# MECHANICAL MATERIAL PROPERTIES EFFECT ON PELLETIZATION

Lucie Jezerska, Ondrej Zajonc, Jan Vyletelek, Jiri Zegzulka Vsb-Technical University of Ostrava, Enet Centre Ostrava, Czech Republic

(Received July 2015)

# ABSTRACT

The relation between the mechanical properties of input materials and the smoothness of material flow from a storage bin, cohesion variability of the compressed powder mixtures, and pelletization process were studied. Three material types were examined: Pyrolysis char from biomass – spruce wood (*Picea abies* L.), compost and spruce sawdust. Increased input material compressibility and cohesion affected the resulting durability and hardness of the pellets. Additional important material parameters affecting the pelletization process and final pellet quality include flowability and wall friction angle: Pellet durability and hardness increases with decreasing flowability (shift to the cohesive materials mode) and wall angle of the incoming materials. Those parameters are taken into account when designing conveyors. Application of the Quality by Design (QbD) approach to the prediction of behaviour in the pelletization process is outlined. The feasibility of inferring acceptable pelletization process behaviour from the mechanical-physical properties of the input materials is demonstrated.

KEYWORDS: Mechanical properties, flowability, pelletization, Quality by Design (QbD).

# **INTRODUCTION**

The conditions of the pelletization of materials have been investigated by a number of authors, who largely focussed on the mechanical - physical properties, major components of the biomass (lignin, cellulose, etc.), and chemical composition of the materials for pelletization. Although the named properties are important with consideration to the material pelletization process, these properties can be assessed only from a physical point of view. Therefore, the mechanical-physical properties were taken into account with respect to the processing of selected materials (Gil et al. 2014). The present work was aimed at assessing those mechanical-physical properties of materials (flowability, wall friction, compressibility and cohesion) that play an important role in the pelletization process. Such mechanical-physical properties of bulk materials are also important as regards the design of engineering structures coming in contact with such

materials. It is appropriate to analyse the relation between the engineering structure and the material from the force action point of view. Due to the forces acting on them and to the existing boundary conditions of the system, materials alter their behaviour based on their mechanical-physical properties. When considering pelletization as a process of material transport through a die, we find that the bulk material's properties have transformed.

Mechanical-physical properties of materials play a key role in the examination of the use of conveying and processing facilities in the complex logistic process. The pelletization process itself can be looked upon as a logistic element where the mechanical-physical properties of the output material are different from those of the input raw material. As a result of the compression change in the volume of the particles and their morphology, the behaviour of the material along the transport routes changes substantially (Rudolfsson et al. 2015) and hence, different equipment must be used at the input and at the output. Arching and blocking can occur when transporting the material, for example through a conveyor system or during discharging from a silo (Chen et al. 2012, Chevanan et al. 2009, Mellmann et al. 2014, Miccio et al. 2013, Oldal et al. 2012). Transport through conveying systems is affected not only by the type and parameters of the biomass but also by the particle size and moisture content of the material (Gil et al. 2013).

The internal friction angle is one of the primary properties of powders, determining friction which exists between the particles, and one of the major parameters describing the flow properties of the material (Zegzulka 2012). For instance, the angle of internal friction will be reduced appreciably by cutting the biomass into pieces (Ileleji and Zhou 2008).

The wall friction angle is a parameter determining the particle behaviour in contact with various surfaces (Wu et al. 2011). Hence, this is a parameter that must be taken into account in the design of silos, hoppers, etc. (Ganesan et al. 2008; Iqbal and Fitzpatrick 2006).

The flow properties of biomass particles depend on the biomass type and on the particle shape and size (Miao et al. 2014). It has been found that herbal biomass particles such as switchgrass, wheat straw and corn stover, 7 - 15 mm particle size, cannot be transported based solely on gravity (Chevanan et al. 2009, 2010).

The feasibility of pelleting materials has been studied by many authors. The major parameters reported to affect the final pellet quality include grain size (Serrano et al. 2011), moisture content (Li, et al. 2012, Serrano et al. 2011), and the amount of binders in the material (Kaliyan and Morey 2010, Stelte et al. 2011, Telmo and Lousada 2011). Of major importance are also material pretreatment procedures (steam pretreatment (Biswas et al. 2011, Lam et al. 2013), Ammonia Fiber Expansion (AFEX) pretreatment (Hoover et al. 2014)), conditions of the pelletization process itself (temperature, pressure, material holdup time in the pellet press die) and of the subsequent pellet cooling procedure (Morey and Vance 2008, Obernberger and Thek 2010).

The present work was aimed at examining the effect of the mechanical properties of the materials on the quality of the pellets too. Important mechanical properties of pellets determining their quality include durability, hardness, water resistance and density.

Durability expresses how resistant pellets are to fragmentation during handling, transportation and storage (Obernberger et al. 2006, Thomas and van der Poel 1996, Vinterbäck 2004). The separated pieces (fragment) of the material may block conveyor routes and disturb the homogeneity of the combustion conditions; also, the dust dispersed in air is harmful (Vinterbäck 2004) and may form explosive mixtures (Hedlund et al. 2014, Lehtikangas 2000).

Hardness is the resistance of the individual pellets during handling, transport and storage (Jiang et al. 2014). It is related to the fragmentation of the pellets (Thomas and van der Poel 1996).

The density of the pelletized material is a measure of densification. Together with durability, density is a primary pellet quality indicator (Temmerman et al. 2006).

Water resistance is a parameter which is important in the transport and storage of the pellets. Moisture absorption reduces the calorific value and may induce mouldering and microbial decomposition resulting in pellet decay (Chico-Santamarta et al. 2011). Increased moisture may bring about auto-ignition of the stored pellets (Obernberger et al. 2006).

# MATERIAL AND METHODS

# Materials

Three types of material were used: pyrolysis char from wood biomass, compost with spruce saw dust, and spruce saw dust alone (Fig. 1-4).



Fig. 1. Pyrolysis char before crushing.



Fig. 3. Crushed compost.



Fig. 2. Pyrolysis char after crushing.



Fig. 4. Spruce sawdust.

# Pyrolysis char from biomass

Pyrolysis char from wood biomass was prepared in a Pyromaniac pyrolysis unit. Spruce pellets were pyrolyzed at 550°C for 45 minutes (Frantik et al. 2013). The resulting pyrolysis char failed to attain the optimum particle composition, the majority of the material disintegrated after pyrolysis but lumps of the pyrolysis char remained and hindered mixing the pyrolysis char with the binder. To avoid this, the pyrolysis char was crushed to smaller particles and homogenized. Pyrolysis char is hydrophobic and contains 2.37 % water. Containing no binder, the pyrolysis char was pelletized with an addition of 20 % starch. The amount of added starch is not according to Standard EN 14961-1 (2010). For the pelletization, the mixture had a moisture content of 17 %.

## Compost

The compost was obtained from the compost plant in Namest nad Oslavou. The compost was after fermentation (90 days maturation). The compost was dried from the initial 48.72 % moisture content to 10 - 15 % and crushed on a Green Energy 9FQ 50 hammer mill. In order to increase its calorific value, the compost was combined with wood saw dust before pelletization.

The pelletized mixture had a moisture content of 25 %.

## Spruce sawdust

Spruce sawdust was dried to 13 % moisture for pelletization. No binders were added.

# Testing the materials' mechanical and flow properties

The materials were tested on a FT4 Powder Rheometer (Freeman Technology) by using various measuring methods. This rotary rheometer serves to describe the flow properties of mixtures based on dynamic measurements. The rheometer includes a shear cell for shear force measurement, a wall friction kit and accessories for measuring the bulk material parameters such as density, compressibility and permeation.

The rotary rheometer consists of a measuring vessel, blade on a shaft and aeration station with a control unit. The measuring vessel is made from a precisely bored borosilicate 50 mm glass tube with flame-polished walls. The blade, 48 mm in diameter, was made of quenched steel. A 1 mm gap was left between the blade edge and the measuring vessel wall.

A shaft with the specifically shaped blade is pushed through the powder bed. The resulting motion of the blade fixed on the shaft is a specific combination of rotary and axial motion, resulting in a predefined helical motion which in turn defines the flow rate and flow type. The motion of the blade induces forces causing powder deformation and flow, which are measured and used to calculate the energy required to bring the powder in motion (flow energy). The rotary and axial motions can be combined in various ways giving rise to different modes of action. Two are shown in (Fig. 5).



Fig. 5: Left: pushing mode, right: Raising mode (Freeman 2007).

The descending, pushing mode shown on the left in (Fig. 5) (in which the blade rotates counter clockwise through the powder) results in powder compaction and large flow. In the ascending, raising mode (in which the blade rotates clockwise through the powder) the degree of compaction is low.

## Flowability and wall friction

The rotary shear module for measuring the friction parameters and subsequently the flowability consists of a vessel containing the powder sample and a shear head inducing both normal and shear stresses. The shear head blades are pushed into the powder mass and the front face of the head starts exerting normal stress on the powder bed surface. The shear head is pushed downwards until an adequate and steady pressure establishes between the head and the powder bed. Now the shear head starts rotating slowly, thereby exerting shear stress in the bulk mass. In this manner a shear plane is created immediately below the blade edges. The powder bed resists the shear head rotation, whereby shear stress is recorded. Flowability is calculated as the ratio of the main stress under which the powder is consolidated to the powder's shear strength. Powder flowability was categorized based on the Jenike flow function (Leturia et al. 2014).

The rotary shear module for the wall friction angle resembles that described above. The contact plane of the shear head (which is made of stainless steel, although other materials can be used as well) travels down to the powder sample and its front face starts exerting normal stress onto the surface and continues to do so until a steady state is attained. Now a shear stress is induced and is increased until slip.

## Compressibility

Compressibility is measured as volume (or density) change in dependence on the normal load. The data are expressed in terms of percent compressibility for a 15 kPa normal load created by a module which is a component of the FT4 powder rheometer.

#### Cohesion

Cohesion was measured through the basic flow energy, which is the energy required to create a flow profile of the powder conditioned by an identical volume. This is calculated from the work done by the blade during its motion through the powder bed, consisting in rotation combined with axial down motion and simultaneous compaction. The values are higher for freely flowing materials and lower for cohesive materials that are easy to compact.

## Pelletization

The materials were pelletized on a KAHL 14 - 175 laboratory pelleting press with a flat die. One and the same die was used in identical conditions for all the pellets. As a consequence, the properties of the pellets were not optimal – in fact, different dies are used in practice for different materials to obtain the best pellets.

# Mechanical properties measurements of the pellets

#### Durability

Durability was determined by alternative procedure according to the following testing instruction with the Holmen NHP 100 tester. In the test, a 100 g  $\pm$  0.5 g pellet sample circulates pneumatically for 60 seconds in a chamber with perforated walls, having the shape of an inversed pyramid. The pellets tumble, colliding and hitting the chamber walls and themselves, and powder and broken-off pieces of the pellets emerge ((Kaliyan and Morey 2009). When the test is over, the sample is sieved on a sieve 3.15 mm mesh. The pellets are weighed and the abrasion in % is calculated. The alternative mechanical durability expressed in term of the Pellet Durability Index (PDI) is 100 % - AR (Obernberger and Thek 2010):

$$AR = \frac{m_E - m_A}{m_E} .100 \tag{1}$$

where: AR - abrasion in weight percent,

 $m_E$  - mass of pellets before treatment,

 $m_A$  - mass of pellets after treatment in grams.

This procedure is performed in five replicate tests and the mean value, rounded to one decimal place, is used in the calculation.

#### Hardness

Hardness of the pellets was expressed in terms of the weight load, in kg, that the pellet withstands without breaking or crushing. A KAHL ak - 14 tester was used to determine the hardness. In the test, the pellet is placed between a steady plane and a flat tip which is

progressively compressed by a spring simulating the weight load. The flat tip is pressed down until the pellet is broken or crushed. The load is then read on a scale. Ten replicate tests are performed and the mean value is used to determine the hardness level.

#### Density

Density of the pelletized material is a measure of densification. A Mettler Toledo JEW-DNY-43 density tester was used to determine the density of the pellets based on Archimedes' law, i.e. on the weight difference between the pellets in air and submerged in distilled water with a small amount of a surfactant. A minimum of 4 pellets are measured in each test. The procedure was repeated ten times to obtain the mean value.

#### Water resistance

Water resistance is expressed by the wettability index (WI). The test consists in submerging the pellet into distilled water for 30 seconds and measuring the difference between the pellet weights before the test,  $m_1$ , and after the test,  $m_2$ . The wettability index is the percent weight increment relative to the initial pellet weight, which is the relative absorbed amount of water (Lindley and Vossoughi 2008):

$$WI = \frac{m_2 - m_1}{m_1} \times 100 \ (\%) \tag{2}$$

The test is repeated ten times and the mean value is used to calculate the wettability index.

## Quality by Design (QbD)

Quality by Design (QbD) is methodology that proposes change in the quality management approach by testing for the approach, quality being directly integrated into the product design and manufacturing process (Freeman et al. 2015). QbD relies on the ability to describe how the powder will behave during the process (i.e. pelletization), and makes use of the possibility to directly measure those mechanical - physical properties that are relevant to the process conditions. Since powders have been processed by the industries for many years now, an extensive knowledge base exists as regards the quality of various powders, bulk materials and their mixtures and their suitability for various industrial processes (Valentine 2013).

# **RESULTS AND DISCUSSION**

Mechanical - physical properties of the materials are listed in Tab. 1. Compressibility and cohesion were found to increase in order: pyrolysis char < compost < spruce sawdust. The wall friction angle and flowability which was determined according to Jenike flow function (Leturia et al. 2014), decreased in that order. The grain size distribution of the materials for pelletization is given in Tab. 2. The smallest grains contained starch ( $d_{90} = 0.650$  mm), which was used as a binder for pelletizing pyrolysis char, of which 90 % of the represention particles is very similar, i.e. 0.666 mm (see chapter 2.1.1). The impact of the granulometry of input raw pelletized material has an effect on the resulting mechanical properties of the pellets (Sultan et al. 2010, Kaliyan and Morey 2009). From a practical point, the use of material with a wider distribution of particulate size is preferable (satisfied for all samples, see Tab. 2). This ensures the mechanical resistance of pellets formed by bonds between the particles and filling the space between coarse particles with fine ones. A larger amount of fine particles, however, may cause cracking due to water absorption

by a relatively larger specific surface area. It is also preferable for this reason to use, if possible, a wider distribution of particulate sizes of the input material.

Pellet hardness and durability (Tab. 3) increased in parallel to compressibility and cohesion. Pyrolysis char pellets containing some amount of starch were more durable than the compost or spruce sawdust pellets. This was due to the presence of starch which plays the role of a binder in the pelletization process, thereby enhancing durability of the final pellets. Pelletization of pyrolysis char from wood biomass alone, without any starch, is very problematic. As found before (Zajonc 2012), the durability of pellets made from pure pyrolysis char is about 70 %. This is why durability was related to the increasing compressibility and cohesion tendency. The highest durability was found for the spruce sawdust pellets, which bears out the favourable effect of the binders present (i.e. lignin) on the durability of the pellets. The mechanical and flow properties of the materials are listed in Tab. 2 while the mechanical properties of the pellets are included in Tab. 3. The results of ultimate and proximate analysis are given in Tab. 4 as auxiliary data. As mentioned in the introduction, an important parameter for pelletization is also the humidity of the input material (Obernberger et al. 2006). Biomass with a lower moisture content will comprise from 5 - 10 % more compact pellets than biomass with 15 % moisture content (Mani et al. 2006). This prerequisite was met for further work. Calorific values were in a wide range from 6.6 -29.5 kJ. kg<sup>-1</sup>. The chemical composition (C, H, N, S) of a material generally determines the final properties of the fuel, for example, the N content is responsible for the formation of NOx, etc. The data in Tab. 4 are particularly informative. The aim of the paper is mainly to outline the QbD (Quality by Design) methodology, which would facilitate pelletizing dissimilar materials based on information about their mechanical - physical properties, as mentioned below.

Sample	Pyrolysis char from biomass	Compost	Spruce sawdust
Wall friction angle (%)	$19.6 \pm 0.2$	$10.05\pm0.1$	$7.8\pm0.1$
Flowability -	10.6	6.7	3.3
Flowability classification	Easy-flowing	Free-flowing	Cohesive
Compressibility factor (%)	$9.50 \pm 0.02$	$10.23\pm0.04$	$19.20\pm0.07$
Cohesion (kPa)	$0.74 \pm 0.01$	$1.4 \pm 0.01$	$1.72\pm0.01$

Tab. 1: Mechanical - physical properties of the raw materials.

Grain size	Pyrolysis char	Pyrolysis char after	Starch	Compost	Spruce sawdust
(mm)	before crushing (%)	crushing (%)	(%)	(%)	(%)
< 0.045	0.135	26.48	9.63	17.91	2.11
0.045-0.063	1.282	4.87	3.21	4.19	1.2
0.063-0.16	5.112	18.41	10.33	15.48	10.15
0.16-0.25	4.371	13.73	15.29	13.03	17.05
0.25-0.5	9.055	20.46	39.7	23.71	39.78
0.5-1.0	7.619	11.93	20.89	17.88	24.79
1.0-1.4	4.364	2.89	0.94	5.32	3.79
1.4-2.0	5.340	1.23	0.01	2.48	1.13
2.0-2.8	6.853	0	0	0	0
2.8-3.15	6.054	0	0	0	0
> 3.15	49.816	0	0	0	0

Tab. 2: Grain size distribution of the materials before and after crushing for pelletization.

d 10	0.140	0.011	0.047	0.021	0.135
d 50	2.776	0.160	0.313	0.247	0.356
d 90		0.666	0.650	0.898	0.801

Tab. 3: Mechanical properties of the pellets.

Sample	PDI (%)	Hardness (kg)	WI (%)	Density (kg.m <sup>-3</sup> )
Pyrolysis char pellets	97.4	39.0	4.2	1.250
Compost with sawdust pellets	96.3	43.6	26.6	1.403
Wood pellets	98.8	57.2	18.8	1.386

Tab. 4: Ultimate and proximate analysis (Laboratory of Geologic Engineering, VSB – TU Ostrava).

Sample	Pyrolysis char	Starch	Compost	Spruce sawdust
Moisture (%)	2.37	9.69	4.05	7.48
Ash (dry matter) (%)	9.81	0.21	61.24	0.45
Volatile flammables (dry matter) (%)	19.83	98.51	34.61	89.32
Fixed carbon (dry matter) (%)	70.37	1.89	4.15	10.23
Combustion heat (dry matter) (kJ.kg <sup>-1</sup> )	30.202	15.006	7.378	18.611
Calorific value (dry matter) (kJ.kg <sup>-1</sup> )	29.500	12.930	6.662	17.198
Calorific value (wet matter) (kJ.kg <sup>-1</sup> )	28.740	11.433	2.226	15.739
C (dry matter) (%)	85.970	43.944	17.85	47.67
H (dry matter) (%)	3.409	10.077	3.31	6.86
N (dry matter) (%)	0.165	0.196	1.69	0.13
S (dry matter) (%)	< 0.01	< 0.01	< 0.01	< 0.01

The QbD methodology, which is fostered mainly in pharmaceutical engineering, was outlined for a proposal of prediction of the process behaviour (pelletization) of the diverse materials (Freeman et al. 2015). Pelletization of bulk materials is a widely used process. The frequent cases during the production of pellets are process problems arising with respect to being a new alternative material. The common technologists question therefore is: "Will this new material process well in existing pellet press?" or "Why do some batches of material process well when others are so difficult?". The below proposed methodology can call attention to problematic materials or define the problematic process parts. Pellet production consists of several steps from drying via interim storage of materials to packing pellets. Also pelleting process itself can be divided into at least four discrete processes:

- discharge of raw materials from the hopper,
- flow into and through the feed frame,
- die filling and compression,
- cutting of pellets.

Each of these processes subjects the bulk materials to a specific set of conditions for example flow rates, stresses, equipment surface properties etc., making different mechanicalphysical properties more significant at different stages. In this paper are highlight the following as especially substantial. The first properties are dynamic flow properties (including basic flowability energy) to optimise the flow regime in the feed frame and the efficiency of die filling or to assess the likehood of attrition, segregation. The second determinations are shear properties for optimising flow from the feed hopper, where shear properties of powder-wall are important (influenced by abrasion, corrosion, moisture) and last but not least are compressibility and aeration for assessing how easily the powder can transmit air and the impact of compression of bulk materials. Both last characteristics are important during the filling and compression steps.

Among the basic process problems of pelletization can belong:

- intermittent material flow from the storage container,
- · variability in the degree of cohesion of the powder mixtures compressed,
- variability of pellet durability and hardness.

If all powder materials are described and their flow properties input to a dedicated database, a relation between the flowability and the process behaviour may be deduced. This will enable the extensive know - how which currently exists to be quantified.

The following basic process problems and mechanical-physical properties of raw materials, which may influenced them, listed in Fig. 6, was defined for the design of the pelletization methodology. For the purpose of outlining the methodology have been selected these three different biomass (sawdust, compost and pyrolytic carbon) that exhibited during the pelletization process running in the same device mentioned process problems (Fig. 6).

Process problem	Sawdust	Compost	Pyrolysis char
Intermittent flow from the storage	bin YES	NO	NO
Variable cohesion of the mixtures	NO	NO	YES
Variable PDI of the pellets	NO	YES	NO
Thich mechanical physical propertie	es of the incoming	natorial are rold	nyant?
Thich mechanical-physical propertion	es of the incoming i	naterial are rele	want?
<i>Thich mechanical-physical propertion</i> <u>Intermittent flow from the storage bin?</u> Angles of internal friction and wall	es of the incoming i Cohesion variability Permeability	naterial are rele	evant? Pellet PDI variability? Compressibility and col
<i>Thich mechanical-physical propertion the storage bin?</i> Angles of internal friction and wall friction and wall triction	es of the incoming r Cohesion variability Permeability Aeration	naterial are rele	evant? <u>Pellet PDI variability?</u> Compressibility and col Wall friction angle

Fig. 6: Definition of the pelletization process parameters and the associated physical properties.

Based on the identification of the key variables affecting each specific process problem, bulk materials can be described in relation to those process problems and rated with respect to them. A scale from 0 to 10 is used (for simplicity and ease of use in the industry), 0 denoting the most serious problem in relation to the specific bulk material and 10 denoting no problem of that type at all.

This means that if the pelletization process has been stopped due to a significant arching of material in the hopper, this bulk material obtains when evaluating value of 0. On the contrary, other material, which does not cause problem in the operation obtains when evaluating the value of the 10. Furthermore, the specific flow properties affecting the occurrence of a given problem (see Fig. 6) are measured (Valentine 2013). Sawdust was associated with the non - uniform storage bin discharging problem; pyrolysis coal was associated with the cohesion problem; and compost was associated with the poorer pellet durability problem. As a result, a set of parameters indicating potential problems in a particular criterion is obtained. For instance, pellet durability, expressed alternatively through the pellet durability index (PDI), is markedly affected by compressibility, cohesion, and wall friction angle in relation to the pelletization process. Based on the relation between the process behaviour and the mechanical - physical (flow) properties, relative acceptable process behaviour can be defined for each of the properties. In other words, the measurements allow us to define a range of mechanical - physical properties within which the bulk material will behave as desired in the given process environment. This must be performed for each segment of the process separately.

Now, based on the example from the pelletization process area where we attempted to outline

the use of the QbD methodology, acceptable behaviour identified from the tests described in Tabs. 1 - 4 can be defined specifically for pellet durability variability. Compressibility as the first of the parameters affecting variability of the pellet PDI (Fig. 6) is a measure of how the volume/ density will change in dependence on normal load -15 kPa in this case.



Fig. 7: Acceptable process behaviour (region indicated by the red arrow) of the input materials for the pelletization process.

The figure for compressibility shows that acceptable behaviour occurs in the upper segment starting from a compressibility factor value of 4.1. Spruce sawdust was assigned the value of 8 indicating a relatively low probability of the pellet durability variability problem.

In other words - because production of spruce sawdust in relation to the mechanical durability of the pellets was (compared with other materials) almost trouble-free, the mean value of the measured compressibility factor 19.20 (Tab. 1) is assigned the value 8 ratings. Pyrolysis char, on the contrary, will require mechanical, physical or chemical treatment to overcome the limit of acceptable behaviour in all the relevant parameters. (To the average value of the pyrolysis char low compression factor 9.5 is assigned 3.96). As mentioned above, addition of binders and moisture is required for the pelletization of pyrolysis char. The values of acceptable process behavior have been established for two values of alternatively PDI. The first value for PDI is 97.5 % (PDI A) according to the EN 14961-2 (2011) Standard i.e Solid biofuels-fuel specifications and classes-part 2: Wood pellets for non-industrial use. The second value is 96.0 % (PDI B) for non-woody pellets according to the EN 14961-6 (2012). The graphs show (Fig. 7) that acceptable behavior for higher value of PDI (97.5 %) would meet only pellets made from spruce sawdust, other input materials have to be adjusted.

However, a larger volume of data, allowing us to identify statistical correlations and hence, make more precise predictions, must be gathered to enable more detailed conclusions following from the QbD approach to be drawn. Validation of the method, permitting this approach to be applied in the industry, should follow as the next step in this context.

As mentioned above, this approach may constitute a powerful tool in the product (pellet) design development stage as well as in the production process (pelletization) stage. Its application may save appreciable costs that would otherwise be associated with the replacement of unsuitable production equipment because of poor properties of the product. This method also helps us gain a deeper insight into the pelletization process and its relations to the material being pelletized.

# **CONCLUSIONS**

The aim of the presented work was a mechanical – physical characterization of different raw materials, biomass, which is suitable for pelletization in connection with subsequent process behaviour and final quality of pellets. On the base of obtained data it was outlined the proposal the QbD application to pelletization process of biomaterials i.e. the prediction of material process behaviour on the base of the measured mechanical – physical properties.

Three types of material were investigated: Pyrolysis char, compost, and spruce sawdust. The parameters that affected the pelletization of the materials the most included the compression factor and the cohesion parameter: Materials with the highest compression factor and cohesion provided the hardest and most durable pellets. Durability is appreciably affected by compressibility, cohesion and wall friction. As a simplifying assumption, granulometry and particle shape as material parameters were included in the effect of cohesion. The binder and water contents of the pelletized materials also affected their mechanical properties, particularly for hydrophilic materials. The optimum binder content cannot be precisely determined, the proposed methodology can only predict binder adding. This issue will be the subject of forthcoming studies.

Generally, durability of the pellets increases with decreasing wall friction and flowability and with increasing cohesion and the compressibility factor. From among the 3 types of material, most suitable from the flowability aspect was pyrolysis char (an easy - flowing material), followed by compost, while spruce sawdust exhibited the poorest flowability.

The Quality by Design (QbD) concept, which is mainly applied in other fields of science and technology, was outlined as a tool showing promise for predicting the process behaviour of the materials. This approach appears to be a suitable tool to ensure smooth production of quality pellets from diverse materials, characterized by their basic mechanical - physical properties. To make it practical, a large volume of data for different materials must be collected to obtain statistically grounded dependences (correlations) that could be validated and then used by engineers in practical everyday activities.

## ACKNOWLEDGMENTS

This paper has been elaborated in the framework of the project New creative teams in priorities of scientific research, reg. no. CZ.1.07/2.3.00/30.0055, supported by Operational Programme Education for Competitiveness and co-financed by the European Social Fund and the state budget of the Czech Republic.

This paper was supported by research projects of the Ministry of Education, Youth and Sport of the Czech Republic: LO1404 - Sustainable Development of Centre ENET.

# REFERENCES

- 1. Biswas, A.K., Yang, W., Blasiak, W., 2011: Steam pretreatment of *Salix* to upgrade biomass fuel for wood pellet production. Fuel Processing Technology 92(9): 1711-1717.
- 2. Chen, P., Yuan, Z., Shen, X., Zhang, Y., 2012: Flow properties of three fuel powders. Particuology 10(4): 438-443.

- Chevanan, N., Womac, A.R., Bitra, V.S.P., Yoder, D.C., Sokhansanj, S., 2009: Flowability parameters for chopped switchgrass, wheat straw and corn stover. Powder Technology 193(1): 79-86.
- Chevanan, N., Womac, A.R., Bitra, V.S.P., Igathinathane, C., Yang, Y.T., Miu, P.I., Sokhansanj, S., 2010: Bulk density and compaction behavior of knife mill chopped switchgrass, wheat straw, and corn stover. Bioresource Technology 101(1): 207-214.
- Chico-Santamarta, L., Humphries, A.C., Chaney, K., White, D.R., Magan, N., Godwin, R.J., 2011: Microbial changes during the on-farm storage of canola (oilseed rape) straw bales and pellets. Biomass & Bioenergy 35(7): 2939-2949.
- 6. Frantik, J., Pavlik, P., Schee, Z., Zapletal, P., 2013: Pyrolysis char from woody biomass pyrolysis unit Pyromatic tests. VSB Technical University of Ostrava, Ostrava (in Czech).
- Freeman, R., 2007: Measuring the flow properties of consolidated, conditioned and aerated powders – A comparative study using a powder rheometer and a rotational shear cell. Powder Technology 174(1-2): 25-33.
- Freeman, T., Birkmire, A., Armstrong, B., 2015: A QbD approach to continuous tablet manufacture. In: New paradigm of particle science and technology proceedings of the 7<sup>th</sup> world congress on particle technology. Procedia Engineering 102: 443-449.
- Ganesan, V., Rosentrater, K.A., Muthukumarappan, K., 2008: Flowability and handling characteristics of bulk solids and powders – a review with implications for DDGS. Biosystems Engineering 101(4): 425-435.
- Gil, M., Teruel, E., Arauzo, I., 2014: Analysis of standard sieving method for milled biomass through image processing. Effects of particle shape and size for poplar and corn stover. Fuel 116: 328-340.
- Gil, M., Schott, D., Arauzo, I., Teruel, E., 2013: Handling behavior of two milled biomass: SRF poplar and corn stover. Fuel Processing Technology 112: 76-85.
- Hedlund, F.H., Astad, J., Nichols, J., 2014: Inherent hazards, poor reporting and limited learning in the solid biomass energy sector: A case study of a wheel loader igniting wood dust, leading to fatal explosion at wood pellet manufacturer. Biomass & Bioenergy 66: 450-459.
- Hoover, A.N., Tumuluru, J.S., Teymouri, F., Moore, J., Gresham, G., 2014: Effect of pelleting process variables on physical properties and sugar yields of ammonia fiber expansion pretreated corn stover. Bioresource Technology 164: 128-135.
- Ileleji, K.E., Zhou, B., 2008: The angle of repose of bulk corn stover particles. Powder Technology 187(2): 110-118.
- 15. Iqbal, T., Fitzpatrick, J.J., 2006: Effect of storage conditions on the wall friction characteristics of three food powders. Journal of Food Engineering 72(3): 273-280.
- Jiang, L., Liang, J., Yuan, X., Li, H., Li, Ch., Xiao, Huang, H., Wang, H., Zeng, G., 2014: Co-pelletization of sewage sludge and biomass: The density and hardness of pellet. Bioresource Technology: 166: 435-443.
- Kaliyan, N., Morey, R.V., 2010: Natural binders and solid bridge type binding mechanisms in briquettes and pellets made from corn stover and switchgrass. Bioresource Technology 101(3): 1082-1090.
- Kaliyan, N., Morey, R.V., 2009: Factors affecting strength and durability of densified biomass products. Biomass & Bioenergy 33(3): 337-359.
- Lam, P.S., Lam, P.Y., Sokhansanj, S., Bi, X.T., Lim, C.J., 2013: Mechanical and compositional characteristics of steam-treated Douglas fir (*Pseudotsuga menziesii* L.) during pelletization. Biomass & Bioenergy 56: 116-126.

- 20. Lehtikangas, P., 2000: Storage effects on pelletised sawdust, logging residues and bark. Biomass & Bioenergy 19(5): 287-293.
- Leturia, M., Benali, M., Lagarde, S., Ronga, I., Saleh, K., 2014: Characterization of flow properties of cohesive powders: A comparative study of traditional and new testing methods. Powder Technology 253: 406-423.
- 22. Li, H., Liu, X., Legros, R., Bi, X.T., Lim, C.J., Sokhansanj, S., 2012: Pelletization of torrefied sawdust and properties of torrefied pellets. Applied Energy 93(C): 680-685.
- 23. Lindley, J.A., Vossoughi, M., 2008: Physical properties of biomass briquettes. Transactions of the ASAE, American Society of Agricultural Engineers 32: 2-5.
- 24. Mani, S., Tabil L.G., Sokhansanj, S., 2006: Specific energy requirement for compacting corn stover. Bioresource Technology 97(12): 1420-1426.
- Mellmann, J., Hoffmann, T., Fürll, Ch., 2014: Mass flow during unloading of agricultural bulk materials from silos depending on particle form, flow properties and geometry of the discharge opening. Powder Technology 253(1): 46-52.
- 26. Miao, Z., Grift, T.E., Hansen, A.C., Ting, K.C., 2014: Flow performance of ground biomass in a commercial auger. Powder Technology 267: 354-361.
- Miccio, F., Barletta, D., Poletto, M., 2013: Flow properties and arching behavior of biomass particulate solids. Powder Technology 235: 312-321.
- Morey, N.K., Vance, R., 2008: Densification of biomass: Mechanisms, models, and experiments on briquetting and pelleting of biomass. Saarbrücken, Germany: VDM Verlag Dr Müller, ISBN 978-363-9051-858.
- 29. Obernberger, I., Thek, G., 2010: The pellet handbook: The production and thermal utilisation of pellets. Washington. DC: Earthscan, XXXII, 549 pp. ISBN 18-440-7631-8.
- Obernberger, I., Brunner, T., Barnthaler, G., 2006: Chemical properties of solid biofuels significance and impact. Biomass & Bioenergy 30(11): 973-982.
- Oldal, I., Keppler, I., Csizmadia, B., Fenyvesi, L., 2012: Outflow properties of silos: The effect of arching. Advanced Powder Technology 23(3): 290-297.
- Rudolfsson, M., Stelte, W., Lestander, T.A., 2015: Process optimization of combined biomass torrefaction and pelletization for fuel pellet production – A parametric study. Applied Energy 140: 378-384.
- Serrano, C., Monedero, E., Lapuerta, M., Portero, H., 2011: Effect of moisture content, particle size and pine addition on quality parameters of barley straw pellets. Fuel Processing Technology 92(3): 699-706.
- 34. Stelte, W., Holm, J.K., Sanadi, A.R., Barsberg, S., Ahrenfeldt, J., Henriksen, U.B., 2011: A study of bonding and failure mechanisms in fuel pellets from different biomass resources. Biomass & Bioenergy 35(2): 910-918.
- Sultana, A., Kumar, A., Harfield, D., 2010: Development of agri-pellet production cost and optimum size. Bioresource Technology 101(14): 5609-5621
- Telmo, C., Lousada, J., 2011: The explained variation by lignin and extractive contents on higher heating value of wood. Biomass & Bioenergy 35(5): 1663-1667.
- Temmerman, M., Rabier, F., Jensen, P., Hartmann, H., Bohm, T., 2006: Comparative study of durability test methods for pellets and briquettes. Biomass & Bioenergy 30(11): 964-972.
- Thomas, M., van der Poel, A.F.B., 1996: Physical quality of pelleted animal feed. 1. Criteria for pellet quality. Animal Feed Science and Technology 61(1-4): 89-112.
- Valentine, N.E., 2013: Optimizing tableting processes with quality by design: An overview. International Journal of Advances in Pharmaceutics 2: 12-16.

- 40. Vinterback, J., 2004: Pellets 2002: The first world conference on pellets. Biomass & Bioenergy 27(6): 513-520.
- 41. Wu, M.R., Schott, D.L., Lodewijks, G., 2011: Physical properties of solid biomass. Biomass & Bioenergy 35(5): 2093-2105.
- 42. Zajonc, O., 2012: Research of the environmental properties of pellets. Dissertation, VSB Technical University of Ostrava, Czech Republic. Ostrava (in Czech).
- 43. Zegzulka, J., 2012: The angle of internal friction as a measure of work loss in granular material flow. Powder Technology 233: 347-353.

Lucie Jezerska\*, Ondrej Zajonc, Jan Vyletelek, Jiri Zegzulka VSB-Technical University of Ostrava Enet Centre 17. Listopadu 15 708 33 Ostrava Czech Republic Phone: +420 597 329 430 Corresponding author: lucie.jezerska@vsb.cz