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The effects of sugar beet molasses on wheat straw pelleting and pellet quality. A comparative study of pelleting by using a single pellet press and a pilot-scale pellet press

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ABSTRACT: The main aim of this paper is to investigate the effects of molasses on wheat straw pelleting and physical pellet quality. Molasses was added at weight fractions of 1.5% and 3%, while pure straw served as a control. The effects of molasses were examined by producing pellets in a single pellet press (SPP) and in a pilot-scale pellet press (PSPP). The second aim of this study was to compare the results obtained from the SPP and the PSPP, i.e., to understand how the information from the SPP can be used for the prediction of material behavior, process adjustments, and improvement of pellet quality in an upscale pelleting process. The production and pellet quality parameters were compared and information from the two pelleting methods was combined by response surface modeling. Pellet density was the response variable, while pelleting pressure and temperature were the independent variables. Large differences in pellet quality were observed between the two pelleting methods. These differences are discussed from the perspective of technical differences in the pelleting procedures and different fiber orientations in the pellets. The results indicate that pelleting temperature is a key factor for achieving good pellet quality of all the samples. Exceeding the glass transition temperature of lignin leads to significantly better pellet quality and facilitates pelleting. The results showed that molasses strengthens pellets produced at temperatures below the glass transition of lignin. Addition of molasses at higher pelleting temperature did not significantly affect the pellet quality and processability.

Keywords: straw pellets, molasses, single pellet press

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1. Introduction

Cereal straw, an agricultural byproduct, is one of the alternatives to forestry biomass, mainly because it is available in large quantities and is widely accessible. Application of agro-residues as a renewable energy source is particularly important in agricultural regions that lack forests because it can be a significant factor for rural development and sustainable agricultural production.

Straw has very low bulk and energy density; thus, densification by means of baling, cubing, pelleting, or briquetting are techniques to overcome these disadvantages. Wheat straw is a lignocellulose material with a higher amount of ash, proteins, and extractives compared to wood [1]. The chemical composition of wheat straw can vary greatly because of differences in varieties, cultivation conditions, and location [2]. Agro-residues, such as straw and energy grasses, are more challenging to densify than wood because of the lower content and different structure of lignin, the lower bulk density, and the presence of the cuticula (a hydrophobic layer of cutin and waxes on the surface of straw) [3-5]. The compaction characteristics of different agro-residues have been examined in different studies [6-8]. Research on pelleting of agro-residues has been focused on establishing a cost-competitive process without compromising pellet quality. Application of various binders and blending with wood are common ways to reinforce pellets and reduce the energy consumption for densification. Common binders are starch, lignosulphonate, crude glycerin, bentonite, and molasses [9, 10]. Molasses can help binding among particles during pelleting [10-12]. Soluble sugars recrystallize after drying and cooling of compacts and form solid bridges. A major challenge and obstacle for wider application of molasses is its high viscosity and sticky nature [13]. Heating and/or dissolving molasses are approaches to facilitate its application and to improve its distribution in powders. The application of molasses as a binder in pellet production is known, but a detailed study investigating the effects of temperature, compacting pressure, and amount of molasses on various pellet quality parameters (e.g., density, strength, hydrophobicity, and water activity) is lacking. The single pellet press (SPP) method for testing a material's compacting properties has been widely used in recent years [14-18]. This method can provide information about the material compressibility and estimate some pellet quality properties (strength, density). Information about the processability is limited to properties linked to the die friction (p_{max}) and yield stress of

materials [19]. p_{max} represents the minimum pressure required to initiate the motion of a pellet in the compressing channel at a certain speed and density [19-22]. Some studies showed how to estimate the energy consumption in a SPP by calculating the area under the pressure-displacement curve [23, 24]. Significant work to understand the information from a SPP has been conducted by Holm et al. [21, 25] and Nielsen et al. [15]. These studies focused on analytical and empirical studies of the pelleting process. Holm et al. [25] found that the pelleting pressure increases exponentially with pellet length in a SPP and that it also depends on the process temperature and biomass properties. Nielsen et al. [15] divided the pelleting process into three sections, aiming to estimate the contribution of each section to the overall energy consumption. The first section involves the process of material pre-compaction that occurs in the nip area of a pellet press (compression component). In the following section, located in the die entry, the material is forced to flow into the die holes (flow component). The final section involves friction between the flowing material and the die wall, as well as the energy required to move the compressed material into the die (friction component). The highest portion of energy is needed to force the compressed layer of material to flow into the die (flow component). Nevertheless, there is still a gap in the knowledge and a lack of practical strategies for applying the information from a SPP in a scaled-up pellet press. Two recent studies, by Shang et al. [26] and Puig-Arnavat et al. [27], aimed to extrapolate the results from a SPP to a bench scale pellet mill to find its optimal process parameters. However, finding more information that can aid in the application of SPP data for scale-up and/or optimization of the roller – die pellet press is important as well.

The present study has two objectives: 1) to investigate the effects of sugar beet molasses as a binder on the processability and quality of wheat straw pellets. Pellets were produced by a SPP method at different temperatures and compacting pressures, and in a pilot-scale pellet press (PSPP); 2) to compare results from SPP and PSPP pelleting, i.e., to understand how the information from a SPP can be used for anticipating material behavior, processing, and product quality in roller-die pellet press pelleting.

2. Material and methods

2.1. Material

The wheat straw (*T. aestivum L.* 'Simonida') was collected after the summer harvest in 2014 in the Serbian northern province of Vojvodina. Straw bales were placed in dry storage until they

were used, for about two months. Before preparing the material for pelleting, the moisture content in the straw was 10.62 ± 0.09%. The moisture content was determined by drying in an oven (Memmert UNB400, Memmert GMBH, Schwabach, Germany) at a temperature of 105°C in air atmosphere until constant mass is achieved (EN 14774-1)[28].

The sugar beet molasses used in the experiment was obtained from Crvenka Sugar Factory a.d. in Serbia. The dry matter content in the molasses was 84%, as determined refractometrically by an Abbe 5 refractometer (Carl Zeiss Jenna, Switzerland) following the SRPS E.L3.020:1963 standard [29].

The wheat straw was ground by a hammer mill (Type 11, ABC Inženjering, Pančevo, Serbia) with a screen size of 3 mm. The particle size distribution (PSD) of the ground straw was determined by a sieving test; the results are presented in Fig. 1. The ground material was moisturized to 21% moisture content by spraying water through a nozzle (Düsen-Schlick GmbH, Germany, Model 970) adapted to a double-shaft pedal mixer/vacuum coater (F-6 RVC, Forberg, Norway). The moisturized material served as a control sample (referred to as PS below). Molasses was added in two different amounts, 1.5% (M_1) and 3% (M_2), by spraying previously dissolved molasses over the bulk straw powder in the mixer. The moisture content was the same (≈21%) for all three mixtures. The moisture content was determined by MB45 thermogravimetric moisture analyzer (Ohaus, USA), following the procedure described in the instruction manual [30]. Bulk densities of the raw materials was measured in a bulk density tester (Tonindustrie, West und Goslar, Germany). The bulk density was determined by measuring the mass of 1 liter of material that has been loosely poured into the tester's cylinder. The water activity value (a_w) was determined by a Rotronic HygroLab C1 (Switzerland) instrument [31]. The average temperature during a_w measurements was 22.3±0.3ºC. The content of ash and volatile matters was determined in a muffle furnace (Nabertherm D-2804, Germany) according to EN 14775 [32] and EN 15148 [33] standard procedure, respectively. The fixed carbon was calculated by difference between 100 and the sum of the volatile matter and ash content. The higher heating value (HHV) was determined by an adiabatic bomb calorimeter IKA C 200 (Germany), following the EN 14918 standard procedure [34].

2.2. Pellet production.

In this study, pelleting of the straw with different molasses levels was tested in a PSPP (Model 14-175, Amandus Kahl GmbH&Co., KG Germany) and a SPP designed and manufactured at the Norwegian University of Life Sciences (Ås, Norway)[14, 17, 19]. Straw was ground, moisturized, and mixed with molasses just before PSPP pelleting. The same powders were stored in plastic bags at +4°C until further usage for single pellet production.

1) Cold pelleting (without steam conditioning) was performed in a pilot-scale production facility at the Institute of Food Technology (FINS) in Novi Sad, Serbia. Pellets were produced in a flat die pellet press with a die compression ratio of 3 (die hole diameter 6 mm, die thickness 18 mm). Samples of pellets were taken when steady-state production was achieved. The temperature of the outer surface of the die and the temperature of the pellets exiting the die hole were recorded. The die surface temperature was measured on the outer die wall by using a PT100 resistance thermometer. The pellet temperature was measured by a contact thermometer placed in the bulk of pellets after they exited the die. The production rate was 5.1 kg/h at a steady temperature of about 80°C. The specific energy consumption of the pelleting process was calculated by the following equation:

$$E_{sp} = \frac{E - E_0}{Q} \cdot 1000 \qquad (1)$$

where E_{sp} is the specific energy consumption (kWh/t); E is the energy consumption during the material pelleting (kW); E_0 is the energy consumption during idle running (0.3 kW); and Q is the material throughput (kg/h).

Pellets were left to cool overnight at ambient conditions and then stored in sealed plastic bags until further testing was conducted.

2) SPP pelleting was performed in a blank die unit with a pellet channel diameter of 6 mm (same diameter as in the Khal pellet press). The design of the SPP and the detailed pelleting procedure were presented in our previous publications [17, 22]. The SPP cannot completely replicate all pellet mill operations. One of the main differences is the absence of particle size reduction that occurs in the roller-die gap and in the nip area of a roller-die pellet press [17]. In order to avoid a difference in PSD, the materials for SPP pelleting were finely ground to attain a

PSD as similar as possible to the PSD of pellets after their production in the Kahl pellet press. The PSD of particles in pellets was determined by wet sieving using an AS 200 Control, Retsch (Germany) laboratory sieving machine following the procedure described by Miladinovic [35]. A total weight of 100 g of pellets was dry sieved using a sieve with an opening size of 4.8 mm to separate dust and crumbles. Then, the pellets were immersed in 500 ml of tap water for 2 h at room temperature and were stirred once after 1 h. The soaked pellets were then poured onto the sieves. The sieving amplitude was set to 1.2 mm, and the sample was washed through the set of sieves for 3 × 3 × 3 min. The PSD was calculated based on the proportion of dry matter left on each sieve after drying overnight at 105°C. The results of wet sieving of pellets (Fig. 1) were used to adjust the PSD.

Figure 1. Particle size distribution of the straw before (dry sieving) and after (wet sieving) PSPP pelleting

SPP pellets were produced at four compacting pressures (75, 150, 225, and 300 MPa) and three pelleting temperatures (60, 120, and 180°C). A wide range of pelleting conditions was selected to follow the changes in pellet quality over different pelleting pressures and temperatures. The force required to initiate pellet discharge from the die was recorded, and the discharging pressure (p_{max}) was calculated, with an aim to estimate the difference in energy requirements for the pelleting of tested materials. Estimation of the "spring back effect" (through axial pellet expansion) was accomplished by measuring the length of pellet in the die immediately after the end of compaction and after cooling of pellets (after 48 h).

In addition to pellets produced to test the compressibility, strength, and pelletability of the material (experimental runs described above), another series of single pellets were made according to a central composite design (CCD) in order to use the data for response surface modeling (RSM). The coded variables, natural values, and corresponding response values are presented in Table 1. RSM was used to model the dependence of the pellet density on the independent variables (pelleting temperature and compacting pressure) and to understand the interactive effect of the process variables on the pellet density. Contour plots were generated from the model and used to correlate the density of pellets from the PSPP with the density of

SPP pellets. The second-order polynomial model used in the response surface analysis was as follows:

$$y = \beta_0 + \beta_1 x_1 + \beta_2 x_2 + \beta_{11} x_1^2 + \beta_{22} x_2^2 + \beta_{12} x_1 x_2 + \varepsilon \quad (2)$$

where y is the response variable (density), β_0 is the intercept, β_1 to β_{12} are the regression coefficients, x_1 and x_2 are the coded independent variables (temperature and pressure), and ε is the error term. The coefficients of the equation and the contour plots were generated by Minitab® 16.2.2 software.

Table 1. Experimental design (CCD) and response data for RSM modeling

2.3. Analyses of pellets produced by two methods.

The analyses performed on the pellets produced by both methods (PSPP and SPP) are listed below.

Durability

PSPP: Pellet durability was determined by a Holmen Pellet Tester (New Holmen NHP100 Portable Pellet Durability Tester, TekPro Ltd., Norfolk, UK). The device consists of a perforated test chamber, which was loaded with 100 g of pellets during the test. Before the durability test pellets were sieved to separate dust and smaller particles on a sieve with an opening size of 4.69 mm. The sieve size was selected according to the recommendation of Thomas et al. [36](0.8 × pellet diameter). The sieved pellets were rapidly fluidized in an air stream. The total testing time was 30 s. Fines formed during the fluidization were separated from the pellets by sieving, and the final weight of the pellets was recorded. Durability was calculated as the mass fraction of the dust-free pellets.

SPP: The low production rate in the SPP (5–6 pellets per hour) makes the durability test unsuitable because it requires a bulk quantity of pellets.

Bulk density

PSPP: The bulk density of the pellets was measured in a bulk density tester (Tonindustrie, West und Goslar, Germany) following the same procedure as described for powdered raw materials.
SPP: The bulk density of SPP pellets was not measured because of the low production rate, and correspondingly small sample size (5-6 pellets per hour).

Moisture content and a_w

PSPP and SPP: The moisture content was determined by drying manually crushed pellets in an oven (Memmert UNB400, Memmert GMBH, Schwabach, Germany) at a temperature of 105°C in air atmosphere until constant mass is achieved (EN 14774-1) [28]. The a_w value was measured at an average temperature of 21.5±0.2°C by Rotronic HygroLab C1 (Switzerland) [31].

Pellet dimensions

PSPP and SPP: The lengths and diameters of pellets were measured using a digital caliper. The lengths of PSPP pellets were not precise enough because of the irregularity of the pellets' ends (not perfect cylinders).

Amount of fines

PSPP: The amount of fines in the pellets at the outlet of the pellet press was determined by sieving the pellets on a sieve with an opening size of 4.69 mm. The amount of fines lost in the separation was determined by weight difference.
SPP: This test is not suitable for SPP pellets, because of the same reasons as stated for durability and bulk density.

Pellet density

PSPP: The pellet density was determined using a density measurement kit (Mettler Toledo GmbH, Switzerland) attached to an analytical scale (MS204S, Mettler Toledo GmbH, Switzerland). The mass of a pellet was measured in air and in vegetable oil sequentially, at room temperature (23°C), and the density was determined by Archimedes' law [37]. Vegetable oil was used instead of water to avoid fast pellet disintegration and dissolution of water-soluble components.

SPP: The density of SPP pellets was determined by measuring a pellet's weight and volume. For pellet volume calculation, the length and diameter of a pellet were measured using a digital caliper.

The densities of the pellets produced in the PSPP and SPP were determined by different methods. The method based on Archimedes' law was chosen for PSPP pellets as an alternative to the density determination by measuring pellet volume and mass, because of the irregular pellet shape and the inability to precisely measure the pellet length (not perfect cylinders).

Pellet strength

PSPP and SPP: The strength of pellets was evaluated through a three-point bending test. The three-point bending test was selected in lieu of the typically used diametric compression test because of the inability to precisely determine the length of PSPP pellets, and thus, to correctly interpret the results. The three-points bending test was used to asses pellet strength in the recent publication of Craven et al. [38]. The test is explained in detail in Li et al. [39]. A pellet was placed on a specially designed holder attached to the Lloyd LR 5K texture analyzer (Fig. 2a). The pellet was loaded at a speed of 1 mm/min until breakage, and the force was recorded. The maximum bending stress (σ) for cylindrical pellets was calculated by the following equation [39-41]:

$$\sigma = \frac{8FL}{\pi d^3} \qquad (3)$$

where F is the load at the point of breakage, L is the support span (8.5 mm), and d is the pellet diameter. This test can replicate the type of loading during storage, transportation, and handling (Fig. 2b).

Figure 2. Three-point bending test: a) Pellet fracture during three-point bending; b) Illustration of the loads on pellets during storage (red circles show examples that confirm the relevance of the three-point bending test).

Contact angle and roughness measurements

PSPP and SPP: Differences in the surface hydration properties of the pellets caused by molasses addition were assessed by measuring the contact angle (θ) of a sessile water drop placed on a diametrically positioned pellet, and its changes over time. The θ measurements were conducted at room temperature with a video-based optical θ-measuring device OCA 15EC (DataPhysics Instruments GmbH, Germany). A drop of distilled water (3 µl) was discharged from an automatic dosing syringe onto a cylindrical pellet surface and a video of the drop absorption was recorded. Videos were afterwards analyzed by SCA 20 software 15EC (DataPhysics Instruments GmbH, Germany) where the initial θ and its changes with time were recorded. The method is fully explained in our recent publications [22, 42]. In those studies θ was measured on the flat top surface of SPP pellets. However, in this study the method was used for the first time to estimate the pellet hydration properties on a curved cylindrical surface, because the ends of PSPP pellets were irregular and not suitable for measuring θ. The test was performed on 10 PSPP pellets and 5 SPP pellets. SPP pellets for this test were produced at 120°C under 300 MPa. An example of the principle for θ determination on the curved surface is given on Fig. 3a. The baseline and drop profile were manually determined.

The roughness of the pellets' surface was measured by a MarSurf SD 26 2D profiler (Fig. 3b). The average surface roughness (R_a) and mean peak-to-valley height (R_z) were recorded.

Figure 3. a) Example of baseline/drop profile extraction and the contact angle measurement of a sessile drop on a cylindrical surface. b) Roughness measurement.

2.4. Data analysis.

One-way analysis of variance was used to test if there was a difference in pellet quality parameters among different types of materials (PS, M_1, and M_2). Tukey's HSD tests were employed to determine which groups differed (95% confidence interval (CI)). Statistical analyses were performed using Minitab® 16.2.2 software.

3. Results and discussion

3.1. Raw materials.

Some physical-chemical properties of the raw materials used for pelleting are presented in Table 2. The low initial moisture content in the ground wheat straw required addition of water. Water

was sprayed over the straw powder, and it was set in all three powders to 21% moisture content. The obtained moisture content was higher than the one needed for woody biomass. Other studies have also shown that agricultural biomasses require higher moisture content for pelleting compared to wood [4, 7, 43]. Additional water had a lubricating effect and reduced the pressure formed in the die. Trials with lower moisture content were not successful, i.e., they resulted in high friction in the die, a sharp increase of the die temperature, and blockage of the pellet press. Compared to wood, straw has a lower bulk density, similar amount of cellulose, higher hemicellulose content, lower lignin content, and higher content of extractives, which are mainly located at the fiber surface [1, 44, 45]. The straw entered the pellet press at room temperature (cold pelleting), which means that the lignin and extractives had not yet been subjected to phase transition; thus, their lubricating effect was not significantly pronounced. The high shear during the initial passage of material through the die [46], suddenly increased the friction, resulting in a high back pressure. Further on, low-density straw required long retention in the die to be compacted into high-density pellets. The long material retention in the press caused a longer exposure to the shear stresses and friction, which baked the material in the press and eventually led to the blockage of the PSPP. Larsson and Rudolfsson [5] showed that high material temperature and moisture content and low die temperature are necessary to achieve stable pellet production. However, the PSPP used in this study does not allow die temperature regulation, which means the die temperature was not a controlled variable. Adding moisture in the form of steam, which would at the same time increase the material temperature, would possibly allow pelleting at lower moisture content without blocking the pellet press. Molasses addition did not significantly change the material bulk density and HHV. The a_w value of moisturized wheat straw was 0.936, which decreased significantly with molasses addition to 0.908 (1.5% of molasses) and 0.912 (3% of molasses). Despite the statistically significant decrease of a_w, all three values are considered high enough to support microbiological deterioration of the raw materials. This indicates that the materials should be thermally treated shortly after their preparation to avoid the growth of microorganisms and possible self-heating of the powder. Proximate analysis of the raw materials showed that the molasses addition did not change the fuel composition greatly, because of the small amount of added molasses. M_1 and M_2 samples had slightly higher volatile contents than control. Ash content was higher than 6%, thus, according to ISO 17225 – 6, its value needs be stated in case of commercial production [47].

Table 2. Raw material characteristics

3.2. Analysis of pellets.

In Table 3 some pellet quality and production parameters are presented. The table combines results from PSPP and SPP pelleting at 120°C. The pelleting temperature in the PSPP was ≈80°C at a production rate of 5.1 kg/h. The pelleting temperature presented here was the temperature of the outer wall of the die. The temperature in the core of the pellet is unknown. The results from SPP suggest that the temperature inside the material was substantially higher compared to the one measured on the die wall (Section 3.2.1). Therefore, SPP pellets produced at 120°C were chosen for comparison with PSPP pellets in Table 3. 120°C was the lowest production temperature in the SPP that allowed comparison of pellet quality and seems to be closest to the one achieved in the PSPP (Fig. 7). The moisture content and a_w values of the pellets were lower than those of the bulk raw materials because of the intensive evaporation of water during the pelleting process. The a_w values of PSPP pellets ranged from 0.461 to 0.578 and are indicative of the material's microbiological stability. The moisture contents of PSPP pellets (from 9.4 to 7.6%) met the standard for non-wood pellets (ISO 17225-6) [47]. SPP pellets produced at 120°C had even lower values of a_w and moisture content. The different a_w and moisture contents between the PSPP and SPP pellets can be attributed to the difference in production temperatures and the long retention time (≈10 min) of the material in the SPP, during which the material is in contact with the hot die, and thus, water can readily evaporate. The durability of the PSPP pellets was high and did not change significantly as a result of adding molasses. The bulk density of the pellets was slightly increased by adding molasses. However, the changes were not large enough to be statistically different with a CI of 95%. Durability and bulk density of PSPP pellets met the quality requirements of ISO 17225-6 standard, while the amount of fines from M_1 pellets (1.03%) was slightly higher than the value stated in the standard (≤1%) [47].

Table 3. Summary of the production and quality parameters of pellets produced in PSPP and SPP

Determination of θ on the curved cylinder surface and its change with time proved to be more challenging than for SPP pellets on flat edges [22]. First, for accurate θ calculation, the center of

mass of the drop needs to be coincident to the axial middle plane of the pellet (Fig. 3a). If not, the left and right θ are not symmetrical, which produces errors in the measurement. Second, because of water absorption, the pellets start to disintegrate; thus, the baseline and the drop profile (Fig. 3a) become difficult to define. The results presented here are the first study on a curved pellet surface and further work should be done to ensure a representative estimation of the water absorption rate. The results here showed that θ does not change significantly as a result of molasses addition. SPP pellets absorbed water much faster compared to PSPP pellets (Fig. 4), indicating poorer adhesion among particles and/or differences in fiber alignment. There was no clear difference between pellets with and without molasses. The absorption curves were more dispersed for PSPP pellets (Fig. 4a) than for SPP pellets (Fig. 4b). This can be explained by the varying quality of PSPP pellets. Some pellets were strongly compacted and absorbed water very slowly, while others were less dense with visible small ruptures, causing the water drop to be easily absorbed. The roughness measurement showed that there was no large difference among all samples, but a big variation in pellet roughness within one batch was also observed, particularly for PSPP pellets. In Fig. 5a, the roughness profile of a PSPP pellet with a smooth surface (straight line) and indication of a pellet rupture (pick) is shown. If a drop was positioned on the rupture or close to it, water would be absorbed easily, while positioning on the smooth pellet surface required a longer time for water to penetrate into the pellet. On Fig. 5b, the roughness profile of a SPP pellet is presented. The pellet profile looks rougher, but at the same time, more uniform compared to PSPP pellets.

Figure 4. Changes of θ with time: a) PSPP; b) SPP

Figure 5. Roughness profile of a pellet: a) PSPP pellet (vertical axis 50 μm); b) SPP pellet (vertical axis 25 μm).

3.2.1. Density

Compressibility curves of the three tested materials produced in the SPP, at different temperatures, are shown in Fig. 6. The pellet density sharply increased as pressure increased from 0 (bulk density) to 75 MPa. Further compaction caused just a minor increase in density. Temperature had a strong influence on pellet density. Pellets produced at 60^{o}C were less dense

than pellets produced at 120 and 180°C. Lignin present in the wheat straw was probably softened at temperatures higher than 60°C; thus, the physical quality of pellets produced at elevated temperatures (120 and 180°C) was higher. The T_g point depends on the moisture content and the type of raw material. Stelte et al. tested lignin's T_g point for straw and extracted wheat straw with about 8% moisture, and found that the T_g points were 53 and 63°C, respectively [4, 48]. Based on the results presented here, the T_g point of this type of wheat straw is estimated to be in the range from 60 to 120°C.

The effect of molasses as a binding agent was seen when pelleting was performed at temperatures below the T_g point of lignin. Pellets produced at 60°C had lower density, most likely because only Van der Waals forces and fiber interlocking contributed to particle binding. Molasses reinforced pellets by forming solid bridges that kept particles bonded [10], and thus, the densities of M_1 and M_2 pellets were higher than those of PS pellets. The difference in densities at 120 and 180°C was not large.

In general, it seems that molasses as a binder is redundant when pelleting occurs at temperatures higher than the T_g point of lignin. Nevertheless, it would be recommendable to test the effect of molasses addition at higher throughputs, where the pellet quality may be lower, and the differences in temperature between the core and external surface of the pellets higher.

Figure 6. Compressibility plot for pellets produced in the SPP compared to density of PSPP pellets

The density of PSPP pellets is also given in Fig. 6. The densities of PSPP pellets were high, but differences due to different materials were not statistically significant. To visualize the range of pressures and temperatures that occurred in the PSPP, the data of SPP pellet density were used to generate a mathematical model and 2D surface plots. The mathematical relationship between the density of SPP pellets and the examined factors (pressure and temperature) was fitted to a second-order polynomial equation (Eq. 2). R^2 indicates how good the model fits the data. The adjusted value $R^2_{(adj)}$ corrects the R^2 value for the sample size and for the number of terms in the model. The adjusted values should be close to 1 and close to R^2 if the model is good. $R^2_{(pred)}$ indicates the predictive power of a regression model. According to Eriksson et al. [49] $R^2_{(pred)}$ is

considered to be good if it is > 0.5 and if the difference between $R^2_{(pred)}$ and R^2 is < 0.2 - 0.3. The models presented here satisfy those requirements.

The coefficients of the model and their significance are presented in Table 4. *p* values lower than 0.05 indicate a considerable effect of these coefficients on the response (density). The results showed that the linear terms of pressure and temperature and the quadratic temperature term have a significant effect ($p < 0.05$) on pellet density. Quadratic coefficients of pressure were not significant ($p > 0.05$) for the samples with added molasses. Effects of pressure-temperature interaction were not significant. The model was used for generating contour plots (Fig. 7). The dashed areas on the contour plots represent the range of density values of the pellets produced in the PSPP.

Table 4. Regression coefficients and model performance estimators

Figure 7. Response contour plots for pellet density produced in the SPP (a – PS; b – M_1; c – M_2). The dashed areas indicate the pelleting conditions needed to achieve the density from the PSPP

The idea of using the RSM procedure was to determine the range of pressures and temperatures that occurred in the PSPP, by correlating the density of those pellets to the contour plots generated from the SPP data. The dashed areas on Fig. 7 are located on the upper right corner of the plots indicating that high pressures and temperatures were generated in the PSPP. Temperature seems to be a limiting factor for producing the pellets. The results here indicate also that measuring the temperature of the wall of the die does not provide an accurate estimate of the temperature of the material in the die hole. Apparently, higher temperatures were generated inside the die holes than those recorded by the temperature sensor. According to Salas-Bringas et al. [46], a significant material temperature rise occurs not only in the die hole, because of friction, but also in the nip area of the pellet press, contributing to the overall higher temperature of the material compared to the temperature measured on the die wall and the pellet temperature after leaving the die. However, other findings show that the material temperature can be lower than the die temperature when the retention time is short [7]. Considering the low production rate in this experiment (long retention time), it would be reasonable to believe that the first scenario is more likely.

3.2.2. Strength

The bending stress of SPP pellets produced at different pressures and temperatures is presented in Fig. 8. PS pellets produced at 60°C hardly kept their shape and broke before the strength test; thus, it was not possible to determine their strength by the three-point bending test. Although the density was just slightly increased when the compacting pressure was increased from 75 to 300 MPa, the bending stress was still found to increase. Similar observations were found for other compacted materials [50, 51]. The strength of the pellets increased with temperature, pressure, and molasses addition. The results are in agreement with the strength of the PSPP pellets produced with different percentages of molasses (Fig. 8). Despite the same trend regarding molasses addition, the differences in strength values are large. PSPP pellets were much stronger and better bonded than those produced in the SPP. However, it is important to notice that the detached surface at breakage for SPP pellets was smaller than for PSPP pellets (Fig. 9e-f); thus, the higher resistance to breakage can be associated with the geometry and differences in fiber orientation in the pellets. Straw pellets can be considered to have anisotropic mechanical properties because their strength depends on fiber orientation. The load that pellets can withstand depends on the angle between the fiber and the applied force [24]. The conic type of fiber orientation (Fig. 9c) found in PSPP pellets follows a similar angle to the one located at the die entry hole (18.8°), while in SPP pellets the fibers were transversally orientated (Fig. 9d).

The high standard deviation for the bending stress indicates a large variation in pellet physical quality. This can be attributed to the differences in material flow rate through the die. It was possible to observe during the production differences in the material retention time in the die holes. The material flowed faster through some die holes (less dense pellets with small ruptures on the surface), while a higher retention time was observed in others (very dense pellets with smooth surfaces). These differences can be a consequence of different wear in the die holes, damage to the die hole entry, or damage to roller surfaces.

Pelleting by two different methods resulted in pellets with greatly different physical properties, particularly bending strength. This can be attributed to the already mentioned differences in fiber orientation and mechanical differences between the two methods such as: 1) Shear stress and strain on the material that occur in the roller-die gap and nip area before the material enters the die hole, which cannot be simulated in the SPP (compression and flow component in Nielsen

description of pelleting process)[15]; 2) In PSPP, material enters the die hole at elevated temperatures because of pre-heating in the housing of the pellet press and the roller-die gap. During pelleting in the SPP, the material is usually poured into the die at room temperature; 3) The retention of materials during pelleting in the PSPP was not the same in all die holes; hence, this parameter could not be taken into account for planning the experiment in the SPP.

Figure 8. Bending stress of SPP and PSPP pellets

Figure 9. Illustration of differences between the two pelleting methodologies. a) drawing of die holes and rollers in the PSPP; b) drawing of the SPP die hole and compressing rod; c) illustration of conic fiber orientation in a pellet and direction of force applied during the three-point bending test; d) illustration of transversal fiber orientation in a pellet and direction of force applied during the three-point bending test; g) typical example of a broken PSPP pellet; h) typical example of a broken SPP pellet.

3.2.3. Axial expansion of SPP pellets

Axial pellet expansion, after the applied pressure is removed, is an indicator of the material's elastic recovery (spring back effect) [52]. As shown in Fig. 10, pellets produced at low temperature showed the strongest tendency to expand, indicating poorer binding among the particles. This can be explained by the elastic and flow behavior of a compacted straw, which changes with temperature. Large axial expansion at 60^{o}C shows that the materials were mostly elastically compressed. The materials did not fully achieve plastic deformation, which usually follows elastic compression at increasing pressures [53]. At 120°C and 180°C, the plastic deformation should become stronger. With increasing pelleting temperature, the axial expansion significantly decreased. Particles were better bonded when molasses was added. The binding effect of molasses was also reduced at higher temperatures. Changes in axial expansion with increasing compacting pressure were not significant ($p>0.05$).

Figure 10. Axial expansion of SPP pellets after 48 h

3.3. Energy consumption and p_{max}

The combined results of pressure required for the discharge of single pellets (p_{max}) and energy consumption during PSPP pelleting are presented in Fig. 11. Molasses reduced the energy consumption of the production process. However, high standard deviations indicate high fluctuations in the energy consumption, so the process was not stabilized by adding molasses. p_{max} directly shows the pressure level required to initiate pellet motion in the die, and indirectly informs about the energy uptake of the pellet press. Molasses addition caused a substantial drop of the discharging pressure. Therefore, it would be reasonable to claim that molasses has a lubricating effect, i.e., it reduces friction in the die holes, which is also in agreement with the results of PSPP pelleting, where energy consumption was lowered by molasses addition. These results indicate that p_{max} can be a good indicator to study the effects of pelleting materials on energy consumption.

Figure 11. Discharging pressure (p_{max}) for the pellets produced in the SPP and energy consumption during the PSPP pelleting

The production temperature had a strong effect on p_{max}. p_{max} was higher at low pelleting temperature. The pressure applied to the "elastic" material produces a radial stress toward the die wall. This resulted in a higher die-wall friction that is observed from high p_{max} at 60 ^{o}C. At elevated pelleting temperatures the materials showed greater plastic compression. Plastic compression would decrease the radial stresses at the die walls as the forces would dissipate with the plastic deformation. This would result in a lower die friction (p_{max} lower). p_{max} depends on the rheological and tribological properties of the biomass, which are affected by temperature, moisture content and particle size [20]. The extractives of a straw (free fatty acids, sterols, waxes, steryl esters, and triglycerides) are concentrated on the fiber surface [54]. Phase transitions of these components cause changes in friction, reflected in the lower p_{max} at higher temperatures [55].

4. Conclusions

The benefits of molasses addition can be summarized as follows: reduced energy consumption, higher pellet strength at low pelleting temperatures, higher pellet density and bulk density, and lower moisture content and a_w value. Molasses as a binder is effective when pelleting is

performed at temperatures below the glass transition point of straw lignin. The pelleting temperature appears to be the major factor that affects pellet quality and energy consumption for the pelleting process. Pellets produced by different methods exhibit greatly different physical quality. PSPP pellets were stronger than SPP pellets. Nevertheless, SPP showed to be useful for predicting the trend in changes of pellet quality of different materials. The bending strength test appears to be an appropriate test for the comparison of strengths of SPP and PSPP pellets, since it replicates the stresses causing failure in pellets during storage. From a processing point of view, p_{max} from the SPP can show differences in energy requirements for the PSPP process.

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Highlights

- Straw pellets with added molasses were produced by two pelleting methods.
- Molasses as a binder is redundant for pelleting at temperatures higher than the Tg of lignin.
- Single pellet press (SPP) and pilot scale pellet press (PSPP) were compared.
- Bending test is suitable for the comparison of strengths of SPP and PSPP pellets.
- Surface hydrophobicity was assessed by measuring contact angle of a water drop.

Table 1

Table 1. Experimental design (CCD) and response data for RSM modeling

Coded variables		Natural variables		Pellet density (kg/m^3)		
X_1	X_2	Temperature (X_1) °C	Pressure (X_2) MPa	PS	M_1	M_2
0	0	120	225	1249.6	1249.5	1241.8
0	0	120	225	1252.6	1247.5	1245.6
-1	1	60	300	851.1	970.1	950.6
-1	-1	60	150	762.3	885.3	913.7
0	0	120	225	1240.6	1237.4	1252.3
0	-1.414	120	118	1132.4	1169.5	1164.4
0	0	120	225	1240.4	1243.7	1248.6
-1.414	0	35.16	225	713.7	880.5	826.2
0	1.414	120	331	1267.7	1285.1	1276.8
1	1	180	300	1297.2	1305.6	1294.7
1.414	0	204.84	225	1290.6	1285.5	1301.8
1	-1	180	150	1226.9	1241.8	1231.0
0	0	120	225	1236.4	1255.6	1239.0

Table 2

Table 2. Raw material characteristics

Property	PS	M_1	M_2
Moisture content, %	21	21	21
Molasses content, %	0	1.5	3
Bulk density, kg/m^3	81.92 ± 1.58^a	79.24 ± 1.55^a	80.73 ± 3^a
a_w value	0.936 ± 0.002^a	0.908 ± 0.002^b	0.912 ± 0.003^b
Volatiles, $\%_{d.b.}$	78.59	78.71	78.86
Fixed carbon, $\%_{d.b.}$	14.25	14.34	14.03
Ash, $\%_{d.b.}$	7.16	6.95	7.11
HHV, MJ/kg	18.55 ± 0.04^a	18.73 ± 0.2^a	18.66 ± 0.12^a

d.b. – on dry basis; ^{a,b} – Means ± SD that do not share a letter are significantly different ($p<0.05$).

Table 3

Table 3. Summary of the production and quality parameters of pellets produced in PSPP and SPP

	PSPP			SPP		
	PS	M_1	M_2	PS	M_1	M_2
Die temperature, °C	81.4	79.4	79.3	120	120	120
Moisture of pellets, %	9.4	7.6	8.3	2.99	2.95	3.01
a_w value	0.578±0.005[a]	0.461±0.002[b]	0.565±0.002[a]	0.253±0.005[c]	0.191±0.01[d]	0.181±0.003[d]
Average pellet diameter (mm)	5.97±0.03[a]	5.96±0.04[a]	5.96±0.03[a]	6.05±0.01[b]	6.05±0.00[b]	6.05±0.03[b]
Average pellet length (mm)*	14.16±0.54[a]	14.16±0.52[a]	14.23±0.57[a]	-	-	-
Durability, %	99.51±0.05[a]	99.56±0.07[a]	99.32±0.05[a]	n.a.	n.a.	n.a.
Fines, %	0.24	1.03	0.318	n.a.	n.a.	n.a.
Bulk density, kg/m^3	627.9±2.7[a]	640.93±11.2[a]	644.5±7.9[a]	n.a.	n.a.	n.a.
Initial θ,°	82.76±2.63[a]	84.21±4.55[a]	80.95±3.48[a]	99.07±1.54[b]	95.27±2.07[b]	92.89±4.09[b]
R_a, µm	1.83±1.1 [ab]	2.37±1.32 [ab]	1.36±0.97 [b]	2.56±0.41[a]	1.72±0.42[ab]	2.14±0.58[ab]
R_z, µm	11.76±7.09 [ab]	17.4±7.86 [ab]	10.18±7.67 [b]	19.18±2.84[a]	14.93±3.3[ab]	19.12±4.3[a]

[a,b] – Means ± SD in the same row that do not share a letter are significantly different ($p<0.05$). * the length of SPP pellets is dependant on amount of material poured into the pellet press; n.a. – not available informattion due to small sample size.

Table 4. Regression coefficients and model performance estimators

Terms	**PS**	**M1**	**M2**
R^2	0.9806	0.9722	0.9852
$R^2_{(adj)}$	0.9667	0.9523	0.9746
$R^2_{(pred)}$	0.8638	0.8057	0.8968
Coefficients			
β_0	1243.93 (0.00)	1246.76 (0.00)	1245.49 (0.00)
β_1	215.80 (0.00)	158.10 (0.00)	166.75 (0.00)
β_2	43.79 (0.02)	39.01(0.01)	32.43 (0.01)
β_{11}	-137.58 (0.00)	-95.48 (0.00)	-101.95(0.00)
β_{22}	-38.61 (0.03)	-23.34 (0.11)^{ns}	-23.66 (0.05)$^{n.s.}$
β_{12}	-4.62 (0.82)$^{n.s.}$	-5.25 (0.76)^{ns}	6.69 (0.62)$^{n.s.}$

[a] p values are given in the brackets. n.s. is not significant term in the model

Figure 1
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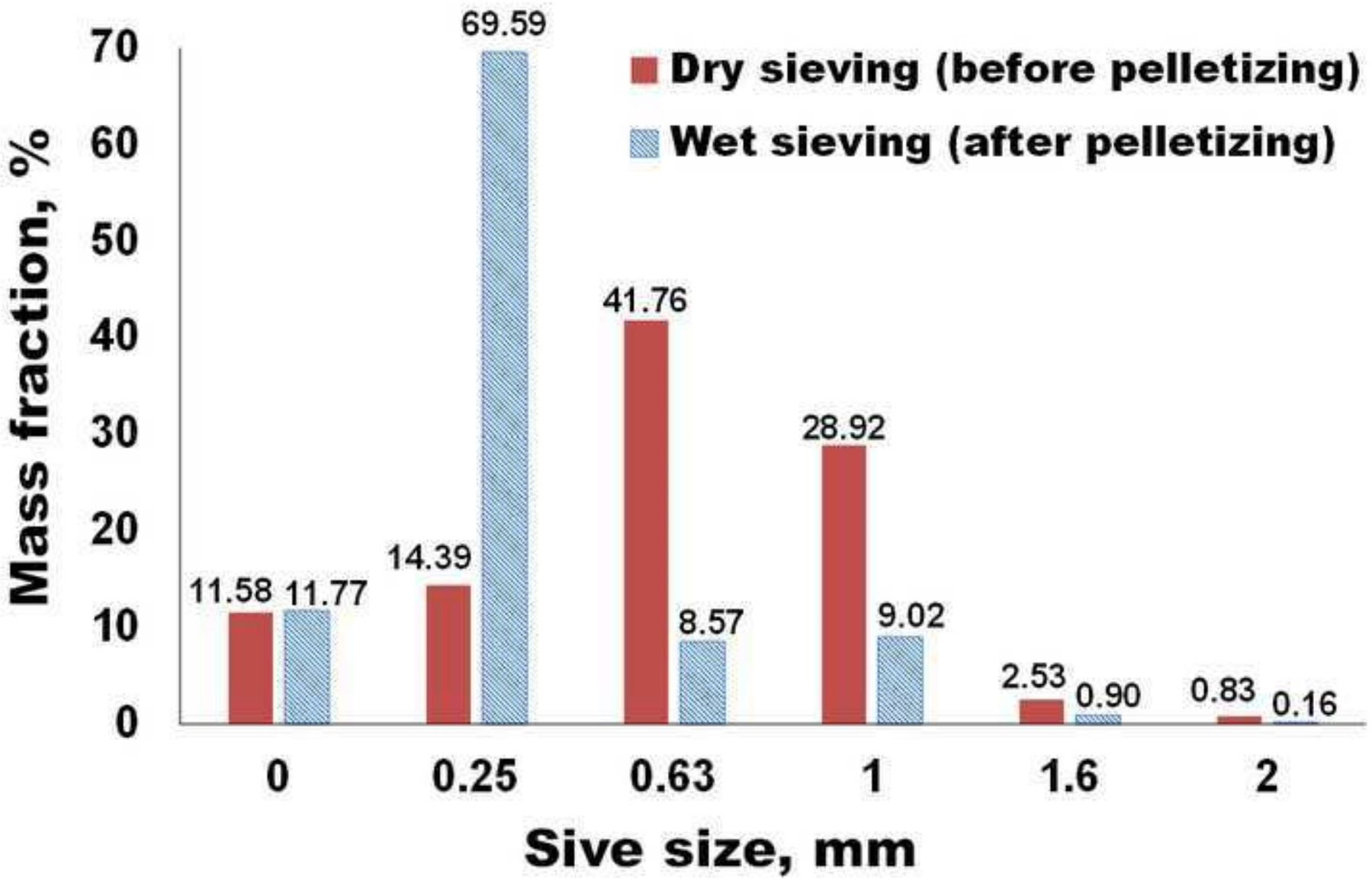

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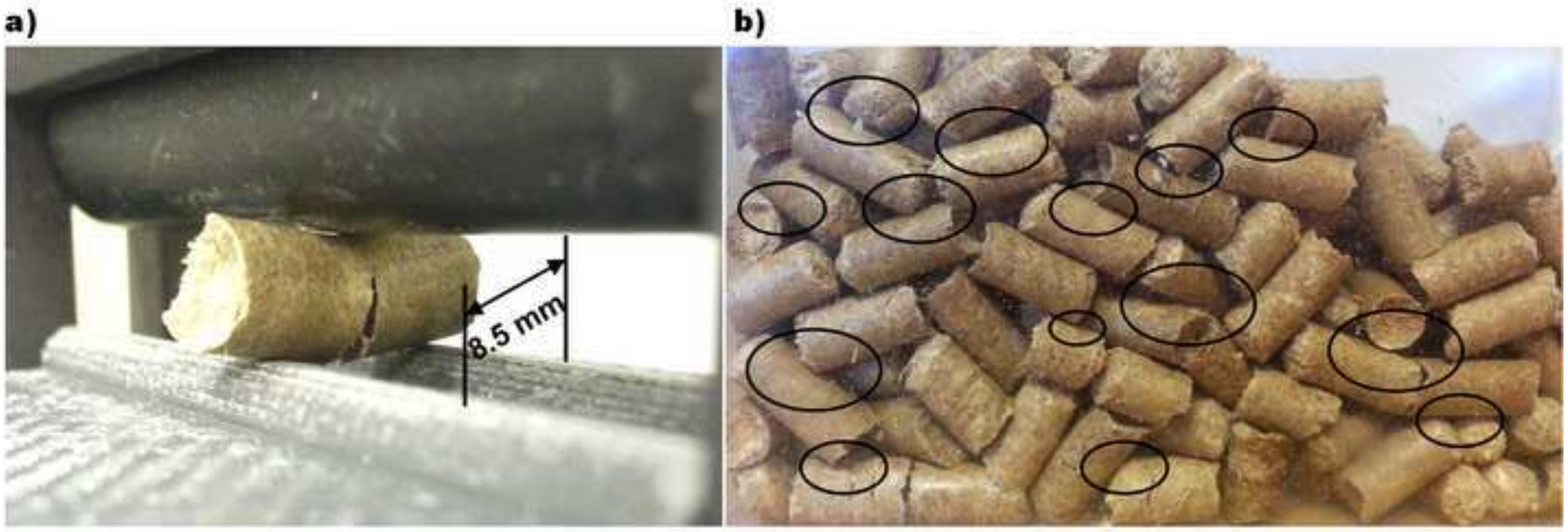

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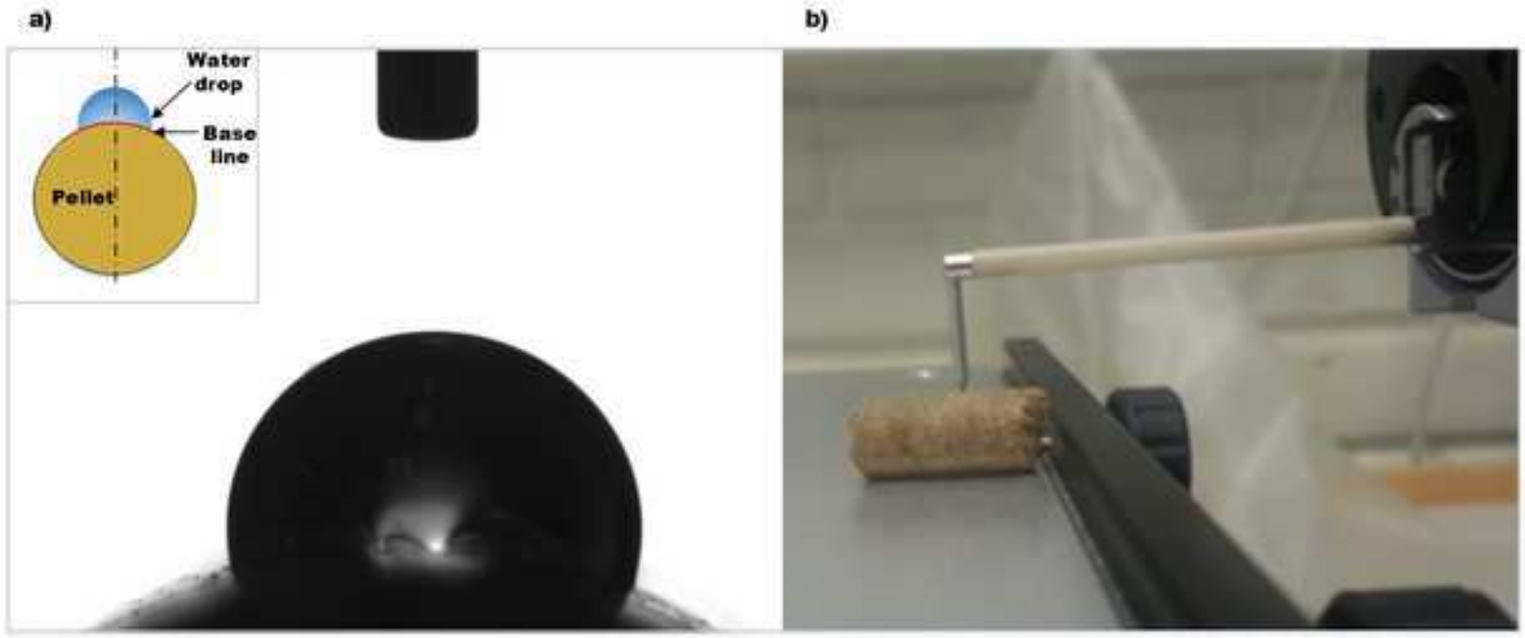

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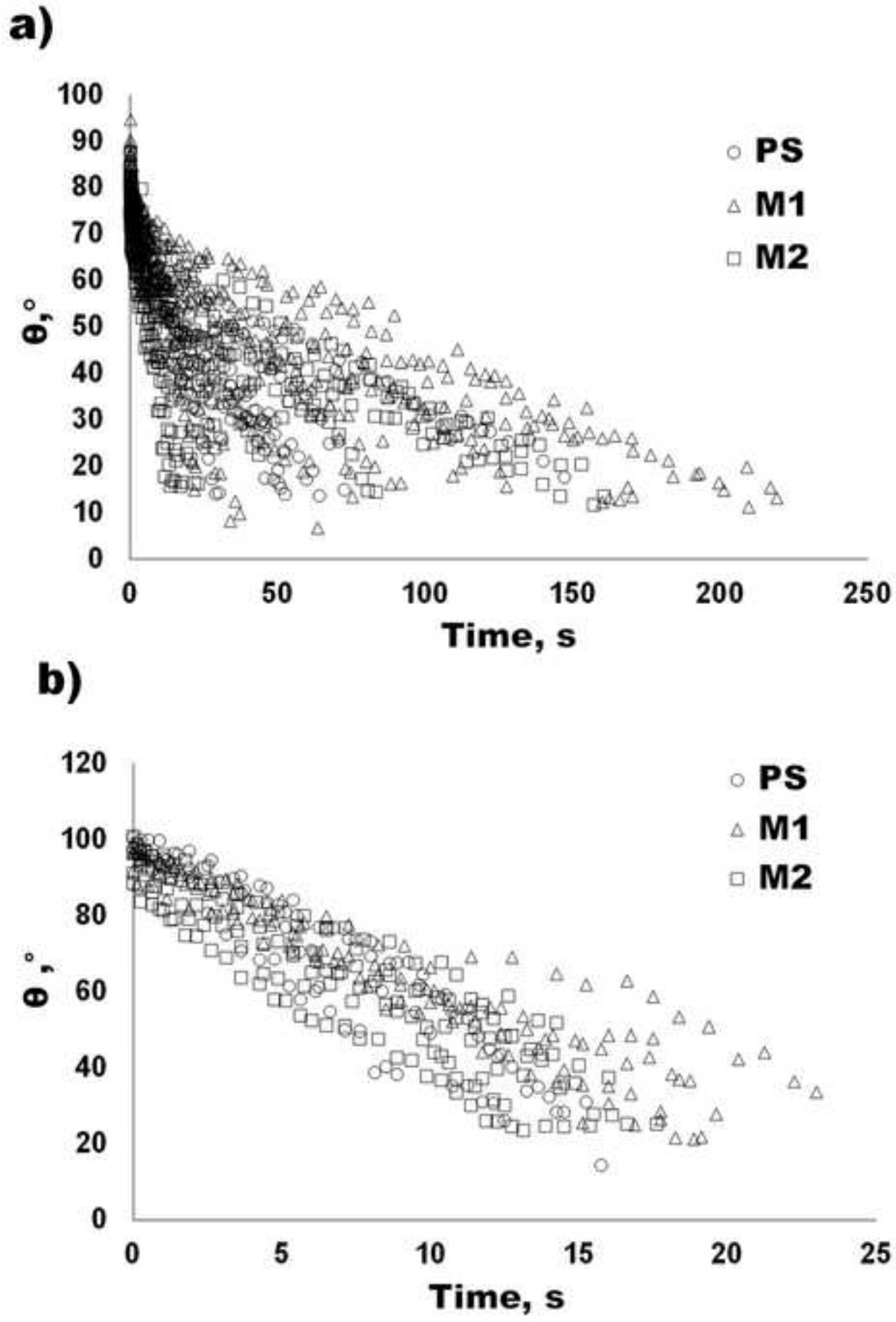

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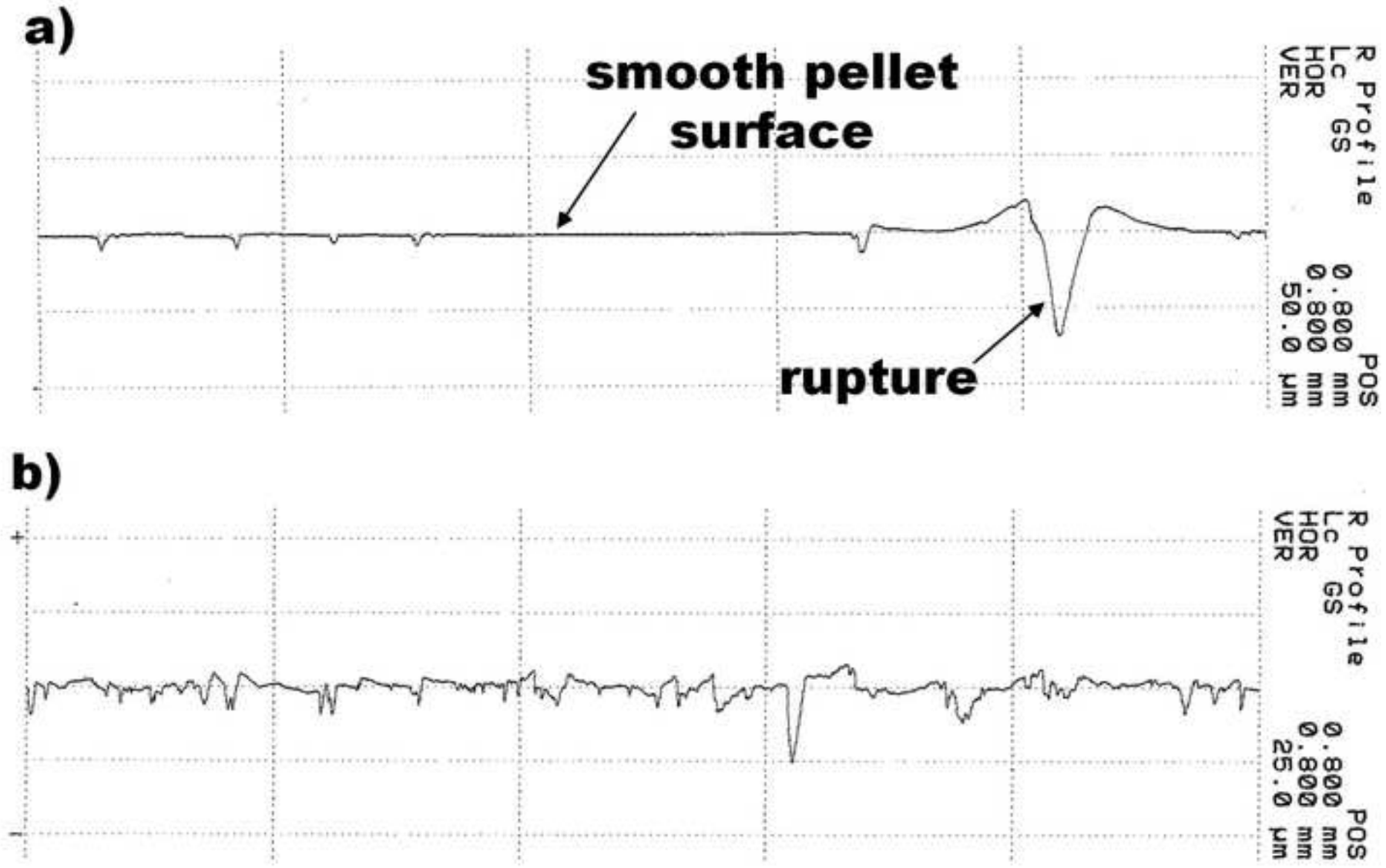

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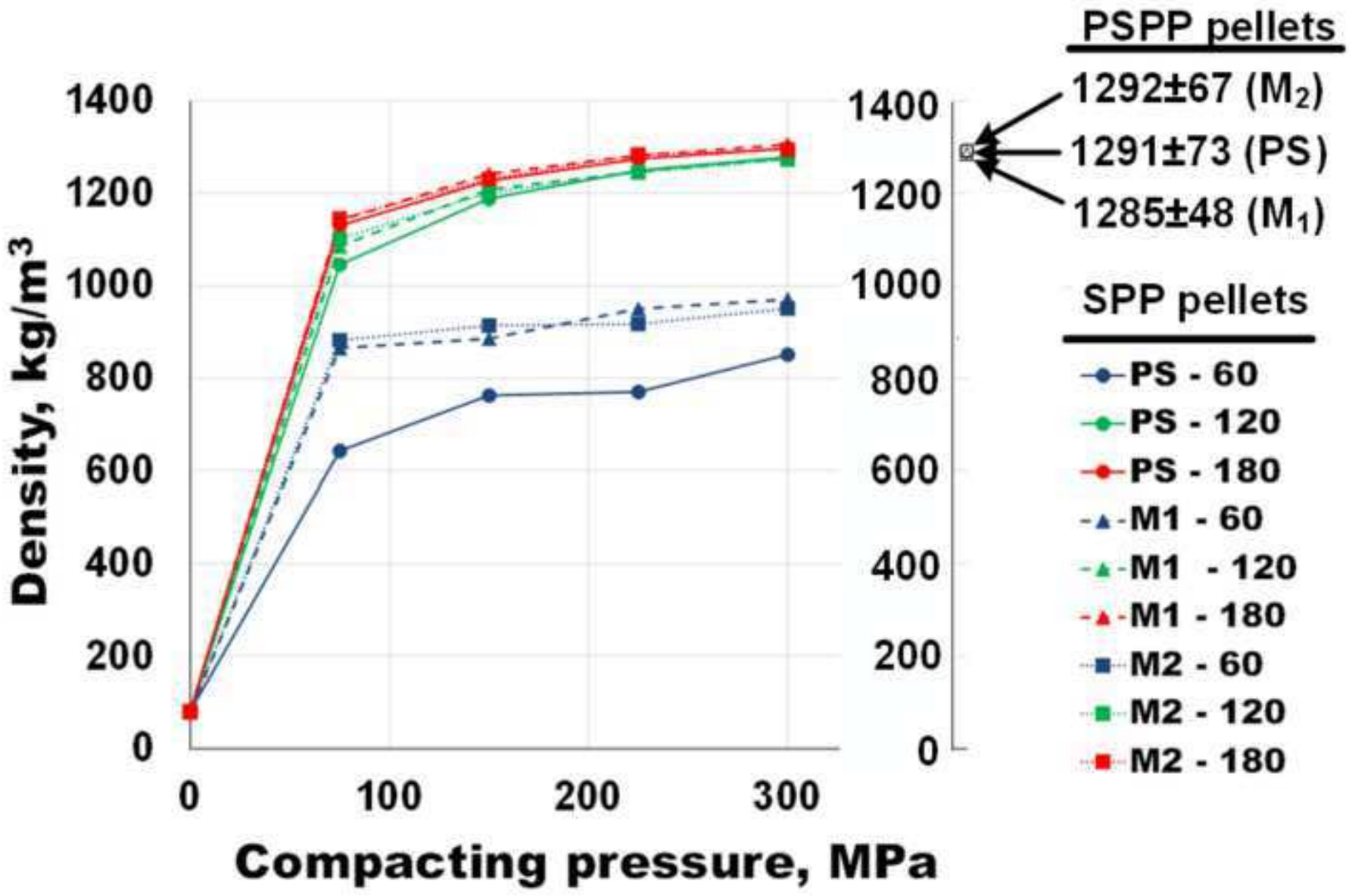

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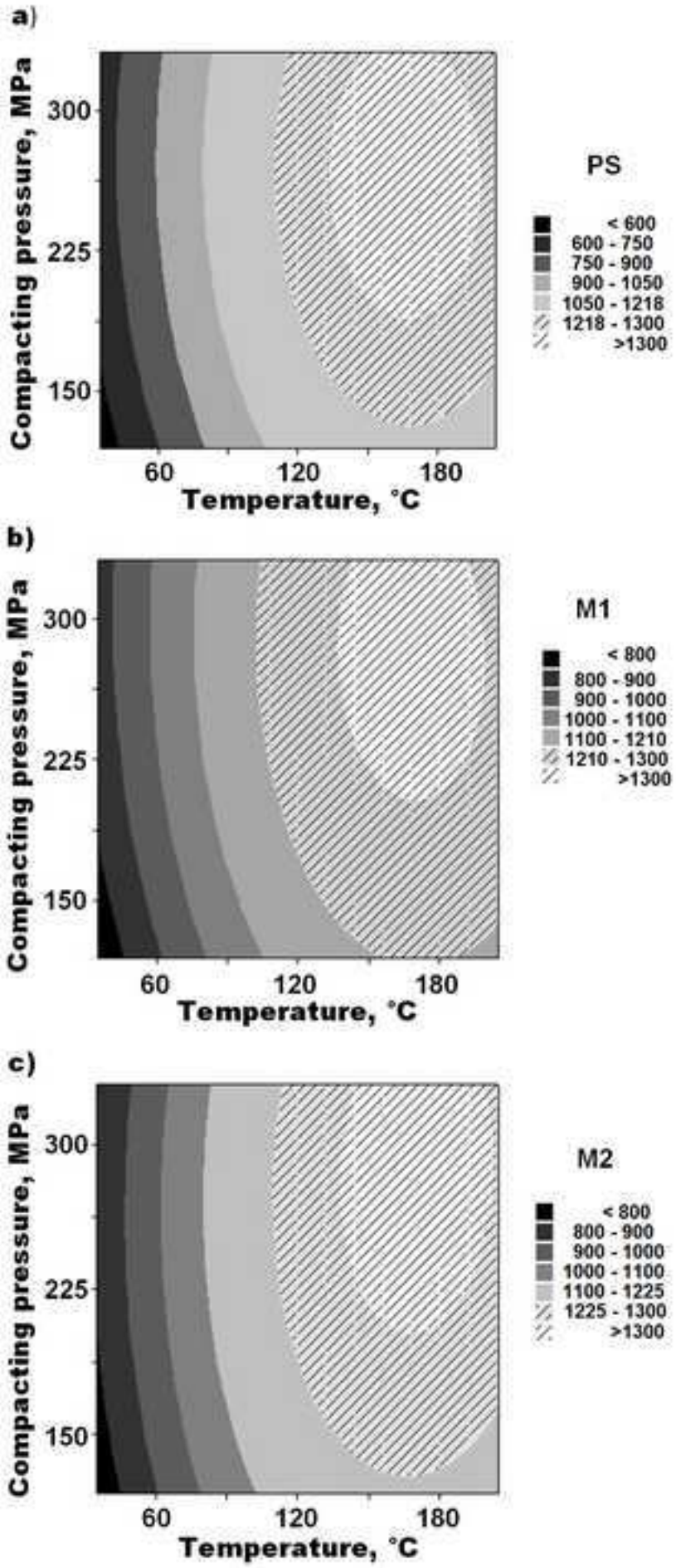

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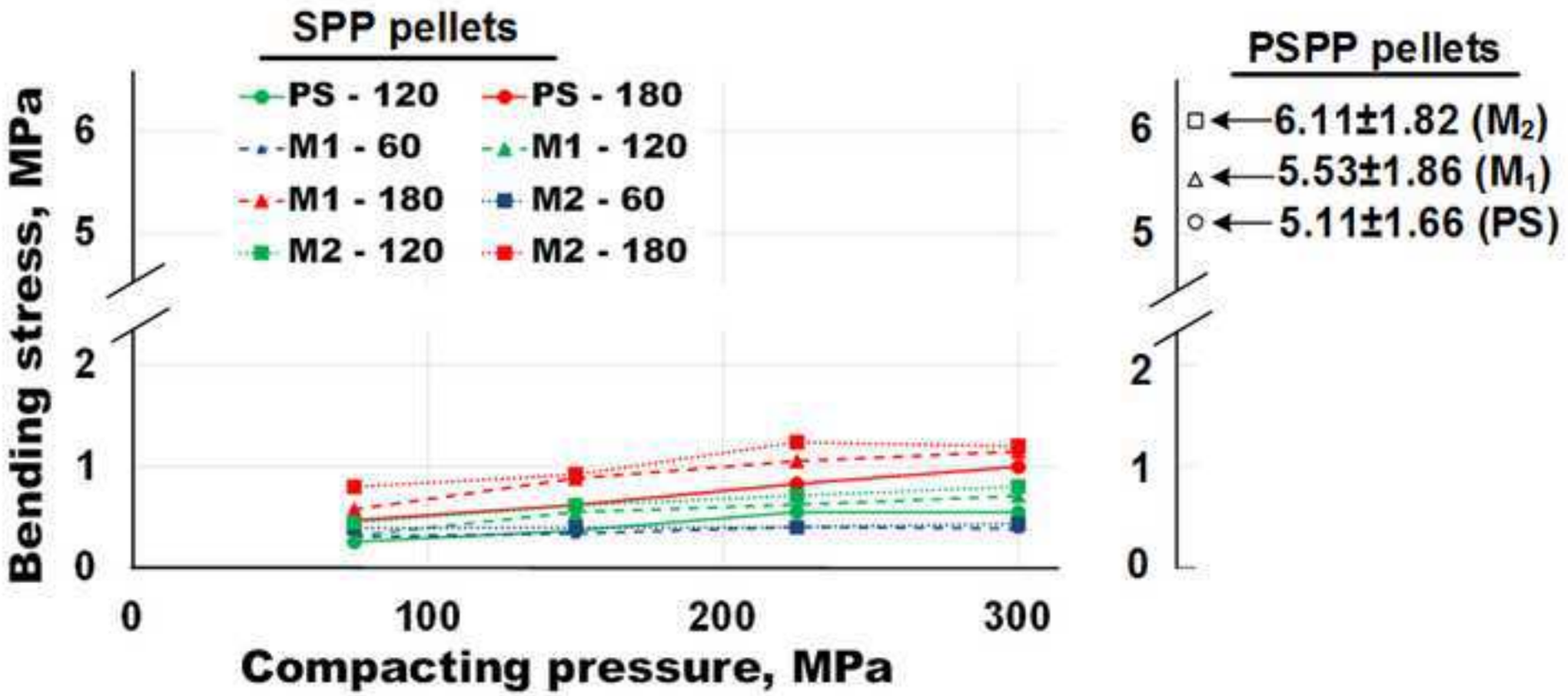

Figure 9
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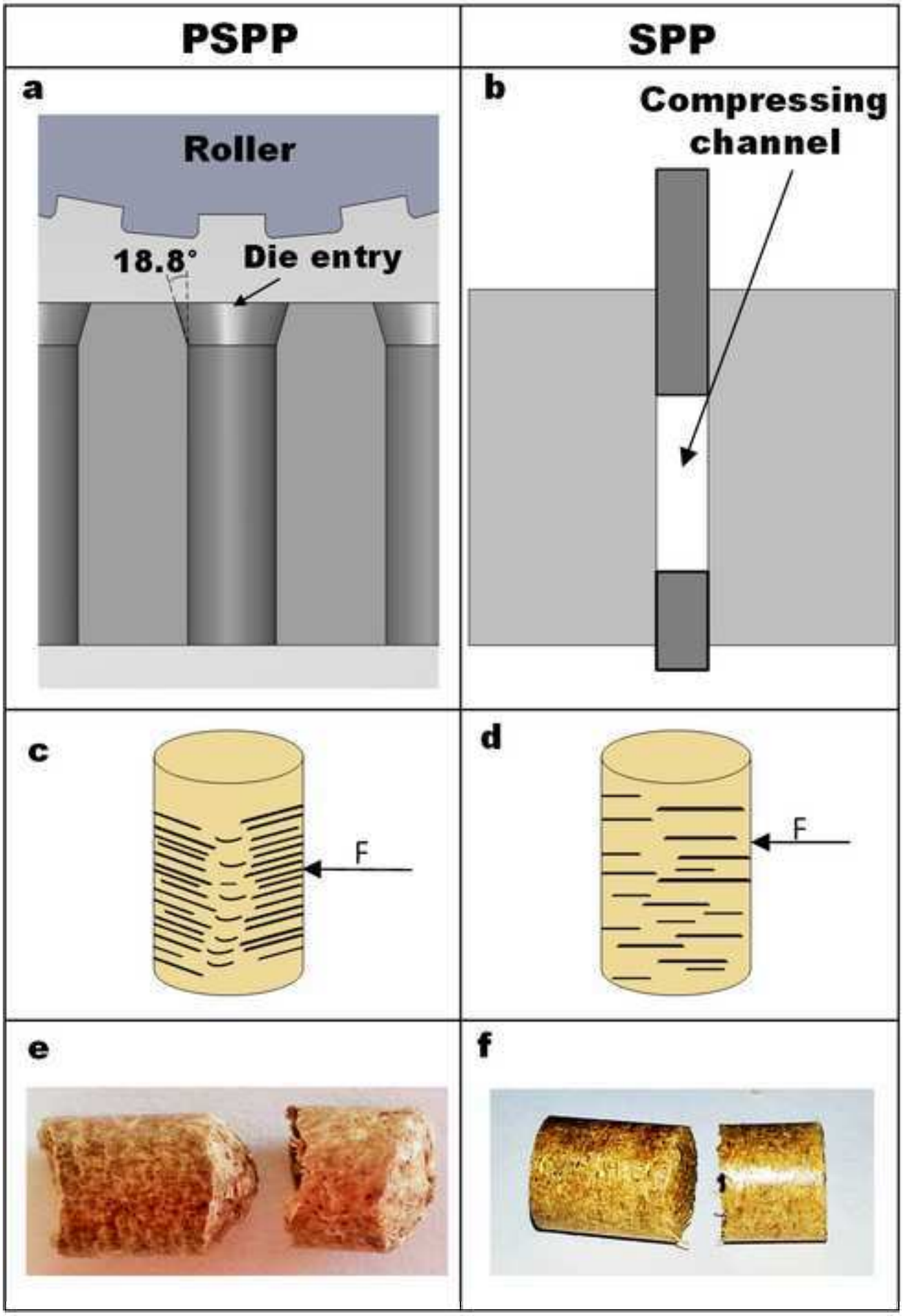

Figure 10
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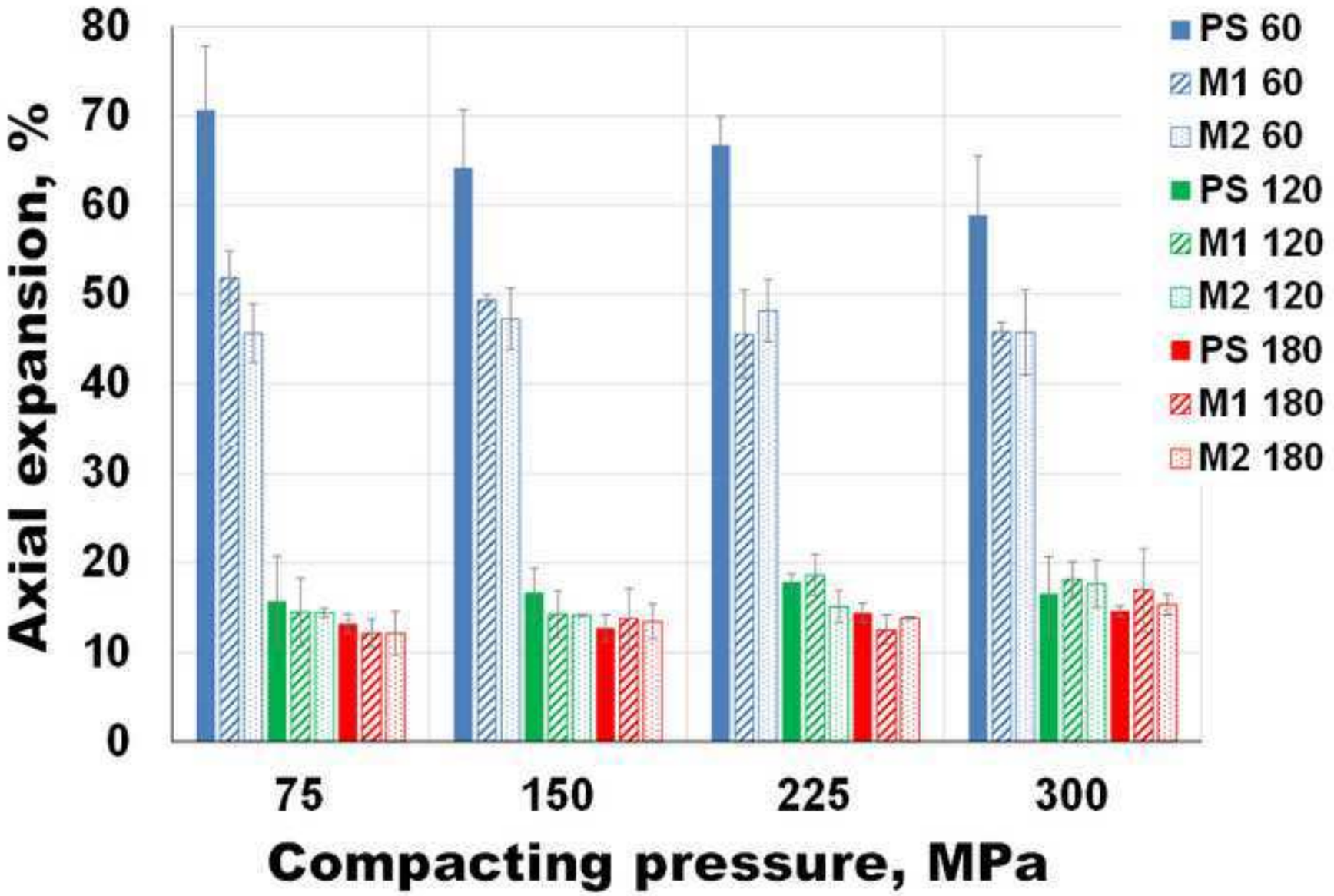

Figure 11
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