Optimization of Parameters Associated with Pellets Made from Biomass Residue from Anaerobic Digestion Using Box-Behnken Design

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Fuel pellets were produced with biomass residue from anaerobic digestion. Single-factor experiment and Box-Behnken design were employed to investigate the effects of pellets-associated variables on the mechanical properties of pellets, and the optimal condition was determined. The results revealed that the pellets-associated variables, including particle size, moisture content, die temperature, and molding pressure had significant influences on the mechanical properties of pellets, such as compressive resistance (CR), durability (DU), and density (DE). The regression models were obtained with the R^2 values of 0.9802, 0.9628, and 0.9610 for CR, DU, and DE, respectively, suggesting that the differences between the actual and predicted values could be explained by the regression models. The optimal values of pellets-associated variables were determined (particle size of 0.4 mm, moisture content of 8.4%, die temperature of 115 °C, and molding pressure of 150 MPa); the corresponding responses were 1470 N, 99.6%, and 1180 kg/m^3 for CR, DU, and DE, respectively. The results of verification showed a good agreement between the predicted data and experimental outputs. In summary, a novel approach was presented for the preparation of pellet fuels made from biomass residue from anaerobic digestion, and a reliable reference was therefore provided for the comprehensive utilization of biomass materials.

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INTRODUCTION

The global consumption of natural energy is increasing due to the rapid industrial variations. However, the storage of natural energy is limited, especially with regard to fossil fuels. Some fossil fuels, such as crude oil, coal, and gas, may be exhausted in the next 50 years (Shafiee and Topal 2009). Therefore, it is essential to explore renewable resources of energy to globally alleviate the energy crisis and meet the energy demand in the future (Callegari *et al*. 2019). In addition to wind energy and solar energy, biofuel is recognized as one of the renewable, sustainable, and environmentally friendly resources of energy, and it can be an appropriate alternative for fossil fuels (Ramezanzade and Moghaddam 2018). Biofuels, including synthetic biofuels, bioethanol, biogas, biodiesel, and bio-hydrogen, are prepared from biomass or their residuals (Lin *et al*. 2014). Compared with the traditional resources of energy, biofuels possess several attractive properties, such as being cost-effective, low emission of greenhouse gases, broadly accessible, *etc*. (Li *et al*. 2015). Hence, further in-depth research on biofuels should be conducted.

In China, more than 3 billion tons of manures and 650 million tons of crop straws are generated annually, which are important raw materials that can be used to produce biological products (Awasthi *et al*. 2019). Although manure and crop straw are high-quality biomass resources, their huge outputs have caused some problems for the development of agriculture and breeding industries. Anaerobic digestion is currently regarded as one of the most environmentally friendly methods for treating various organic substances, including sewage sludge (Stefaniuk and Oleszczuk 2015). Due to its potential in producing renewable energy (*i.e.* methane content of biogas), anaerobic digestion has attracted attention (Baetge and Kaltschmitt 2018). With the rapid development of biogas industry, China's biogas consumption is annually 19 billion cubic meter (m^3). Correspondingly, a large amount of biomass residue from anaerobic digestion (BRAD), which are difficult to dispose and may easily lead to secondary pollution, are produced during anaerobic digestion (Meng *et al*. 2018). Generally, the most common method of utilizing BRAD is to use it as organic fertilizer in agriculture, as it possesses several merits of providing nutrients for plants, enhancing the moisture and buffering capacities of soil (Bai *et al*. 2020). However, the direct application of BRAD to soil may cause leaching of the nutrients from the soil and the deterioration of water, due to the mobility property of its water-soluble contents and an excessive content of heavy metals produced by the use of animal feed additives, resulting in serious environmental concerns (Govasmark *et al*. 2011). Therefore, it is highly essential to develop a novel and environmentally friendly approach for the resourceful utilization of BRAD.

Given the drawbacks of traditional treatment for BRAD, the conversion of BRAD into compressed solid biofuels may be an alternative technique for addressing this challenge. BRAD can be converted into pellets or briquettes using densification technology, including pelletization, briquetting, and extrusion (Liu *et al*. 2014). The purpose of densification is to agglomerate small particles into larger particles by a mechanical process combined with moisture, heat, and pressure (Gilvari *et al*. 2019). Densification increases the bulk density of the biomass materials while reducing the expenses of handling, transportation, and storage; it has been applied widely to the biomass industry in the developed countries (Prawisudha *et al*. 2012). A relevant study showed that the densification of the biofuel pellets could increase the density up to 1,000 to 1,200 kg/m^3; the volume was reduced by 8 to 19 times compared with the raw biomass (Mostafa *et al*. 2019). In addition, biofuel pellets possess a large number of advantages, such as a low level of pollution, a high heating rate, and less emission of odors. Thus, they can be applied directly to residential heating stoves, power plants, and heating boilers (Chen *et al*. 2011). In summary, using BRAD to produce biofuel not only can provide an approach for the treatment of BRAD, but also they can be helpful to meet the large demand for energy in the future. Meanwhile, biofuel pellets from BRAD could improve the utilization value of BRAD. Hence, it is essential to investigate the effects of different parameters on physicochemical properties of pellets from BRAD, as well as the optimal conditions for producing pellets with BRAD.

The physical quality of a produced pellet is mainly affected by the properties of raw materials, including chemical composition, moisture content, particle size, and parameters associated with manufacturing processes, such as forming temperature, applied pressure, holding time, and die geometry (Lestander *et al*. 2012). Numerous researchers have explored the effects of associated parameters and properties of biomass materials on the physical and chemical properties of pellets made from single or mixture of different types of biomass materials under different conditions. Rhén *et al.* reported that the density and compression strength of the pellets from sawdust were elevated by increasing the pelletizing temperature and lowering the moisture content of raw materials (Rhén *et al*. 2005). Puig-Arnavat *et al*. (2016) found that the moisture content of 10 wt.% was optimal for 6 biomass feedstocks, in which the friction increased first and then declined with the increase of die temperature. Stasiak *et al.* (2017) revealed that the density of pellets made from a blend of pine sawdust, wheat straw, and rapeseed straw increased with the raised percentage of the two straws in the mixture and the compaction pressure.

Response surface methodology (RSM) is an effective method to optimize process conditions, and it can determine the influence of various factors and their interactions on the indexes under investigation (Alhajabdalla *et al*. 2021). Generally, it is used for mapping a response surface over a particular region of interest (ROI), optimizing the response, or for selecting operating conditions to achieve target specifications or a customer's requirements (Cheng *et al*. 2021). In particular, RSM based on a Box-Behnken design (BBD) is advantageous for the analysis on the effects of various factors on the responses by simultaneously modulating the effective parameters, formulating second-order polynomial equations of multiple factors, and describing interactions among independent variables (Belgada *et al*. 2021). Compared with other response surface designs, BBD possesses attractive advantages in the necessity of performing a small number of experiments and a higher efficiency. To date, BBD has been successfully utilized in several engineering fields.

In the present study, BRAD were selected as raw materials to produce a type of solid biofuel pellet using heating molding process. This study explored the effects of parameters, such as particle size and moisture content of raw materials, die temperature, and molding pressure on the mechanical properties of the pellets produced using BRAD. The optimal values of the mentioned parameters under different conditions were determined.

EXPERIMENTAL

Materials

Biomass residue from anaerobic digestion (BRAD), as used in the current study, mainly consisted of corn straw and cow manure. These components were obtained from the Biomass Energy Laboratory of Heilongjiang Bayi Agricultural University (Daqing, China) after the fermentation process, as shown in Fig. 1a. First, the collected residues were air-dried at room temperature for 10 days. The dried residues were ground using a hammer mill of YB-1000A (Yongkang Sufeng Gongmao Co., Ltd., Zhejiang, China) and then passed through a sieve with different screen sizes to obtain desired particle sizes (0.2, 0.4, 0.6, 0.8, 1.0, 1.2, and 1.4 mm) (Fig. 1b). The chemical compositions of BRAD and other biomass (wheat straw, corn stover, rice straw, Chinese fir sawdust and camphor sawdust) are presented in Table 1.

Before pelletizing, the grounds were dried in a convection oven at 40 °C until the weight of samples remained constant. The moisture content of the feedstocks was adjusted by addition of purified water (predetermined amount) to the grounds. They were subsequently calculated on a wet-weight basis. After that, the feedstocks were put into a plastic sealed container and stored at 4 ± 0.5 °C for 2 days for equilibration of the moisture (Zhang and Guo 2014).

Table 1. Chemical Compositions of Biomass Species

Biomass	Components			Reference
	Lignin (%)	Cellulose (%)	Hemicellulose (%)	
BRAD	25.76	33.77	13.28	Current
Wheat straw	7.61	42.51	22.96	Mani et al. 2006
Corn stover	3.12	31.32	21.08	
Rice straw	9.22	41.33	24.60	Jiang *et al.* 2015 and Li *et al.* 2014
Chinese fir	27.61	36.22	20.82	
Camphor	24.40	38.87	20.82	

Fig. 1. Experimental materials of (a) initial BRAD and (b) ground BRAD

Pelletizing Apparatus and Procedures

A pelletizer used in the test was manufactured at Heilongjiang Bayi Agricultural University (Fig. 2). The single press consisted of a cylindrical die lagged with an electromagnetic heating unit, a spiral water-cooling unit, and a computer control system. The inside diameter and length of cylinder were 15 and 50 mm, respectively, and the diameter and length of piston were 15.3 and 70 mm, respectively. A thermocouple placed into a cylinder was used to ensure that the temperature reached the desired value. The die temperature can be detected and controlled by a computer system in the range of 0 to 400 °C.

(1) Frame; (2) Temperature sensor; (3) Molding unit; (4) Water cooling unit; (5) Temperature controller; (6) Controlling computer; (7) Thermocouple; (8) Heating unit; (9) Base; (10) Pellet; (11) Cooling unit; (12) Cylinder; (13) Piston

Fig. 2. Photo and schematic illustration of the single press unit

The preparation of the pellet was conducted as follows. Prior to each test, the heating system was run for 15 to 20 min for the desired die temperature. A total of 4 g of ground residues (dry basis) with a specific moisture content was placed in the die. The piston with a preset load applied by the universal testing machine began to compress the materials (loading speed, 5 mm per min). The loading force was controlled by the computer, which recorded the force and displacement curves. After the loading force preset was reached, the pressure was kept at a full pressure for twenty seconds. When 25 °C or below was reached for the die temperature, the cooling system was closed, and the pellet was pressed out from the mold. The produced sample was cooled at an ambient temperature for 10 min. The sample was stored in a plastic bag at 4 ± 0.5 °C.

Measure of Compressive Resistance

Compressive resistance, sometimes called crushing resistance or hardness, is defined as the maximum crushing force of a pellet or a briquette that can withstand before cracking or breaking (Kaliyan and Morey 2009). It is an essential property of densified pellets because of its effects on the mechanical strength and efficiency during the processes of handling, transportation, and storage. Compressive resistance can be measured using different measuring devices with the same working principle. There is currently no standard method to assess the compressive resistance of densified biomass pellets or briquette; however, the compressive resistance of the densified biomass materials can be evaluated by the maximum force that can withstand until failure or breakage (Kambo and Dutta 2014; Hu *et al*. 2016). In the present study, the compressive resistance of the pellets was measured using a WDW-200E Computerized Electronic Universal Testing Machine (Jinan Time Shijin Testing Machine Co., Ltd., Shandong, China). The sample was placed between two horizontal steel plates, and an axial force was applied to the sample at a constant rate of 10 mm per minute until failure and breakage. In the compression process, force-displacement curves were drawn by the computer, and the peak of the force was determined as the compressive resistance of the pellet.

Measure of Durability

Durability is a measure to assess the ability of densified biofuel pellets to withstand against destructive forces of compression, impact, and shear during handling and transportation processes (Gilvari *et al*. 2019), because breakage of pellets during processing and transportation has a negative effect on the supply chain. Previously, different devices of rotating drum, tumbling can, and Holmen and Ligno testers have been applied to measure the durability of the densified products. The tumbling can method simulates the mechanical handling of pellets and predicts the possible fines produced by collision of pellets against each other and against the walls of a defined rotating chamber. In the current report, the tumbling can method was employed to determine the durability of the densified pellets. Before each test, a known mass of 10 pellets was applied to the rotating chamber. The rotation speed was fixed to 50 ± 2 rpm, and the revolution lasted for 10 min. After stopping the test, pellets were sieved using a sieve with the screen size of 0.8 times a pellet's diameter. Finally, the durability was calculated using Eq. 1,

$$DU = M_e/\mathrm{M_a} \times 100 \quad (1)$$

where DU refers to the durability of the densified pellets (%), M_e refers to the initial mass of the tested pellets before the test (g), and M_a refers to the mass of the tested pellets after the test (g).

Measurement of Density

The density of densified pellets is mainly considered as an essential index related to management and transportation processes. It presents the ratio of the sample mass to its volume with inclusion of inner porosity. After 24 h of pelletizing, each pellet was weighed using an analytical balance with an accuracy of 0.001 g. The dimensions of the pellets, including diameter and length, were gauged with a Vernier caliper with a precision of 0.01 mm. The density of the densified pellet was calculated as the mass of the pellet divided by its volume. As reported previously, an optimal density of a single pellet is in the range of 1,000 to 1,400 kg/m^3 (Emadi *et al*. 2016).

Table 2. Variables and Levels for Single-Factor Experiment

Variables	Levels						
Particle sizes (mm)	0.2	0.4	0.6	0.8	1.0	1.2	1.4
Moisture content (%)	4	6	8	10	12	14	16
Die temperature (˚C)	60	80	100	120	140	160	180
Molding pressure (MPa)	80	100	120	140	160	180	200

Experimental Design and Optimization

The particle size, moisture content, die temperature, and molding pressure were selected as variables, and were coded of x_1, x_2, x_3, and x_4, respectively. The compressive resistance, durability, and density, with the codes of y_1, y_2, and y_3, respectively, were deemed as responses. Single-factor experiments were performed to determine the reasonable ranges of variables in the BBD experiment. Table 2 lists the variables and their values in the single-factor experiments. The principle of changing one factor, while other factors were fixed at a middle level was used during the single-factor experiments.

Based on the results of single-factor experiments, BBD was employed to assess the main, interaction, and quadratic effects of many variables on the responses, and the optimal condition in the pelleting process was determined. The variables and their values in the BBD experiment are listed in Table 3.

Table 3. Variables and Levels Used in BBD Experiment

Variables	Symbols	Levels		
		-1	0	+1
Particle sizes (mm)	x_1	0.4	0.7	1.0
Moisture content (%)	x_2	8	10	12
Die temperature (˚C)	x_3	100	120	140
Molding pressure (MPa)	x_4	120	140	160

For BBD, the required number of runs can be calculated by Eq. 2 (Rakhmania *et al*. 2021),

$$N = 2k_0(k_0-1)+C_0 \tag{2}$$

where k_0 is the number of variables, and C_0 is the number of center points, which is equal to 5 in this study.

Thus, in the current research, a total of 29 runs were carried out with the purpose of minimizing the effect of unexplained variabilities on the response observed. The BBD method has the potential to analyze several factors with the minimum experimental trials compared with other response surface methods. Based on analysis of variance (ANOVA) of the experimental data of selected points, the model of objective function on the variables was obtained, and the response surface was plotted. Finally, optimal conditions could be calculated from the final model and verified by an actual experiment (Le *et al*. 2019).

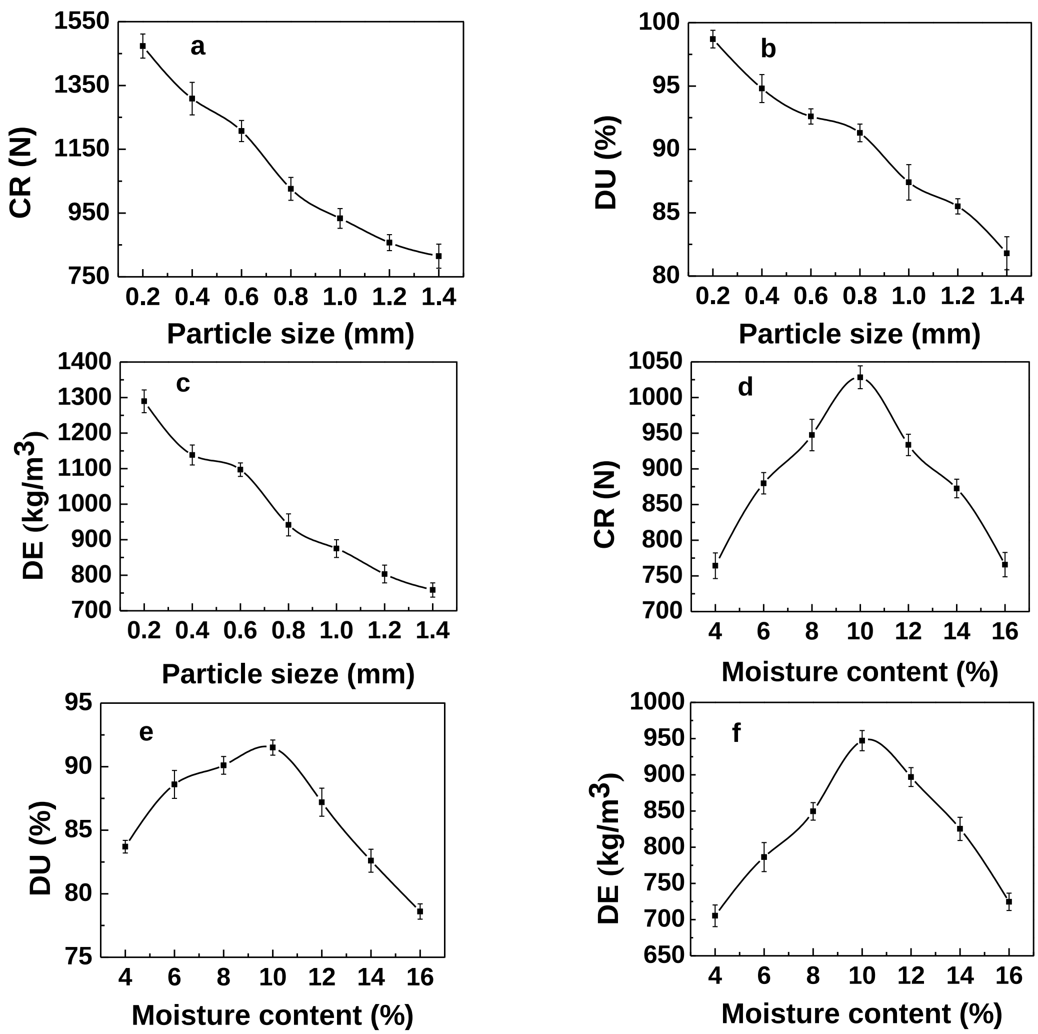
Statistical Analysis

Origin 8.5 software (OriginLab Corporation, Northampton, MA, USA) was used for graph-based analysis. Using Design Expert 8.0.6 software (Stat-Ease Inc., Minneapolis, MN, USA), ANOVA was performed and the 3D response surface was plotted. Duncan's multiple range test was used to calculate the least significant difference ($P<0.05$). All experiments were repeated three times.

RESULTS AND DISCUSSION

Single Factor Experiment

The effects of particle size, moisture content, die temperature, and molding pressure on the mechanical properties of CR, DU, and DE of the pellets are shown in Fig. 3.



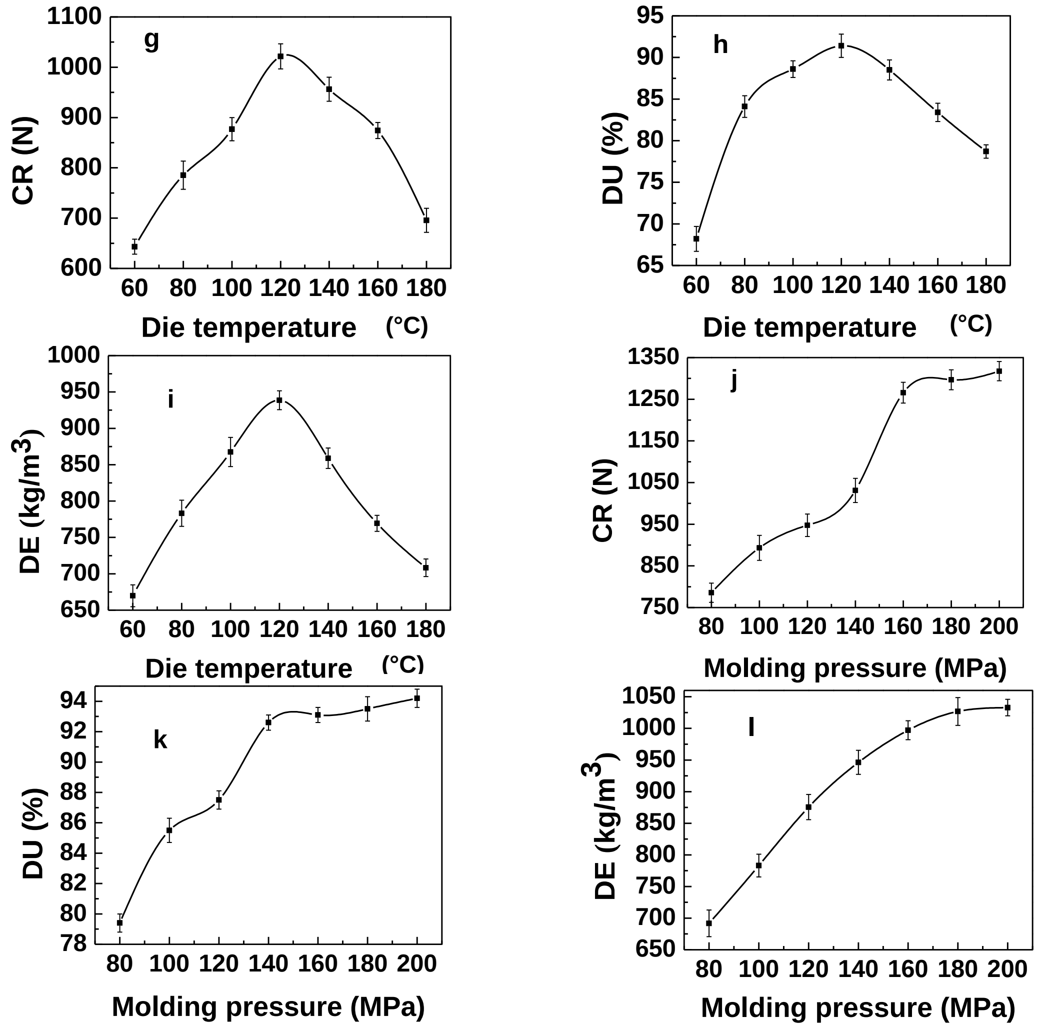


Fig. 3. The effects of independent variables on CR, DU and DE. (a) - (c) Particle size, (d) - (f) Moisture content, (g) - (i) Die temperature, and (j) - (l) Molding pressure. (CR: compressive resistance, DU: durability, DE: density)

Effect of particle size on compressive resistance, durability, and density

The effects of particle size on mechanical properties of the pellets are depicted in Fig. 3a-c. The experiment was conducted at a moisture content of 10%, die temperature of 120 °C, and molding pressure of 140 MPa. The relationships between CR, DU, and DE and particle size were negative. Furthermore, the association between particle size and the three responses was statistically significant ($P<0.05$) according to the one-way ANOVA of the experimental data. This result confirmed that particle size is an important factor influencing the mechanical properties of the pellets. When particle size increased from 0.2 to 1.4 mm, CR, DU, and DE decreased by 658.8 N, 16.9%, and 531 kg/m^3, respectively. Several reasons can be used for interpreting the variation. First, finely ground biomass can greatly increase the inter-particle contact area and decrease the inter-particle distance, contributing to the development of the solid bridge between adjacent particles at the points of contact. Common inter-molecular attractive forces, including hydrogen bonds, van der Waals forces, and magnetic forces, were produced during the densification process. The inter-molecular forces can cause particles to adhere to each other and generate strong interlocking bonds between adjacent particles, which are advantageous for improving the quality of the pellets. A previous study demonstrated that van der Waals forces can be strongly affected by the particle size of the biomass materials (Stelte *et al*. 2011). Generally, the finer the grind, the higher the quality of pellet. Fine particles usually accept more moisture than large particles and, therefore, undergo a higher degree of conditioning. Also, large particles are fissure points that cause cracks and fractures in pellets (Kaliyan and Morey 2009). Second, reducing the particle size can enhance the mobility of the feedstock in the cylinder during the pelletizing, causing flow of smaller particles into the void fractions of biomass particles, thereby resulting in an increase in strength and density of the pellets. Moreover, decreasing the particle size could provide a greater surface area, promoting absorption of moisture and heat, which is highly significant for activating the binding properties of some natural binders (*e.g*., lignin, starch, cellulose, protein, and hemicellulose) (Carone *et al*. 2011). A previous study recommended a particle size of 0.5 to 0.8 mm to produce pellets with a promising quality (Kaliyan and Morey 2009). Though fine particles produce high-quality biomass pellets with higher hardness, durability, and density, fine grinding is undesirable due to the raised production cost. Therefore, in the present study, the particle size of the BRAD used for the BBD experiment was determined to be in the range of 0.4 to 1 mm.

Effect of moisture content on compressive resistance, durability, and density

The effects of moisture content on the mechanical properties of CR, DU, and DE are displayed in Fig. 3d-f. The data could be obtained from the tests carried out at a particle size of 0.8 mm, die temperature of 120 °C, and molding pressure of 140 MPa. The values of CR, DU, and DE increased first and then decreased with the raising of moisture content from 4% to 16%. As moisture content increased from 4% to 10%, the CR, DU, and, DE, increased from 764.3 to 108.4 N, 83.7% to 91.5%, and 705.3 to 947.2 kg/m^3, respectively. With a further increase in the moisture content, CR, DU, and DE decreased by 262.6 N, 12.9%, and 222.6 kg/m^3, respectively. This result was consistent with that reported previously, which demonstrated that the peak values of CR, DU, and DE were observed at an optimum moisture content of 10%, whereas higher values of moisture content negatively influenced the densification quality of the pellets as it exceeded the reasonable range (Kanliyan and Morey 2009; Zainuddin *et al*. 2011). Based on the one-way ANOVA of the data, the moisture content had an extremely significant effect ($P<0.01$) on CR and a significant effect ($P<0.05$) on DE; however, its effect on DU was not statistically significant ($P>0.05$). The moisture content, which is generally regarded as the most important factor affecting the mechanical properties of the pellets, acts as a lubricant and a binder in the pelleting process (Li *et al*. 2015). A thin film of water around the particles would exhibit bonds via capillary sorption between particles. In addition, the increase of water content is conducive to the briquetting process, as some water-soluble compounds (*e.g*., starch, sugar, soda ash, sodium phosphate, potassium salt, calcium chloride, *etc*.) are present in the feed. Once the protein and carbohydrate were squeezed out from the materials, they flowed into the gaps between particles with water in response to capillary pressure, which increased the inter-particle bonding forces, leading to the improvement of the pellet quality. After pelletizing, many chemical and physical changes, *e.g*., crystallization of some ingredients, chemical reactions, hardening of binders, and solidification of melted components are induced. However, the mechanical properties of the pellets decreased as the moisture content of the feed exceeded the optimum value. The decrease of CR, Du, and DE at a higher content of moisture may be attributed to the fact that excessive water molecules within the particles may generate a thick water layer on the surface of the particles because of the incompressibility of water, preventing the release of natural binder from the particles and resulting in less creation of the hydrogen bonds between polymers of particles. Additionally, the water layer caused an extra particle-to-particle sliding, thereby weakening the adhesion and cohesion forces from one particle to another, which may help to explain the decrease of quality under high moisture content conditions. Therefore, the optimum moisture content of the materials needs to be determined to achieve high-quality pellets. In the present research, the value of moisture content applied to the BBD experiment was finally identified as 8%-12% to maximize the responses.

Effect of die temperature on compressive resistance, durability, and density

The quality of pellets with respect to the die temperature at a particle size of 0.8 mm, a moisture content of 10%, and a molding pressure of 140 MPa was compared, and the results are shown in Fig. 3g-i. The CR and DU first increased when die temperature raised from 80 to 140 °C, and then reduced with the raised die temperature. Moreover, the peak values of CR (1021.6 N), DU (91.4%), and DE (938.7 kg/m^3) were observed at a die temperature of 120 °C in the current research. A similar result has been reported previously, which revealed that the quality of pellets increased with the elevation of die temperature from 30 to 110 °C (Li *et al*. 2014). Contrary to this conclusion, the negative effects of die temperature on the quality of pellets were demonstrated in some previous studies (Ishii *et al*. 2014; Said *et al*. 2015). The effects of die temperature may be due to the differences in the pellets-associated parameters, type of materials, and test conditions. The positive effect of die temperature may be attributed to its induced function on some chemical ingredients, such as lignin, cellulose, hemicellulose, starch, and protein, which are considered as natural binders. In general, the synergistic effects of lignin softening and protein denaturation, contributing to the formation of a solid bridge between and within particles, are the principal binding mechanisms in the pelletization process. Under a lower die temperature, the binding role of the natural binders contained in the materials was limited; consequently, the bonding forces related to the pellets are mainly short-range forces, including van der Waals forces, hydrogen bonds, and mechanical interlocking. Nevertheless, when die temperature reached the glass transition, which has been reported at a temperature of 75 to 150 °C for a variety of biomass materials, the lignin and hemicellulose inside the materials were softened and squeezed out, and filled in the gaps of particles under the interaction of elevated temperature and pressure.

Once exiting the pelletizer, the pellets were left to cool down, lignin was hardened, and protein was re-associated, significantly enhancing the strength, durability, and density of pellets. In addition, as the molding temperature was in an appropriate range, the increase in die temperature led to the plasticization of lignocellulosic fibers, resulting in the reduction of the modulus of elasticity of biomass particles, which made the material more flexible. Consequently, it caused the reduction of empty spaces within and between particles, which contributed to improvement of the pellets' quality. In contrast, when die temperature exceeded glass transition or even more, the higher temperature decreased the quality of pellets. This may be due to that an elevated temperature, nearly all of the moisture inside the materials would be evaporated, which negatively influenced the quality of pellets. A further increase in die temperature would cause brittle protein after excessive denaturation, which may be detrimental to the pelletization. Consequently, aiming to obtain high-quality pellets, the rational values of die temperature used in the BBD were set within the range of 100 to 140 °C.

Effect of molding pressure on compressive resistance, durability, and density

The effects of molding pressure on CR, DU, and DE of the pellets are illustrated in Fig. 3j-l. During the densification process, the molding pressure applied in this study was 80, 100, 120, 140, 160, 180, and 200 MPa when particle size, moisture content, and die temperature were kept at 0.8 mm, 10%, and 120 °C, respectively. The maximum values of CR, DU, and DE were 1314.4 N, 93.4%, and 1072 kg/m^3, respectively. Significant correlations between molding pressure and CR and DE were observed by one-way ANOVA. Additionally, molding pressure showed to play an extremely significant role in DU. It has been reported that applied pressure positively influences the quality of pellets during the pelletization process, as it can activate different binding mechanisms within the feed particles (Carone *et al*. 2011; Poddar *et al*. 2014; Said *et al*. 2015; Guo *et al*. 2016). At a low pressure, the particles rearranged and maintained their original physical properties, reducing the volume of the raw materials to a certain extent; however, these changes had slight effects on CR and DU, because there was no effective molecular binding force between particles, which may be the possible reason for the lower mechanical properties of the pellets under a low pressure. Nevertheless, at a high pressure, elastic and plastic deformation of the particles occurred, which caused flow of the smaller particles into the empty spaces between particles. When particles were close together, the inter-particle bonding was formed. Moreover, the softened natural binding components in the materials, *e.g*., lignin, water soluble carbohydrate, cellulose, protein, and starch, were pressed out and diffused from one particle to another at the points of contact to act as a binder. These features facilitate formation of van der Waals forces and electrostatic forces and hydrogen bonds. Subsequently, the higher molding pressure applied, the higher pellet quality obtained. To date, it has been a common industrial technique to improve adhesion by increasing pressure to enhance molecular contact between adjacent molecules. Several studies have suggested that the expected pressure in a pellet mill should be in the range of 100 to 150 MPa, as well as 100-200 MPa in a roll press (Thomas *et al*. 1997; Dec *et al*. 2003). Considering that the molding pressure is associated with the production cost, the final values of the molding pressure used in BBD were in the range of 120 to 160 MPa.

Box-Behnken Design Experiment

The BBD experimental results are listed in Table 4.

Fitting of data to the models and statistical analysis

The data were fitted individually to multiple polynomial mathematical models, which are shown in Table 5. Based on the four selected factors, the possible model could be linear, 2FI, quadratic, or cubic. Results showed the P-value (0.2878) for cubic versus quadratic model was more than 0.05, the P-value for quadratic versus 2FI was less than 0.0001, and the P-value (0.9648) for 2FI versus linear was more than 0.05. Generally, a low order model is chosen if the comparison of low order and high order model is not statistically significant, otherwise a higher order model is chosen. Therefore, the quadratic model was chosen in the work.

Table 4. Values of Compressive Resistance, Durability, and Density in the BBD Experiment

Run	Variable levels				CR, y_1(N)	DU, y_2(%)	DE, y_3(kg/m^3)
	x_1	x_2	x_3	x_4			
1	0.4	8	120	140	1328.55	96.4	1142.1
2	1	8	120	140	1056.5	88.5	929.6
3	0.4	12	120	140	1198.5	92.4	1017.3
4	1	12	120	140	865.3	90.1	814.2
5	0.7	10	100	120	1007.8	86.4	881.4
6	0.7	10	140	120	747.5	85.2	788.3
7	0.7	10	100	160	1278.5	93.3	998.6
8	0.7	10	140	160	998.6	89.5	986.6
9	0.4	10	120	120	1195.7	93.8	1018.1
10	1	10	120	120	987.3	90.5	907.8
11	0.4	10	120	160	1516.5	99.6	1239.5
12	1	10	120	160	1103.6	90.5	1011.4
13	0.7	8	100	140	1075.3	89.6	933.2
14	0.7	12	100	140	890.5	85.6	866.7
15	0.7	8	140	140	886.5	87.2	798.4
16	0.7	12	140	140	751.3	82.2	816.4
17	0.4	10	100	140	1359.5	94.2	996.7
18	1	10	100	140	1023.7	89.5	895.6
19	0.4	10	140	140	1159.8	91.5	1007.6
20	1	10	140	140	825.6	84.6	827.1
21	0.7	8	120	120	975.5	90.1	916.5
22	0.7	12	120	120	965.6	87.1	906.4
23	0.7	8	120	160	1254.9	94.3	1108.6
24	0.7	12	120	160	1100.6	89.8	985.1
25	0.7	10	120	140	1148.6	92.1	976.8
26	0.7	10	120	140	1108.7	90.8	925.4
27	0.7	10	120	140	1107.9	92.7	954.2
28	0.7	10	120	140	1133.6	91.6	937.3
29	0.7	10	120	140	1125.3	91.4	954.8

Table 5. Model Summary Statistics

Response	Models	R^2	Adjusted R^2	Predicted R^2	S.D.	Remarks
CR	Linear	0.7188	0.6719	0.5668	102.5	
	2FI	0.7381	0.5926	0.1646	114.22	
	Quadratic	0.9802	0.9605	0.8918	35.57	Suggested
	Cubic	0.9937	0.9708	0.2872	30.59	Aliased
DU	Linear	0.5333	0.4555	0.2834	2.72	
	2FI	0.5858	0.3556	-0.3085	2.96	
	Quadratic	0.9628	0.9256	0.8085	1.01	Suggested
	Cubic	0.9932	0.9681	0.7907	0.66	Aliased
DE	Linear	0.6298	0.5681	0.4225	67.04	
	2FI	0.67	0.4867	-0.0837	73.09	
	Quadratic	0.961	0.922	0.7974	28.48	Suggested
	Cubic	0.9932	0.9681	0.7624	18.21	Aliased

ANOVA was performed to identify the significance and accuracy of the chosen model. The results are shown in Tables 6 through 8. These results would be used to examine the significance of the model and the independent variables on the response based on the

F-value and P-value. Important variables are commonly rated using the F-value or P-value at a 95% confidence interval. The larger the F-value and the smaller the P-value obtained, the more significant the corresponding coefficient achieved (Ashraf *et al*. 2021).

Table 6. ANOVA Results of the Regression Models for Compressive Resistance

Source	Sum of squares	df	Mean square	F-value	P-value
Model	8.79E+05	14	62783.59	49.63	< 0.0001
x_1	3.00E+05	1	3.00E+05	236.93	< 0.0001
x_2	54062.48	1	54062.48	42.73	< 0.0001
x_3	1.34E+05	1	1.34E+05	105.57	< 0.0001
x_4	1.57E+05	1	1.57E+05	124.23	< 0.0001
x_1x_2	934.83	1	934.83	0.74	0.4045
x_1x_3	0.64	1	0.64	5.06E-04	0.9824
x_1x_4	10455.06	1	10455.06	8.26	0.0122
x_2x_3	615.04	1	615.04	0.49	0.4971
x_2x_4	5212.84	1	5212.84	4.12	0.0618
x_3x_4	96.04	1	96.04	0.076	0.7869
x_1^2	36919.93	1	36919.93	29.18	< 0.0001
x_2^2	45218.18	1	45218.18	35.74	< 0.0001
x_3^2	1.04E+05	1	1.04E+05	82.11	< 0.0001
x_4^2	1342.78	1	1342.78	1.06	0.3204
Residual	17711.72	14	1265.12		
Lack of fit	16522.78	10	1652.28	5.56	0.0564
Pure	1188.95	4	297.24		
Total	8.97E+05	28			
C.V.%	3.31				
Adeq Precision	30.64				

Table 7. ANOVA Results of the Regression Models for Durability

Source	Sum of squares	df	Mean square	F-value	P-value
Model	366.57	14	26.18	25.88	< 0.0001
x_1	97.47	1	97.47	96.35	< 0.0001
x_2	29.77	1	29.77	29.43	< 0.0001
x_3	28.21	1	28.21	27.89	0.0001
x_4	47.6	1	47.6	47.06	< 0.0001
x_1x_2	7.84	1	7.84	7.75	0.0146
x_1x_3	1.21	1	1.21	1.2	0.2926
x_1x_4	8.41	1	8.41	8.31	0.012
x_2x_3	0.25	1	0.25	0.25	0.6268
x_2x_4	0.56	1	0.56	0.56	0.4682
x_3x_4	1.69	1	1.69	1.67	0.2171
x_1^2	20.11	1	20.11	19.88	0.0005
x_2^2	20.47	1	20.47	20.24	0.0005
x_3^2	83.56	1	83.56	82.6	< 0.0001
x_4^2	0.68	1	0.68	0.67	0.4266
Residual	14.16	14	1.01		
Lack of fit	12.09	10	1.21	2.34	0.2142
Pure	2.07	4	0.52		
Total	380.73	28			
C.V.%	1.11				
Adeq Precision	23.64				

Table 8. ANOVA Results of the Regression Models for Density

Source	Sum of squares	df	Mean square	F-value	P-value
Model	2.80E+05	14	20002.22	24.65	< 0.0001
x_1	89372.28	1	89372.28	110.15	< 0.0001
x_2	14861.44	1	14861.44	18.32	0.0008
x_3	10080.4	1	10080.4	12.42	0.0034
x_4	69205.64	1	69205.64	85.3	< 0.0001
x_1x_2	22.09	1	22.09	0.027	0.8713
x_1x_3	1576.09	1	1576.09	1.94	0.1851
x_1x_4	3469.21	1	3469.21	4.28	0.0577
x_2x_3	1785.06	1	1785.06	2.2	0.1602
x_2x_4	3214.89	1	3214.89	3.96	0.0664
x_3x_4	1644.3	1	1644.3	2.03	0.1765
x_1^2	17075.97	1	17075.97	21.05	0.0004
x_2^2	2660.96	1	2660.96	3.28	0.0916
x_3^2	36478.38	1	36478.38	44.96	< 0.0001
x_4^2	12541.19	1	12541.19	15.46	0.0015
Residual	11358.68	14	811.33		
Lack of fit	9833.76	10	983.38	2.58	0.1872
Pure	1524.92	4	381.23		
Total	2.91E+05	28			
C.V.%	3.00				
Adeq Precision	21.76				

To ensure the robustness of the proposed models, the significance and fitness of the models were examined based on the P-value and F-value. The results of ANOVA revealed that all the P-values for CR, DU, and DE models were strictly lower than the significant level of 0.05, indicating that the developed mathematical models were efficacious and the correlation between responses and independent variables was promising. The reliability of the model could be checked *via* the lack-of-fit test, and insignificant lack-of-fit illustrates a favorable correlation between the model and the data. In the present study, all the P-values of lack-of-fit for the three responses (0.0564 for CR, 0.2142 for DU, and 0.1872 for DE) were higher than 0.05, which indicated that the proposed model was valid and it could be well used to reveal the relationship between independent variables and responses, and therefore, it was highly accurate to describe the experimental data. Other parameters, such as the coefficient of determination (R^2), the adjusted determination coefficient (Adj-R^2), the coefficient of variation (C.V.%), and adequate precision (Adeq Precision), should also be considered to investigate the quality of the regression models (Issa *et al*. 2020). The R^2 values of the three models were 0.9802, 0.9628, and 0.961 for CR, DU, and DE, respectively.

These results demonstrated that more than 98.02%, 96.28%, and 96.1% of the sample variance for the three responses could be attributed to the variables, supporting a satisfactory accuracy and a noticeable capacity of the proposed models. Meanwhile, Adj-R^2 is frequently used as the measure of the goodness-of-fit. In the present study, the values of Adj-R^2 for the three responses were 0.9605, 0.9256, and 0.922, confirming a high correlation between the observed and the predicted values. Additionally, the reproducibility of the model can be measured by C.V.% and the values of 3.31%, 1.11%, and 3% confirmed the higher reproducibility of the developed models. Adeq Precision was originally used to evaluate SNR (signal-to-noise ratio), in which SNR > 4 is optimal (Rakhmania *et al*. 2021). In this report, the Adeq Precision values from the three responses were 30.64, 23.64, and 21.76, respectively, indicating an adequate signal with the experimental data.

The correctness of the proposed models was further assessed by the residual analysis. The actual responses *versus* the predicted responses for CR, DU, and DE are shown in Fig. 4. The data points were equally distributed on both sides of the line and close to the center line, demonstrating that the regression model was accurate. In other words, a good fitness between the developed model and the observed data was achieved, making a good estimate of response for the system in the target range.

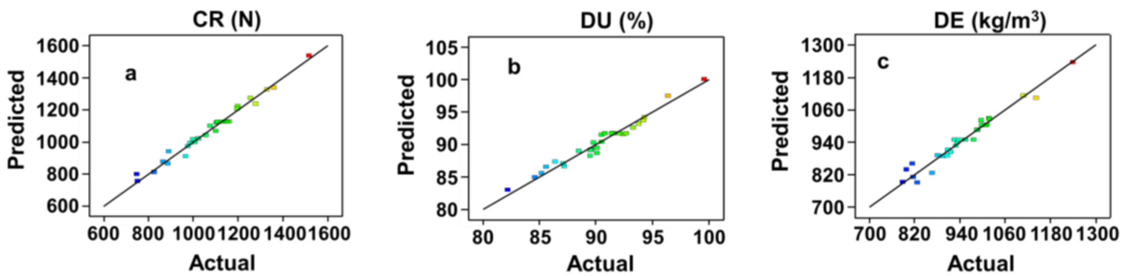


Fig. 4. Predicted versus actual response for (a) CR, (b) DU, and (c) DE (CR: compressive resistance, DU: durability, DE: density)

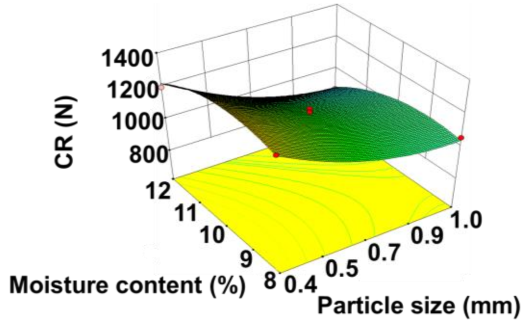
Interaction effect of variables on compressive resistance

Generally, factors with a P-value higher than 0.05 are considered as significant and important terms, and they can be applied to formulate a regression equation. The ranking of significant regression terms is based on F-value, and regression terms with a higher F-value are regarded as the most significant terms. According to the results of ANOVA (Table 6), it was evident that all the four independent variables had positive effects on CR. Among all the significant factors, particle size was found to be the most influential factor that had the major effect on CR, because it had the highest F-value of 236.93, followed by molding pressure, die temperature, and moisture content. Meanwhile, x_1x_4, x_1^2, x_2^2, and x_3^2 were noted as significant quadratic terms ($P<0.05$). After removing insignificant terms at the confidence level of 95%, the second-order polynomial equation in terms of actual factors was formulated to predict the relationship between variables and CR as a function of particle size, moisture content, die temperature, and molding pressure, as given in Eq. 3.

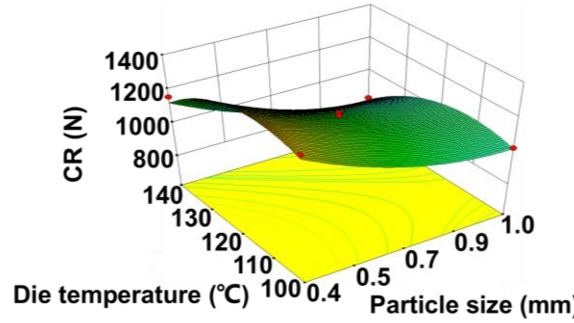
$$y_1 = -5971.35-260.69x_1+490.89x_2+69.22x_3+12.11x_4-8.52x_1x_4 +838.27x_1^2-20.87x_2^2-0.32x_3^2 \quad (3)$$

The coefficients of the regression equations with a positive sign indicate a positive effect, meanwhile, terms with a negative sign represent the negative impact (Alhajabdalla *et al*. 2021). For instance, in Eq. 3, the linear terms of x_2, x_3, x_4 and the quadratic terms of x_1^2 reflected their influence on increasing the CR, whereas the other terms of x_1, x_1x_4, x_2^2 and x_3^2 with negative signs had a negative effect on the response.

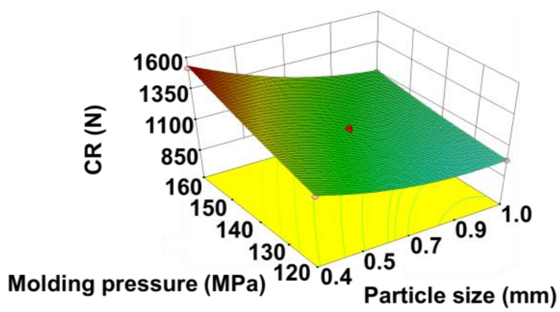
To intuitively investigate the quadratic influence of variables on CR, 3D (three-dimensional) response surface plots are shown in Fig. 5a-f. These plots were developed by drawing two factors over their respective ranges, while the other two factors were kept at a constant range, which facilitated the study on the synergistic effects of each pair of independent variables on the response.



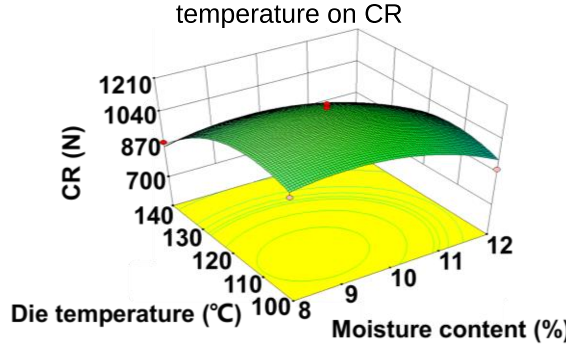
(a) Interaction effect of particle size and moisture content on CR



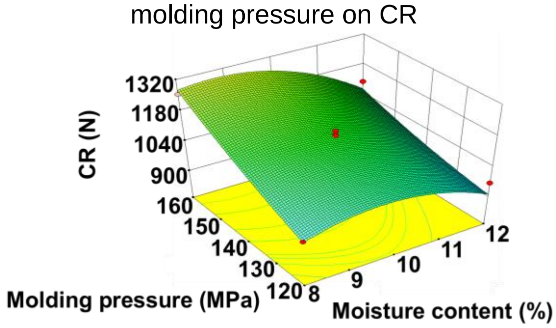
(b) Interaction effect of particle size and die temperature on CR



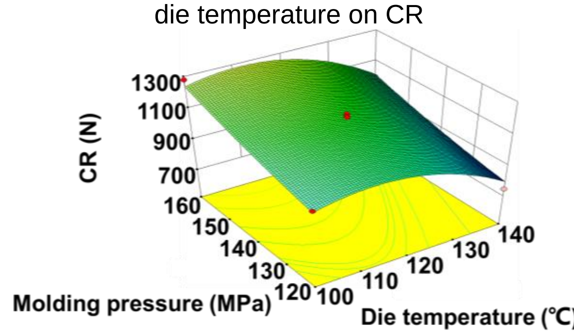
(c) Interaction effect of particle size and molding pressure on CR



(d) Interaction effect of moisture content and die temperature on CR



(e) Interaction effect of moisture content and molding pressure on CR



(f) Interaction effect of die temperature and molding pressure on CR

Fig. 5. 3D response surface plots of CR versus (a) Ps and Mc; (b) Ps and Dt; (c) Ps and Mp; (d) Mc and Dt; (e) Mc and Mp; and (f) Dt and Mp (CR: compressive resistance)

Figure 5a presents the coupled action of particle size and moisture content on the CR when die temperature and molding pressure were kept at a zero level. Particle size had a negative impact on CR, that is, with the enlargement of particle size from 0.4 to 1 mm, CR decreased from 1326.65 to 1043.99 N. The similar effect of particle size on CR can be observed in Fig. 5b,c. These findings confirmed that a smaller particle size was associated with a higher CR. The maximum CR of 1362.06 N was observed at the moisture content of 9.5%, and when the moisture content was higher or lower than this value, a slight decrease of approximate 100 N would be noted in CR. The similar variation of CR caused by the moisture content can be observed in Fig. 5d,e. The influence of the moisture content on CR can be explained by the fact that a reasonable moisture content can improve the formation of a solid bridge and short-range forces between particles during the densification process, which led to high-quality pellets. The effects of die temperature and molding pressure are represented by the 3D surface plots (Fig. 5f). Enlargement of the die temperature to a certain level was associated with a noticeable elevation in CR, and further increase in die temperature led to a decrease in CR (Fig. 5b,d). At a rational value of approximately 112 °C, the optimum CR could be achieved due to the enhanced generation of binding forces between particles, resulting from the adhesion of the natural binders caused by the cooperative effects of temperature and pressure. Besides, CR significantly increased along with increasing the molding pressure, as depicted in Fig. 5c,e. This may be due to the fact that, increasing the molding pressure was advantageous to the spreading of the natural binding components, which had been softened and pressed out from the materials, from one particle to another at the points of contact to act as a binder, leading to the enhancement of the strength of pellets.

Interaction effect of variables on density

Based on the ANOVA results present in Table 7, all the linear terms were significant for DU ($P<0.05$), which confirmed that the selected factors of particle size, moisture content, die temperature, and molding pressure strongly influenced DU. Of all the linear terms, particle size was the most significant factor due to its highest F-value and lowest P-value, followed by molding pressure, moisture content, and die temperature. For the interactive terms, apart from the interaction between particle size and molding pressure and moisture content (x_1x_2 and x_1x_4), the other interaction terms (x_1x_3, x_2x_3, x_2x_4, and x_3x_4) were statistically insignificant. This demonstrated that any variation in particle size coupled with either of moisture content or molding pressure affected CR appreciably. Except for the quadratic term of x_4^2, the other quadratic terms (x_1^2, x_2^2, and x_3^2) were statistically significant ($p<0.05$). After model reduction, the regression model for DU related to different factors and interactions in terms of actual values is formulated using Eq. 4.

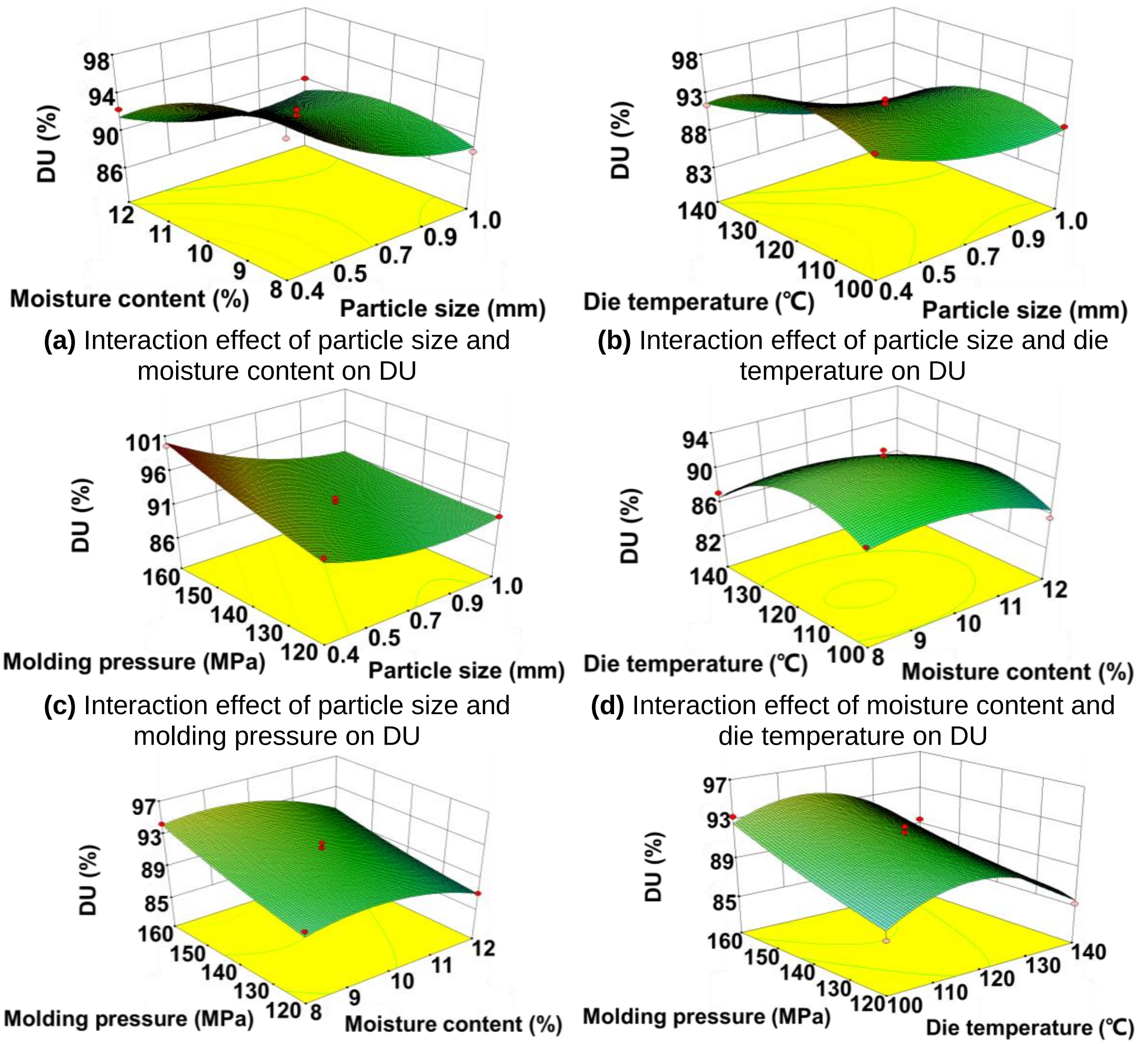
$$y_2 = -109.67-15.39x_1+8.53x_2+2.43x_3+0.33x_4+2.33x_1x_2$$

$$-0.247x_1x_4+19.55x_1^2-0.44x_2^2-0.009x_3^2 \qquad (3)$$

To visualize the combined effects of the two factors on DU, the 3D response surface plots are depicted in Fig. 6a-f.

As shown in Fig. 6a-c, DU significantly decreased with the enlargement of particle size. Decrease of particle size can enhance the contact area and reduce the distance of the inter-particle spaces. Meanwhile, an increase in fluidity of the particles resulting from the smaller size led to the reduction of empty spaces between particles. The association of moisture content with other independent variables is illustrated in Figs. 6a, 6d and 6f. DU declined first, and then it increased with the raised moisture content in feedstock, and the maximum DU was achieved at a moisture content of around 9.3%. As a result, DU was closely correlated with the moisture content of the compacted materials. This indicated that when moisture content was in the reasonable range, the bonding act of the natural binders (*e.g.*, lignin, starch, cellulose, sugar, *etc*.) can be greatly stimulated, leading to the improvement of DU.

The main effects of die temperature and molding pressure on DU were represented by the 3D response surface plots (Fig. 6b-f). It was revealed that the molding pressure had a positive effect on DU, as an increase in pressure could enhance the contact forces between particles, resulting in the elevation of DU. The effect of die temperature on DU was similar to that of moisture content. The maximum DU was observed at the die temperature of approximately 120 °C, and this could be related to the fact that when die temperature was in an optimum range, the natural binders inside the materials were softened, pressed out and diffused from one particle to another, which was advantageous for the generation of the solid bridge, hydrogen bonds, and van der Waals forces between adjacent particles.



(a) Interaction effect of particle size and moisture content on DU

(b) Interaction effect of particle size and die temperature on DU

(c) Interaction effect of particle size and molding pressure on DU

(d) Interaction effect of moisture content and die temperature on DU

(e) Interaction effect of moisture content and molding pressure on DU

(f) Interaction effect of die temperature and molding pressure on DU

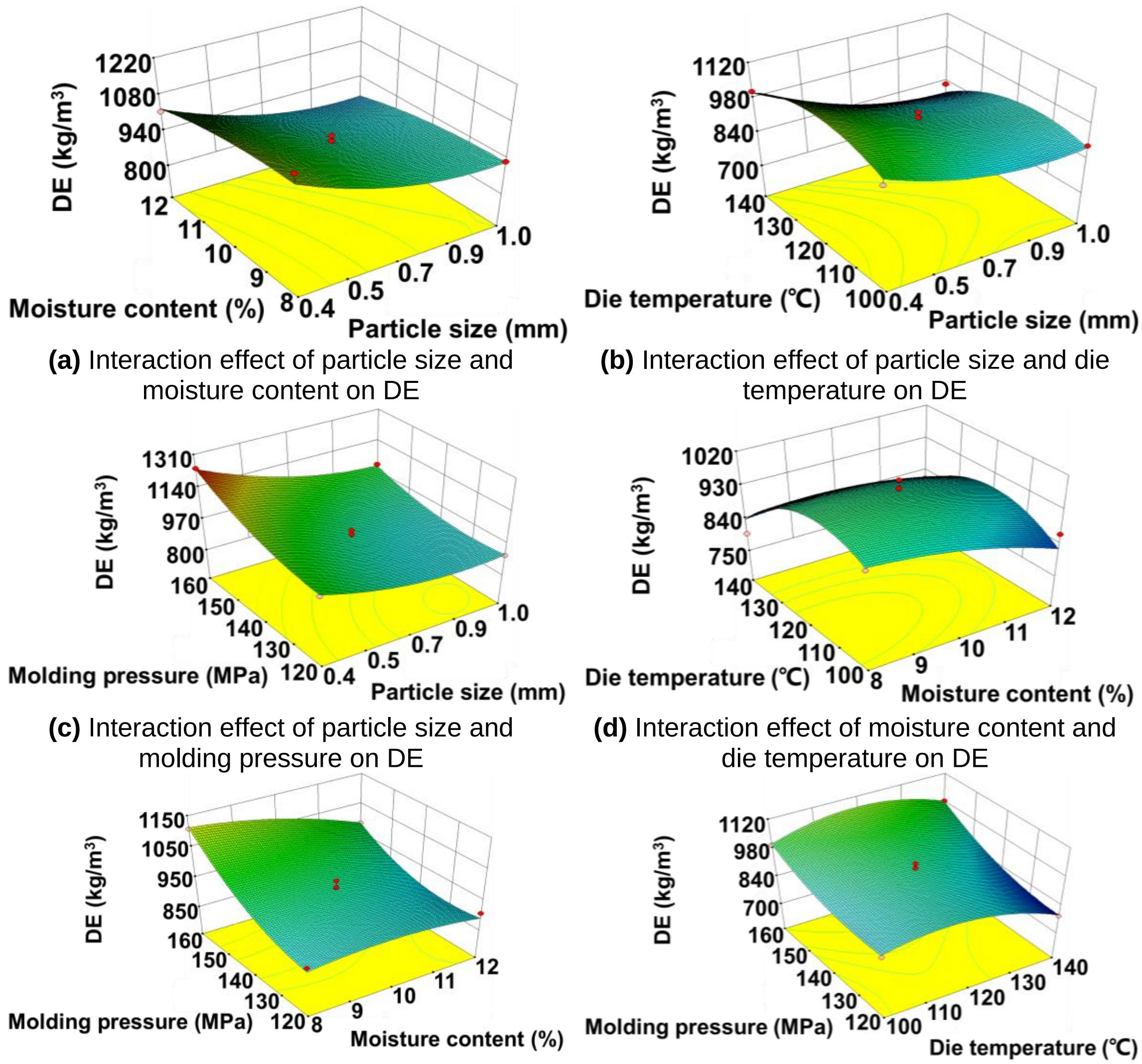
Fig. 6. 3D response surface plots of DU versus (a) Ps and Mc; (b) Ps and Dt; (c) Ps and Mp; (d) Mc and Dt; (e) Mc and Mp; and (f) Dt and Mp (DU: durability)

Interaction effect of variables on density

According to the ANOVA results listed in Table 8, all the chosen factors had a significant influence on DE ($P<0.05$). With the F-values compared, the effect of the chosen factors on DE followed the descending order: particle size > molding pressure > moisture content > die temperature. There was no significant interaction term for DE ($P>0.05$). In addition to x_2^2, the other quadratic terms of x_1^2, x_3^2, and x_4^2 were significant for DE. The quadratic regression equation for DE based on the actual values of the input variables was formulated as follows:

$$y_3 = -41.25-40.80x_1+116.78x_2+33.48x_3-22.54x_4+51.31x_1^2-74.99x_3^2+40.97x_4^2 \quad (3)$$

The coupled influences of independent variables on DE are depicted in Fig. 7a-f.



(a) Interaction effect of particle size and moisture content on DE

(b) Interaction effect of particle size and die temperature on DE

(c) Interaction effect of particle size and molding pressure on DE

(d) Interaction effect of moisture content and die temperature on DE

(e) Interaction effect of moisture content and molding pressure on DE

(f) Interaction effect of die temperature and molding pressure on DE

Fig. 7. 3D response surface plots of DU versus (a) Ps and Mc; (b) Ps and Dt; (c) Ps and Mp; (d) Mc and Dt; (e) Mc and Mp; and (f) Dt and Mp (DE: density)

Figure 7c shows the interaction effect of particle size and molding pressure, and it can be seen that both factors significantly affected DE, which was consistent with the results of ANOVA presented in Table 8. DE was negatively influenced by particle size, whereas a positive correlation was noted between DE and molding pressure. As discussed above, a decrease of the particle size could greatly enhance the flowability of the particles, which could reduce the empty space between particles and make the particles more tightly bind together, leading to the increase of DE. As mentioned above, the natural binding components, *e.g.*, protein, lignin, and water-soluble carbohydrates in biomass materials, can be squeezed and they can form a solid bridge, hydrogen bonds, and van der Waals forces between adjacent particles under the function of pressure, which may be related to the increase of DE with the elevation of molding pressure. The coupled effects of die temperature and moisture content on DE were observed (Fig. 7d), as particle size and molding pressure were kept constant. The optimum DE of about 973 kg/m^3 was obtained at a medium moisture content of 8% and a low die temperature of about 115 °C within the design limits. This phenomenon can be explained by the fact that in the reasonable ranges of moisture content and die temperature, the binding between particles was more pronounced. Kaliyan and Morey found that the natural components in the materials can be activated as die temperature increased from 75 to 150 °C (Kaliyan and Morey 2010).

Optimization and Verification

The principle of maximizing CR, DU, and DE was employed to determine the optimal values of parameters. All the proposed regression models were employed to investigate the specified conditions of optimization. During optimization, all the variables, including independent variables and response variables, were equally weighted. Moreover, the desirability function approach, which has been widely used to assess the optimization response, was applied. Then, the optimal conditions for producing pellets from BRAD were determined (particle size of 0.4 mm, moisture content of 8.4%, die temperature of 115 °C, and molding pressure of 150 MPa), meanwhile, the corresponding predicted CR, DU, and DE were 1468.71 N, 99.58%, and 1178.67 kg/m^3, respectively. To confirm the validity of the regression models, verification experiments were conducted in triplicate under the determined optimal conditions, and Table 9 shows the results. Since the ash content is one of the pivotal parameters to measure the quality of the pellet, the ash content of the pellet under the optimal condition was measured to be 6.32%, which is slightly higher than that of 5.5%-6.4% of barley pellets and lower than 6.15% to 6.8% of wheat straw pellets. This may be caused by the difference of materials, molding parameters, and the adhesive addition of linear low-density polyethylene (Emadi *et al.* 2016).

Table 9. Test and Predicted Responses Values under the Optimal Conditions

Responses	Values		Durability
	Predicted	Experimental	
y_1 (CR, N)	1468.71	1438.69	0.932
y_2 (DU, %)	99.58	99.62	
y_3 (DE, kg/m^3)	1178.67	1207.54	

The test values obtained from the verification experiment were close to the predicted values, demonstrating that data of the experiment were precisely predicted by the regression models. Moreover, the desirability of 0.932 further testified the practicability and accuracy of the regression models.

Table 10 lists the properties of pellets made from different biomass. It can be seen that great differences were observed in the characteristics of pellets made from different biomass. This phenomenon may be due to the differences of compositions of material, the shape and dimension of pellets and the densification parameters.

Table 10. Properties of Pellets Made from Different Biomass

Pellet	Properties				Reference
	Compressive resistance (N)	Durability (%)	Density (kg/m³)	Meyer hardness (N/mm²)	
BRAD	1438.69	99.62	1207.54	/	Current
Wheat straw	/	/	800-1000	/	Mani *et al.* 2006
Corn stover	/	/	1000-1200	/	
Sewage sludge + rice straw	/	/	1150-1200	3.5-5.5	Jiang *et al.* 2015 and Li *et al.* 2014
Sewage sludge + Chinese fir	/	/	1050-1125	3.5-4.5	
Sewage sludge + camphor	/	/	1000-1025	3.5-4.8	

/ data not available

CONCLUSIONS

1. Pellets were produced with biomass residue from anaerobic digestion (BRAD) using densification, and the mechanical properties of the pellets were investigated.
2. Response surface methodology (RSM) based on Box-Behnken Design (BBD) experimentation was conducted to optimize the process parameters, and the regression models were obtained with suitably high R^2 values of 0.9802, 0.9628, and 0.9610 for compression resistance (CR), durability (DU), and density (DE), respectively. The ANOVA results showed that all the influential factors of particle size, moisture content, die temperature, and molding pressure significantly influenced the responses. For CR, the most influential factor was particle size, followed by molding pressure, die temperature, and moisture content. For DU, the effect of variables followed the order below: particle size > molding pressure > moisture content > die temperature. For DE, the effect of variables followed the descending order: particle size, molding pressure, moisture content, and die temperature.
3. The optimal values of these factors were 0.4 mm for particle size, 8.4% for moisture content, 115 °C for die temperature, and 150 MPa for molding pressure; meanwhile, the predicted values of CR, DU, and DE were 1,469 N, 99.6%, and 1,180 kg/m^3, respectively. The results of verification test were consistent with the predicted values under the specified condition. The desirability of 0.932 further verified the accuracy of the regression model.

ACKNOWLEDGMENTS

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