In this paper, we will present shortly evaluation of experiment for research of significant parameters’ impact on final briquettes quality. The final briquettes quality (quality of solid high-grade biofuels) is mainly evaluated by briquettes density. In the introduction, we present a short description of pressed material properties. At densification process exist parameters which significantly influencing the final briquettes quality. One of the goals of our research was to detect the influence of some parameters on final briquettes quality. Experimental investigation which had been carrying out at our institute consists of several phases. This paper deals with experimental research history of briquettes production in details at our faculty and publishing several significant output data in regard to briquettes quality and decrease of power consumption. From briquettes production point of view and also from densification machines constructions point of view is very important to know mutual interaction of these technological parameters on final briquettes quality. The main goal of this paper is to present results of analysis of the significance of the technological parameters at densification of selected two species of dendromass feedstock – hardwood and softwood. The experimental findings presented here show the importance of these parameters in the densification process.

KEYWORDS: Densification, mathematical model, briquettes density, pressing pressure, pressing temperature, fraction size, material moisture.
INTRODUCTION

The densification process is a very interesting biomass treatment process. It is a very complicated process, because many parameters influence the process and also the quality of the final briquettes. Briquette quality is defined by EU standards, and is evaluated by mechanical and chemical-thermic indicators.

Solid biofuels are produced by densification machines most usually from wood wastes, (chips, piece of wood waste, sawdust, topwood, coarse wood after mining, waste from forest management, tree stumps, roots, etc...). If we use solid biofuels with accumulated energy values the combustion process is as effective as burning a piece wood waste. At the present time we know many producers of densification machines with a wide range of machines. But not every machine can produce biofuels (briquettes or pellets) to the required quality Standards. This fact demonstrates the many problems which have to be solved.

The properties of the pressed material as well as technological demands are both very important to the briquette production process. The input material needs to be reduced to the optimal particle size, to dry to optimal moisture content level and we have to provide optimal technological parameters throughout the process of densification (Serrano et al. 2011). Every type of material requires an independent approach. Each small change in the properties of the input materials can influence the final quality of the briquette. Different material properties cause different conditions during the densification process (Lehtikangas 2001). In this contribution factors influencing briquettes quality were specified and assigned.

Briquette quality is evaluated mainly by density. Briquette final density is influenced by many parameters. On the basis of the experience that we have acquired and the analyses that we have made, we can divide the parameters into the following three groups (Križan 2009):

► Material parameters,
► Structural parameters,
► Technological parameters.

The material parameters emerge from the properties of the pressed material, i.e. moisture, fraction size, chemical composition of the material, etc. (Požgaj et al. 1997, Križan 2009). We know the following basic properties of the material: The chemical composition of the material; the material density; the material weight; the moisture content of the material; the material contexture and structure; the fraction porosity; the fraction size; the material’s caloric value and others (Požgaj et al. 1997, Križan 2009). After analyses carried out in our institute we know that final briquette quality is significantly affected by the chemical composition of the material, its density, its moisture content, the material contexture and structure which is shown mainly by the size of the chips. Some words about these material properties.

The chemical composition of biomass or wood differs between individual plants. On average plants contain approximately 25 of lignin and 75 % of hydrocarbons or sugars (Križan 2009). Hydrocarbon units consist of many molecules of sugars joined in long chains of polymers. Two important units of hydrocarbons are cellulose and hemicellulose. Nature uses the long polymers of cellulose for fibre building which gives plants the strength they need. Lignin acts as glue which holds together cellulose fibers. Differences occur not only in the chemical composition between groups of softwoods and hardwoods (Fig. 1), but also between individual species of wood (Tab. 1).
Every type of material has its own, specific density, different from each other’s. Density significantly influences the mechanical and physical properties of the wood (Križan et al. 2010a, Križan 2009). The higher the specific density of the input material, the better the densifying of the final briquette. We can say that wood density is important in the mechanical and chemical treatment and its importance increases as the amount of wood increases. For example heavier wood is firmer, stronger and more wear resistance and more resistant to working than soft wood.

Wood density and also wood weight is influenced mainly by moisture content, by the width of the year rings and by the ratio of summer wood, the position in the stem and the age of the tree (Požgaj et al. 1997). Wood density and weight increase with moisture content, whilst wood weight and volume are unchanged. Wood weight increases with the increase of moisture content till it is fully saturated. Wood volume increases to the point of fiber saturation. By controlling the moisture content increases the bound wood volume is unchanged. When the pressed material moisture content is very low, and also when it is very high, that is falling outside of the optimal range of 8 – 15 %, the material particles aren’t consistent and the briquettes fall apart (Križan et al. 2010a, Križan 2009). At lower moisture values the material isn’t sintered, which is needed to produce consistency of the material particles in the briquette. Research and experiment proved that moisture content also has an impact on lignin plasticisation. The temperature of lignin softening depends also on the type of wood from which it is isolated and also on the isolation method. The temperature at which lignin switches from a solid to a plastic state is in direct proportion to its molecular weight, and in indirect proportion to its moisture content.
From the pressing point of view it is very important to know the material composition, its structure and the particle size of the material. Particle structure and size significantly influence the binding mechanisms and those with their amount (force) influence the lonely joining - pressing (Lehtikangas 2001, Li and Liu 2000). They influence the fluency of the densification process and the final quality of densification. Material particles are the result of deformation and strain exerted by external forces, mainly in contact areas. From the densification point of view it is very important that the described force bindings mechanisms increase according to particle size (Mani et al. 2006, Serrano et al. 2011). When the particle size is bigger, then the input power needed for densification also has to be bigger, with the briquette having lower homogeneity and hardness. When the particle size increases, the binding forces decrease, which results in the briquette falling to pieces quickly during the burning process (briquette burns faster, which represents its disadvantage) (Serrano et al. 2011). Briquette quality decreases and the required pressing pressure increases with an increase in the material particle size. During pressing, but mainly during pressing without binding, the material surface planes have to touch over the largest possible area. The size of the contact plane of grains increases with the increase of material smoothness and pressing pressure.

**Technological parameters of densification process**

If we respect mentioned material properties and patterns we are able to produce quality briquettes from each type of wood or material. Therefore is very important to know which technological parameters need to be provided during the densification process for producing briquettes with EU Standards given quality. Every type of material has different mechanical and thermal properties and different chemical composition. Therefore is needed to do experiment step by step for all types of materials. All described material properties influence the quality of the densification process and briquette quality which is evaluated mainly by density. During the pressing process there are many parameters which influencing the final briquettes quality.
density. On our department we made some analyses and experiments to detect the impact of these parameters.

After theoretical analyses we decided that we have to pay attention to the following technological parameters: Pressing pressure, pressing temperature, pressing speed, residence time, etc. These parameters can be changed in the course of pressing according to the capabilities of the briquetting machine.

With research of technological parameters we are dealing on our institute for many years. We are trying to describe mathematically impact of technological parameters (pressing pressure, pressing temperature, material moisture, fraction size) changes on final briquette quality. Created mathematical model helps us by briquette density prediction before lonely real densification process. We are able to say which parameters we have to set for achieving the density given by Standards. Also this mathematical model helps us by engineering of some mechanical parts of briquetting (pelleting) machines. Very important thing was specifying what will be the main goal of the experiment. The primary goal was to detect the impact and to analyze the significance of lonely parameters. But the conditions at densification process are very complicated. There are many external parameters which can influence the researched parameters impact and their mutual activity.

MATERIAL AND METHODS

First phase

So because the impact of input (pressed) material is very influencing, from this reason we had to divide our experiment to 3 main phases. In first phase we have done experiment according to designed levels of measured parameters (Tab. 2). These levels come from our analyses, further experience and pressing stend possibilities. We realized experiment by form of full factorial experiment $2^4$ according to Tab. 2. Goal of the experiment was to follow up the briquettes quality in dependence with pressing temperature, pressing pressure, fraction size and material moisture. At first step we have done experiment by using one chosen type of softwood. We choose pine sawdust for first step of experiment. We designed experimental plan for detecting of impact of changes of technological parameters at densification of pine sawdust. For this type of experiment was designed and made experimental pressing stend (Križan et al. 2010a, Križan 2009) on which we were able to provide all changes of investigated parameters setting according to experimental plan. Briquettes quality was evaluated by briquettes density as is given in EU Standards about solid biofuels. According to EU Standards briquette have very good quality if density is from 1 to 1.4 kg.dm$^{-3}$. In every setting we pressed 7 briquettes. We measured briquette’s dimension, length and weight. These measured values were the base for density calculation. Briquettes density values were processed by various mathematical and statistical methods (e.g. Bartlett’s Test, ANOVA, etc...) with the help of software Stathgraphic S Plus. For closely determination of parameters impact and also impact of their mutual interaction we used method of parameters effect (Križan et al. 2010a, Svátek and Križan 2014). The next step was execution of selection of variables for mathematical model. For this selection we used three widely known criterions: Index of multi-launching determination ($R^2$), Akaike’s criterion (AICc) and Root mean squared error criterion (RMSE).
Tab. 2: Levels of measured parameters (Križan 2009, Križan and Svátek 2010).

<table>
<thead>
<tr>
<th>Phase</th>
<th>Pressure $p$ (MPa)</th>
<th>Temperature $T$ (°C)</th>
<th>Size $L$ (mm)</th>
<th>Moisture $w_r$ (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>First</td>
<td>95 - 159</td>
<td>85 - 115</td>
<td>1 - 4</td>
<td>8 - 12</td>
</tr>
<tr>
<td>Second</td>
<td>63 - 191</td>
<td>55 – 130</td>
<td>0.5 - 4</td>
<td>5 - 15</td>
</tr>
</tbody>
</table>

Second phase

Second phase of experiment was very similar. Also the experiment procedure was very similar but the differences were extended parameters intervals where was experiment done. Comparison of these phases from parameters intervals point of view you can see in previous Tab. 2. The second phase of experiment was done also with the pine sawdust. The goal of the second phase was to design the mathematical model which will describe behavior material (pine) at densification process and to compare the methods of data evaluation. But this model will valid in extended parameters intervals as the mathematical model (Eq. 2). Similarly as in first phase we set pressing pressure $p$, pressing temperature $T$, material moisture $w_r$ and fraction size $L$ as independent variables. Briquette density $\rho$ was set as dependent variable.

In this phase we had to analyze measured data. At first was needed to make Goodness-of-Fit test by Shapir-Wilk W test. This test was realized for each level of each parameter. In the next step we analyzed measured data from outliers´ data point of view. We had to determine if we had some extreme data in our file of data. For testing we used Dean-Dixon non-parametric test of outliers and Grubbs test. If some data are outlying we need to exclude them from measured data for following processing. Also these tests were done for each level of measured data. From file of measured data at pine sawdust we excluded some values from following data processing. We also tested equal variances in groups of measured data because it is very important for following processing. The most famous tests for equal variances testing are Bartlett’s test, Brown-Forsythe test, Leven and O’Brien tests. We used these tests through much known statistical software SAS JMP 8. Results of these tests said us that we could accept the hypothesis about variances equal at 95 % interval of probability. For final design of mathematical model and regression parameters selection was used index of multi-launching determination ($R^2$), Akaike’s criterion (AICc), Malowo’s criterion (Cp) and Root mean squared error criterion (RMSE).

Third phase

The third phase of experiment was done with some chosen types of Slovakian domestic wooden – oak, spruce and beech. Values from mentioned wooden densification were obtained with the same procedure as above and obtained data were processed with the above presented methods.

RESULTS AND DISCUSSION

Method of parameters effect (Fig. 4) in first phase determined that the biggest impact have pressing temperature and material moisture content. Also their interaction has very significant impact on final briquette density. This is very important and helpful result.

Pressing temperature is influencing the lignin plasticisation in cellular structures of material (Nielsen et al. 2009). Lignin is in briquette as nature “glue”. Helps to briquettes be stronger and to have higher density. Pressing temperature significance is important also from the machines engineering point of view. If we are able to provide higher pressing temperature we can use not so
high pressing pressure (as usual). This is influencing the final price of briquetting machine and of course final price of production hour. Because higher pressing temperature is cheaper to provide as higher pressing pressure. But very important is also the interaction between temperature and material moisture content. When is the pressed material moisture content very low and vice-versa very high that means out of some optimal interval (8 – 15%), material particulars aren’t consistence and briquettes is falling to pieces.

![Graph showing individual parameters impact](image)

**Fig. 4:** Individual parameters impact – Pareto’s effects diagram (Križan 2009, Svátek and Križan 2014, Križan and Svátek 2010).

At lower moisture content values material isn’t sintered what is needed for consistence of material particulars into the briquette. Researches and experiment proved that moisture have impact also on lignin plasticisation. As we wrote above pressing temperature is influencing the lignin plasticisation, therefore is also very important mutual interaction between temperature and moisture content. Temperature of lignin softening is depending also on type of wood from which is isolated and also on isolation method. Temperature of lignin phase switching form solid to plastic state is direct proportion to its molecular weight and indirect proportion to its moisture content. On the following figure you can see also the graphical results.

![Graph showing dependence of briquettes density on pressing pressure](image)

**Fig. 5:** Dependence of briquettes density on pressing pressure by various pressing temperatures for pine wood sawdust (wr=10 %; L=2 mm) (Križan 2009, Križan et al. 2011).

![Graph showing output example of Goodness-of-Fit test](image)

**Fig. 6:** Output example of Goodness-of-Fit test for given level of parameters at pine sawdust (Svátek 2010).
In Fig. 5 the briquette density change is shown due to changes in temperature and pressure. If we look closely on the change of briquette density, pressed at 95 MPa, then only under the effect of the change of temperature from 85 to 115°C the density changed from around 25.8% from 0.9378 to 1.1797 kg.dm⁻³. If we press a material of the same characteristic with a pressure of 159 MPa, then under the effect of temperature from 85 to 115°C, the density changed around 15.6% from 1.0604 to 1.2255 kg.dm⁻³. This means that although, we pressed with a 67.4% higher pressure than in the first case, we achieved very small gains in density at the mentioned temperature values. At a temperature of 85°C the change in briquette density was around 13%, while at a temperature of 115°C the change in density was no more than 4%. This proves the assumption that pressing at lower pressure and higher temperature is more effective.

Our aim was not only obtain a graphical result but also to design a mathematical model. According to results of method of parameters effect we choose model with containing of all parameters and also with their most important mutual interactions (Eq. 1). This model had the best results after selection according to criterions.

\[ \rho = f(A, B, C, D, AB, AC, BC, ABC) \]  

(1)

We were able to design the mathematical model and also we were able to calculate or estimate the regression parameters values (\( \beta_0...\beta_8 \)). The final designed form of mathematical model for the first phase you can see in following Eq. 2, where the "p" is briquette density. This model is valid only for briquetting of pine sawdust and is valid only by parameters intervals listed in Tab. 2.

\[ \rho = e^{(\beta_0 - \beta_1 p - \beta_2 T - \beta_3 w_t - \beta_4 L + \beta_5 p.T + \beta_6 p.w_t + \beta_7 T.w_t - \beta_8 p.T.w_t)} \]  

(kg.dm⁻³)  

(2)

Experiment in second phase was done with extended parameters intervals. It was necessary to make Goodness-of-Fit test by Shapir-Wilk W test. This test was realized for each level of each parameter. On the following Fig. 6 you can see outputs of these analyses for one level of parameters at pine sawdust densification. As you can see - measured data for given level of parameters approaching to the line of normality. Also the histogram shows for normality of measured data. A result of Shapir-Wilk W test of normality was that we did not reject the testing hypothesis.

Analysis of variance examines interaction between dependent variable and one or more independent variables. File of data was divided into the groups according parameters changes and was tested if differences between group's averages were random or statistically significant. If the differences are significant, parameter is statistically significant – we can say between dependent variable and parameter exist some interaction. This analysis was done with software SAS JMP 8. You can see on following Fig. 7 that parameters which are overhung the given interval (two full vertical lines). These parameters have significance impact on final briquette quality from pine sawdust.

With help of software SAS JMP 8 we have done estimation of regression parameters (\( \beta_0...\beta_4, \beta_5, \beta_7, \beta_8 \)), evaluation and generated all possible models. We knew also all parameters of each generated model. We choose model containing of all significance parameters and also with their most important mutual interactions (Eq. 3). This model had the best results after selection by criterions. This model is valid only for pine sawdust densification and in the parameters level specified in Tab. 2.

\[ \rho = \beta_0 + \beta_1 p + \beta_2 T - \beta_3 w_t - \beta_4 L + \beta_5 (p - \beta_6 T - \beta_7 w_t - \beta_8 (p - \beta_9 L - T.w_t)) - \beta_9 (p - \beta_5 p.T - \beta_6 p.w_t - \beta_7 (p - \beta_8 T.w_t)) - \beta_8 (p - \beta_9 T.w_t) + \beta_2 w_t (w_t - \beta_{10}) \]  

(kg.dm⁻³)  

(3)
On Fig. 7 you can see simple graphic comparison of measured density values and calculated density values for pine sawdust. This comparison confirmed that our experiment and measured data processing were correct. With these designed mathematical models we obtained the tool for effective and fast prediction of final briquettes density values, pressing temperature values, pressing pressure values, material moisture values and fraction size values. This model can be used at engineering of some important parts of densification machines.

As we can see on the Figs. 4 and 8 the pressing temperature has the largest impact on the whole process of densification. The pressing temperature affects the plasticization of lignin during the pressing process. Lignin, when in liquid state, can bind better to the pressed material. The effect of the pressing pressure, followed by cooling, creates a solid briquette. Based on the experiment results it can be stated that the pressing temperature, from a quality point of view, is more important than the pressing pressure. From a briquette quality point of view the optimal temperature is more important than the pressure. From practical experiences we know, that obtaining the optimal temperature in a pressing process of a machine is cheaper than reaching an optimal value of pressing pressure. It is then logical, when constructing pressing machines; we would use a device having the ability to regulate temperature. Another effect of the pressing temperature on the process could be the fact, which was observed during the experiments, that during the pressing process and using the optimal pressing temperature, less force for removing the briquette from the pressing chamber was required when compared to pressing with no applied heat in the process. The reason is, when densification under higher temperatures, plasticization of the lignin occurred followed by its subsequent mixing within the material particles, a smooth shiny film is formed on the surface of the briquette, which causes a smaller friction coefficient between the briquette and chamber wall.

![Fig. 7: Graphic comparisons of measured density values and calculated density values - pine wood sawdust.](image)

Finding functional dependence \( \rho = f(p,T) \) is the „alpha and omega“ of the whole densification process for the optimal construction of a pressing machine. Not one manufacturer of these machines knows to precisely say what the appropriate pressing pressures for making a quality briquette according to standards are. Too often these densification machines are oversized and thus economically demanding. Finding the functional dependence can help when constructing a densification machine, sizing of the drive, mechanism design, sizing of the shape and dimensions of the pressing chamber etc.

Above described experiment was executed only with pine wood sawdust but we wanted to know behavior of several types of woods during densification. We tried to achieve results also with some hard wood therefore we chose for this experiment oak wood sawdust.
Fig. 8: Analysis of significance of individual parameters – Pine wood sawdust (Švátek 2010).

Structure and the particle size of the oak sawdust are totally different according to pine sawdust. Different material properties cause different conditions during densification and this fact proves following Fig. 9. On this figure is shown effect of investigated parameters at pine and oak sawdust densification. Effects of parameters and also of parameters interaction is not the same. It comes from material properties. Here you can see why is important to know material and its behavior during densification. According to presented results and facts about different parameters impact at pine and oak wood densification you can imagine that for different material have to be designed different mathematical model.

If we want to look closely on the change of briquette density, we have to use the “density response surface”. Response surface is a great and useful tool for presenting the process in maximum amplitudes of individual parameters. As presented in functionality Fig. 5, it is limiting to present the functionality with fixation of several parameters. Using the response surface we can get clear presentation of the process within the given extend by the means of surfaces. These represent set-up levels. Fig. 10 shows the effects of interacting pressing pressure and temperature on the density of the pine briquette at various moisture levels.
The following Fig. 11 shows the effects of interacting pressing pressure and temperature on the density of the oak wood briquette at various moisture levels.

The presented response surfaces are shown at constant value of fraction size – 2 mm, and various values of the compacted material moisture content (5, 8, 10, 12 and 15 %). The initial value (point zero on the axis) is in this case pressing pressure of 63 MPa and pressing temperature of 55°C.

As it can be seen, at lower levels of moisture content and with increasing pressing pressure the final briquette density increases. At higher levels of moisture content, when the pressing temperature is spent on evaporating of the excessive moisture content, with the increased pressure the final value of briquette density increases too. It can be noticed that increasing the pressing temperature has more positive impact on the briquette density increase than increasing the pressing pressure. This response surface graph confirms the hypothesis about using the suitable temperature during densification and at the same time about the suitable initial level of
compacted material moisture content. Such graphic presentations offer complex overview of the selected parameters mutual interaction within the densification process and the influence on the briquettes final density. Based on the calculated and proposed mathematical model it is possible to get specific values for each monitored parameter at any point on the presented surfaces. This provides a very simple tool for briquettes density prediction for any specific setting.

On the Figs. 12 – 16 (Križan et al. 2011, 2010b) we would like to present you the graphical results. You can see very clearly differences between briquettes density from each type of material. Material properties in combination of technological parameters of densification process very significantly influence the final briquettes quality. For a better comparison we enclose also the results for pine wood sawdust.

**Fig. 12:** Dependence of briquettes density from pine wood sawdust from pressing pressure by various pressing temperatures (wr=10 %; L=2 mm).

**Fig. 13:** Dependence of briquettes density from spruce wood sawdust from pressing pressure by various pressing temperatures (wr=10 %; L=2 mm).

**Fig. 14:** Dependence of briquettes density from beech wood sawdust from pressing pressure by various pressing temperatures (wr=10 %; L=2 mm).

**Fig. 15:** Dependence of briquettes density from oak wood sawdust from pressing pressure by various pressing temperatures (wr=10 %; L=2 mm).
From previous and subsequent results it is clear how important the type of material is during the densification process – both briquetting and also pelleting. Among the other significant quantifiable parameter that are also important, the pressing temperature and material moisture content have the most significant influence. It is true that pressing temperature is not a direct parameter of the pressed material but it significantly influences some material properties, it is changing and influencing also the material structure and chemical composition during the densification process. Briquettes from materials with higher lignin and cellulose contents – softwoods (pine or spruce woods) have evidently higher density than briquettes from hardwoods (oak or beech woods).

CONCLUSIONS

The main aim of the experiment was to detect and identify the effect rate of monitored parameters on the final briquettes quality evaluated by briquettes density. By the individual steps we discovered that the most significant effect on briquettes quality has pressing temperature and then material moisture and mutual interaction of these two parameters. The results our hypothesis that pressing pressure, which may seem to be a parameter having the biggest effect on the final briquettes quality, is minor in analyze of effects on briquettes quality. With usage of mathematical and statistical tools we were able to design mathematical model of single axis pressing of pine wood sawdust. From results of our experiment you can see that pressing temperature and material moisture content are most significant parameters also at briquetting of softwoods and also at briquetting of hardwoods. Temperature and moisture content are influencing the lignin plasticisation. Lignin is at biomass densification as natural “glue” and from briquette density and strength point of view is a very important component.

On the base of presented executed analyses and results obtained from the experiment we clearly proved the importance of the pressed material type during the densification process. The parameters which influence and enter the densification process come not only from the pressing process itself but also from the foregoing processes - processing of the material treatment and its preparation - disintegration, separation and drying. Therefore it is very important to know the optimal parameter values which influence final briquette quality for various types of materials.
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