biomass densification. However, direct observation of its internal structure has not yet been realized, so the study remains to be explored in the future.

CONCLUSIONS

1. The binder addition method was identified as the most critical factor, with 100% importance being assigned to it for the normalization of the relaxation ratio, and the pressurized spraying method outperformed the stirring method.
2. The optimal process parameters were a densification pressure of 50 MPa, a moisture content of 14%, a binder ratio of 4%, and the spraying addition method.
3. The minimum densification pressure was reduced drastically to 2 MPa by the spraying method, and pellet durability and relaxation density were markedly improved, especially under low pressure.
4. Finer and more uniformly distributed solid bridges were observed through microscopic analysis, which were confirmed to enhance inter-particle bonding and mechanical strength.
5. The overall quality of biomass pellets was effectively promoted, and densification was facilitated by the spraying introduction of an optimal quantity of binder, with solid bridge formation being enhanced during the process.

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Conflict of Interest

The authors declare that they have no known competing financial interests or personal relationships that could have influenced the work reported in the article.

Use of Generative AI

The authors confirm that no artificial intelligence (AI) tools were used in any stage of the preparation of this manuscript, including text generation, data analysis, or reference collation. All content was solely prepared by the authors.

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