Process optimization of combined biomass torrefaction and pelletization for fuel pellet production – A parametric study

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Abstract

Torrefaction of plant biomass has the capacity to produce a fuel with increased energy density and homogeneity, but there are reports that it changes the pelletizing properties of the biomass, making it more difficult to obtain high quality pellets. A parametric study was therefore conducted in which three key qualitative parameters, degree of torrefaction (250-300°C), moisture content (0-10%) and pelletizing temperature (125-180°C), were varied according to a five level fractional factorial design, also including particle size as a qualitative parameter. Pelletizing at 300 MPa (pellet densities: 1.0-1.2 mg/mm^3) was undertaken using a single pellet press and the responses recorded were compression work (W_{comp}), maximal force to overcome static friction (F_{max}), kinetic friction work (W_{fric}), single pellet dimensions and strength. Small particles had reduced W_{comp} and F_{max}, but increased strength. As expected, all other parameters also had significant effects. In general, less energy was required for W_{comp}, W_{fric} and F_{max} at lower degrees of torrefaction and higher moisture contents and when pelletizing was conducted at higher temperatures. The process window to optimize pellet strength was narrow and, surprisingly, somewhat higher moisture content at higher degrees of torrefaction increased strength. This narrow production window in combination with feedstock variations may, in practical pelletizing situations, result in varying quality. Furthermore, the study illustrates that factorial experiments using single-pellet devices provide new insights that are of importance for the next generation of pelletizing of torrefied biomass.

Key words: Densification, thermotreated, wood.

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Introduction

Heat and power production from renewables such as lignocellulosic biomass represent an increasing business sector and will result in strong growth of the global biomass trade, in particular biomass pellets [1]. Lignocellulosic biomass has, compared to conventional fuels, a relatively low bulk and energy density and a high degree of inhomogeneity. Thermal and mechanical pre-treatment technologies such as torrefaction and pelletization can increase energy density and homogeneity of biomass and reduce handling costs at the same time, as well as reducing transport costs [2,4].

During torrefaction, biomass is roasted in an oxygen depleted environment at temperatures between 240 and 320 °C (depending on the reactor type and technology), resulting in the removal of moisture and some of the volatiles, and leading to a reduction of the atomic ratios O/C and H/C in the resulting torrefied product [2, 3]. In practice, this means that the majority of the calorific value of the biomass is retained within a fraction of the original mass, resulting in a biobased product with high specific energy, typically around 30 % more energy per mass unit dry weight [2,5]. The physical properties of biomass fibres change significantly during torrefaction. Thermal degradation of the cell wall polymers, i.e. hemicelluloses, cellulose and to some extent lignin, transforms the biomass into a brittle material with hydrophobic properties [2]. In combination with pelletization, the aim is to produce a durable biobased fuel pellet of high energy density, with a high degree of homogeneity and hydrophobic characteristics that can, ideally, be handled and stored outdoors without weather (rain) protection. The brittleness and the reduced oxygen content of torrefied pellets make them an ideal candidate to replace coal with biomass in existing heat and power plants [2,6]. A number of studies have shown that torrefaction increases the efficiency of biomass combustion [7,8] and gasification [9] processes.

While pelletization of biomass is an established technology, with the annual global production of wood pellets estimated to be about 24.5 million tons in 2013 [10], torrefaction is still a new technology for the production of solid energy carriers, and is in a pre-commercial phase. Technological development has made significant progress during recent years and there are a number of initiatives and private companies in the process of scaling up production and starting to produce torrefied biomass pellets commercially [6]. Major technical challenges that have been identified regarding the development of torrefaction technologies are predictability and consistency of product quality, densification of torrefied biomass, heat integration and the flexibility associated with using different input materials [6].

During torrefaction the biomass polymers, especially hemicelluloses, are degraded mainly by depolymerization, demethoxylation, bond cleavage and condensation reactions [11]. An increasing degree of torrefaction has been shown to result in an increasing wall friction in the press channels of a pellet press and poorer mechanical properties [12-14].

Mechanical interlocking, solid bridges and intermolecular forces during pelletizing have been noted as important factors for bond formation, affecting the mechanical properties of a biomass pellet [15-18]. It has been suggested that the moisture content of the biomass is an important factor in this context due to its plasticizing effect and ability to reduce the glass transition temperature of cell wall polymers [19]. The modification of cell wall polymers, the removal of moisture and polar hydroxyl groups from the biomass during torrefaction, as well as reduced interlocking due to the brittleness of particles, are probably important factors decreasing the bonding properties of torrefied biomass when densified.

It has previously been shown that pelletizing parameters such as press channel dimension, moisture content, particle size and temperature have a significant effect on the friction generated in the press channel and thus on the energy required for pelletization [20]. These parameters also affect the pellets' mechanical properties. Different strategies are applied to counteract the effects of torrefaction on pelletizing properties such as increasing pelletizing temperature, adding moisture [21] and the addition of processing aids with lubricating properties to improve pellet quality and ease processing [22]. Overall, the aim is to reduce energy consumption while maximizing the capacity and quality of the pellet production processes.

Most process optimization today is undertaken experimentally in either a lab or in pilot scale units and is based on trial and error. This is mainly due to a lack of understanding of the correlations between torrefaction and pelletization parameters and their effect on pellet quality (strength) and process energy consumption (compression and friction). To address this, the present study maps the combined effects of key parameters of torrefaction and pelletization on the pelletizing process and the resulting pellet quality.

Materials and Methods

Materials
Norway spruce (*Picea abies* Karst.) grown in Värmland, Sweden was used for the torrefaction. The spruce was harvested during fall 2011 and then sawn and dried in a wood kiln. Thereafter the sawn timber was trimmed and the pieces trimmed off were shredded. The shredded material was then sieved (Sizer typ E0554, Mogensen, Sweden) to separate it into different sizes; the material used for torrefaction was larger than 4mm but less than 8 mm. The material was stored dry for 12 months before torrefaction.

Experimental design

A D-optimal fractional factorial experiment with four parameters was designed. First, a qualitative parameter "particle size" was used with two classes ("small" < 0.5 mm, and 0.5 mm < "big" < 2 mm). The following were quantitative parameters at five levels: torrefaction degree (within the range 250-300 ˚C resulting in mass yields from 90.5% down to 71.1% based on dry matter); moisture content of materials entering the pellet press (dry to 10%); and, finally, die temperature during pelletizing (125-180 ˚C). The centre point for the larger particles was repeated three times. In all, 29 separate experiments were run (Table 1). The experimental design allowed analysis of the dependency between different process parameters on pellet quality and forces occurring in the press channel of a pellet press.

Table 1. Fractional factorial experimental design with four parameters (the target torrefaction temperature is here replaced by recorded real values of mass yield)

Experiment name	Run order	Particle size	Torrefaction temperature (˚C)	Mass yield (d.b.) (%)	Moisture content (%)	Pelletizing temperature (˚C)
N12	1	small	287.5	75.9	0	180
N11	2	small	262.5	85.7	0	125
N4	3	small	300	71.1	10	180
N6	4	small	250	90.5	6.7	125
N25	5	big	275	80.3	5	180
N3	6	small	300	71.1	10	125
N7	7	small	250	90.5	10	143.2
N29	8	big	275	80.3	5	152.5
N28	9	big	275	80.3	5	152.5
N18	10	big	250	90.5	10	125
N23	11	big	287.5	75.9	10	125
N13	12	small	262.5	85.7	10	180
N5	13	small	250	90.5	3.3	125
N14	14	big	250	90.5	0	125
N10	15	small	300	71.1	0	161.7
N22	16	big	300	71.1	10	143.3
N26	17	big	275	80.3	0	152.2
N17	18	big	300	71.1	0	180
N19	19	big	250	90.5	10	180
N1	20	small	300	71.1	0	125.5
N9	21	small	300	71.1	3.3	180
N16	22	big	250	90.5	0	180

N15	23	big	300	71.1	0	125
N2	24	small	250	90.5	0	180
N8	25	small	250	90.5	10	161.7
N27	26	big	275	80.3	5	152.5
N24	27	big	250	90.5	5	152.5
N20	28	big	300	71.1	10	180
N21	29	big	300	71.1	6.7	125

Torrrefaction

A bench scale torrefaction reactor was constructed to enable the production of materials with different degrees of torrefaction. A box made of stainless steel, with a volume of about one litre, and fittings for the inflow and outlet of gasses was used as a reactor. The reactor was inserted in a programmable muffle furnace with a maximum output of 3000W (Carbolite furnaces, Carbolite UK) and flushed with nitrogen during the whole process at a rate of 0.5 L/min using a pressure and flow regulator. For temperature monitoring, a thermocouple was installed at the centre of the reactor and connected to a logger (TESTO 735-2, Testo, Germany), and another thermocouple was installed in the furnace to control the heating of the furnace. The heating was controlled by the furnace thermocouple instead of the thermocouple placed in the middle of the reactor. This minimized the risk that the furnace temperature would overshoot, which could result in a torrefaction degree gradient in the reactor caused by higher temperatures at the reactor sides. Five different temperatures were chosen resulting in different degrees of torrefaction (Table 1). The degree of torrefaction is here defined as mass yield in percentage based on dry mass before and after torrefaction.

The material was dried at 105˚C for 16 h before torrefaction. A sample consisting of about 130 g of the dried material was placed in the reactor. The reactor was heated to the set temperature at a heating rate of 3.8-7.3 ˚C/min depending on the temperature set (a higher temperature resulted in a higher heating rate) and with a declining heating rate closer to the set temperature. Then the set temperature was maintained for 60 minutes. Directly thereafter the reactor was quenched with cold tap water to stop the torrefaction process. Amount of C, H, O, N, S and ash in the torrefied materials as well as their calculated gross calorific values are presented in Table 2.

Table 2. Chemical composition in % of dry weight and gross calorific value (GCV) for the different torrefied materials used in the study.

Mass yield (%)	C (%)	H (%)	O (%)	N (%)	S (%)	Ash (%)	GCV[1] (kJ/g)
71.1	59.2	5.5	35.0	<0.1	<0.01	0.33	23.5
75.9	57.2	5.8	36.7	<0.1	<0.01	0.31	23.0
80.3	55.9	5.8	38.0	<0.1	<0.01	0.30	22.4
85.7	54.4	5.9	39.4	<0.1	<0.01	0.29	21.9
90.5	53.1	5.9	40.7	<0.1	<0.01	0.29	21.3
100	50.7	6.1	42.9	<0.1	<0.01	0.28	20.4

[1]Estimated according to equation by S.A. Channiwalla [23].

Milling and sieving

After torrefaction, the material was milled in a knife mill (Retsch SM2000, Retsch, Germany) over a 6 mm sieve. To achieve the two different fractions of particle sizes, the milled material was sieved using an oscillating sieve (Fritsch Analysette 3, Fritsch , Germany) to achieve the fractions <0.5 mm (small particles) and <2 mm but >0.5 mm (big particles). The material was sieved for 15 min and the amplitude was set to 1.5 Hz

Adjusted moisture content

The experimental design required five levels of moisture content from dry to 10 %. The dry samples were dried at 105 ˚C for 16 h and then packed and sealed in gaseous tight plastic bags. The moisture content of the other samples was adjusted with water, using a spray bottle and mixing, to achieve the correct moisture content according to the experimental design. All these samples were then put in gaseous tight and sealed plastic bags for equilibration lasting more than one week before pelletizing. The moisture content was measured just before pelletizing by analysing subsamples of about 5 g using a moisture analyser (Mettler Toledo; Greifensee, Switzerland) equipped with an infrared drying device at 105˚C. The moisture contents used in the modelling were the average of the adjusted values and the measured values just before pelletizing (the mean difference was just 0.017% or expressed as mean error 0.692%). These moisture values were close to the set targets in Table 1.

Pelletizing

The pelletizing test of torrefied material was undertaken using a single pellet press tool widely used for testing and evaluating the pelletizing properties of biomass feedstock [10-12]. The experimental set-up and working principle of the tool is shown in Figure 1. The pellet press tool consisted of a metal cylinder with a press channel and a backstop. To press a pellet, the press channel was closed with the backstop and the die was filled with biomass (phase 1) then compressed into a pellet (phase 2). Once the pellet was pressed, the backstop was removed and the pellet was extruded from the die

(phase 3). The die was made from hardened steal and the press channel had a diameter of 8 mm. The single pellet press tool was equipped with thermocouples used in combination with a material test system (AGX, Shimadzu, Japan) with a 200 kN load cell. For pelletization, the die was pre-heated until the temperature set-point was reached and kept stable at $\pm 2 \degree C$. A mass of 750 mg $\pm$ 5 mg torrefied materials was fed into the opening of the press channel in one step and compressed at a rate of 100 mm min^{-1} until a maximum pressure of 300 MPa was reached. The pressure was held for 5 seconds. Next, the bottom piston was removed from the die, and the pellet was pressed out of the channel at a rate of 100 mm min^{-1}. The system recorded the force and the distance continuously and these readings were used to calculate the response variables (Table 3).
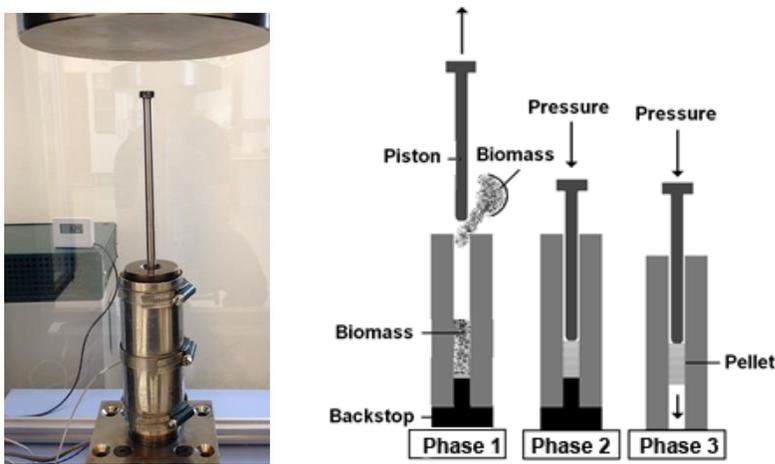
Figure 1. Single pellet press tool used to determine the forces generated in the press channel during compression and extrusion of the pellet. The die can be heated and forces are detected using a material test system and a load cell.

After pelletization the pellets were cooled overnight and compression tested the following day. Compressive resistance (or crush resistance) is a measure of strength, revealing the maximum crushing load a pellet (perpendicular to the length of the pellet) can withstand before cracking or breaking. This test, described previously by Stelte et al. [25], has been shown to be related to standardized pellet quality parameters i.e. durability [24]. In this test, the pellet was laid on its side and crushed between two metal plates at a compression rate of 20 mm min^{-1}. A force distance curve was recorded and the point at breaking was used as a figure that described the strength of single pellets. Furthermore, the unit density of the pellets was determined by measuring the dimensions using a vernier caliper. For all data collected, the average value of at least five measurements was determined and used in the modelling.

Table 3. Response variables determined for the single pellet press tested.

Response	Unit	NB
W_{comp}	Joule	Work required to compress biomass into a pellet (750 mg). The value was calculated by integrating the area under the force-distance curve until the maximum pressure of 300 MPa was reached
F_{max}	Newton	The initial force to overcome the static friction between pelletized biomass and the press channel surface
W_{Fric}	Joule	The energy required to push the pellet 25 mm through the press channel (dynamic friction work)
Strength	Newton	Force required to crush the pellet between two metal plates
Weight	mg	Material weight before and after pelletization
Length	0.01 mm	Pellet length before crush resistance test
Diameter	0.01 mm	Diameter before crush resistance test

Modelling and diagnostics

The software MODDE 9.0 (Umetrics AB, Sweden) was used for the multiple linear regression (MLR) and ANOVA modelling of the fractional factorial design, see Tables 1 and 3. All parameters and responses were mean centred and the design involving particle sizes as the quality parameter was analysed in coded form (small and big, respectively). Non-significant parameters or their non-significant 2-way interactions as well as non-significant squared parameters ($p > 0.05$) were excluded from the modelling. The diagnostics were based on the residuals between observed (y_{obs}) and predicted (y_{pred}) responses for each run. The cross-validated coefficient of multiple determination (Q^2) that expresses how much of the variance in the response variable can be predicted (0: no predictive capability; 1: all variance can be predicted) was used to find the most reliable model and was calculated as follows:

$$Q^2 = 1 - PRESS(SS)^{-1}$$

where *SS* is the square sum of the mean centred responses (y_c) in the calibration set ($SS = y_c^T y_c$) [26] and *PRESS* (prediction error sum of squares) is calculated as follows:

$$PRESS = \mathbf{1}[(\mathbf{f}^T\mathbf{f})](\mathbf{1}^T - \mathbf{h})^{-1}]$$

where **1** is a vector of ones (dimension $1 \times N$; $\mathbf{1}^T$ has the dimension $N \times 1$) (^{T} denotes a transposed vector or matrix), vector **f** is the residuals $\mathbf{f} = \mathbf{y}_{obs} - \mathbf{y}_{pred}$ between the observed and predicted response values ($N \times 1$), vector **h** ($N \times 1$) is the diagonal elements of the Hat matrix, $\mathbf{X}(\mathbf{X}^T\mathbf{X})^{-1}\mathbf{X}^T$, and the **X**

matrix here constitutes the model coded parameters for size and the quantitative parameters and interactions for each observation in the calibration set.

Residual standard deviation (*RSD*) was calculated as follows:

$$RSD=[\mathbf{1}(\mathbf{f}^{\mathrm{T}}\mathbf{f})](N\text{-}p)^{-1}]^{0.5}$$

using the residual vector **f** between the observed ($\mathbf{y}_{\mathrm{obs}}$) and predicted ($\mathbf{y}_{\mathrm{pred}}$) response vectors. Here the row vector of ones (**1**; dimension $1 \times N$) is used in matrix algebra notation just to summarize the squared residual column vector ($N \times 1$). The scalars N and p are the number of observations and the degrees of freedom used in the model, respectively.

The condition number represents a measure of the orthogonality of the design. In a fully orthogonal factorial design the condition number is 1. However the set design points may not be reached so the design will not be fully orthogonal. Numbers <3 indicate a good model design whereas numbers >6 a bad design [27]

The software SIMCA 10.0. 0 (Umetrics AB, Sweden) was used for principal component analysis (PCA) to gain an overview of the data. A cross-correlation matrix of parameters and responses was calculated using Matlab (The Mathwork, Inc., USA).

Results and discussion

Overview of the data
An overview of all observations, parameters and responses is presented in Figure 3 as a PCA biplot showing score and loading values for the first two model components. It should be noted that one observation (N15) contains missing values regarding pellet dimensions, density and mass lost. (The material at this set-point had, in most cases, disintegrated when extruded from the pellet press die.) These PCA components explained 82,2% of the variation and the first component was spanned by density on one side and F_{max} and W_{fric} on the other side the second component was spanned by weight and moisture (meaning that these responses are negatively correlated). See also the correlation matrix in Supplementary Table 1. It also shows that the pelletizing responses W_{comp}, F_{max} and W_{fric} are closely grouped, as were length and diameter, and that they are negatively correlated to density as expected when a fixed mass is used for each pellet. Moisture content are negatively correlated to weight. This was also expected as more moisture in the material will lead to great mass loss due to the high pelletizing temperatures.
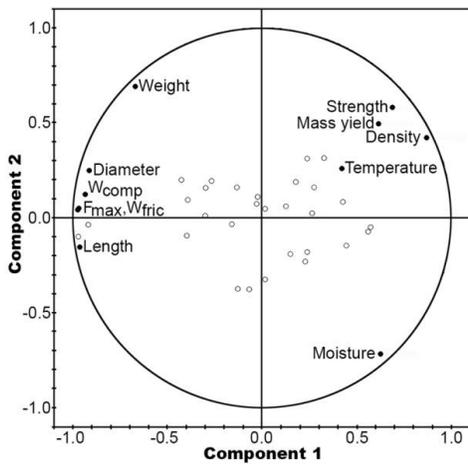
Figure 3. PCA biplot showing normalized score and loading values of the first two model components (open circle: observations; filled circle: parameters and responses).
A cross-correlation matrix for all parameters and responses is presented in the supplementary material and these correlations confirm the groupings revealed by the PCA biplot.

MLR models

The predictive capacities of the MRL models presented in Table 4 were excellent for the three responses W_{comp}, F_{max} and W_{fric} within the pelletizing process and for pellet length. They had the highest Q^2 values with respect to predictive capacity (> 0.926) whereas the one for pellet diameter had the lowest. It is natural that the pellet diameter is constrained, due to the fixed die diameter, irrespective of other parameters, making it difficult to explain on the basis of the tested parameters. In addition, the pellet dimensions weight and density exhibited high predictive capacity (0.87-0.89) together with pellet strength (0.81). The condition number varied between 2.0 and 4.9, indicating good to acceptable experimental designs.

Table 4. Pelletizing (W_{comp}: compression work; F_{max}: static friction; W_{fric}: friction work) and pellet quality (strength, weight, length, diameter and density) responses for the torrefaction experiment modelled by MLR.

Model components	W_{comp}	F_{max}	W_{fric}	Strength	Weight	Length	Diameter	Density
Unit	J	N	J	N	mg	0.01 mm	0.01 mm	mg/mm^3
Mean value	37.13914	1131.089	23.05401	378.1211	717.6	1202	807	1.17
Standard deviation	5.71131	479.1591	11.83677	107.8424	16.01	65.34	3.49	0.06
Maximum value	50.0747	2278.81	52.7523	612.225	756	1398	822	1.26
Minimum value	28.1347	318.959	4.87626	95.0042	681	1071	801	1,00
Number of observations	29	29	29	29	28	28	28	28

Degrees of freedom	21	20	22	21	23	18	19	23
R^2 adjusted	0.974	0.968	0.963	0.859	0.912	0.964	0.827	0.903
Q^2	0.959	0.946	0.944	0.801	0.889	0.926	0.627	0.874
RSD	0.924	86.26	2.292	40.44	4.742	12.37	1.45	0.0184
Condition number	4.042	4.063	3.888	4.853	3.925	4.73	1.955	3.831
Constant	37.9619	1229.63	25.1462	462.986	721.875	1177.9	806.943	1.19118
Main factors								
Size (small)	-0.90398a	N.S.	N.S.	49.2637a	-2.72827b	-7.03723c	N.S.	N.S.
Size (big)	0.90398a	N.S.	N.S.	-49.2637a	2.72827b	7.03723c	N.S.	N.S.
Mass yield, % (d.b)	-3.84031a	-320.616a	-7.8544a	68.8735a	N.S.	-51.9967a	-1.67341a	0.054861a
Moisture content, % (w.b)	-4.82127a	-379.209a	-9.59034a	N.S.	-19.9005a	-46.1585a	-3.73399a	0.020537a
Temperature, ˚C	-1.93788a	-247.995a	-5.54268a	64.5506a	-5.66733a	-43.9745a	-1.64775a	0.03513a
2-way interactions[1]								
Size (small)×Mass yield	N.S.	-45.7644a	N.S.	N.S.	N.S.	N.S.	N.S.	N.S.
Size (big)×Mass yield	N.S.	45.7644a	N.S.	N.S.	N.S.	N.S.	N.S.	N.S.
Size (small)×Moisture content	0.57172c	-45.2558c	N.S.	N.S.	N.S.	N.S.	N.S.	N.S.
Size (big)×Moisture content	-0.57172c	45.2558c	N.S.	N.S.	N.S.	N.S.	N.S.	N.S.
Size (small)×Temperature	N.S.	N.S.	N.S.	N.S.	N.S.	8.03419c	0.736723c	N.S.
Size (big)×Temperature	N.S.	N.S.	N.S.	N.S.	N.S.	-8.03419c	-0.736723c	N.S.
Mass yield× Moisture content	N.S.	122.185a	3.41829a	-31.7379c	N.S.	10.6749c	1.42117b	N.S.
Mass yield×Temperature	N.S.	N.S.	N.S.	N.S.	N.S.	10.5858c	0.90457c	N.S.
Moisture content×Temperature	2.73442a	148.558a	4.47319a	N.S.	N.S.	N.S.	1.62153b	N.S.
Squared factors[1]								
Mass yield×Mass yield	-1.26991b	-142.045b	-3.1565a	-49.8991c	N.S.	N.S.	N.S.	N.S.
Moisture content×Moisture content	N.S.	N.S.	N.S.	-81.161c	-7.36376b	27.3002c	N.S.	-0.04668a
Temperature×Temperature	N.S.	N.S.	N.S.	N.S.	N.S.	21.4536c	N.S.	N.S.

[1] Non-significant interactions and squared factors not shown; N.S. non-significant (p>0.05) and therefore not included in the modelling; a: significant at p<0.001; b: significant at p<0.01; c: significant at p<0.05;

Table 4 also shows that all responses except for W_{fric} and density were influenced by particle size as a main or interacting factor. The presence of more fine particles lowered W_{comp}, but increased pellet strength and also raised F_{max} somewhat. An increased mass yield, i.e. a lower torrefaction degree, and elevated pelletizing temperature improved pellet strength and reduced the energy requirements for the compaction and particle to wall friction. Because of the influence of significant interactions and squared factors, surface plots give a better description of how different parameters influence the work and force responses, see Figs 4 and 5.
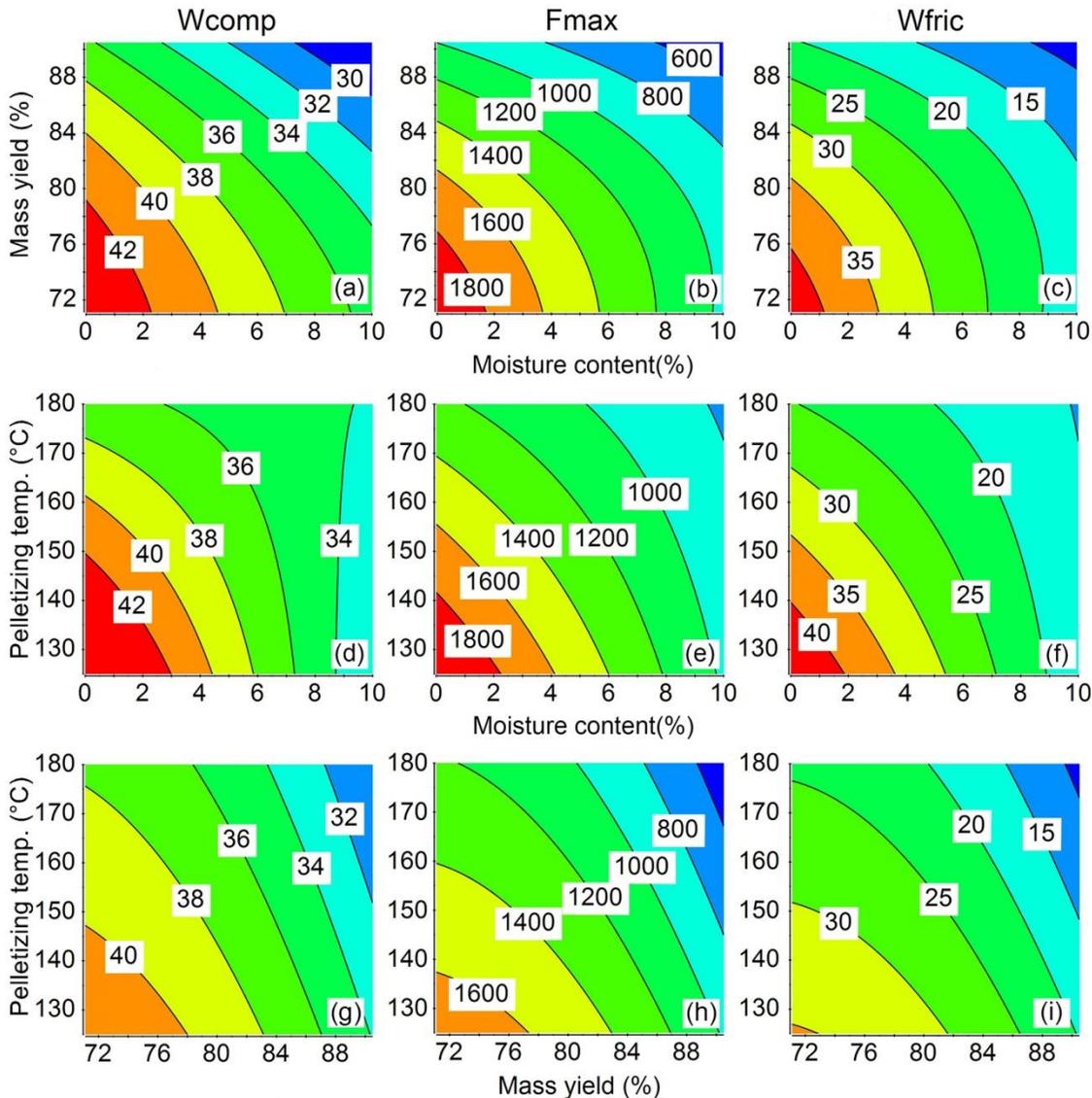
Fig. 4 a-i. Influence of mass yield, moisture content and pelletizing temperature on W_{comp} (Joule) (4a, d, g), W_{fric} (Joule) (4c, f, i) and F_{max} (Newton) (4b, e, h). Contour surfaces shown for the small particle size and other parameters at their intermediate values: mass yield at 80.3%; moisture content 5.0%; pelletizing temperature 152°C.

The general pattern in Fig 4 (a-i) seems to be that increased mass yield (4a-c), moisture (4d-f) and pelletizing temperature (4g-i) reduce pelletizing energy to compress (W_{comp}) the material and to overcome static (F_{max}) and kinetic friction (W_{fric}) in the pelletizing press channel. The more the material is torrefied, the more pelletizing energy is consumed, i.e. when mass yield decreases (4a-c and 4g-i). This is in accordance with other studies [25]. As indicated by the surface plots in Fig. 4, this energy requirement can to some extent be reduced by increasing the temperature (4g-i) and moisture (4a-c) content. However, pellet quality (strength) may be adversely affected by increased

temperature and moisture content. At the moment there are no special criteria demanded by the end users regarding torrefaction degree. Different end users could have different requirements depending on how the pellets are to be used e.g. gasification or co-firing with coal. But in all cases it is likely that the goal is durable pellets that can be stored outdoors and can withstand rain. This means that, at the moment, there are no clear guidelines for required pellet strength, but most probably this will be similar to that for wood pellets. However, if pellet strenght is too pronounced in the pelletizing then this may influence downstream processes, e.g. increased energy consumption when milled to a powder prior to being fed into a pyrolyser or gasifier.

Fig. 5 indicates process routes that can influence pellet strength and Fig. 5a suggests that somewhat more moisture is needed within the material as it is more torrefied. Furthermore, Fig. 5b and 5c show clearly that increasing pelletizing temperature results in increased strength regardless of torrefaction degree and moisture content. Thus the strength of pellets made from material of low mass yield may be improved by a higher pelletizing temperature and about 2% increased moisture.
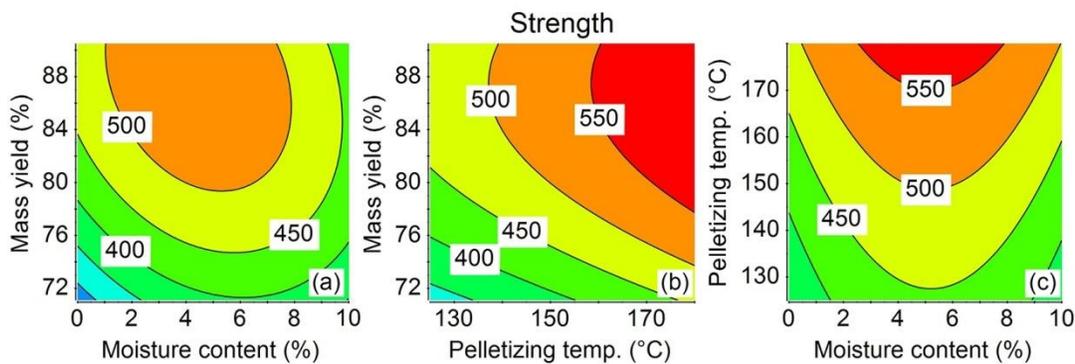
Fig. 5a-c. Surface plots showing influence of mass yield, moisture and pelletizing temperature on pellet strength (Newtons at breakage). (Contour plot settings see legend in Fig.4.)

The optimization of strength according to Fig 5a-c seems to follow a very narrow production window, indicating a delicate balance required during continuous, industrial pelletizing. In practice, friction between compressed particles and press channel walls during continuous pelletizing operations will produce additional heat in the press die. When this heat is rising more water will vaporize from the torrefied material, and this will occur more rapidly. This then raises a fundamental question: How is it possible to keep and control moisture in the pelletizing material at high pelletizing temperatures? One idea is to compensate for moisture in the original material, i.e. more moisture if the pelletizing temperature is higher than optimal and *vice versa*. In conventional wood pelletizing, steam is used, but how is it possible to differentiate between the heat transferred to the material and the moisture as these co-vary in a complex way. Here it is almost the same problem: to create differences between temperature and moisture and control them separately. Thus,

inventions are still required in order to fine-tune the simultaneous control of water content in the material and pelletizing temperature at given degrees of torrefaction.

The model of pellet strength may give some clues about how to proceed with higher moisture levels and temperatures. When simulating the model components at moisture ranges of >14%, clearly above the range studied in the current factorial experiment, then the models indicate that F_{max} and W_{fric} increase at higher temperatures, as found at >8.8% for W_{comp}. This contradicts the common conception of the current model as shown by Figs 4 and 5. This change also occurs for increased moisture contents, starting to increase W_{comp} at temperatures >197°C, W_{fric} at >221°C and F_{max} at >240°C i.e. above these elevated temperatures, energy requirements increase regardless of torrefaction degree although moisture content is increased. As these simulations are clearly outside the investigated range of moisture contents and pelletizing temperatures, they are just speculations, but will be interesting to study in the future.

In this study, the initial pressure to compress different materials was constant and set to 300 MPa for 5 seconds. The resulting density of the pellets produced ranged from 1.00 to 1.26 mg/mm^3. As these densities are in the lower spectrum for conventional wood pellets, it will be important to include higher initial compression pressures as a parameter in future research.

Conclusions
In conclusion, it was clearly shown that factorial experiments using single-pellet devices can provide insights that otherwise are either difficult or expensive to obtain. All parameters tested – particle size, torrefaction degree (mass yield), moisture content and pelletizing temperature – were shown to have significant influence on compression and friction work as well as pellet dimensions and strength. The results of the factorial experiment indicate that, for strength, there is a narrow process window to optimize one of the most important pellet quality factors. This may be hard to ensure during practical pelletizing due to feedstock variations in relation to particle size, torrefaction degree and moisture content and, therefore, the quality of the pellets produced may vary. Further experiments are needed to extend the range of the parameters and explore whether new process routes are possible.

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Supplementary material

Table 1. Correlation coefficient matrix for factors and response variables.

		Mass yield	Moisture	W_{comp}	F_{max}	W_{fric}	Strength	Weight	Length	Diameter	Density
Parameters	Temperature	-0.107	-0.075	-0.183	-0.328	-0.293	0.469	-0.238	-0.433	-0.263	0.387
	Mass yield		0.032	-0.589	-0.570	-0.567	0.520	0.011	-0.648	-0.359	0.755
	Moisture			-0.642	-0.621	-0.626	-0.092	-0.877	-0.459	-0.685	0.218
Responses	W_{comp}				0.926	0.943	-0.386	0.697	0.807	0.831	-0.691
	F_{max}					0.994	-0.458	0.648	0.898	0.884	-0.808
	W_{fric}						-0.461	0.648	0.876	0.886	-0.786
	Strength							-0.038	-0.691	-0.441	0.776
	Weight								0.543	0.735	-0.258
	Length									0.840	-0.949
	Diameter										-0.713

Cross-correlations of all parameters and responses (regardless of particle size) are presented in Table 1 The experimental parameters mass yield, moisture content and pelletizing temperature were set independently of each other and should, therefore, exhibit low and non-significant correlations and this was, indeed, the case (<0.11). Of the experimental parameters, moisture content of the torrefied material showed the highest correlation (0.877) to the response variables and especially to weight as this response was directly influenced by moisture content. Mass yield and density were positively correlated (0.755) meaning that density increased when mass yield increased i.e. when the degree of torrefaction decreased. Other correlations between parameters and responses were less than 0.69. It is notable that all experimental parameters, especially moisture, exhibited low correlations to pellet strength. As expected, length and diameter were highly correlated (>0.94). In addition, the pellet quality measures strength and density were positively correlated (0.776). The pelletizing responses W_{comp}, F_{max} and W_{fric} were also highly correlated to each other (>0.92), and they were also correlated to pellet length and diameter (>0.80, but <0.90). The friction force (F_{max}) was negatively correlated to density (-0.808) indicating that when high quality pellets were formed in phase 1 of the process, these experienced lower static friction in the die channel. A higher torrefaction degree (lower mass yield) was associated with more energy needed for pellet formation (W_{comp}, F_{max} and W_{fric}) and with lower pellet strength and density.