Research of interaction between technological and material parameters during densification of sunflower hulls

Peter Križan*, Miloš Matúš, Juraj Beniak and Ľubomír Šooš
Institute of Manufacturing Systems, Environmental Technology and Quality Management, Faculty of Mechanical Engineering, STU in Bratislava, Nám. Slobody 17, 812 31 Bratislava, Slovakia

* Corresponding Author: peter.krizan@stuba.sk

Abstract. During the biomass densification can be recognized various technological variables and also material parameters which significantly influences the final solid biofuels (pellets) quality. In this paper, we will present the research findings concerning relationships between technological and material variables during densification of sunflower hulls. Sunflower hulls as an unused source is a typical product of agricultural industry in Slovakia and belongs to the group of herbaceous biomass. The main goal of presented experimental research is to determine the impact of compression pressure, compression temperature and material particle size distribution on final biofuels quality. Experimental research described in this paper was realized by single-axis densification, which was represented by experimental pressing stand. The impact of mentioned investigated variables on the final briquettes density and briquettes dilatation was determined. Mutual interactions of these variables on final briquettes quality are showing the importance of mentioned variables during the densification process. Impact of raw material particle size distribution on final biofuels quality was also proven by experimental research on semi-production pelleting plant.

1. Introduction
The pelletization of biomass is a mass and energy densification process for materials that possess low bulk densities. The quality of biofuels (final product of densification) is determined by the end-user’s requirements on the heating system and the handling properties [1, 2]. In general, the biofuels quality depends on the properties of the raw material (feedstock) – in terms of biomass type, moisture content and particle size; and quality management of the manufacturing process – in terms of operating conditions, technological variables, pelletizer type and binding agent [3, 4]. Low-quality pellets can cause operational problems in combustion systems, including undesired effects in the equipment such as slugging, fouling or corrosion, and may originate substantial amounts of gaseous and particulate matter (PM) emissions [1,2,4]. There are known many projects and applications where we can see energy recovery of different types of raw materials. With the development of biomass energy recovery technologies and with the gradual decrease of the quantities of usable wood biomass, increases the requirements for the processing of other types of biomass, e.g. agricultural - herbaceous biomass[5]. Sunflower hulls as an unused source is a typical product of agricultural industry in Slovakia belongs to the group of herbaceous biomass. Sunflower as herbaceous plant which is widely and usually cultivated on Slovakia has a high potential of usage in energy sector. In case of sunflower hulls densification (in general in case of herbaceous biomass) there are some important differences with comparing the wooden biomass. There are some problems come from the material (sunflower hulls)
composition and also from material structure. These parameters influencing also lonely treatment, densification and also the combustion of final biofuels from sunflower hulls [3].

The main goal of presented experimental research is to determine the impact of technological variables and impact of mechanical treatment of raw feedstock (sunflower hulls) on final biofuels quality. Each type of raw feedstock has its own specific properties, chemical composition and behavior during mechanical treatment and densification. Each small change in the properties of the raw feedstock can affect the final biofuels quality [5]. The variety of raw feedstock properties requires different approach and technological conditions [6]. Due to the design of an economically efficient method of biomass treatment process, it is necessary to know and determine the optimal parameters for densification with respect to the type of raw feedstock [5, 6]. On the other hand, the applicable technical standards for the quality of biofuels are valid and must be complied with. According to the valid EN standards [7,8] the quality of biofuels is evaluated from mechanical indicators of quality (bulk density, mechanical durability, particle density, etc.) points of view and from thermal-chemical indicators of quality (moisture content, heating value, etc.) points of view. According to the main goal it was important to determine not only the impact of technological variables and raw material treatment variables on final biofuels quality but also on biofuels dilatation. The experimental findings presented here are significant from biofuels production point of view and also from densification machines constructions point of view [5].

The solution of this project comes directly from the requirement of practice, where the company demands to increase the quality of the produced pellets.

2. Materials and Methods

2.1. Monitored dependent variables
On the basis of our knowledge and experiences from executed measurements we can identify the impact of technological variables (compression pressure, compression temperature) and the impact of raw feedstock parameters (type of raw feedstock, moisture content and particle size) on biofuels quality [5, 6]. During this experimental study only the impact of compression pressure, compression temperature and particle size distribution on final biofuels quality was determined. All mentioned variables influences the conditions in pressing chamber during densification and thus influences the final biofuels quality and also stabilization time. Biofuels stabilization time is the time interval during which dilatation occurs [9]; it is also the time interval during which the biofuel stabilizes. Biofuel stabilization time takes approximately 24 hours according to the type of the raw feedstock and densification technology used but it can also take longer. Standard DIN 52182 [10] describes the conditions and process for detecting biofuels density after stabilization time. Biofuel dilatation is an effect during which the biofuel dimensions and weight are changing (diameter, length and weight). These changes come from the raw feedstock properties and also external variables of the densification technology. Dilatation directly influences the biofuel quality [9].

With a suitable combination of the mentioned variables and parameters we can significantly impact the densification process, mainly from the final biofuels quality point of view [5]. We will be able to find the optimal conditions for each specific input setting (type of raw feedstock and its parameters).

2.2. Experimental methodology and process
According to the main goal it was important to divide the experimental research into the two phases:

1.) experimental research with using of experimental pressing stand (see figure 1)
2.) semi-operation tests with using of production pelleting plant (see figure 2)

Experiment research was carried out in laboratory conditions at temperature 23°C, with and also without additional pressing temperature. The main goal of first phase was to determine the impact of compression pressure and compression temperature on final biofuels quality and on biofuels dilatation. These results were obtained without influence of additional pressing temperature [11, 12] because additional pressing temperature influences acting radial pressures and friction conditions in pressing
chamber [12, 13]. Experiment was realized on available experimental pressing stand (see figure 1) which uses a vertical single axis pressing. Pressing stand is inserted to the hydraulic press which is a source for pressing pressure in various levels. According to the designed experimental plan within the range from 63 MPa to 191 MPa of compression pressure was experimental research executed. The raw material was fed into the pressing chamber and compression with a hydraulic piston was commenced. Due to an aim of this research pressing chamber with 140 mm length and ø 20 mm diameter was used in pressing stand during experiment. Also the influence of compression temperature was determined, when the 100°C additional temperature was used.

To maintain the experimental plan and ensure correct results, 7 briquettes with ø 20 mm diameter had to be pressed for each setting. This is the minimum number of values for processing using known statistical and mathematical methods [5]. Holding time in pressing chamber was chosen according to our previous experiences, it’s mean the compression pressure was applied on each briquette for 7 seconds. The quality of the briquettes is defined by a number of quality indicators, but in this case (briquettes with a diameter greater than ø 8 mm) we used as the evaluation parameter the briquettes density. The briquettes density was calculated on the basis of the briquettes weight and volume ratio. For each briquette, its weight, length and diameter were measured. The length and diameter of the briquettes were determined according to EN ISO 17829 [14] and the volume of the briquettes was then calculated. For each setting, the average value of the density obtained was calculated. After the briquettes has been stabilized, i.e. after the expansion of the briquettes has stabilized, the briquettes have been reweighed and measured. In this way, it is generally possible to define the dilatation of the briquettes and to determine the influence of the said technological parameters on the briquettes dilatation [9].

The main goal of second phase was to determine the impact of particle size distribution on final biofuels quality during semi-operational tests. For the pellets production a vertical pelletizing machine KAHL 33-390 with flat round matrix was used (see figure 2). During the pelletizing process, a matrix with ø 6 mm holes was used. The constant operating parameters for the pelletizing press were the pressing pressure (12 MPa), the pressing temperature (100 °C) and the circumferential speed of rolls (2.2 m.s⁻¹). The interaction between the raw feedstock moisture content, the compression pressure, and the compression temperature during the pelletizing of the biomass is of major importance [12, 13]. If the moisture content of the pressed material is very low, or very high for that matter, the particles become inconsistently arranged, and the resulting pellets becomes unstable [11,15]. Past research showed that moisture has an effect on the plasticization of the lignin. In the present study the initial moisture content of sunflower hulls was low (9.24%), so that additional water was added to the
biomass to increase the raw feedstock moisture content before pelleting. Adequate compression temperature ensures that the lignin plasticizes in the pressed material, and the optimal compression pressure provides densification of raw material [12, 13]. A certain moisture level causes a higher temperature in the densification process and thus a higher quality of pellets [12].

Semi-operational tests for pelleting of sunflower hulls of both prepared particle size distribution (sample 1 and sample 2) were carried out without technical and technological complications. The pelleting process was pretty continuous. During constant feeding of raw feedstock the water flow was increased respectively reduced until the pellets with the highest mechanical quality were reached. The diameter of the pellets produced was ø 6 mm. Subsequently, pellets were subjected to laboratory testing and the determination of mechanical quality indicators and some chemical-thermal quality indicators according to the technical EN standards [7, 8].

2.3. Raw sunflower hulls properties
In this study the sunflower hulls were used. Sunflower hulls as an unused source is a typical product of agricultural industry in Slovakia belongs to the group of herbaceous biomass. Sunflower as herbaceous plant which is widely and usually cultivated on Slovakia has a high potential of usage in energy sector. Initial bulk density of used sunflower hulls according to EN ISO17828 [16] was measured. Bulk density of sunflower hulls is 0.1614 kg.dm$^{-3}$.

Initially, the particle size distribution was analysed with the aid of a Retsch Vibrating Sieve Equipment AS 200. Other different particle size distribution were processed by a hammer mill Stoza ŠV 5 with ø 4 mm sieve. For pellets production, untreated sunflower hulls (sample 1) and sunflower hulls treated by single level shredding (sample 2) were used. Figures 3 and 4 show the prepared samples before and after shredding. Figure 5 shows the raw material particle size distribution of both samples used.

Moisture content of sunflower hulls before disintegration and pelleting was measured with the aid of a Kern MRS 120-3 balance. This measurement consisted in heating the raw feedstock (gravimetric method of moisture content measuring) [17] at 105 ± 2°C until a constant weight was achieved. Due to the complex process of moisture content change, only one moisture content level was used in this study. Sunflower hulls with 9.24% of moisture content was used for disintegration, for pelleting and for whole experimental research.

![Figure 3. Untreated sunflower hulls - sample 1.](image1)

![Figure 4. Treated sunflower hulls - sample 2.](image2)
3. Results and Discussion

All the results in this paper, showing the interrelation between the different variables, were depicted graphically, as seen in the figures below. The results of this experimental research are applicable to the choice of dimensions of densification machines, and gave a comprehensive overview of the behavior of different variables during the sunflower hulls densification process.

3.1. Effect of compression pressure and temperature on briquettes quality and dilatation

During the following experimental research only the untreated sunflower hulls (sample 1) were used. Briquettes with ø 20 mm diameter for these purposes on the experimental pressing stand were produced and can be seen in figures 6 and 8. The interdependence of briquettes density from compression pressure can be seen in figures 7 and 9. These figures describe results related to production also with and without additional compression temperature. In general, as the compression pressure increases so too does the briquette density.

These interdependences also highlights the importance of briquette stabilization time. This effect can significantly distort the results of the experiment. The dilatation effect is produced when the compression pressure is removed, and, as a result of the relationship of the interactions between the compression temperature, moisture content, and input particle size [9,18]. Two of these interdependencies can be seen in figure 7 and also in figure 9. One is valid for densities evaluated...
immediately after releasing the briquettes from the pressing chamber, \textit{i.e.}, before stabilization, and the second for densities evaluated after the stabilization time. During the pressing without any additional temperature (see figure 7), the briquette density changed and dilatation negatively influenced the briquette density. Average density difference of 4.82 % was determined.

![Figure 8. Briquettes produced at T = 100\textdegree C.](image)

![Figure 9. Briquettes density, related to compression pressure at T = 100\textdegree C.](image)

Similar measurement was done also with additional compression temperature of 100 \textdegree C. The briquette density changed but dilatation positively influenced the briquette density. Average density difference of 3.39 % was determined. During densification, lignin is releasing from the biomass which acts as a natural glue. If the briquette isn’t created in optimally adjusted conditions, dilatation of the briquette can also significantly influence the absorption of moisture from surroundings. In practice, the stabilization time can be decreased by cooling. In figure 10 the graphical dependence of differing density before and after stabilization on the compression pressure and compression temperature can be observed. This figure clearly proves the impact of compression pressure and temperature on briquette dilatation and means that with increasing of compression pressure and also temperature, briquettes dilatation is decreasing.

![Figure 10. Graphical density difference dependent on compression pressure at both levels of compression temperatures.](image)

The positive impact of compression temperature on the final briquette dilatation was proven. In general (in case of wooden) the compression temperature has a positive impact on the lignin component of the compressed feedstock [18, 19]. Lignin, cellulose and hemicelluloses content are
very important during densification and influences creation of binding mechanisms between material fibers and particles. The compression temperature influences lignin plasticization [19, 20]. With increased compression temperature during densification, the lignin becomes more and more liquid, plastic. Temperature significantly affects the binding mechanism forming between the solid particles of the compressed material [18, 20]. A more effective binding of the cellular structure of the compressed feedstock and the required plasticization of lignin is possible only under induced compression temperatures [18, 19]. Binding of raw feedstock particles in sunflower hulls briquettes can be seen in figure 11.

![Figure 11. Binding of raw feedstock particles in sunflower hulls briquettes.](image)

The results presented in figure 12 were obtained as a result of densification at different compression temperature levels (23 and 100 °C) and the displayed interdependencies are valid for case after briquettes stabilization. This figure shows the relationship between briquette densities the compression pressure at different levels of compression temperatures during densification of sunflower hulls.

![Figure 12. Binding of raw feedstock particles in sunflower hulls briquettes.](image)

Results displayed in figure 12 are shown that in case of sunflower hulls densification were higher densities achieved during densification without additional compression temperature. This fact can be attributed to the nature and chemical composition of sunflower hulls. With low contents of lignin and hemicelluloses in the raw feedstock, the additional temperature during densification has no significant influence on the final briquettes density [5,15,19]. Average values of densities achieved during densification without additional compression temperature were higher about 3.57 % in comparing with densities achieved during densification at 100 °C of compression temperature.
3.2. Effect of particle size distribution on pellets quality

The main aim of following part of research was to determine the effect of particle size distribution during semi-operational tests with using of production pelleting plant. As was mentioned above two different particle size distribution were processed by a hammer mill. For pellets producing, untreated sunflower hulls (sample 1) and sunflower hulls treated by single level shredding (sample 2) were used. Pellets with ø 6 mm diameter from both samples representing different particle size distribution (see figure 5) were produced and can be seen in figures 13 and 14.

![Figure 13. Sunflower hulls pellets - sample 1.](image1.png) ![Figure 14. Sunflower hulls pellets - sample 2.](image2.png)

In general the particle size of the input sawdust has an important effect on the densification process because larger particles increase the energy needed for densification. Moreover, pellets densified from large particles have lower homogeneity and strength [11]. A large portion of fine particles allows for better raw material densification. In general (case of wooden), with increasing of particle size, the bond strength between particles decreases, causing them to crumble [18]. In terms of densification, it is very important that the binding forces are created between particles. The strength of the resultant bonds increases with decreasing of particle size [6,19]. It is known that the size of bindings between particles and the particle size distribution affects the physical properties of the pellets.

When pelletizing, especially without additives, the surfaces of the raw material particles must contact with the greatest possible area [20]. The size of this contacting area increases as the particle size becomes smaller and higher compression pressures are applied.

| Parameter               | Pellets (obtained from company) | Pellets (sample 1) | Pellets (sample 2) |
|-------------------------|--------------------------------|-------------------|-------------------|
| Moisture content (%)    | 8.61                           | 5.60              | 5.99              |
| Heating value (MJ.kg⁻¹) | -                              | 19.07             | 19.06             |
| Bulk density (kg.dm⁻³)  | 0.422                          | 0.611             | 0.552             |
| Particle density (kg.dm⁻³) | 0.856                         | 1.237             | 1.231             |
| Mechanical durability (%)| 81.63                         | 87.08             | 79.26             |
| Pellets abrasion (%)    | 32.79                          | 12.63             | 20.51             |
| Hardness (N)            | 197.5                          | 190.3             | 207               |
Table 1 presents the physical properties of the produced pellets from both samples. It is seen that with decreasing of the particle size, the physical properties of pellets changed. The resulting pellets are uniform but of medium quality. What we can attribute to the properties of the raw feedstock - mainly to its chemical composition (low content of lignin, hemicelluloses, etc.). According to presented results we can see that in case of sunflower hulls densification the higher pellets quality was reached from the untreated sample. It means that positive influence of particle size reduction is not in this case significant.

4. Conclusion
The main conclusions that can be withdrawn from this study are as follows:

• With a suitable combination of the influencing variables can be significantly (positively) affected the densification process of sunflower hulls, mainly from the final biofuels quality point of view.
• Increasing of compression pressure positively influences the biofuels final density and dilatation.
• Increasing of compression temperature positively influences only the biofuels dilatation.
• Reducing of particle size does not significantly affect the final pellets quality.

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