Pelletization of Biomass Feedstocks: Effect of Moisture Content, Particle Size and a Binder on Characteristics of Biomass Pellets

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Research Article

Pelletization of biomass feedstocks: Effect of moisture content, particle size and a binder on characteristics of biomass pellets

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Abstract

Pelletization of low value added biomass materials such as furfural residue (FR) and sawdust was performed by using a lab scale pelletizer. Effects of moisture content (MC), particle size and a binder on quality parameters (e.g. pellet density, strength and hardness) and on energy consumption were investigated. Quality of pellets was analysed and compared. MC was found to be the more dominant parameter affecting pellet density, strength and hardness of furfural residue pellets (FRPs) and sawdust pellets (SPs), followed by particle size and a binder. Highest particle density of 1.419 g/cm^3 for FRPs (0.5-1.41 mm) and 1.243 g/cm^3 for SPs (0.25-0.5 mm) was achieved at MC of 8% and 18%. Highest decrease in relaxed density was observed at MC of 13% for FRPs and 28% for SPs. True density of FRPs and SPs made from particles of 0.25-0.5 mm was found higher than 0.5-1.41 mm. The highest strength and hardness (6.29 MPa and 401.3 N/mm^2) for FRPs was achieved at 5.5% MC and particles 0.25-0.5 mm. Optimum strength (6.03 MPa) and hardness (96.06 N/mm^2) for SPs was obtained at 18% MC and particles 0.25-0.5 mm. The lowest energy consumption (16.16 J/g) for FRPs (0.25-0.5 mm) and 20.22 J/g for SPs (0.5-1.41 mm) was achieved at MC of 13% and 28%. Addition of binding agent to FR sawdust decreased energy consumption of FRPs and SPs. SPs quality was enhanced with the use of a binder. Heating value of FRPs were found higher than SPs.

Keywords: Biomass pellets; Process parameters; Biofuel quality; Energy consumption

1 Introduction

Environmental pollution due to use of fossil fuels has become the major concern all over the world [1]. Energy production from biomass sources such as forestry residues, woody biomass, dedicated energy crops and sewage sludge has been renowned as promising way to reduce environmental pollution. After coal, oil and gas, biomass is considered as the fourth largest source of energy and has a potential to be used as clean and green fuel to substitute fossil fuel (e.g., coal) in many applications [2–4]. Due to intrinsic properties of raw biomass such as higher moisture content (MC), low density, scattered distribution etc., application of raw biomass are limited. Recently, biomass pelletization has been renowned as attractive way to addresses these issues more effectively, producing biomass pellet with higher density and uniform structure. Further, pelletization decreases the MC (less than 10%) and improves the quality characteristics such as density (bulk or particle), strength and hardness etc., in an environmental friendly manner [5, 6]. Thus, pelletization significantly affect the transportation, handling and storage characteristics of the product [6]. For example, pelletization facilitates its utilization by enhancing the pellet density. Hence, it significantly reduces handling, transportation and storage costs [7–9] and dust emission during handling and transportation.

Biomass pelletization is a proven technology, which produces fuel (e.g. pellets) with uniform size and structure, enhances the pellet quality (e.g. high density and strength) and lowers the MC of extruded pellets (e.g. $\leq 10\%$) depending upon the type of feedstock and its MC being used for pellet production [5, 6]. Pelletization process improved the bulk density of pellets produced from woody biomass and agricultural residues (e.g. straws and grasses). Bulk density was found in the range of 600-800 kg/m^3 (for wood pellets) and 40-200 kg/m^3 (for straw pellets) [10]. Neal A. Yancey et al. [11] reported that pelletization of biomass materials had enhanced the bulk density of pellets. The bulk density of 645 kg/m^3 (corn stover pellets), $\geq$700 kg/m^3 (switchgrass and lodgepole pine pellets), and 754 kg/m^3 (eucalyptus pellets) were

found higher than raw biomass materials. Furthermore, the highly dense fuel pellets can be transported for longer distance, hence the cost for handling and transportation may possibly be reduced [12]. Biomass pellets also cause very less or no dust emission during their transportation. without causing the dust emission [13].

Quality of pellet is highly dependent on feedstock properties and type such as MC, ash particle size and ash content, operating variables (i.e., temperature, pressure and holding time) and binder(s) [14]. In current study, influence of moisture content (MC) particle size and a binder on pellet quality was investigated. MC is considered as a critical factor for biomass pellet production as it is important in pelletizing performance, process configurations (e.g., storage, size reduction, drying, feeding), process economics and combustion characteristics of pellets [15]. MC has both positive and negative influence on pellet quality (e.g., particle density, strength and hardness) and an optimum level must be selected for feed material prior to pelletization. Water acts as the lubricating and binding agents, reducing the friction and increasing the pellet durability [10, 16, 17]. On the other hand, water is not compressible, limiting the final density of the pellet [18], and higher MC increases the extent at which pellets 'relax' after formation, which can lead to poor quality of pellets [19]. Moreover, MC reduces the glass transition temperature of lignin, which increases bonding between particles [20]. At MC >20%, steam is generated and reaches relatively high pressure depending on operation temperatures. The steam pressure reduces compression [21] and/or hydrogen bonds between wood polymers are replaced by bonds to water molecules, resulting in weaker pellet [16, 20]. Previous studies had recommended the optimum MC of 5% for olive [18], 6-13% for Scots pine [16, 22, 23], 8-15% for wheat straw [23–25], 20-25% for miscanthus [26], 30-33% for corn stover [27] and 10–15% for sewage sludge [28, 29], respectively, to achieve better pellet qualities.

Size reduction of biomass particles prior to pelletization is a crucial step and the degree of crushing required depends upon the type of feedstock [30]. Particle size of biomass greatly influences the pore size, total surface area and points for inter particle bonding that is essential for producing pellets of higher quality [31, 32]. During compaction, particles of smaller sizes rearrange and fill in the void spaces between larger particles, resulting in improvement of pelletization process, and more denser, strong and durable pellets [25]. Smaller particles (e.g., 0.25-1 mm) provide larger surface area for adsorption of MC during conditioning, facilitate better absorption of heat during compaction, resulting in better binding of biomass particles [18, 33]. At the same time, small particles dry out quickly in arid seasons or areas , leading to difficulties in pelletizing process [30]. Particle densities of pellets made from woody (e.g., willow and poplar sawdust) and herbaceous biomass (e.g., corn stover, barley straw, wheat straw and switchgrass) were found higher for small particle sizes [34–36]. Criteria for selecting optimum particle size depends upon the densification process and types of biomass. Optimum level of particle size should be selected to produce pellet of higher quality.

Sometimes, addition of binder to biomass materials are suggested to improve the binding properties of feed material during pelletization and reduce the energy consumption. Various organic and inorganic binders are available in the market, and selection criteria of binders depends upon the type of biomass, cost and environmental affability [37].

Many studies have investigated the influence of MC, particle size and a binder on quality of biomass pellets. For example, Quy Nam Nguyen et al. [38] investigated that particle density and strength was dropped with increasing MC and particle size. The highest density (1115 kg/m^3) strength 66 N/mm were found at MC of 8.1% and particle size (<0.25 mm) and temperature of 125°C. Hamid Rezaei et al [39] found that at MC of 20% and particle size of 6 mm, pellets produced from refuse-derived fuel had better quality (e.g. higher strength and durability) and lower energy requirement. Another study evaluated that particle density of

fallen leaves pellets dropped by 6% with increasing MC from 10 to 20% and 10 % with increasing glycerol from 0 wt% to 10 wt% [40]. For obtaining the high quality of olive pruning residues pellets i.e. higher density and durability, the authors suggested MC of <10%, particle size of 0.25-0.5 mm, temperature of 150°C and pressure of ≥170 MPa [18]. Addition of binder such as recovered polyvinyl alcohol (2 to 6 wt%) to biomass materials (e.g. groundnut shell, sawdust and leaf litter waste) enhanced the strength of pellets [41]. With MC increasing from 5 to 20% significantly decreased the energy consumption of sludge mixed pellets [42]. Donghui Lu et al. [43] investigated that using binders (e.g. bentonite, crude glycerol, lignosufonate and wood residue) improved the pellet density and strength and lowered the energy consumption. Energy consumption of pellets made from poplar energy crop reduced by 43% by employing maize starch as a binder [44].

However, many studies have been performed on pelletization of different kinds of biomass sources. Still there is need to explore more about the new biomass sources (e.g. FR, sewage sludge and or mixture) rather than depending upon woody biomass or agricultural residues. Biomass pellet industry can face several challenges such as discrepancy in supply of raw biomass feedstock and rising prices. To address the sustainability issues and meeting the global pellet demands, finding and researching new renewable energy sources is essential. Pelletization of FR can be a topic of greater interests and is expected to open the new pathways for the researchers.

Pellets were produced from biomass materials such as FR and sawdust. Various characteristics (particle, relaxed and density, strength, hardness etc.), energy consumption, and their dependency on MC, particle size and a binder were studied. According to our knowledge, no any literature on quality comparison between these two types of pellets has been reported. Satisfactory findings of this research are expected to be very informative and beneficial for

pellet industry and utilization of low value added materials (e.g., FR and sawdust) to more value added fuels (e.g., pellets) from both academic and industrial perspectives.

2 Materials and Methods

2.1 Biomass materials

Biomass materials i.e. FR was obtained from a local furfural production plant located in Hebei province, and sawdust from a furniture plant situated in suburb of Beijing, respectively. Biomass materials were air-dried at room temperature before pelletization until the materials moisture reached to equilibrium (i.e. MC of FR ~5 wt.%, sawdust ~4 wt.%). Later, FR and sawdust were crushed. After that, biomass materials were sieved to obtain the required particle size (e.g., 0.25-0.5 mm and 0.5-1.41 mm). Biomass materials were sieved for 10 minutes. A similar method of sieving was adopted by previous study [38]. MCs of FR and sawdust were adjusted to 5.5, 8, 13, 18, 28 wt.% respectively by mixing predetermined amount of deionized water with raw biomass consistent to previous study [45]. Table 1 represents the bulk density, proximate analysis (volatile matter, ash, and fixed carbon), elemental analysis (carbon, hydrogen, nitrogen and sulfur) and chemical analyses of raw biomass materials. A binder i.e. synthetic resin (SR) used in current study was received from a local laboratory in Beijing. SR was used in proportion of 1:9 (10% binder+90% biomass material).

Table 1. Properties of raw biomass materials and heating value of pellets

Analysis					
Ultimate analysis [a] (wt.%)			Proximate analysis [b] (wt.%)		
	FR	Sawdust		FR	Sawdust
C	50.78±0.002	48.66±0.001	Volatile matter	59.3±0.002	75.4±0.002
H	4.98±0.001	5.5±0.001	Fixed carbon	23.9±0.002	16.7±0.001
N	0.57±0.002	3.19±0.002	Ash	10.6±0.001	3.1±0.002
S	1.26±0.001	0.53±0.004	Moisture content	6.2±0.003	4.85±0.003
O [c]	31.79±0	42.12±0			
Chemical analysis (wt.%) [a]					
	Lignin [c]	Cellulose	Hemicellulose	Acid-insoluble ash	Impurity
FR	29.81±0	34.89±0.002	5.04±0.003	2.97±0.001	27.28±0.002

Sawdust	11.98±0	50.17±0.003	21.3±0.002	1.36±0.005	15.18±0.003
Bulk density (kg/m³)					
FR	0.25-0.5 mm	0.5-1.41 mm	Sawdust	0.25-0.5 mm	0.5-1.41 mm
	496±0.01	530±0.006		202±0.007	193±0.02
Heating value (MJ/kg) of pellets					
FRPs	23.36±0.02	22.88±0.012	SPs	20.6±0.052	21.21±0.04

[a] Oven dry basis, [b] air dry basis, [c] Calculated by difference

2.2 Pellet production

A lab scale single pelletizer (Model LYWN-W50KN, Jinan lingyue precision instrument Co. Ltd) with diameter 12 mm was employed for making pellets. The cylindrical die was first heated to the preset temperature (130°C). After reaching to preset temperature, approximately 2-3 g of biomass sample i.e., FR and sawdust was filled and compressed by piston at a speed of 200 N/min to the chosen pressure (100 MPa). A holding time of 300 seconds was employed at full pressure for each pellet. After completing the compression, the pressure was released and the pellet was pressed out of the cylinder by removing the plate. A similar method of pellet production was employed by previous studies [46–48]. Force-displacement data was noted during pelletization by the MaxText control system for the complete cycle of compression and ejection of pellets. Table 2 represents the experimental design of the current research. At least 8 experiments on same condition were performed to ensure the repeatability. Preventing pellets from ambient moisture, both kinds of pellets were safely stored in airtight plastic bag at room temperature.

Table 2. Experimental design for pelletization

Parameters					
Moisture (%)	5.5	8	13	18	28
Particle size (mm)		0.25-0.5		0.5-1.41	
SR/FR [a] and SR/SD [b]			1:9		
Temperature (°C)			130		
Pressure (MPa)			100		

[a] SR/FR is synthetic resin/furfural residue ratio and [b] SR/SD is synthetic resin/sawdust ratio

2.3 Measurement techniques

Table 3 manifests the different measuring techniques for characterization of biomass materials and pellets. Immediately after ejection from the die, mass, diameter and length of pellet were measured to calculate initial particle density of pellets. The method to determine the particle density of pellets was given in Table 3. After storage time of two weeks, mass, diameter and length were again measured to calculate relaxed density of individual pellet. Strength and hardness of pellets were determined by employing a computer controlled testing machine. A pellet was placed horizontally on a plate and was radially compressed at compression speed of 6 mm min^{-1} by another plate. The machine was stopped automatically until the failure of pellet. Hardness of pellets was measured by penetrating a hemisphere-end rod of small diameter into the pellet. Compressive force was applied to the center of the pellet, which was placed horizontally on a steel plate. The rod moved downward at a speed of 2 mm min^{-1} and stopped after the pellet was cracked. The maximum force at which pellet cracked was noted. Further details for determining the strength and hardness of pellets was given in Table 3.

Energy consumption related to compaction and extrusion was calculated by integrating the force-displacement curve. The energy consumption in (J/g) was determined by dividing total energy (compression plus extrusion) to the mass of pellet.

Table 3. Characterization parameters and measuring techniques

Characterization parameters	Methods/Standard	Analytical Instruments
Moisture content (%)	MC of biomass sample was determined in accordance with (EN ISO 18134-1). MC was determined according to mass change before and after drying. $$MC = \frac{original\ mass - mass\ after\ drying}{original\ mass} \times 100 \quad (1)$$	Analytical balance (ME104E, Metler Toledo, Switzerland) Oven (DHG-914OA, Shanghai, Yiheng Scientific instrument Co.)
Proximate analysis	Proximate analysis of biomass materials was performed according to the standard (GB/T 28731-2012) proximate analysis of solid biofuels, China. Three replicates were made to obtain the average value.	

Elemental analysis	Carbon, hydrogen, nitrogen and sulfur were determined using elemental analyzer. The O content was obtained by employing O=100-C-H-N-S-ash. At least three replicates were made to obtain the average value.	Elemental analyzer (VARIO EL cube, Germany)
Particle density (g/cm^3)	Particle density was determined by using equation 2. A similar method was adopted by previous study [38, 49]. $$\rho_p = \frac{m_p}{V_p} \qquad (2)$$ ρ_p = particle density m_P = mass of pellet (g) V_P = volume of pellet (cm^3) Particle density was determined for at least eight pellets and an average value was obtained.	Analytical balance (ME104E, Metler Toledo, Switzerland)
Relaxed density (g/cm^3)	Relaxed density was determined according to a same method used for particle density after storage time of two weeks. Eight replicates were made for relaxed density to obtain the average value.	Analytical balance (ME104E, Metler Toledo, Switzerland)
True density (g/cm^3)	True density was determined after two hours. True density was determined for eight pellets and an average value was obtained. A similar technique was used by previous studies for measuring the true density of pellets [48, 50].	Analytical balance (ME104E, Metler Toledo, Switzerland) A pycnometer (Quantachrome, Boyton Beach, FL, USA)
Chemical composition	Chemical composition (cellulose, hemicellulose and lignin) of biomass materials was determined according to previous studies [51, 52] using fiber extraction analyzer. Three replicates were made to ensure the consistency of data.	Fiber analyzer (ANKOM 2000, USA)
Strength (MPa)	Strength of pellet was calculated by adopting the method suggested by Lisowski et al. [53], Liu et al. [9] and Bazargan et al. [54]. Three replications were made for strength test and an average data was obtained in (MPa) using equation 3. $$Strength\ (MPa) = \frac{2F}{\pi l d} \qquad (3)$$ F = Maximum breaking force (N) L = length of pellet (mm) D = diameter of pellet (mm)	A computer-controlled single pellet press (Model LYWN-W50KN, Jinan lingyue precision instrument Co. Ltd)
Hardness (N/mm^2)	The hardness, defined as the applied force divided by the projected indentation area, was calculated by using equation 4. Same method was employed by previous study [55]. $$Hardness = \frac{F}{[\pi(Dh - h^2)]} \qquad (4)$$ F is the maximum breaking force (N) h is the indentation depth (mm) D is the diameter of rod (mm). Hardness test was repeated three times.	A computer-controlled single pellet press (Model LYWN-W50KN, Jinan lingyue precision instrument Co. Ltd)
Heating value (MJ/kg)	Heating value of biomass sample and biomass pellets was determined using a bomb calorimeter on dry basis according to BS EN ISO 18125-2017. Pellets crushed after performing strength test were used for	A bomb calorimeter (ZDHWA9, Henan Sanbo Instruments Co., Ltd, China)

2.4 Statistical analyses

Analysis of variance (ANOVA) and Bonferroni range tests were performed to analyse the statistics of experimental data. Results of ANOVA are tabulated in Table 4. The obtained results were statistically evaluated using Origin pro 9.0 software. All experimental results were presented in mean values with standard deviation. Polynomial regression analysis was successfully performed only for particle and relaxed density of FRPs. Model equation was created from the factor such as MC as shown in Equations 5.

$$Y = a + B_1 X + B_2 X^2 \qquad (5)$$

where Y is the dependent variable, a, B_1 and B_2 c are model constants, and X is MC, respectively.

Table 4. Analysis of variance (ANOVA) results for quality parameters and energy consumption of pellets at different moisture content and particle size

Quality parameters	Moisture content (%)	Furfural residue pellets					
		0.25-0.5 mm			0.5-0.141 mm		
		SS [a]	F-value	P-value	SS [a]	F-value	P-value
Particle density (g/cm^3)	5.5-13	0.001	3.59	0.03	0.007	161.31	0
Relaxed density (g/cm^3)	5.5-13	0.002	6.42	0.004	0.01	173.75	0
True density (g/cm^3)	5.5-13	0.002	9.04	7.4 E-4	0.01	49.58	0
Energy consumption (J/g)	5.5-13	302.5	381.49	0	270.47	384.44	0
Strength (MPa)	5.5-13	30.17	114.7	0.0001	7.67	60.6	0.001
Hardness (N/mm^2)	5.5-13	152500	183.89	0.00001	85339	125.3	0.0001
Sawdust pellets							
Quality parameters	Moisture content (%)	0.25-0.5 mm			0.5-0.141 mm		
		SS [a]	F-value	P-value	SS [a]	F-value	P-value
Particle density (g/cm^3)	5.5-28	0.03	80	0	0.025	58	0
Relaxed density (g/cm^3)	5.5-28	0.02	112	0	0.01	26.89	0
True density (g/cm^3)	5.5-28	0.05	93	0	0.03	35	0
Energy consumption (J/g)	5.5-28	269	116	0	275	115	0
Strength (MPa)	5.5-28	16.4	18.45	0.001	1.68	4.9	0.02
Hardness (N/mm^2)	5.5-28	2290.5	53.8	0	2017.8	15.63	0.002

[a] Sum of squares

3. Results and discussion

3.1 Particle, relaxed and true density of pellets

3.1.1 Influence of moisture content

Figure 1a-1b represents the effect of MC on particle and relaxed density of FRPs and SPs. MC was found to be the most significant parameter, affecting the pellet density (particularly relaxed density) greatly (Table 4). MC was found more dominant parameter for particle density of SPs compared to FRPs. For FRPs, particle density increased slightly (1.392±0.01 to 1.398±0.01 g/cm^3) for particles 0.25-0.5 mm and (1.414±0.01 to 1.419±0.01 g/cm^3) for 0.5-1.41 mm with MC increasing from 5.5 to 8% (Figure 1a). The particle density decreased and reached to 1.383±0.01 g/cm^3 and 1.385±0 g/cm^3 at 13% MC for the corresponding particle sizes. FR comprised of higher lignin (29.81%) and ash (10.6%) content and impurity, which might be reason that MC had shown no considerable enhancement of particle density. At MC > 13%, FRPs could not produced due to higher ash and lignin content, which enhanced the FR flowability and the material was flowing out of the die. For SPs, particle density increased from 1.19±0 to 1.243±0 g/cm^3 and 1.178±0 to 1.230±0.01 g/cm^3 with MC increasing from 5.5 to 18% for the corresponding particle sizes (Figure 1b). MC >18% led to decrease in particle density of SPs. Higher MC filled the pore spaces between the biomass particles, resulting in increase of pellet mass, thus increasing the particle density consistent to previous study [45]. Furthermore, moisture role as a lubricating and binding agent, and interaction between moisture and lignin during compression, resulting in more condensed structure and increase in binding forces between the individual particles, consistent to previous study [56]. In addition to this, moisture acted as a film type binder with hydrogen bonding and lowered the glass transition temperature of lignin during pelletization and increased the particles contact area [57]. At higher MC i.e., 13% and 28% (for FR and sawdust) decreased the particle density of pellets. Moisture higher than its optimum level occupied the volume of biomass materials and increased the expansion and resistance to compaction thus decreased the particle density of

pellets. These findings were in line with previous studies [58, 59]. Further, incompressible nature of water, higher moisture prevented the complete release of natural binding components from biomass particles as it stuck within the particles [18]. Additionally, higher MC increased the degrees at which pellets relaxed after ejection from the die, which could significantly influence the particle density of pellets [19]. FRPs were found more denser at MC 8%, the particle density was 11.08% (0.25-0.5 mm) and 13.3% (0.5-1.41 mm) higher than SPs (at 18% MC).

MC had considerably influenced the relaxed density of FRPs and SPs after 2 weeks storage time. Relaxed density of FRPs decreased by 1.08% (0.25-0.5 mm) and 3.12% (0.5-1.41 mm) with MC increasing from 5.5 to 13%, found lower than those of initial particle density (Figure 1a). Relaxed density of SPs reduced by 1.43% (0.25-0.5 mm) and 1.16% (0.5-1.41 mm) at 28% MC (Figure 1b). Relaxed densities of SPs at higher MC were found lower than those of initial particle density. Decrease in relaxed density of pellets with increasing MC could be due to inadequate adhesion and cohesion of biomass particles particularly at higher MC (13% for FR and 28% for sawdust), leading to more absorption of water and expansion of pellets during storage period. Relaxed densities of FRPs were found higher than the initial and relaxed density of SPs at different MC and particle size.

Polynomial regression analysis was performed to model the relationship between the MC and pellet density (particle and relaxed) of FRPs using Equation 5. The model was well fitted to the experimental results of pellet density with R^2 ranging from 0.99-1 for 0.25-0.5 and 0.5-1.41 mm, respectively.

True density of FRPs (0.25-0.5 mm) decreased with MC increasing from 5.5 to 8% (Figure 1c). Above MC of 8%, true density slightly increased for same particles but were found near to those of MC 5.5%. The highest true density for corresponding particles was found as 1.504 g/cm^3 at MC of 5.5%. For particles 0.5-1.41 mm, true density increased with MC increasing

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from 5.5 to 13%, the highest (1.496±0.02 g/cm^3) was achieved at MC of 13%. Moreover, FRPs (0.5-1.41 mm) were found with lower true density at all MCs as compared to 0.25-0.5 mm. More condensed structure of pellets made from larger particles, expansion in pellets volume due to absorption of more moisture and higher pressure (100 MPa) might be the reasons for lower true densities of corresponding pellets.

For SPs (0.25-0.5 mm), true density enhanced slightly with MC increasing from 5.5 to 8%. True density decreased at MC of 13 and 18% (Figure 1d). SPs (0.5-1.41 mm) followed the same trend. Higher MC (e.g., 28%) led to a slight increase in true density of SPs produced from both particles.

Comparing the pellet quality, FRPs had higher pellet density than those of SPs, indicating the potential value of FR for cleaner fuel production.

3.1.2 Influence of particle size

Particle size plays an important role in biomass pelletization and pellet quality is affected by particle size. Particle size considerably affect the biomass compression, contact between the adjacent particles flow of material and friction in the pellet press. We found interesting results for pellet density of FRPs. FRPs (0.5-1.41 mm) were found more denser, achieved higher pellet density than FRPs (0.25-0.5 mm) as shown in Figure 1a. Strong mechanical interlocking of larger particles of FR led to higher particle density, consistent to previous studies [36, 60, 61]. Further, it could be possible that the adsorbed water accrued on small particles (0.25-0.5 mm) surface, forming a thin film, thus inhabiting the intermolecular forces between the adjacent particles and resulting in poor binding. Therefore, FRPs (0.25-0.5 mm) achieved lower particle density as compared to FRPs (0.5-1.41 mm). A similar trend was observed for the relaxed density of FRPs made from both particle sizes. True density of FRPs fabricated from particle size of 0.25-0.5 mm was found higher than those made from particles of 0.5-1.41 mm (Figure 1c).

SPs fabricated from particle size of 0.25-0.5 mm achieved the higher pellet density i.e. particle, relaxed and true density than SPs made from particles of 0.5-1.41 mm (Figure 1b-1d). This could be due to small size particles had filled the spaces and voids of large size particles by rearrangement of particles during pelletization, provided larger specific surface. Thus intensified the binding between the biomass particles and hence resulted in more compact structure with higher density.
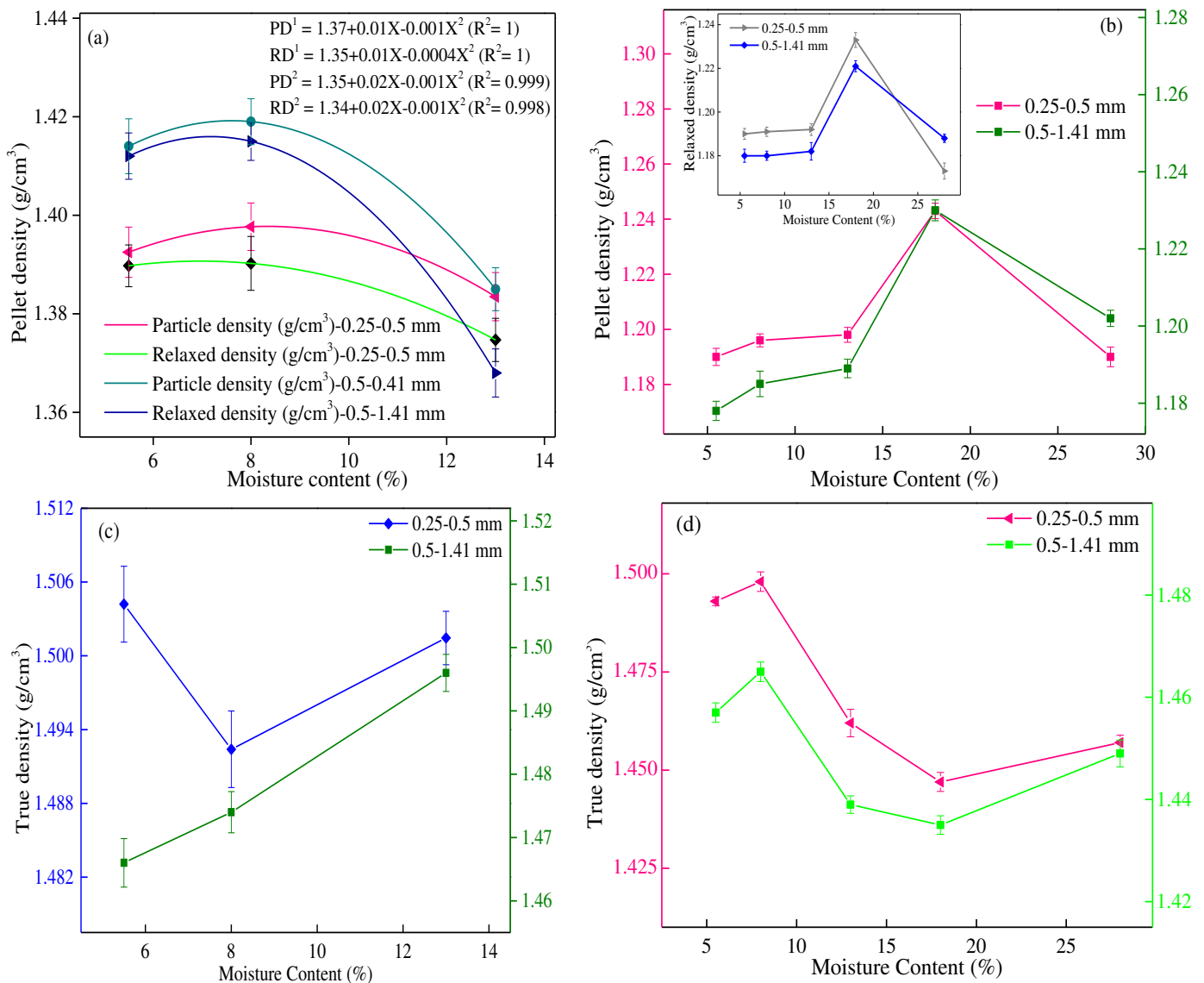

Figure 1. Influence of MC and particle size on particle and relaxed density (a) FRPs, (b) SPs and true density (c) FRPs and (d) SPs

3.1.3 Influence of a binder

Biomass materials containing lower content of lignin, protein, starch etc., may require addition of binder to enhance the binding characteristics and pellet quality (e.g., density

strength and hardness) by promoting the strong inter particle bonding and to reduce energy consumption. For selecting an appropriate binder, consideration must be paid on the price and environmental affability of a binder. Using SR as a binder, showed no any significant enhancement in particle density of FRPs. FRPs with no use of binder were found with higher particle densities compared to those with use of binder at the same process conditions (Table 5). SR limited the moisture and lignin to expel out of FR, reduced compressibility, contact area of FR particles, and hence resulted in poor binding between the adjacent particles. However, relaxed density FRPs remained quite similar as of initial particle density, indicating the dimensional stability of the formed pellets. True density decreased with use of binder and were found lower than no use of binder due mainly to condensed structure of pellets.

SR was found beneficial for compressing sawdust, enhanced the binding characteristics of pellets. For SPs (0.25-0.5 mm), particle density enhanced from 1.19 ± 0 to 1.240 ± 0.02 g/cm^3 and from 1.17 ± 0.01 to 1.219 ± 0.02 g/cm^3 (for 0.5-1.41 mm), respectively. SPs remained dimensionally stable and their relaxed density by employing SR as a binder was found nearly same with initial particle density. Addition of binder decreased the true density of SPs.

Table 5. Effect of binder on pellet density

| SR/FR [a] | Furfural residue pellets | | |
	Particle size	Particle density (g/cm^3)	Relaxed density (g/cm^3)	True density (g/cm^3)
1:9	0.25-0.5 mm	1.373±0.01	1.373±0.01	1.423±0.02
	0.5-1.41 mm	1.374±0.01	1.375±0.01	1.404±0.02
SR/SD [b]	Sawdust pellets			
1:9	0.25-0.5 mm	1.24±0.02	1.238±0.02	1.391±0.01
	0.5-1.41 mm	1.219±0.02	1.239±0.01	1.376±0.01

[a] SR/FR/SD = synthetic resin/furfural residue/sawdust

The current study implies that pelletization of FR require no any additional binder to enhance the pellet quality, provided that energy consumption is compromised with quality and hence is beneficial from economic perspective.

3.2 Strength and hardness of pellets

3.2.1 Effect of MC, particle size and a binder

Table 6 manifests the mechanical characteristics of FRPs and SPs and their dependency on MC. Strength and hardness of pellets are the quality indicators of pellets, representing the deformation and breaking resistance of an individual pellet during handling, transportation and storage [62]. Biomass pellets are vulnerable to the external forces [8], and pellets with lower strength and hardness are easy to break and produce dust emission during handling, transportation and storage. The strength of FRPs decreased from 6.288±0.71 to 2.069±0.43 MPa (0.25-0.5 mm) and 4.395±0.51 to 2.496±0.32 MPa (0.5-1.41 mm) with increasing MC from 5.5% to 13%. The decrease in strength might be due to incapability of fibrous biomass. Accumulation of moisture on the surface of FR particles caused the extra particle-to-particle lubrication, consistent to previous study [63]. An opposite trend was observed for SPs. The strength of SPs increased from 3.797±0.25 to 6.03±0.09 MPa and 3.19±0.48 to 3.877±0.06 MPa with MC increasing from 5.5 to 18%. At MC of 28%, strength of SPs decreased. Release of excessive water during pelletization prevented the expulsion of natural binding agents from biomass particles due to incompressibility of water [10], led to lower strengths of pellets particularly at higher MC (e.g. 8-13% for FR and 28% for sawdust). FRPs were found more strong, the strength was 3.9% (at 5.5% MC) and 11.8% (at 5.5%MC) consistent to particles of 0.25-0.5 and 0.5-1.41 mm, respectively higher than SPs at same particle size and MC of 18%.

Table 6 shows the hardness of FRPs and SPs made at different MCs. Hardness of FRPs decreased with MC increasing from 5.5 to 13%, the highest (401.331±13.75 N/mm^2) for 0.25-0.5 mm and 328.042±15.29 N/mm^2 for 0.5-1.41 mm was achieved at MC of 5.5%. Meanwhile, the hardness of SPs increased with MC increasing from 5.5 to 18% and at MC of 28% it decreased. Increase in hardness of SPs may possibly be due to decrease in porosity with increasing MC, creating a large bonding area, hence increased the hardness.

Pellets made from FR were conceivably found with higher hardness than those of SPs. It is worth to mention that, the FRPs made from MC of 8-13% were found with higher hardness as compared to SPs made at MC of 5-28% (Table 6).

Particle size considerably affected the strength and hardness of pellets. Small particles (0.25-0.5 mm) were found with higher values of strength and hardness. This could be due to enhancement of natural binder (e.g. lignin) flow, large contact areas of particles, better heat transfer rate, and better interactions between steam and natural binder (e.g., lignin). For FRPs, 5.5% MC was optimum for achieving the higher strength and hardness and 18% for SPs, respectively.

Addition of SR to FR and sawdust increased the strength and hardness of pellets made from particles of 0.25-0.5 and 0.5-1.41 mm (Table 6). SR was only added to biomass materials, which had MC of 5.5%, and pellets were only made from corresponding materials. SR is waste cooking oil, lubricated the material (e.g., FR), and thus had shown no any considerable effect on strength and hardness of pellets. However, addition of SR to sawdust has positive effect on strength and hardness of pellets. Addition of SR filled the voids of sawdust particles and created strong mechanical interlocking of particles, thus increased the strength and hardness of SPs.

Table 6. Effect of MC, particle size and a binder on strength and hardness of pellets

Process parameters	Furfural residue pellets		Sawdust pellets	
	0.25-0.5 mm	0.5-1.41 mm	0.25-0.5 mm	0.5-1.41 mm
MC (%)	Mean (N/mm^2)	Mean (N/mm^2)	Mean (N/mm^2)	Mean (N/mm^2)
Strength (MPa)				
5.5	6.288±0.71	4.395±0.51	3.797±0.25	3.19±0.48
8	2.64±0.22	2.383±0.33	3.149±0.18	3.476±0.07
13	2.069±0.43	2.496±0.32	3.3±0.16	3.02±0.5
18	N/A		6.03±0.09	3.877±0.06
28	N/A		3.661±0.07	3.812±0.06
Use of SR				
5.5/1:9	6.354±0.42	4.48±0.31	4.368±0.53	3.898±0.36
Hardness (N/mm^2)				
	Mean (N/mm^2)	Mean (N/mm^2)	Mean (N/mm^2)	Mean (N/mm^2)
5.5	401.331±13.75	328.042±15.29	62.575±9.67	55.203±9.07
8	183.86±1.83	157.626±2.94	63.781±1.54	56.644±2.61
13	90.655±2.85	98.315±2.98	69.484±1.23	67.257±1.63
18	N/A		96.064±0.96	87.288±1.66

28	N/A	78.585±0.64	71.226±1.62
Use of SR			
5.5/1:9	412.331±16.36	342.042±19.32	78.564±11.67	8.443±12.26

<div align="center">N/A= not available</div>

3.3 Effect of MC, particle size and binder on energy consumption

Minimum energy consumption for pellet production is highly desirable in pellet industry and is one of critical issue. MC, particle size and a binding agent significantly affected energy consumption of FRPs and SPs. Table 7 presents the energy consumption of pelletizing FR and sawdust.

Energy consumption of FRPs and SPs made from both particle sizes decreased with MC increasing from 5.5 to 13% (for FRPs) and 5.5% to 28% for SPs (Table 7). The lowest energy consumption of 16.16±0.61 J/g (0.25-0.5 mm) and 14.99±0.66 (0.5-1.41 mm) was achieved at MC of 13%. The decrease in energy consumption was due to plasticity of biomass material at 130°C temperature and 100 MPa pressure and reduction of friction between the biomass particles and wall of the die. Further, due to lubricating and adhesive action between the particle when water was added to biomass materials and substantial interaction between the moisture and natural binder (e.g., lignin), resulted in decrease of energy consumption consistent to previous study [42].

Energy consumption of SPs decreased from 25.44±0.4 to 20.50±0.6 (for 0.25-0.5 mm) and 26.24±0.4 to 20.22±0.72 (for 0.5-1.41 mm) with MC increasing from 5.5 to 28% (Table 7). Higher MC decreased energy consumption by promoting the better boding of sawdust particles and made particle flow easily, lowered the friction between sawdust particles.

Energy consumption of FRPs made at all MC (5.5 to 13%), was found much lower as compared to SPs made at 5.5-28% MC. Difference in glass transition behavior of FR and sawdust under different MC might be reason for lower energy consumption of FRPs.

Furthermore, difference in chemical composition and compaction characteristics of FR and sawdust the energy consumption was different. Pelletization of FR required less energy

consumption compared to compressing sawdust. Lower energy consumption of pelleting FR indicating the higher feasibility of FR for pellet production, opening more pathways for its utilization more economically.

Particle size had considerably affected the energy consumption of pellets. For FRPs, smaller particles (0.25-0.5 mm) achieved lower energy consumption than those of larger particle size (0.5-1.41 mm) (Table 7). Energy consumption of SPs made from 0.5-1.41 mm was slightly higher than those made from particle size of 0.25-0.5 mm. However, energy consumption values for SPs made from both particles were very close to each other (Table 7). Mechanical resistance for compressing sawdust of larger particles (0.5-1.41 mm) was high as compared to particles 0.5-0.5 mm, increasing the friction between the biomass and pelletizer die, decreasing the effects of lubricant (e.g., water) and binding agents (e.g., lignin) and hence higher energy consumption. Furthermore, moisture evaporated quickly from the larger particles due to the large void fraction as compared to 0.25-0.5 mm particles, consequently required more energy for pelleting sawdust.

Adding additional binder to FR and sawdust reduced their energy consumption (Table 7). Energy consumption of FRPs decreased by 15.76% (0.25-0.5 mm) and 12.1% (0.5-1.41 mm), respectively. Addition of binder promoted the lubricating affect and decreased the friction between the particles and the particles and walls of the die, thereby reduced the energy consumption of corresponding pellets. Similarly, using SR as a binder decreased the energy required for pelleting sawdust. Energy consumption of SPs decreased by 8.6% (0.25-0.5 mm), and 10.9% (0.5- 1.41 mm), respectively.

Table 7. Effect of MCs and a binder on energy consumption of pellets

Process parameters	Furfural residue pellets		Sawdust pellets	
	0.25-0.5 mm	0.5-1.41 mm	0.25-0.5 mm	0.5-1.41 mm
MC (%)	Mean (J/g)	Mean (J/g)	Mean (J/g)	Mean (J/g)
5.5	22.96±0.59	21.14±0.35	25.44±0.4	26.24±0.4
8	18.61±0.64	16.66±0.81	25.25±0.4	25.86±0.87
13	16.16±0.61	14.99±0.66	25.39±0.32	25.66±0.39

18			22.65±0.59	24.62±0.88
28			20.50±0.6	20.22±0.72
Use of binder				
SR/FR/SD [a]	Mean (J/g)	Mean (J/g)	Mean (J/g)	Mean (J/g)
1:9	19.34±0.65	18.58±0.24	23.25±0.58	23.38±0.34

[a] SR/FR/SD = synthetic resin/furfural residue/sawdust, N/A= not available

3.4 Higher heating value (HHV) of pellets

Table 1 represents the HHV of FRPs and SPs. HHV of 23.36 MJ/kg and 22.88 MJ/kg for FRPs made from particles 0.25-0.5 and 0.5-1.41 mm were found very close to each other. HHV of corresponding pellets were found similar to the previous studies, for example 22.93 MJ/kg for Caragana torrefied pellets [46], 16.34 for cow dung pellets [64] and 17.5-22.65 MJ/kg for torrefied pellets, depending on feedstock type and composition [65–67]. For SPs, HHV of 20.6 MJ/kg and 21.21 MJ/kg were found for particles of 0.25-0.5 and 0.5-1.41 mm, respectively. FRPs had higher HHV compared to SPs.

3.5 Pellet quality comparison with previous studies

Jaya Shankar Tumuluru et al.[68] reported that, the highest pellet density was found in the range of 1.056-1.080 g/cm^3 at lower MC i.e. 5.1% vs 11.8% (temperature 50-100°C). Nguyen et al. [38] found that increasing MC and particle size had shown negative impact on the particle density of pellets made from woody biomass. The highest particle density of >1.107 g/cm^3 and strength of >62 N/mm ^{2} were obtained at >8.1 % MC and 125°C. Highest particle density of 1.062 g/cm^3 (cob pellet), 1.097 g/cm^3 (stalk pellet) and 0.995 g/cm^3 (husk pellet) was found at MC of 10%, temperature of 80°C, pressure of 150 MPa and particle size of 0.5-0.8 mm [69]. Our findings were supported by these studies. For FRPs, the highest particle density of 1.398 g/cm^3 and 1.419 g/cm^3 consistent to particle size of 0.25-0.5 and 0.5-1.41 mm was achieved at MC of 8%. For SPs, highest particle density of 1.243 g/cm^3 for 0.25-0.5 mm and 1.230 g/cm^3 for 0.5-1.41 mm was found at MC of 18%. Particle density of FRPs were found higher than the above-mentioned studies found for different pellets.

Increasing particle size and MC decreased the particle density of wood pellets and highest particle density of 1.201 g/cm^3 was obtained at 3.175 mm vs 6.35 mm and 17.5% vs 20%. However, the authors found the opposite trend for strength of pellets, where strength of pellets increased from 5.2 to 7.1 MPa with increasing particle size from 3.175 to 6.35 mm at MC of 17.5% and 2.1 to 3.5 MPa at 20% MC [34]. Similar observations were found in the current study, where strength decreased with increasing MC from 5.5 to 13 % and particle size from 0.25-0.5 to 0.5-1.41 mm for FRPs and >18% for SPs. Lisowski et al. [70] reported that, at higher MC i.e. >20% strength of biomass pellets decreased, due to incapability of fibrous biomass material to absorb water. Maximum strength of spruce pellets (higher than 10 N/mm) was found at MC of 8.3% (at pressure of 300 and 400 MPa)[45]. Another study found the maximum strength of 2.68 MPa for wheat straw pellets at MC of 23.2%[71]. Mohammad Ramezanzade et al. [72] recommended the MC of 11.7%, particle size of 1.65 mm and temperature of 100°C for obtaining the higher quality of biomass pellets (i.e. density 1.085 g/cm3 and strength 41.57 N/mm). Highest strength of 6.3 MPa (0.25-0.5 mm), 4.4 MPa (0.5-1.41 mm) for FRPs, 6.03 MPa (0.25-0.5 mm), and 3.88 MPa (0.5-1.41 mm) for SPs were found comparable to previous studies.

Increasing MC from 5 to 20% decreased the energy consumption of sludge mixed pellets. Similar observations were noted in current study for FRPs and SPs. Use of binders significantly reduce the energy consumption of pellets. For example, Donghui Lu et al. [43] found that addition of binder to biomass materials lowered the energy consumption and improved the pellet quality. Mediavilla et al. [44] added adopted maize starch as a binder and reported that that energy consumption of poplar energy crop pellets decreased by about >42%, depending upon the ratio of binder being used. In current research, energy consumption of FRPs decreased by 15.76% (0.25-0.5 mm) and 12.1% (0.5-1.41 mm) and by 8.6% (0.25-0.5 mm), and 10.9% (0.5- 1.41 mm) for SPs, respectively with the use of binder.

4 Conclusion

Biomass pellets are considered as clean and environmental friendly fuels for various applications. Effects of MC, particle size and a binder on pellet density, strength and hardness were investigated. MC had considerable influence on density of FRPs and SPs, up to certain level (5.5-8% for FRPs) and 5.5 to 18% for SPs, where density enhanced. Higher MC i.e., 8-13% for FRPs and >18% for SPs negatively affected the relaxed density, strength and hardness of both FRPs and SPs. The lowest energy consumption of FRPs and SPs were found at 13% and 28%. Addition of binder to FR had shown no any considerable influence on FRPs quality, whereas it had improved the SPs quality. Adding binder to biomass materials abridged the energy consumption of both kinds of pellets. FRPs were found with better quality and minimum energy requirement than SPs.

Satisfactory findings of current study implied that pelletization of FR and sawdust revalue these low value added materials for obtaining cleaner solid fuels.

Bulk density, durability and final MC of pellets were not taken into account in current study. Further, economics of pelletizing FR would be a topic of greater interest. Future studies may perform co-pelletization of FR with other biomass sources (woody biomass, agricultural residues and sewage sludge) to investigate more about potential applications of FR for fuel (e.g. pellets) production.

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Declarations

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Figures

The equations shown in panel (a):

$$PD^1 = 1.37 + 0.01X - 0.001X^2 \ (R^2 = 1)$$
$$RD^1 = 1.35 + 0.01X - 0.0004X^2 \ (R^2 = 1)$$
$$PD^2 = 1.35 + 0.02X - 0.001X^2 \ (R^2 = 0.999)$$
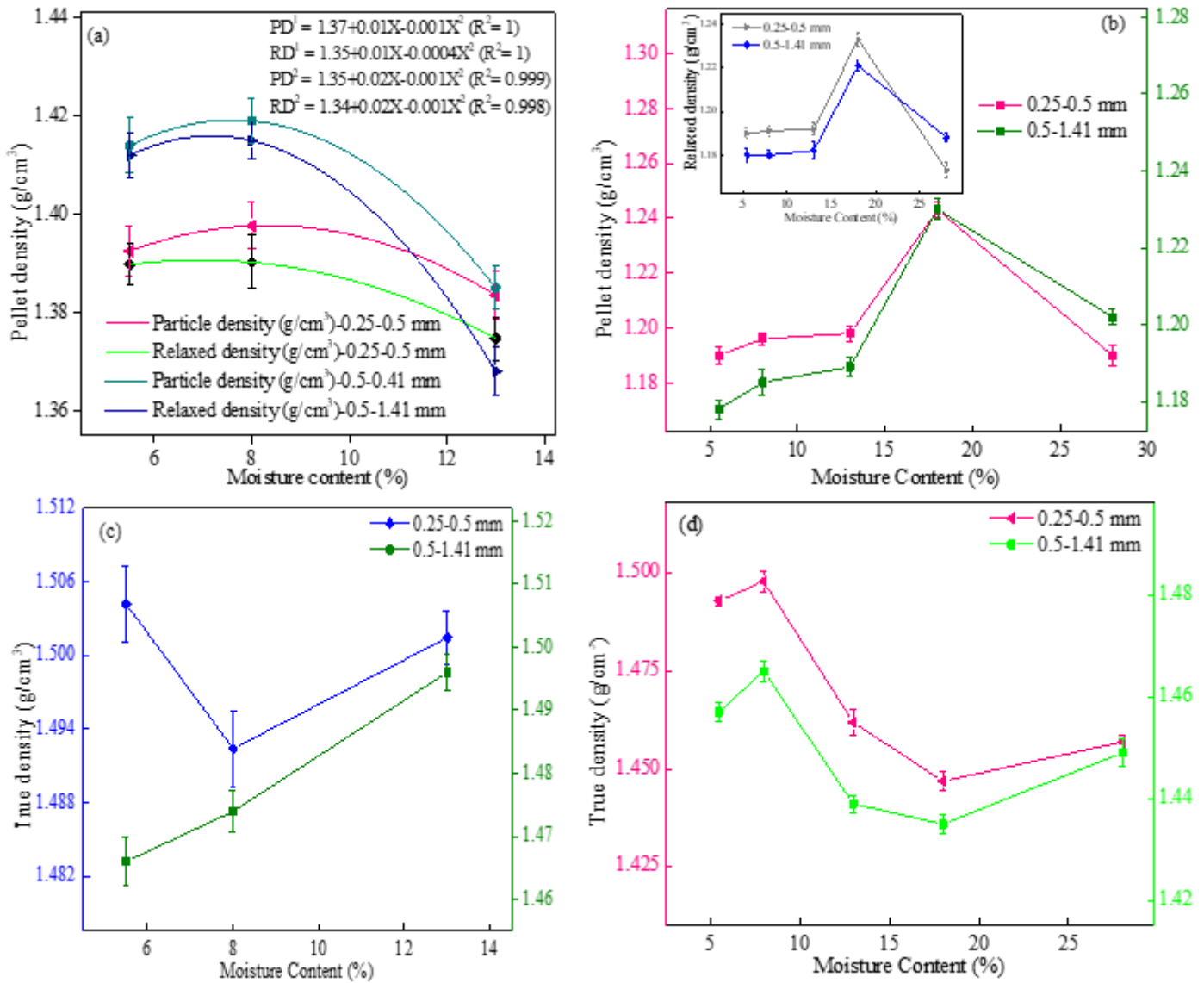$$RD^2 = 1.34 + 0.02X - 0.001X^2 \ (R^2 = 0.998)$$

Figure 1

Influence of MC and particle size on particle and relaxed density (a) FRPs, (b) SPs and true density (c) FRPs and (d) SPs