The Effect of Waste Vegetable Oil Addition on Pelletability and Physical Quality of Wood Pellets

Nevena Mišljenović¹, John Mosbye¹, Reidar Barfod Schüller², Odd-Ivar Lekang¹ and Carlos Salas-Bringas¹

Dep. Of Mathematical Sciences and Technology, Norwegian University of Life Sciences, P.O. Box 5003, N-1432 Ås, Norway

² Dep. Of Chemistry, Biotechnology and Food Science, Norwegian University of Life Sciences, P.O. Box 5003, N-1432 Ås, Norway

ABSTRACT

The aim of this study is to produce alternative wood pellets with addition of waste vegetable oil and to examine how the oil affects pelletability and pellet's physical properties. The oil addition was 2.2% and 5.8% on dry solids. The results show that oil addition significantly increases energy content in wood pellets (p<0.05). Energy content was increased from 19.69 MJ/kg (control) to 20.05 MJ/kg (2.2% of oil) and 20.64 MJ/kg (5.8% of oil). Strength of pellets and the pressure required to initiate pellet discharge from the die were reduced by oil addition.

INTRODUCTION

The EU is currently a leader in world pellets market. Demands for pellets in Europe have already exceeded production capacities, thus significant amounts of pellets have been imported mainly from US. Moreover, European Commission predicts that pelleted solid biofuels will play important role in meeting renewable energy targets in Europe¹. All this indicate further growth in pellet production and necessity for utilization of new, cheap, alternative raw materials especially because woody biomass is almost exclusively used as a feedstock for pellets production which makes it scarce on

a global level. Alternative feedstock of particular interest are residues and side products from agriculture and food/feed processing². Market integration of alternative and mixed pellets still meets various constrains, but recent study showed importance of this sector because it helps sustainable rural development and add value on locally available cheap raw materials³. Alternative biofuel pellets are typically produced from sawdust, which can be considered as a carrier, and additional nonwoody biomass in smaller amounts⁴. The amount of alternative raw material depends on process and product quality limitations.

Apart from other fibrous materials like straw, hay and grass, usage of starch and lignin based materials is very common because those materials help binding particles together⁵⁻⁷. Vegetable oils are mentioned in literature as an additive which reduce die wall friction and reduce energy consumption for pelleting process due to lubricating effect⁷, but there is no systematic study about the influence of oil on pelletability and physical quality of pellets. In this study waste vegetable oil (WVO) was selected as a raw material for production of alternative biofuel pellets.

WVO is discarded by restaurants, food manufacturers (potato chips, breaded fish

sticks, doughnuts, etc.) and other facilities (schools, hospitals. households. etc.) because it cannot be further used in human or animal nutrition. Inappropriate disposal of WVO can be environmentally hazardous; therefore its further utilization is preferable. WVO has a considerable potential as a component of pelleted biofuels, because it is non-fossil oil, has high calorific value and does not require any pretreatment. Oil addition can improve wood fuel properties, but densification of wood with added oil can be more challenging. Application of oil can reduce dust formation during pelleting and later during pellet handling. Availability of WVO is not a limitation since large amounts available from restaurants, food are processing industry and households⁸. Price of WVO oil is 2.5 - 3 times lower than virgin vegetable oil⁹. Costs of handling WVO before usage include iust transportation. storage, and costs of mechanical cleaning (filtration), which is usually done at production place¹⁰.

The main purpose of this article is to produce alternative wood pellets with addition of WVO and to examine how the oil addition affects pelletability and pellet's physical properties. Oil has been added in two different amounts (2.2 and 5.8% on dry solids). Mixed and control pellets (no oil addition) were produced by single pellet press method at 120°C under four different compacting pressures (75, 150, 275 and 300 MPa).

MATERIAL AND METHODS Preparation of the raw materials

Spruce stumps (*Picea abies*) were selected as raw material and cut from a local forest in Ås, Norway in December 2012. In order to preserve woods physicochemical properties the stumps were stored in a freezer until further usage. Prior to the pelleting the wood was defrosted, the bark was removed, stumps were cut in smaller pieces, pre-dried and grinded in a grinder (Brabender, Duisburg, Germany) with screen opening size of 1.5 mm.

Adopting literature recommendation for pelleting process¹¹, the wood powder was moisturized to $10 \pm 1\%$ moisture content by spraying water in an intensive laboratory mixer (Diosna P1/6, Germany). The final moisture content in spruce powder was $10.81\pm0.34\%$ (Table 2.). This material served as control sample and as a base raw material for the mixtures with oil. The moisturized powder was stored in vacuum packed plastic bags at ambient conditions until further usage (1 - 2 weeks).

The WVO was obtained from McDonald's restaurant in Vestby, Norway. Before spaying, the WVO was filtrated trough filter paper. Basic physicochemical properties of the WVO are presented in Table 1.

rable 1. roperties of waste vegetable off.				
Property	Value			
HHV, MJ/kg	37.60±0.07			
Density*, kg/m ³	910.7			
Dynamic viscosity ¹² ,	0.117 (at 20°C)			
Pa s	0.026 (at 60°C)			
	0.007 (at 120°C)			
Ash content ⁹ , %	0.006			

Table 1. Properties of waste vegetable oil.

*Measured at room temperature

The WVO at two different levels (L_1 and L_2) was added to the powder by spraying in a high shear mixer having three impellers and a tulip-form chopper (Diosna P1/6, Germany). Intensive mixing (mixer 250 rpm; chopper 500 rpm), manipulating oil drop size in the spraying lance (Düsen-Schlick GmbH, Germany, Model 970) and preheating of the WVO to 60°C were used to facilitate an even distribution of the oil in sawdust. Final content of oil in sawdust was calculated from difference in the higher heating value (HHV) of control powder and powder with oil. Powders L_1 and L_2 contained 2.2% and 5.8% of WVO on dry solids, respectively. The materials were pelletized at least 48 hours after the mixing in order to allow WVO to penetrate from fiber surface into pores.

Characterization of the raw materials

Following analyses on raw materials were performed:

• The particle size distribution of spruce sawdust was measured by Mastersizer 3000 optical unit combined with Aero S dry dispersion unit (Malvern Instruments, U.K.).

• The bulk density was determined by measuring the mass of a known volume of material that has been loosely poured into a graduated cylinder. The density of oil was determined by measuring the mass of know volume of oil in a graduate cylinder (± 0.2 ml) on analytical balance (± 0.0001 g)

• The water activity value (a_w) was measured by a Rotronic HygroLab C1 (Switzerland) instrument. Average temperature during the a_w measurements was 20.16±0.10°C.

• The moisture content was determined by overnight drying in an oven (Termaks, Norway) at 105°C.

• The content of ash and volatile matters was determined in a muffle furnace (Nabertherm D-2804, Germany) according to EN 14775 and EN 15148 standard procedure, respectively.

• The fixed carbon $(\%_{d,b})$ was calculated by difference between 100 and the sum of the volatile matter and ash content.

• The HHV was calorimetrically determined (Parr 1341 Oxygen Bomb Calorimeter, Moline IL) following the EN 14918 standard procedure.

Pellet production

Pellets were produced in a single pellet press following the same procedure as described in Mišljenović et al.¹³ Detailed description of the pelleting unit was previousely presented by Salas–Bringas et al.¹⁴ The pellets were produced at 120 °C under four different compacting pressures -75, 150, 225 and 300 MPa. The pressure required to initiate pellet discharge from the die i.e. the pressure at incipient flow (P_{max}) was recorded to determine differences in friction generated on 'die – pellet' contact area among the materials.

Testing the pellets

The produced pellets were stored in sealed plastic bags at ambient conditions until further testing. The strength of pellets was measured 48 hours after production. Before the strength measurement. dimensions (length and diameter) and mass of each pellet were measured to calculate pellet density. The strength measurement was performed by a diametric compression test using a 60mm diameter probe connected to Lloyd LR 5K texture analyzer (Lloyd Instruments, U.K.). Maximum normal force during the pellet breakage was recorded. Compression test was performed with speed of 1 mm/min.

<u>Data analysis</u>

Experiment was designed around two factors; oil content at three levels (C – control; $L_1 - 2.2\%$; $L_2 - 5.8\%$) and compressing pressure at five levels (0, 75, 150, 225 and 300MPa). Minimum 3 replicates were tested per each combination of the factors. Results were analyzed using ANOVA – General Linear Model for unbalanced data (p<0.05). Tukey's tests were employed to determine which groups differed (95% confidence interval).

RESULTS AND DISCUSSION

The addition of WVO caused changes in the spruce sawdust properties. Small changes in the powders appearance were observed. The powders with oil were less dusty, slightly darker and less flowable during handling and filling of the die channel. The changes in powder flow are particularly important for potential industrial application of the oily raw materials.

Parameter	Control	L ₁	L ₂				
Bulk density, kg/m ³	119.44 ± 1.54^{a}	119.35±1.95 ^a	122.05±1.81 ^a				
a _w value	0.517 ± 0.009^{a}	$0.540{\pm}0.007^{a}$	0.461 ± 0.018^{b}				
Proximate analysis							
Moisture content, %	10.81±0.34	10.60*	10.24*				
Ash, % _{d.b.}	0.28 ± 0.02^{a}	0.27 ± 0.05^{a}	0.28 ± 0.04^{a}				
Fixed carbon [*] , % _{d.b.}	12.14	8.19	7.62				
Volatile matter, % _{d.b.}	87.58±0.75	91.54±0.15	92.10±1.08				
HHV, MJ/kg _{d.b.}	19.59±0.16 ^a	19.99 ± 0.10^{b}	20.64±0.18 ^c				

Table 2. Raw material properties.

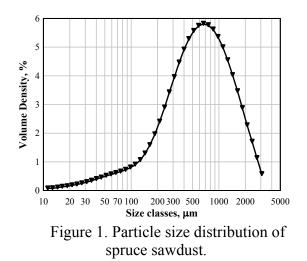
^{abc} different letters within the same raw indicate significant differences according to post – hoc Tukey's test (95% confidence interval); ^{*}calculated values

Good flowability of powders can be achieved by careful design of silo and equipment or by modification of powder flow properties¹⁵. The most important properties of tested materials are presented in Table 2. The bulk density varied between $119.35 - 122.05 \text{ kg/m}^3$. The oil addition did not cause significant changes in the bulk density of spruce sawdust (p<0.05).

A_w value describes availability of free water in different products (typically food and feed). Free water affects products microbiological, enzymatic and chemical stability. The aw value of tested materials was significantly reduced only when the WVO was added in higher amount (L₂). This can be explained by covering the water with oil layer and making it in that way not accessible for the measurement. The a_w values of tested powders were very low indicating stability of materials in respect of their microbiological activity. Microbiological activity can be a reason of self-ignition, deterioration material of material and health problems. In this range of a_w values (0.461 - 0.540) growth of microorganisms is not possible¹⁶.

The proximate analysis of powder showed expected changes such as increasing of the volatiles content with WVO addition and no significant change of the ash in powder. Tested powders had low ash content and according to EU legislation (EN 14961-1) can be categorized in the A group

of pellets. The most important change caused by WVO addition is reflected in increased energy content. Significant increasing was observed (p < 0.05). Energy content was increased from 19.69 MJ/kg (control) to 20.05 MJ/kg (L_1) and 20.64 MJ/kg (L₂). Compared with other method of biomass energy enrichment like torrefaction, addition of 2.2% oil increased final energy content in powder similarly as torrefaction at $225^{\circ}C^{13}$. This way of energy enrichment is definitely easier and less energy demanding. In addition to the tabular data which describe used materials, the volume based particle size distribution of spruce powder is presented in Fig. 1. 90% of the particles had diameter lower than 1750 µm and 10% had higher than 166 µm. The mean volume diameter of particles D [4;3] was 819 μm.



Pellets produced in single pellet press have perfect cylindrical shape. In this experiment the control pellets were wellformed, with shiny surface, while the L_1 and L₂ pellets had more porous structure and could easily disintegrate, indicating poor binding between the particles. Physical characterization of the pellets was performed by measuring their density and strength. Statistically, the type of material did not have significant influence on pellet density while the compacting pressure and their interaction had (Table 3.). However, from Fig. 2 is possible to see changes in compressibility of the oiled materials compared to control. The densities of control pellets significantly increase with increasing compacting pressure from 75 to 300 MPa, which was not the case for L_1 and L_2 pellets. The densities of control pellets changed from 1089 kg/m³ (75 MPa) to 1243 kg/m³ (300 MPa), while the densities of L_1 and L_2 pellets were practically pressure independent (L₁: 1142 \rightarrow 1196 kg/m³; L₂: $1164 \rightarrow 1191 \text{ kg/m}^3$). Additionally, L₁ and L_2 lines overlap indicating that the amount of oil between 2.2 to 5.8 % did not have influence on pellet density. Control pellets had higher density than L_1 and L_2 pellets except for those produced at 75 MPa.

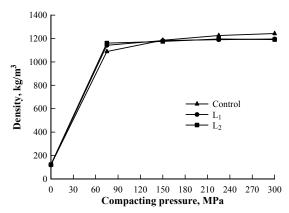


Figure 2. Compressibility of spruce sawdust with different oil levels.

The results show that WVO made the material less compressible, probably due to

filling the fiber lumen and inter-particle space with incompressible liquid and because of poorer binding among the particles.

Table 5. Results of statistical analysis.							
Parameter	df	SS	MS	F	р		
Strength							
Material	2	888.03	444.0	137.7	0.00		
Pressure	3	894.14	298.0	90.40	0.00		
Interact.	6	182.91	30.48	9.25	0.00		
Error	41	135.17	3.3				
Density							
Material	2	667	334	1.75	0.18		
Pressure	3	62035	20678	108	0.00		
Interact.	6	29319	4886	25.56	0.00		
Error	55	10513	191				
DP							
Material	2	0.1835	0.092	52.49	0.00		
Pressure	3	0.0184	0.006	3.51	0.02		
Interact.	6	0.0044	0.001	0.42	0.86		
Error	55	0.0961	0.002				

Table 3. Results of statistical analysis.

The strength of the pellets was measured by a diametric compression test following the same procedure as described in Mišljenović et al.¹³ The peak force was recorded and the results are expressed as the maximum force per length of pellet (N/mm). Results of the statistical analysis are presented in Table 3. Both factors and their interaction had statistically significant impact on pellet strength. Also, statistically there were no differences in strength between L_1 and L_2 pellets produced at the same pressure, but both differed compared to control (according to Tukey's test). The strength of pellets significantly decreased with addition of WVO indicating poorer binding among particles (Fig. 3). Binding mechanisms during pelleting can be divided in five groups: solid bridges, adhesion and cohesion forces, surface tension and capillary pressure, attraction forces between solids and interlocking bonds¹⁷. The oil layer on the sawdust particles led to the surface deactivation, which resulted in

deactivation of all the mechanisms that include contact sites and attraction forces between surfaces. Similar effect was observed by Nielsen et al.¹⁸ in sawdust with higher extractive content which deposition on particle surface deactivated their binding ability. Only interlocking and capillary attraction between the particles due to entrapped oil gave the strength to pellets. Even the interlocking was poor because of the oil's lubricating effect which caused sliding between the particles. Capillary attraction due to oil layer is lower compared to water since the force is directly proportional to surface tension¹⁹. Surface tension of oil is about half of water value and decreases with increasing temperature 20 .

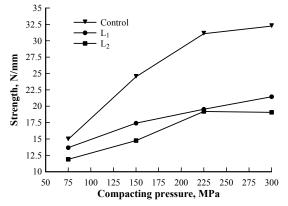


Figure 3. Strength of pellets produced under different compacting pressures.

Pelleting in single pellet press is a method that simulates compaction that occurs in flat or ring die pellet press. This method is widely used for testing material pelleting properties and can be considered as first step in process and experimental design, but it cannot be a replacement for pilot or large scale experiments. Energy consumption cannot be measured in single pellet press but can be indirectly estimated²¹. In this paper the pressure required to initiate pellet discharge (P_{max}) was used as an indicator of the friction generated in the die - pellet contact area. During pellet discharge, P_{max} never

exceeded the value of the pressure used to produce pellets. Absolute value of this parameter does not indicate the extent of the power requirements, but it can show the difference between materials and rank how easy or difficult material will flow through die. The material's resistance to be moved in a die hole is in large extent responsible for the power requirements in commercial pellet presses.

Results showed that P_{max} values were significantly reduced by oil addition (p<0.05). Moreover, higher oil content led to a lower P_{max} . This is a consequence of the lubricating effect of WVO. P_{max} was slightly higher when higher pressure was used for pelleting (Fig. 4). Both factors had significant influence on P_{max} , but their interaction did not (Table 3).

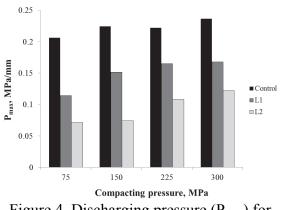


Figure 4. Discharging pressure (P_{max}) for pellets produced at different compacting pressures.

In this experiment the pelleting was performed 48 hours after the oil spraying in order to allow better oil incorporation in fiber particles. In an industrial case this could be done in a silo. During pelleting, it was possible to observe some oil retention on the bottom rod and on the bottom part of pelleting machine. Also, pellets the produced at higher pressure had 'oil gradient' which can be a result of pressing out the WVO from fiber lumen and interparticle space (Fig. 5). This leakage definitely is not desirable. In context of real

pellet press where material continuously flows through die 'oil gradient' probably would not appear. For a large scale production the main concern is about the previously mentioned powder flowability and the fact that temperature in the die is generated by friction between material and the wall, so if the friction is too low temperature will not be high enough to ensure good pellet quality. This article shows the effects of oil in wooden materials, but an investigation in a pilot or commercial pellet press is needed to find the optimum between processing and physical quality.



Figure 5. Example of the 'oil gradient' in L₁ pellet produced at 150 MPa.

CONCLUSION

Waste vegetable oil has been added in two different amounts in spruce sawdust, which has been pelletized in a single pellet press under four compacting pressures. Presented results led to the conclusion that oil addition significantly increases energy content in biofuel, make material less compressible, reduce pellet strength and reduce friction on the pellet – die contact area.

REFERENCES

1. Flach, B., S. Lieberz, K. Bendz, and B. Dahlbacka. <u>EU Biofuels Annual 2013</u>, Global Agricultural Information Network (GAIN), NL3034, 2013

2. Pollex, A. and T. Zeng. *Initiators Handbook* MIXBIOPELLS, 2012

3. Pollex, A. and T. Zeng. Summary of the MixBioPells project results, MIXBIOPELLS, 2012

4. Nunes, L.J.R., J.C.O. Matias, and J.P.S. Catalão, (2014), "Mixed biomass pellets for thermal energy production: A review of combustion models", *Applied Energy*, **127**(0): p. 135-140.

5. Ahn, B.J., H.-s. Chang, S.M. Lee, D.H. Choi, S.T. Cho, G.-s. Han, and I. Yang, (2014), "Effect of binders on the durability of wood pellets fabricated from Larix kaemferi C. and Liriodendron tulipifera L. sawdust", *Renewable Energy*, **62**(0): p. 18-23.

6. Berghel, J., S. Frodeson, K. Granström, R. Renström, M. Ståhl, D. Nordgren, and P. Tomani, (2013), "The effects of kraft lignin additives on wood fuel pellet quality, energy use and shelf life", *Fuel Processing Technology*, **112**(0): p. 64-69.

7. Tarasov, D., C. Shahi, and M. Leitch, (2013), "Effect of Additives on Wood Pellet Physical and Thermal Characteristics: A Review", *ISRN Forestry*, **2013**: p. 6.

8. Toop, G., S. Alberici, M. Spoettle, and H.v. Steen. *Trends in the UCO market*, Ecofys, 2013

9. Demirbas, A., (2009), "Biodiesel from waste cooking oil via base-catalytic and supercritical methanol transesterification", *Energy Conversion and Management*, **50**(4): p. 923-927.

10. Cvengroš, J. and Z. Cvengrošová, (2004), "Used frying oils and fats and their utilization in the production of methyl esters

of higher fatty acids", *Biomass and Bioenergy*, **27**(2): p. 173-181.

11. Leaver, R.H. "Wood Pellet Fuel and the Residential Market". in *The Ninth Biennial Bioenergy Conference - Bioenergy 2000 -Moving Technology into the Marketplace*. 2000. Bufalo, New York.

12. Mišljenović, N., R.B. Schüller, E.-O. Rukke, and C.S. Bringas, (2013), "Rheological Characterization of Liquid Raw Materials for Solid Biofuel Production", *Annual Transaction of the Nordic Rheology Society*, **21**: p. 61 - 68.

13. Mišljenović, N., Q.-V. Bach, K.-Q. Tran, C. Salas-Bringas, and Ø. Skreiberg, (2014), "Torrefaction Influence on Pelletability and Pellet Quality of Norwegian Forest Residues", *Energy & Fuels*, **28**(4): p. 2554-2561.

14. Salas-Bringas, C., T. Filbakk, G. Skjevrak, O.-I. Lekang, O. Høibø, and R.B. Schüller, (2010), "Assessment of a new laboratory die pelleting rig attached to a texture analyzer to predict process-ability of wood pellets. Energy consumption and pellet strength.", *Annual Transactions of the Nordic Rheology Society*, **18**: p. 77 - 85.

15. Schulze, D., (2013), "Shear Testing of Powdera for Process Optimization", *Annual Transactions of the Nordic Rheology Society*, **21**: p. 99 - 106. 16. Figura, L.O. and A.A. Teixeira, (2007) "Food Physics Physical Properties -Measurement and Applications": Springer -Verlag Berlin Heidelberg

17. Pietsch, W., (2002) "Agglomeration Processes - Phenomena, Technologies, Equipment". Weinheim: Wiley - VCH Verlag GmbH

18. Nielsen, N.P.K., L. Nørgaard, B.W. Strobel, and C. Felby, (2009), "Effect of storage on extractives from particle surfaces of softwood and hardwood raw materials for wood pellets", *European Journal of Wood and Wood Products*, **67**(1): p. 19-26.

19. Naidich, Y.V., I.A. Lavrinenko, and V.Y. Petrishchev, (1965), "Study on the capillary adhesive forces between solid particles with a liquid layer at the points of contact", *Soviet Powder Metallurgy and Metal Ceramics*, **4**(2): p. 129-133.

20. Esteban, B., J.-R. Riba, G. Baquero, R. Puig, and A. Rius, (2012), "Characterization of the surface tension of vegetable oils to be used as fuel in diesel engines", *Fuel*, **102**(0): p. 231-238.

21. Nielsen, N., D. Gardner, T. Poulsen, and C. Felby, (2009), "Importance of Temperature, Moisture Content, and Species for the Conversion Process of Wood Residues Into Fuel Pellets", *Wood and Fiber Science*, **41**(4): p. 414-425.