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Pressure agglomeration of biomass with additive of rapeseed oil cake or calcium carbonate

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Summary. The aim of this study was to explain the effect of the 2.5% additive of calcium carbonate or 5% additive of rapeseed oil cake for chopped plant material of topinambour, prairie spartina, multiflora rose, polygonaceous and Virginia mallow (sida) for durability, calorific value and density of the pellets. The additive of calcium carbonate and rapeseed oil cake increased the durability of pellets to 6.1% and 13.6%, respectively, and slightly decreased the density of pellets – 0.7% and 1.7%, respectively. The use of rapeseed oil cake in the pellets improved their calorific value and calcium carbonate had practically no effect on the change in this value. The most marked effect for additives was obtained for topinambour, prairie spartina and Virginia mallow, however for polygonaceous and multiflora rose no significant changes in pellet durability were observed.

Key words: pressure agglomeration, pellet, calcium carbonate, rapeseed oil cake, mechanical durability.

INTRODUCTION

The use of alternative energy sources, including biomass, is becoming wider in recent years [6]. Biomass combustion technology is not complicated and thanks to the availability of cheaper raw material its use for energy purposes is more competitive to currently used conventional fuels [3, 11]. One way of converting biomass is pressure agglomeration, which improves the properties of solid biofuels [4].

One of the stages in the process of pressure agglomeration is conditioning. This covers a range of activities and treatments that are designed to activate natural binders in the material [12]. During conditioning it is possible to add water or steam to the material in order to soften the fiber of densificated particles [20], and particularly the lignin and hemicellulose, which improves the pelletisation process and results in more durability and better physical properties of the obtained pellets [13, 14]. In addition, conditioning also involves adding specific binding agents or other additives.

Although producing pellets without any additives is common [21], there has appeared an interesting possibility to look for a more efficient method of densification of agglomerates improved with appropriately selected binders.

Substances that increase pellet consistency are binders, which means that they support the pressure agglomeration process and, at the same time, they improve their quality and environmental performance [9]. They bind the plant material in a sustainable product that meets standards and does not significantly increase the production cost of the agglomerate , including pellets [1]. Also, binders offer the possibility of developing a new technology and obtaining significantly better agglomerate properties (extra dry, proof against the absorption of moisture from the air, with reduced energy consumption) than before , which may not only affect the combustion process itself, but to a large extent reduce emissions of SO_2, $N2O_5$, dust, CO_2, etc. [8].

Additives, both liquid and solid, produce strong bindings between molecules in the densificated material. Additives are often used in order to improve the quality of pellets according to current standards [10]. Different types of additives are used: binders that improve bindings between particles and those that reduce energy intensity of pellets and improve their combustible properties. There is also an interesting possibility to use waste substances from agricultural production as additives for the production of pellets [15]. In addition to utilising these additives, it can be assumed that rational dosing of those substances can improve physical and chemical parameters of the obtained granules [16, 17].

Adding inert materials, for example, calcium compounds (calcium hydroxide, calcium carbonate) improves plasticity of organic substance which is to undergo pelleting and makes it easier to get forms [19], and a small additive of such a material often significantly reduces dust. In turn, on the basis of agricultural practice, it can be concluded that increasing the participation of protein improves pellet durability, while a too high proportion of fats significantly worsens it, but positively affects granulate energy [5, 18, 22].

For example, calcium carbonate increases ash melting point and thus reduces the risk of ash pollution on grades [7]. Because it is a mineral, the quantity of ash increases. Such additive absorbs heat to decompose, at the same time decreasing the temperature of combustion, which in consequence leads to a reduction in the amount of NOx. The process of pelletization leads, however, to binding of the water contained in plant material. With little moisture of organic substance, the durability of bond between particles is lower, which results in the reduction of mechanical durability of pellets. In order to compensate for these defects binders that increases the durability of bindings and calorific value, eg. rapeseed oil cake, should be used in the mixtures.

The aim of the study was to determine the impact of the additive of calcium carbonate and rapeseed oil cake into energy plants shredded material on the physical properties of pellets produced from these mixtures in the pressure agglomeration process.

MATERIAL AND METHODS

The research material was obtained from rose multiflora (*Rosa multiflora*), prairie spartina (*Spartina pectinata*), topinambour (*Helianthus tuberosus*), Virginia mallow – sida (*Sida hermaphrodita*) and polygonaceous (*Polygonum sachalinense*) plantations. The raw material moisture content (wet basis) was determined using the dry-and-weighing method according to PN-EN 15414-3: 2011 Standard with accuracy 0.1% (Fig. 1).

The plant material was broken up on a stationary stand by means of a forage harvester and then it underwent pressure agglomeration on the pellet machine ZLSP-200B whose basic parameters are summarized in Table 1.

Table 1. Technical parameters of the pellet machine ZLSP-200B

Efficiency	Power	Weight	Dimensions	Die		
				ø die	ø hole	hole length
[kg·h⁻¹]	[kW]	[kg]	[mm]	[mm]	[mm]	[mm]
80–120	7,5	250	1000/430/950	200	8	20

Tests were carried out for each of the plants without the binder and with 2.5% of calcium carbonate or 5% of rapeseed oil cake. The total final weight of each sample was 2 kg. After each trial the material was cooled to ambient temperature.

Mechanical durability coefficient of pellets was determined at a stand made in accordance with the requirements of the PN-EN 15210-1 Standard. During the tests the bin rotational speed was 50 rpm·min^{-1}, and test time – was 10 min. After the durability test the material was sieved through a sieve with a hole diameter of 3.15 mm and the obtained fractions were weighed with an accuracy of 0.01 g while durability was calculated according to the formula:

$$\Psi = 100\frac{m_{pt}}{m_p}, \qquad (1)$$

where:
Ψ – durability coefficient, %,
mp – pellet mass before trial, g,
mpt – pellet mass after trial, g.

Combustion heat was determined by the calorimetric method using KL-10 calorimeter and the calorific value was calculated. Milled samples of 1 g were burned at a pressure of 3 MPa. Taking into account the hydrogen content in the material, which was established in the Analytical Center of Warsaw University of Life Sciences, the calorific value was calculated using the following formula:

$$W_u = W_t - 2454(W_w + 9H), \qquad (2)$$

where:
Wu – calorific value, MJ·kg^{-1},
Wt – combustion heat of sample, MJ·kg^{-1},
Ww – relative moisture of fuel,
H – relative proportion of hydrogen in fuel.

Pellet density measurement was made on a randomly selected representative sample of 10 pellets from each group. Two series of measurements were performed within 1 minute after the agglomeration process as well as after 15 minutes. Pellet diameter was measured in two perpendicular planes in the middle of the pellet length. Linear measurements were made by means of an electronic digital caliper with an accuracy of 0.01 mm, the measurement of mass of each pellet was carried out with an accuracy of

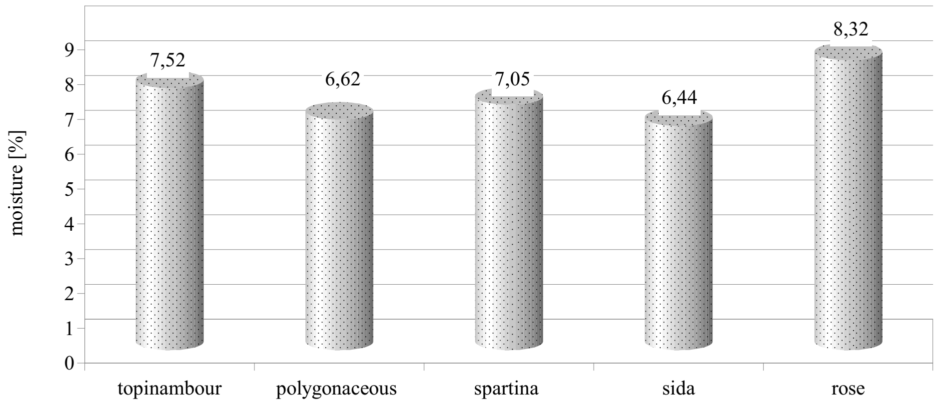


Fig. 1. Moisture content of the material during the tests

0.01 g on laboratory scales RADWAG WPS 600 / C, and the density of the pellets was determined by the formula:

$$\rho = \frac{m}{V}, \tag{3}$$

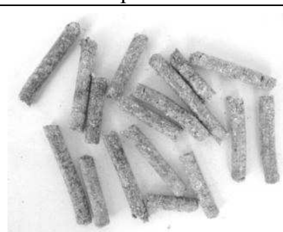
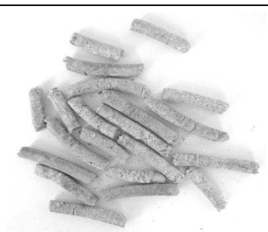
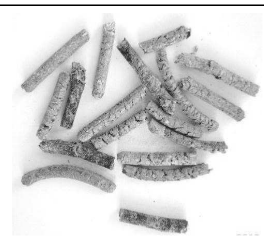
where:
ρ – pellet density, kg·m^{-3},
m – pellet mass , kg,
V – pellet volume , m^3.

Data analysis was performed using the Statistica computer program version 10, using the procedure of analysis of variance and Duncan's test.

RESULTS AND DISCUSSION

With additives, pellets of similar size and shape changed their colors and appearance (Table 2). Pellets produced with the additive of calcium carbonate were characterized by a slightly cracked outer surface and were matte, rough, without the characteristic glassy surface which is formed as a result of thermal conversion of lignin. Pellets with rapeseed oil cake were characterized by a darker color and a shiny outer surface with burnt lignin. Rapeseed oil cake reduced dust in pressure agglomeration, whereas the additive of calcium carbonate significantly increased dust in that process.

Table 2. Pellets after pressure agglomeration process

Pellets without additive	Pellets with additive of rapeseed oil cake	Pellets with additive of calcium carbonate
topinambour		
		
polygonaceous		
		
sida		
		
spartina		
		
rose		
		

An analysis of variance showed that both the energy plant species, the type of the additive, and their interaction had a statistically significant effect on the pellets mechanical durability, as in all cases, the value of the critical level of significance of $p < 0.0001$ (Table 3). The results of the Duncan test (Table 4) allow the conclusion that in the case of both types of additives and pellets without any additive three distinct homogeneous groups of pellet mechanical durability values were formed.

Table 3. The results of the analysis of variance of factors affecting the mechanical durability of pellets from energy plants material

Source	Sum of squares	Degrees of freedom	Mean square	F factor	p – value
Plant: A	1360.8	4	340.2	311.4	<0.0001
Additive: B	735.6	2	367.8	336.6	<0.0001
Interaction: A x B	472.9	8	59.1	54.1	<0.0001
Error	32.8	30	1.1		

Table 4. The results of the Duncan test of the analysis of mean values of the mechanical durability of pellets for homogeneous groups of plant species and type of additive

Factor	Ψ, %	Homogenous group			
Plant					
Topinambour	73.59	x			
Polygonaceous	74.36	x	x		
Sida	74.78		x	x	
Spartina	75.45			x	x
Rose	88.21				x
Type of additive					
None	72.50	x			
Calcium carbonate	76.96		x		
Rapeseed oil cake	82.38			x	

Plant species formed four homogeneous groups, which included pairs of plant species of mixed system; this means that there were no clear differences between the coefficients of mechanical durability of pellets for the investigational species. Mean values of the mechanical durability, their standard deviations and 95% range of variation are shown in Table 5, a graphic interpretation of the interaction of the durability is shown in Figure 2.

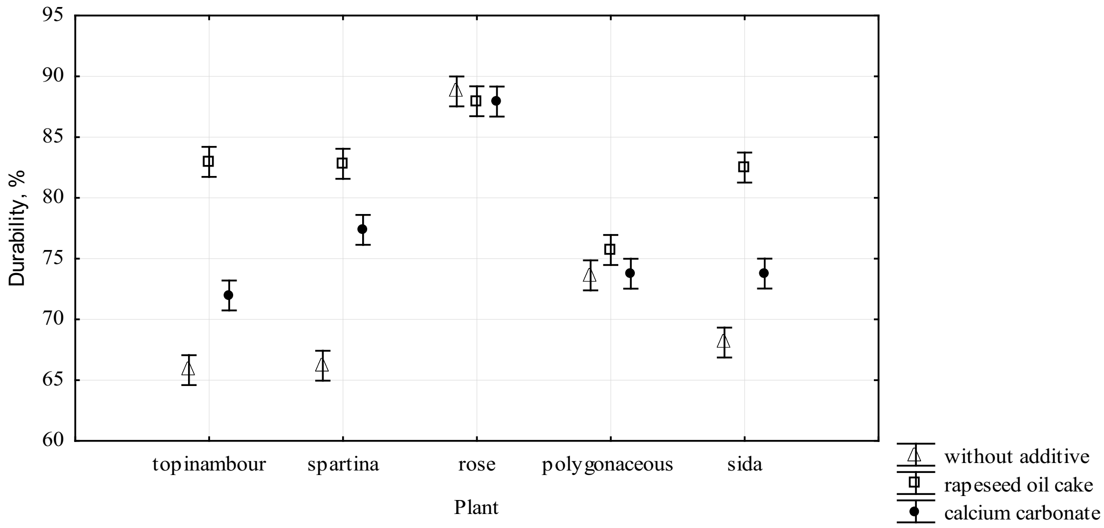


Fig. 2. The interaction of the additive with the species of energy plants on mechanical durability of pellets

Table 5. The average values of mechanical durability, standard deviation, SD and 95% confidence intervals for the plant species and type of additive or its lack

Factor	Ψ, %	SD Ψ, %	-95% Ψ, %	+95% Ψ, %	Population
Plant					
Topinambour	73.59	0.35	72.87	74.30	9
Spartina	75.45	0.35	74.74	76.16	9
Rose	88.21	0.35	87.50	88.92	9
Polygonaceous	74.36	0.35	73.65	75.07	9
Sida	74.78	0.35	74.07	75.50	9
Additive					
None	72.50	0.27	71.94	73.05	15
Rapeseed oil cake	82.38	0.27	81.83	82.94	15
Calcium carbonate	76.96	0.27	76.41	77.51	15

Rose pellets had the highest durability (87%–89%), but the effect of additives on the value rate change of mechanical durability was the lowest and ranged within statistical error (Fig. 2). For other plants, additives improved pellet durability more effectively. The additive of rapeseed oil cake increased durability to a greater extent than the additive of calcium carbonate (Fig. 2). The additive of calcium carbonate and rapeseed oil cake allowed to increase the durability of pellets by 6.1% and 13.6% respectively. To sum up, the greatest effect of additives was obtained for topinambour, spartina and sida, but for rose and polygonaceous practically no change in the pellets durability was recorded. Therefore the durability of the produced pellets was mainly affected by the type of densificated material and type of additive. This observation confirms the inference of Niedziółka et al. [16], who also found that the percentage of additives in prepared mixtures has a significant influence on the durability of pellets. Another factor influencing the value of durability coefficient is also the moisture of material [2]. In our study, material from multiflora rose had relatively the highest moisture (8.32%), but the lowest moisture for sida (6.44%) was not lower significantly enough in this area to draw any conclusions about its impact on the durability of pellets, especially since the moisture content of the material was covariate and was associated with plant species.

Pellet calorific values ranged from 15.1 MJ·kg^{-1} for polygonaceous without an additive to 19.9 MJ·kg^{-1} for spartina without an additive (Fig. 2). For other plants calorific values were similar and were approximately 17 MJ·kg^{-1}. Calcium carbonate had no significant effect on the calorific value of pellets and the additive of rapeseed oil cake generally caused a increase in combustion heat and calorific value.

An analysis of variance showed that pellet density of was affected in a statistically significant way by the main factors: species of energy plants, the type of the additive used, the time of measurement and all of their double and triple interactions with a critical significance level p <0.0001 (Table 6). On the basis of an analysis of the Duncan test (Table 7) four homogeneous groups for the plant species were identified, including a common group created by spartina and rose. An additive of calcium carbonate did not significantly differentiate the density of pellets because it formed homogenous groups with the additive of rapeseed oil cake and without any additive. Table 7 summarizes the mean density of the pellets determined after 1 min and after 15 min from the time of pellet production for comparison purposes only, because for two factor levels variance analysis results are sufficient (Table 6). A smaller value of 2.9% for density of the pellets after 15 min indicates the expansion of the pellets associated with stress relaxation of the material between the particles during their storage. Average densities of pellets with their standard deviations and 95% range of variation are shown in Table 8, and the graphic interpretation of factors interaction on the density of pellets is presented in Figure 4.

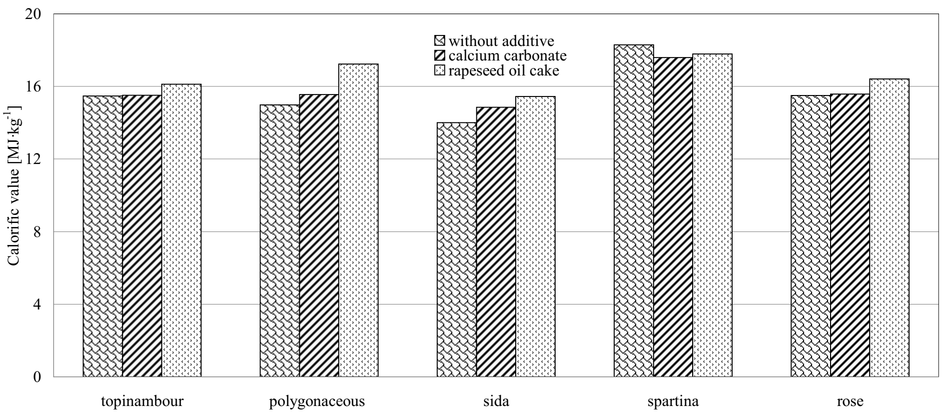


Fig. 3. Calorific values of pellets

Table 6. The results of the analysis of variance of factors affecting the density of the pellets from energy plants material

Source	Sum of squares	Degrees of freedom	Mean square	F factor	p – value
Plant: A	1143889	4	285972	93.6	<0.0001
Additive: B	19681	2	9841	3.2	<0.0001
Time: C	80988	1	80988	26.5	<0.0001
Interaction: A×B	1160604	8	145076	47.5	<0.0001
Interaction: A×C	403876	4	100969	33.0	<0.0001
Interaction: B×C	131004	2	65502	21.4	<0.0001
Interaction: A×B×C	1151864	8	143983	47.1	<0.0001
Error	824974	270	3055		

Table 7. The results of the Duncan test of the analysis of mean values of the density of pellets for homogeneous groups of plant species, type of additive and measurement time

Factor	ρ, kg·m⁻³	Homogenous group			
Plant					
Spartina	1050.47	x			
Rose	1054.53	x			
Topinambour	1112.61		x		
Polygonaceous	1137.88			x	
Sida	1218.51				x
Type of additive					
Rapeseed oil cake	1104.21	x			
Calcium carbonate	1116.31	x	x		
None	1123.88		x		
Measurement time, min					
1	1131.23	x			
15	1098.37		x		

After 1 min from the pellets production, sida without additive was characterized by the highest density of pellets, whereas spartina with rapeseed cake had the lowest pellet density (Fig. 3). The density of pellets after 15 min was smaller, but more stable than after 1 min. After 1 min from the pellet production, the additive of calcium carbonate increased the density of pellets made of spartina material and after 15 min also of the material of topinambour and sida. Generally, however, the additive of calcium carbonate and rapeseed oil cake contributed to a slight decrease in pellet density, by 0.7% and 1.7% respectively. Confirmation of this inference requires an extension of research on the participation of additives, extended time of stress relaxation in the pellets, increasing the moisture content and diversity of agglomerate pressure.

Table 8. The average values of density, standard deviation, SD and 95% confidence intervals for the plant species, type of additive or its lack and measurement time

Factor	ρ, kg·m⁻³	SD ρ, kg·m⁻³	-95% ρ, kg·m⁻³	+95% ρ, kg·m⁻³	Population
Plant					
Topinambour	1112.61	7.14	1098.56	1126.66	60
Spartina	1050.47	7.14	1036.42	1064.52	60
Rose	1054.53	7.14	1040.48	1068.58	60
Polygonaceous	1137.88	7.14	1123.83	1151.93	60
Sida	1218.51	7.14	1204.46	1232.56	60
Additive					
None	1123.88	5.53	1113.00	1134.76	100
Rapeseed oil cake	1104.21	5.53	1093.33	1115.10	100
Calcium carbonate	1116.31	5.53	1105.42	1127.19	100
Time					
1	1131.23	4.51	1122.346	1140.12	150
15	1098.37	4.51	1089.485	1107.26	150

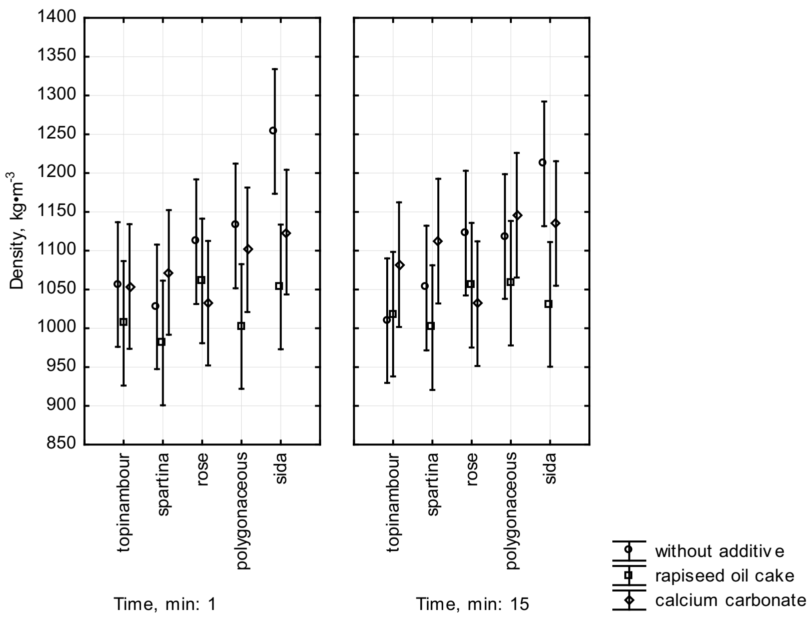


Fig. 4. The interaction of the additive with the species of energy plants and measurement time on the density of pellets

CONCLUSIONS

1. The additive of calcium carbonate or rapeseed oil cake allowed to increase mechanical durability of pellets by 6.1% and 13.6%, respectively, and slightly decreased pellet density by 0.7% and 1.7%, respectively. Also, the calorific value of this solid fuel changed slightly.
2. The density of the pellets after 15 minutes was smaller by 2.9%, but more stable than after 1 minute from production time, which indicates that expansion of the pellets is associated with stress relaxation between the particles of the material during their storage time.
3. The most marked effect of using additives was obtained for topinambour, spartina and sida, but for rose and polygonaceous practically no change in the pellets durability was recorded. However, pellets from rose were characterized by the highest values of the mechanical durability coefficient (87%–89%), which may be due to the structure and physical properties of that material.
4. Pellets produced with the additive of calcium carbonate were characterized by a slightly cracked outer surface and were matte, rough, without the characteristic glassy surface, formed as a result of thermal conversion of lignin. Pellets with an additive of rapeseed oil cake were characterized by a darker color and a shiny outer surface with burnt lignin.

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AGLOMERACJA CIŚNIENIOWA BIOMASY Z DODATKIEM MAKUCHÓW RZEPAKOWYCH LUB WĘGLANU WAPNIA

Streszczenie. Celem pracy było wyjaśnienie wpływu dodatku 2,5% węglanu wapnia lub 5% makuchów rzepakowych do rozdrobnionego materiału topinambura, spartiny, róży, rdestowca i ślazowca na trwałość, wartość opałową i gęstość peletów. Do-

danie węglanu wapnia i makuchów pozwoliło na zwiększenie trwałości peletów, odpowiednio o 6,1% i 13,6% oraz nieznaczne zmniejszenie gęstości peletów, odpowiednio o 0,7% i 1,7%. Zastosowanie makuchów poprawiło wartość opałową peletów, a węglan wapnia praktycznie nie miał wpływu na zmianę tej wartości. Największy efekt wpływu dodatków uzyskano dla topinambura, spartiny i ślazowca, a dla rdestowca oraz róży praktycznie nie zarejestrowano zmian w trwałości peletów.
Słowa kluczowe: aglomeracja ciśnieniowa, pelet, węglan wapnia, makuchy rzepakowe, trwałość mechaniczna.