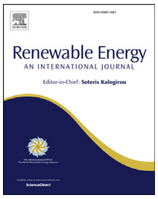
Renewable Energy 178 (2021) 766–774

Contents lists available at ScienceDirect

Renewable Energy

journal homepage: www.elsevier.com/locate/renene



Pelleting torrefied biomass at pilot-scale – Quality and implications for co-firing

David A. Agar [a, *], Magnus Rudolfsson [a], Simon Lavergne [b, f], Thierry Melkior [c], Denilson Da Silva Perez [d], Capucine Dupont [e], Matthieu Campargue [b], Gunnar Kalén [a], Sylvia H. Larsson [a]

[a] Swedish University of Agricultural Sciences, Department of Forest Biomaterials and Technology, SE-90183, Umeå, Sweden
[b] Laboratoire RAGT Energie, Zone Innoprod, Chemin de La Teulière, 81000, Albi, France
[c] Université Grenoble Alpes, CEA-LITEN, DTBH, 17 Avenue des Martyrs, 38054, Grenoble Cedex 09, France
[d] FCBA, 10 Rue Galilée, 77420, Champs-sur-Marne, France
[e] IHE Delft Institute for Water Education, Department of Water Supply, Sanitation and Environmental Engineering, Delft, Netherlands
[f] Bioenergy Europe, Place du champ de Mars 2, 1050, Bruxelles, Belgium

ARTICLE INFO

Article history:
Received 25 March 2021
Received in revised form
7 June 2021
Accepted 21 June 2021
Available online 26 June 2021

Keywords:
Torrefaction
Durability
Energy density
Densification
Pelletization
Bio-coal

ABSTRACT

The co-firing of solid biofuels in coal plants is an attractive and fast-track means of cutting emissions but its potential is linked to biomass densification. For torrefied materials this topic is under-represented in literature. This pilot-scale (121–203 kg h^{-1}) pelleting study generated detailed knowledge on the densification of torrefied biomass compared to untreated biomass. Four feedstock with high supply availability (beech, poplar, wheat straw and corn cob) were studied in their untreated and torrefied forms. Systematic methods were used to produce 180 batches of 8 mm dia. pellets using press channel length (PCL) and moisture content (MC) ranges of 30–60 mm and 7.3–16.6% (wet basis) respectively. Analysis showed that moderate degrees of torrefaction (250–280 °C, 20–75 min) strongly affected pelleting behaviour. The highest quality black pellets had a mechanical durability and bulk density range of 87.5–98.7% and 662–697 kg m^{-3} respectively. Pelleting energy using torrefied feedstock varied from −15 to +53 kWh t^{-1} from untreated with increases in production fines. Optimal pelleting MC and PCL were reduced significantly for torrefied feedstock and pellet quality was characterised by a decrease in mechanical durability and an increase in bulk density. Energy densities of 11.9–13.2 GJ m^{-3} (as received) were obtained.

1. Introduction

The push to limit global temperature rise to one and a half degrees above pre-industrial levels signals the phase out of coal within a generation [1]. The realisation of European Union energy policy, for instance, will cut emissions by 40% by 2030 [2] and China has committed to carbon neutrality by 2060 [3]. With such short time spans, sustainable transitional fuel use in existing coal power plants makes a good economic argument.

Sixteen years ago, bio-coal entered the renewable energy scene. *Bio-coal* is a fossil coal substitute produced from renewable biomass resources. A more precise name is *pellets made from torrefied lignocellulosic biomass* because *torrefaction* (from French usage for *coffee roasting*) is the partial pyrolysis process at the heart of the fuel [4]. Interest in bio-coal stems from the approach taken for thermal conversion (e.g. the combustion of solid fuels to produce heat and electricity), two of which are: 1) Design the power plant according to the fuel or 2) Design the fuel according to the power plant. Bio-coal technology takes the second approach and takes advantage of existing pulverised-fuel power plants, which are plentiful [5,6]. This is the primary application of torrefied pellets; to co-fire them with coal in existing power plants. Large reductions in net carbon dioxide emissions are enabled [7,8] because energy sector emissions make up about one third of anthropologic emissions [9].

The co-firing potential of a fuel is linked to its degree of similarity to coal. Torrefaction induces three key physical changes in lignocellulosic biomass: 1) an enhanced grindability, 2) an

---

* Corresponding author.
E-mail address: david.agar@slu.se (D.A. Agar).

https://doi.org/10.1016/j.renene.2021.06.094

improved effective heating value and 3) a reduced equilibrium moisture content. The high specific energy requirements of milling wood to powders is reduced by 80–90% after torrefaction [10] and laboratory studies have shown that torrefaction improves wood [11–16] and agricultural biomass [12,13,17] heating values by 7–21% and 7–15% respectively. The increase is partly a result of a lower equilibrium moisture content [14]. These results are for torrefaction conditions at 240–300 °C for 10–80 min and are beneficial for large-scale coal replacement [18].

Yet these benefits of torrefaction bring no improvement in the bulk energy density of the feedstock (i.e. energy content per cubic metre) which remains as low as for the untreated materials [19]. Energy density is important for the *direct co-firing* of a fuel, by which it is blended with coal upstream of the coal feeders, pulverised and then pneumatically conveyed to the burners [20]. The poor grindability of untreated biomass [21,22] impedes the throughput of coal pulverisers. For example, when milling wood chips in an industrial vertical roller mill, the maximum capacity mass throughput is 0.25 t h^{-1} while that of coal is 2.2 t h^{-1} [23]. This is a mass-flow difference of almost nine times. In terms of fuel energy capacity, the mill throughput difference is more dramatic; with the effective heating value differences between wood and coal (16 MJ kg^{-1} versus 28 MJ kg^{-1} [24], for example) the energy flow difference is 15 times. This exemplifies why direct co-firing of untreated wood is limited to about 5% energy basis [25].

Improving energy density is achieved by pelleting which also provides easier handling, more compact storage and better flow properties; important factors for economical logistics and transport [26]. The pelleting of wood is a mature industrial process [27] and herein lies the catch; for years, it has been assumed that the technical knowledge and experience in this sector is sufficient for torrefied feedstock. As will be shown, this assumption is far from valid. Pelleting success and product quality is primarily measured by mechanical durability (DU) of pellets, expressed as the fraction of a pellet sample that survives a tumbling test intact (EN 15210). The minimum DU is 97.5% for A1 quality (Table 2) and with untreated wood feedstock this is routinely achieved.

Other technical and efficiency-based considerations of co-firing torrefied fuels in pulverised-coal boilers are not foreseen as problematic. Replacing 15% coal with torrefied wood under conditions of constant mass-flow, requires about the same energy to mill [28]. After torrefaction, wood is still rich in volatile content [4] and only slightly less reactive than untreated wood [29]. Ignition times (volatiles and char) decrease with the degree of torrefaction [30] but particle burnout times are still rapid compared to coals. This means that the optimal particle size for combustion is marginally lower than for untreated wood [31] but otherwise not an issue in practice. Indeed, results from co-firing in a 0.5 MW boiler have demonstrated that combustion efficiency remains constant or increases slightly when co-firing from 30 to 70% (mass basis) torrefied (pine) pellets with coal [32]. This was accompanied by a reduction in coal-mill performance with increasing fractions of torrefied wood (i.e. the particle size distribution shifted to larger diameters). Further modelling shows that from 50 to 100% coal substitution (mass basis) is technically feasible without losses in boiler efficiency yet with significant reductions in ash and NO$_x$ emissions [33,34].

Pelleting of torrefied biomass is then a vital step in the main application of bio-coal but, of the two thousand peer-reviewed journal publications on *torrefaction*, only five have investigated pelleting at scales representative of industrial pellet production (i.e. pilot scale). Table 1 summarises the findings from these studies, which show that low durability of torrefied pellets is very much the norm, especially when torrefaction conditions are anything but mild. The reported DU ranges from 46 to 92% for the majority of feedstock, with two glaring exceptions: 97% for beech and 99% for eucalyptus, after torrefaction at 270 °C/45 min and 265 °C/22 min respectively. The heating value increase, a measure of torrefaction degree, ranged from 4 to 15%; roughly half of the values reported at laboratory scale where pelleting was not investigated. One can conclude that the ability to pellet with high DU restricts the degree of torrefaction.

In contrast, the reported bulk densities in Table 1 are high, ranging from 557 to 725 kg m^{-3}, well above ENplus quality for the most part. These contribute to energy densities from 10 to 17 GJ m^{-3}. Note, however, that several values are on a dry mass basis (i.e. not as received).

What unites these studies is the necessary limitations they shared on the breadth and depth of investigation; their experimental matrix was small. The reason is the high cost of research at this scale as it requires equipment investments, specially designed facilities, technicians and large material volumes. Experimental designs with several feedstock types and parameters lead to a large experimental matrix, requiring more trials and analysis while inflating the project budget. This sets a high price tag for pilot-scale scientific study in academia. Luckily, commercialisation of

Table 1

A summary of previous results on pilot-scale pelleting of torrefied lignocellulosic biomass.

Feedstock	Torrefaction conditions	Heating value increase^{a} (%)	Mechanical durability (%)	Bulk density (kg m^{-3})	Energy density (GJ m^{-3})	Reference
Spruce wood	270/300 °C 16.5 min.	4.2–11.1	80–90^{b}	648–713	12.3–14.7^{c}	[35]
Pine wood	235/245/250 °C 30–45 min.	4.1–9.7	80–92	557–634	9.8–12.2^{d}	[36]
Logging residues^{e}	240/250 °C 30–45 min.	6.8–7.4	87–89	643–681	12.0–12.7^{d}	[36]
Beech wood	270 °C 40–45 min.	9.8	97	702	13.4^{d}	[36]
Pine wood^{f}	291–315 °C 6–12 min.	4.4–13.6	46–87	558–725	12–17^{c}	[37]
Eucalyptus globulus wood^{g}	245/265 °C 22 min.	6.4–10.7	99	671–715	13–15^{c}	[38]
Logging residues	308 °C 9 min.	15	64–90^{h}	610–686	13.5–15.2^{c}	[39]
Willow	308 °C 9 min.	4.1	83–88^{i}	601–683	12.2–13.9^{c}	[39]

Symbols refer to (a) calculated as 100 x (HHV$_T$ - HHV)(HHV)$^{-1}$ where HHV$_T$ and HHV are the higher heating values of torrefied and untreated feedstock respectively, (b) production fines range 10–30%, (c) value for dry matter, not as received, (d) at 22 °C and RH 85%, (e) with and without 3% wheat flour binder, (f) production fines range 4–86%, two PCLs used, (g) 1% starch binder, (h) production fines range 3–22%, (i) production fines range 3–17%, (na) not available.

Table 2
Torrefied feedstock used in the study and their torrefaction conditions.

Feedstock	Torrefaction temperature (°C)	Residence time (min.)	Dry mass yield (%)	Energy yield[a] (%)
Beech stem wood including bark	250	75	77	81
Poplar stem wood including bark	250	75	81	87
	280	30	64	80
Wheat straw	250	20	74	75
Corn cob	260	45	77	88

[a] Calculated as (Dry mass yield %) x (HHV_T/HHV) in which HHV_T and HHV are the higher heating values (MJ kg^{-1}) of the torrefied and untreated feedstock, respectively.

torrefaction technologies has also driven development in torrefied pellet production.

Since 2005, commercial activity has churned out scores of companies and torrefaction process technologies [40] aiming to keep up with the projected market for the new green (black) gold. One decade on, the International Energy Agency (IEA) concluded the following in their 2015 report entitled Status overview of torrefaction technologies [41]: *"The maturation and market introduction of torrefaction technologies has gone slower than anticipated 5 years ago, when it was expected that a significant fraction of the biomass pellets supplied today could have been replaced by torrefied pellets. It has been hard to fully prove the claims made earlier on product characteristics, and several companies have gone bankrupt due to inability to produce good quality product or due to a lack of buyers."*

This may have come as a shock at the time but the conclusions of the IEA report were acknowledging what was, in fact, widely known in the industry for years; that pelleting of torrefied materials was not trivial. This is why representative pelleting research is so important. It is intimately linked to the political and social desire to mitigate carbon dioxide emissions. Without pelleting, large fast-track emission reductions in co-firing cannot be realised. This is why the presented study has so much relevance.

Torrefied pellet production at representative scales is clearly under-represented in literature. Up until now, scientific literature have failed to report with the required breadth on even the most common feedstock types (e.g. wood and straws) with the necessary attention paid to the pelleting process. There is a clear need for in-depth pilot studies on torrefied pellet production in order to better describe quality differences compared to untreated feedstock.

This study investigates how torrefaction affects the pelleting of feedstock. It does this by looking at the influence of moisture content and press channel length on the pelleting responses of energy input, extruded pellet temperature, production fines and quality of pellets. Additionally, this study aims to illuminate the energy-density implications of torrefied pellets in the heat and power sector. It attempts to answer the broader question: What needs to be understood in order to achieve a good quality product when pelleting torrefied lignocellulosic biomass? The novelty is three fold: 1) It focuses on the key obstacle to enabling emission reduction through bio-coal use 2) It compares torrefied pellet quality directly with untreated pellets through extensive production amounts that have great relevance for industrial production 3) It builds knowledge on how torrefaction affects pelleting with respect to production energy, pellet quality (durability and bulk density), production rate and the implications on fuel energy density and use.

2. Materials and methods

Systematic pilot-scale pelleting of lignocellulosic biomass was carried out using four highly available and representative feedstock materials, which were pelleted in their untreated form and after torrefaction. The feedstock were beech (*Fagus* ssp.) wood, poplar (*Poplar* ssp.) wood and corn cob (*Zea* ssp.), originated from Toulouse, France, and wheat straw (*Triticum* ssp.) from Mälardalen, Sweden. All feedstock were prepared for pre-treatment by shredded to 15 mm in size with an industrial shredder (Micromat 2000, Lindner Recyclingtech GmbH, Austria).

Torrefaction of feedstock was done at CEA (*Commissariat à l'énergie atomique et aux énergies alternatives*) and the chemical analysis at FCBA (*l'Institut Technologique Forêt Cellulose Bois-construction Ameublement*) in Grenoble, France. Pelleting and quality characterisation were performed at the Biomass Technology Centre of SLU (Sveriges lantbruksuniversitet) in Umeå, Sweden.

2.1. Torrefaction of lignocellulosic feedstock

Torrefaction of feedstock was carried out in the CENTORRE pilot-scale torrefaction unit (Fig. 1a). CENTORRE is a vertical six-stage multiple-hearth furnace (MHF) which runs under inert conditions at atmospheric pressure [41]. The MHF is directly heated with internal natural gas burners. The combustion gases (CO_2, CO) and vaporised H_2O maintain inert conditions in the furnace with N_2 available for emergency injection (Figure S1). The installation is fully instrumented for determining mass and energy balances. The furnace has an operating temperature range of 200–350 °C and a mass-flow capacity of 50–150 kg h^{-1}. Feedstock were torrefied in the range of 250–280 °C using residence times spanning 20–75 min (Table 2).

2.2. Pelleting

The pellet mill (Fig. 1b), the production process, the pilot-scale setup, the selection process of the appropriate PCL and the evaluation of pellet temperature and specific pelleting energy followed a systematic procedure and has been described in earlier work [42]. All pellet feedstock were reduced in size using a hammer mill (Vertica DFZK, Bühler Nordic, Sweden) with a 4 mm circular sieve size. Two PCLs and up to four moisture levels were used for each feedstock. The exception was torrefied straw and corn cob for which only one PCL was used. Pellet sampling was done in triplicate. In total, 180 batches of 8 mm diameter pellets were produced (84 batches of torrefied pellets and 96 batches of untreated pellets) and characterised for quality. The pellet production rate ranged from 121 to 188 kg h^{-1}. The reported production fines (expressed as mass per cent of produced pellets) were the amount of fine non-densified feedstock material exiting the pellet press. No fine tuning of production was done. Steam and other quality enhancing additives were not used, although permitted by ENplus standards up to two per cent level [43].

2.3. Quality analysis

Produced pellets were tested to comply with ENplus international pellet quality standards (Table 3).

Feedstock and pellet quality was analysed according to European standard (EN) methods for solid biofuels which included bulk density

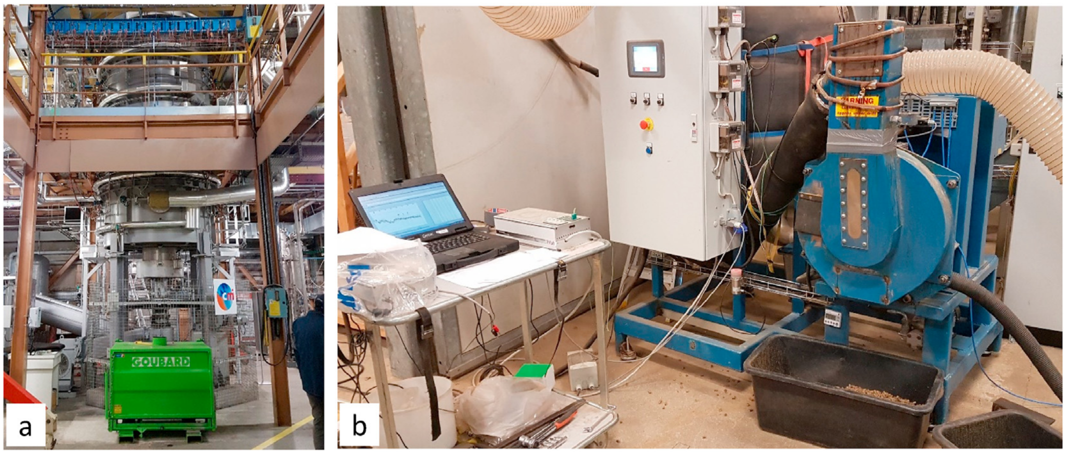


Fig. 1. The pilot-scale (a) CENTORRE torrefaction unit and (b) pellet mill and data acquisition system.

Table 3
ENplus pellet quality requirements [43].

Property	Unit	ENplus A1	ENplus A2	ENplus B
Diameter	mm	6–8		
Length	mm	$3.15 < L \leq 40$		
Moisture Content	% a.r	≤10		
Ash Content	% a.r	≤0.7	≤1.2	≤2.0
Mechanical Durability	% a.r	≥98.0	≥97.5	
Fines (<3.15 mm)	% a.r	≤1.0		
Net calorific value	MJ kg^{-1} a.r	≥16.5		
Bulk density	kg m^{-3}	≥600		
Additives	% a.r	≤2.0		
Nitrogen	% d.b	≤0.3	≤0.5	≤1.0
Sulphur	% d.b	≤0.04	≤0.05	
Chlorine	% d.b	≤0.02		≤0.03
Ash Deformation Temperature	°C	≥1200	≥1100	

Symbols refer to a.r = as received, d.b = dry basis.

(EN 15103), mechanical durability (EN 15210) and moisture content (EN 14774). Quality analysis of pellets was done the day after pelleting. Presented values on quality are averages from three replicated measurements. The energy density of pellets (as received) σ_{ar} (MJ m^{-3}) was calculated (Equation (1)) as a function of heating value (as received) LHV_{ar} (MJ kg^{-1}) and bulk density (as received) ρ_{ar} (kg m^{-3}).

$$\sigma_{ar} = LHV_{ar} \times \rho_{ar} \tag{1}$$

2.4. Chemical analysis

Chemical analysis of feedstock (Tables 4 and 5) was carried out according to standard methods for heating value determination (EN 14918), ultimate analysis (P CEN/TS 15104), inorganic constituents (P CEN/TS15290), ash content (XP CEN/TS 14775), monosugar composition (TAPPI T249 cm-85/ASTM E1758 – 2007) and lignin (TAPPI standard T222 om-83). The extractive content of the feedstock was determined using an accelerated solvent extractor both with water and acetone. The reported total extractives are the sum of extractives found with these two methods. The details of analysis procedures have been described earlier [44].

3. Results and discussion

The durability (DU) and bulk density (BD) of produced pellets as a function of feedstock moisture content are presented for the four investigated feedstock (Fig. 2). To assist discussion on torrefied pelleting results, there are general observations to be made regarding the pelleting of untreated lignocellulosic biomass. The first is the existence of an inverse relationship between pellet durability, whose data series have positive slopes, and pellet bulk

Table 4
Proximate and ultimate analysis of untreated and torrefied feedstock.

Feedstock	HHV (MJ kg^{-1})	LHV (MJ kg^{-1})	Ash (%)	C (%)	H (%)	N (%)	O (%)	S (%)	Cl (%)
Beech	18.2	16.9	0.8	49.1	6.0	0.2	43.0	<0.1	<0.02
Poplar	18.0	16.7	2.8	49.0	5.9	0.2	43.5	<0.1	0.1
Straw	18.9	17.8	9.3	44.6	5.6	0.5	39.9	0.1	0.5
Cob	18.5	18.0	1.9	47.4	6.1	0.4	44.1	<0.1	0.1
Beech 250	19.0	17.8	0.9	51.4	5.8	0.2	40.9	<0.2	<0.02
Poplar 250	19.3	18.2	NA	51.2	5.8	0.2	41.7	<0.1	NA
Poplar 280	22.6	21.5	NA	55.3	5.6	0.2	37.8	<0.1	NA
Straw 250	19.1	18.1	NA	55.8	3.1	0.6	40.4	0.1	NA
Cob 260	21.2	20.1	NA	50.3	5.9	0.3	42.3	<0.1	NA

Table 5
Macromolecular composition of untreated and torrefied feedstock.

Feedstock	Cellulose (%)	Lignin (%)	Hemicellulose (%)	Extractives, water (%)	Extractives, acetone (%)	Total extractives (%)	Glucan (%)	Xylan (%)	Mannan (%)	Galactan (%)	Arabinan (%)	Klason lignin (%)	Soluble lignin (%)
Beech	44.4	26.5	27.3	1.4	0.5	1.9	45.8	20.9	2.2	1.4	1.5	22.8	3.6
Poplar	45.6	26.9	23.2	1.7	2.7	4.4	47.4	15.6	2.9	1.4	1.5	24.5	2.4
Straw	42.3	22.4	27.4	1.1	6.7	7.8	42.6	22.6	0.2	1.1	3.2	21.1	1.3
Cob	40.5	15.9	36.7	0.5	6.4	6.9	40.5	30.4	0	1.9	4.4	12.6	3.3
Beech 250	35.1	31.9	19.4	3.7	2.6	6.3	37.0	13.4	3.1	0.6	0.4	29.9	2.0
Poplar 250	44.1	38.4	10.4	2.7	4.4	7.1	44.1	6.9	0.0	0.1	0.1	37.4	1.1
Poplar 280	34.1	56.7	4.91	1.7	3.4	5.1	34.7	2.0	0.6	0	0	56.0	0.8
Straw 250	26.2	46.1	10.8	6.6	3.6	10.2	29.2	10.8	0	0	0	45.2	0.9
Cob 260	39.7	33.5	18.1	6.5	4.0	10.5	39.7	13.1	0	0.3	1.4	31.8	1.7

Table 6
Pelleting data, pellet quality indicators and their relative variation of untreated and torrefied pellets, based on those pellets with the highest observed mechanical durability (EN 15210).

Feedstock	Pelleting data					Pellet quality indicators					
	MC (%)	PCL (mm)	E (kWh t^{-1})	T_p (°C)	fines (%)	DU (%)	BD (kg m^{-3})	MC_{ar} (%)	LHV_{ar} (MJ kg^{-1})	σ_{ar} (GJ m^{-3})	ENplus A1* deficiencies
Beech 250	7.9 (−5.2)	30 (−7.5)	85.5 (+18)	140 (+30)	1.3 (−0.5)	98.7 (+1.9)	669 (+63)	3.4 (−4.2)	18.2 (+3.1)	12.2 (+3.0)	ash
Poplar 250	11.7 (−2.9)	45 (−5.0)	56.0 (−15)	112 (−6)	1.4 (+0.9)	95.6 (−3.1)	682 (+43)	7.5 (−2.3)	16.3 (+2.0)	11.1 (+2.0)	DU, (ash)
Poplar 280	9.6 (−5.0)	38 (−12)	77.6 (+6.2)	120 (+2)	20 (+19)	87.5 (−11)	697 (+58)	4.9 (−4.9)	20.2 (+5.9)	14.1 (+4.9)	DU, (ash)
Straw 250	11.7 (−3.0)	30 (−25)	99.6 (+53)	131 (+31)	1.5 (+0.4)	94.5 (+1.0)	671 (+120)	5.2 (−6.1)	17.7 (+2.7)	11.9 (+3.6)	DU, (ash, Cl)
Cob 260	8.4 (−1.1)	30 (−25)	82.1 (+19)	135 (+12)	2.0 (+1)	94.2 (−2.7)	662 (−56)	4.3 (−1.8)	19.0 (+3.3)	12.6 (+1.3)	DU, (ash)

Variation between untreated and torrefied pellets are shown in parenthesis. Symbols refer to PCL = press channel length, E = relative pelleting energy, T_p = pellet temperature, fines = production fines, MC = moisture content of feedstock, DU = mechanical durability, BD = bulk density, MC_{ar} = moisture content of pellet (as received), LHV_{ar} = lower heating value (as received), σ_{ar} = energy density (as received), ash = ash content, Cl = chlorine content. Deficiencies in parentheses indicate assumed deficiency, based on untreated feedstock analysis. * A1 pellet quality (REF).

density, whose data series have negative slopes (Fig. 2). Pellet durability benefits from feedstock with higher moisture content (MC) because water acts as a binder between particles [45]. That durability is a function of feedstock moisture content originates from the role of water in facilitating hydrogen bonding. Chemical binding takes place on free OH groups [14] being most numerous in hemicellulose and cellulose (non-crystalline) components [46]. Water also lubricates the press channel, relieving the frictional forces that compact the feedstock so that a high moisture content hinders densification and bulk density.

Secondly, as the optimal moisture content is a compromise between the durability and density of pellets there exists a range of MC, in which pellet formation (production) is possible. For the untreated feedstock, this range was 8.7–14.6% for the hardwoods, 12.2–16.6% for straw and 7.4 and 13.1% for cob. The fact that pellets must be first be formed in order to evaluate their durability, defines the lower limits of these ranges. The upper limits are set by the final (equilibrium) MC of pellets which is defined by pellet quality standards; 10% for ENplus A1 (Table 3). Cereal straws and grasses (e.g. wheat straw, barley straw, corn stover and switchgrass) generally require a higher MC in pelleting than that of wood [47]. This trait and a high sensitivity to MC variation (i.e. the data series have a large slope compared to other feedstock) were seen in the results for untreated straw. This is likely due to poorer sorption of water due to both a high extractive (7.8%, Table 5) and high ash (9.3%, Table 4) content. This amplifies the influence of water on particle binding and has also been observed with forest residues which have similar amounts of these constituents [42].

Thirdly, the formation of pellets within the above MC ranges was possible using PCLs with two different lengths. In general, both higher durability and bulk density were obtained with longer PCL, this being due to greater resistance to extrusion through the longer channels. For hardwoods, high pellet quality (DU and BD) is strongly correlated with pellet temperature (T_p) [42]. As pellet formation becomes more successful, fewer production fines are observed, accompanied with physical indicators of good pellet quality. This includes low mechanical vibrations, low noise operation and stabilised power consumption of the press.

With the aforementioned observations in mind, the focus now shifts to the pelleting of torrefied feedstock. The specific pelleting data of torrefied pellets, quality indicators and their relative variation from untreated pellets are presented in Table 6 for those pellet batches which had the highest durability (Fig. 2).

3.1. Pelleting of torrefied feedstock

The durability of torrefied pellets was generally lower than that of their untreated counterparts, with beech being the exception. When considering only peak durability, straw was also exceptional. Bulk density improved markedly with specific increases from 43 to 120 kg m^{-3} with only corn cob showing a slight decrease. The peak durability values of torrefied pellets presented in Table 6 occurred at 3.4% lower feedstock MC on average compared to untreated feedstock. The hemicellulose content of feedstock decreased by 29–79% through torrefaction (Table 5). Destruction of the OH groups from hemicellulose degradation lowers the number of free binding sites and the equilibrium moisture content of feedstock [48]. The shift in peak pellet durability to lower MC can be attributed to this. For the hardwoods, there is a corresponding MC shift of the bulk density peak but for straw and cob, DU and BD do not peak in the same pellet batches. The shift to low MC accompanied a shift to shorter PCLs because frictional forces were enhanced with less water lubricating the channels. The pelleting energy reflected this by increasing from 6 to 53 kWh t^{-1} (9–114%) along with a 2–31 °C rise in extruded pellet

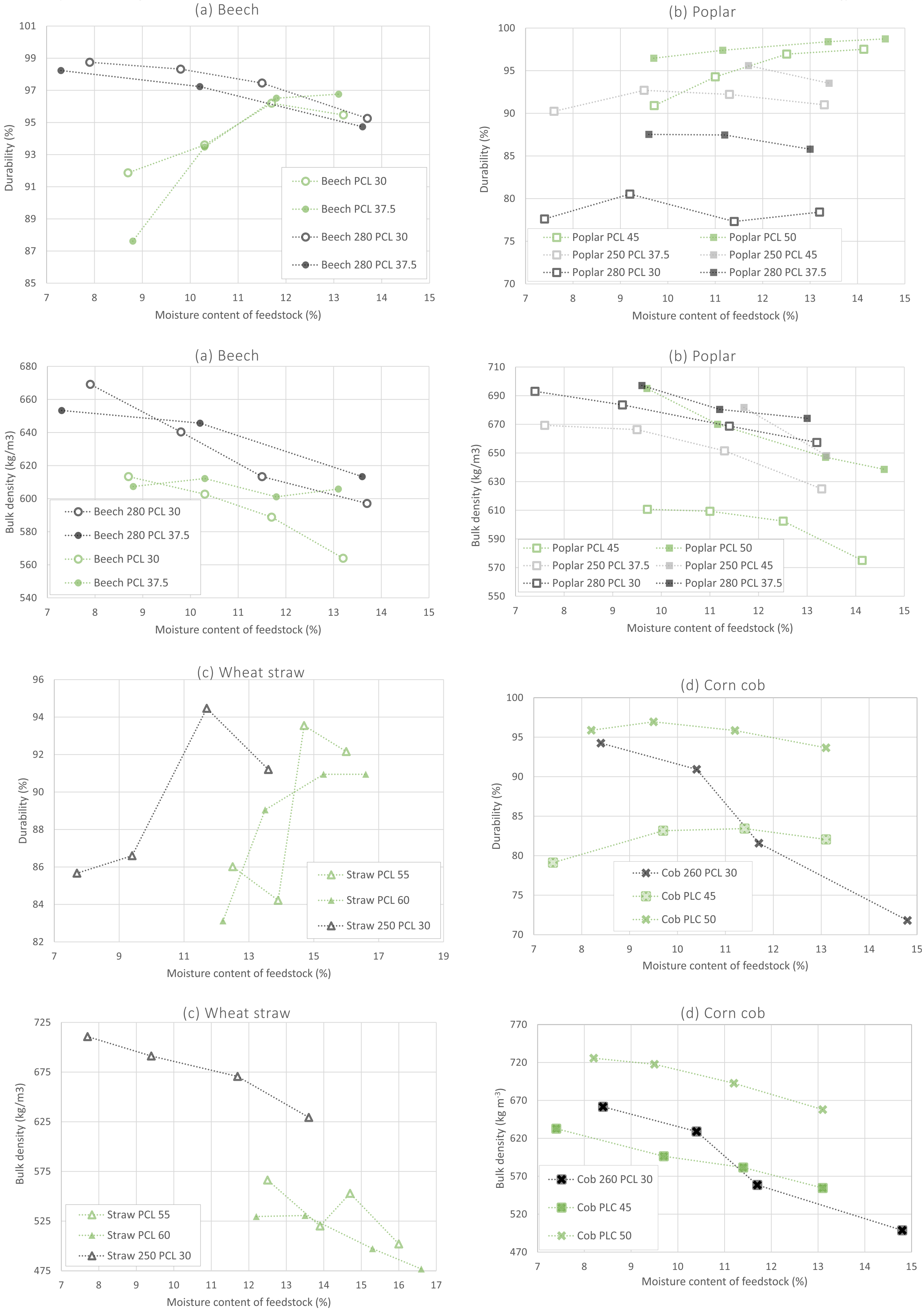


Fig. 2. Mechanical durability (EN 15210) and bulk density (EN 15103) of torrefied and untreated pellets of (a) Beech, (b) Poplar, (c) Wheat straw and (d) Corn cob at selected press channel lengths and as a function of feedstock moisture content. Each data point is an average of three replicate pellet batches. PCL = press channel length, expressed in millimetres.

temperature – itself a measure of heat attributed to friction and MC. Poplar 250 did not display this trend, instead showing a decrease in both pelleting energy and temperature. The two hardwood feedstock showed very different pelleting behaviour.

A high lignin content and its softening during heating is thought to play an important role in wood pellet durability [49]. The lignin content of the feedstock approximately doubled after torrefaction (Table 5) but this is a consequence of the Klason lignin method [50]. Unlike in the wood pellet case, the higher lignin content of torrefied material does not indicate it will exhibit good pellet binding nor high durability.

3.2. Behaviour of beech and poplar

Beech 250 pellets demonstrated a maximum durability of 98.7% and a bulk density of 669 kg m^{-3}, an increase from untreated pellets of 2% and 50 kg m^{-3} respectively. Torrefied poplar pellets had a maximum durability of 95.6% (poplar 250) but it worsened remarkably, from 3 to 21% from the untreated case – poplar 280 leading to the greatest decline. Indeed, poor durability was a trademark of poplar and contributed to a high production fines content (20%). Although the relatively high bulk density was moderately improved, peaking at 697 kg m^{-3}, it would appear that the higher degree of torrefaction was not beneficial to this feedstock. In contrast, the beneficial pellet properties of torrefied beech, under a comparable degree of torrefaction, have previously been observed [36] including a better carbon yield than poplar [51]. Beech 250 were the only pellets fulfilling the durability requirements of ENplus pellet standards (Table 3) yet the ash content of all feedstock was a barrier to A1 quality.

Why is beech well-suited to torrefaction and pelleting when poplar is not? The two species have strikingly similar elemental composition (Table 4) yet important differences in their macromolecular structure (Table 5). Despite the fact that beech has a greater hemicellulose fraction than poplar (27 versus 23%), more of which is xylan (21 versus 16%), the most thermally reactive of the hemicelluloses [52], it loses only 29% of its hemicellulose through torrefaction compared to a 55 and 79% loss in poplar 250 and 280. Torrefied poplar's poor durability can be explained primarily as an extreme case of OH-group degradation but there may be secondary factors. Compared to beech, poplar has a larger extractive content (1.9 versus 4.4%), a known inhibitor of pellet binding [53], and also a relatively high ash content (0.8 versus 2.8%) compared to temperate wood species [54]. As an inorganic material, ash does not participate in binding. The sum of the above factors may contribute to the observed quality differences between the hardwoods.

3.3. Behaviour of straw and corn cob

Straw showed the greatest improvements through torrefaction as straw 250 pellets had significantly higher durability (94.5%) and bulk density (711 kg m^{-3}) than pellets of untreated straw. Cob 260 pellets benefited solely through an LHV_{ar} increase (+3.1 MJ kg^{-1}) which bolstered its energy density (+1.2 GJ m^{-3}). Otherwise, pellet quality decreased substantially from untreated cob whose properties were comparable to previous densification studies [55]. Corn cobs, as a food crop residue, contain starches, water soluble carbohydrates and crude fats [56]. In addition to the lignocellulosic modifications in cob from torrefaction, the benefit of these natural thermally-sensitive [57] binders is likely neutralised. This may explain the good pelleting behaviour of untreated cob and the divergence of BD seen in comparing straw 250 and cob 260 pellets – a difference of some 180 kg m^{-3} (Table 6).

3.4. Future perspective

The produced torrefied pellets had an energy density (a.r) ranging from 11.9 to 14.1 GJ m^{-3}, increases from 1.2 to 4.9 GJ m^{-3} (11–54%) from untreated pellets (Table 6). This represents a large spatial concentration of fuel energy and agrees well with earlier findings (Table 1), which reported a torrefied pellet energy density (a.r) range of 10.7–14.4 GJ m^{-3} using woody feedstock; the highest value for beech wood torrefied at 270 °C for 40–45 min [36]. The results differ appreciably from earlier reports of 14.9–18.4 GJ m^{-3} (a.r) [58] which appear to be the source of the high expectations referred to by the IEA. The peer-reviewed findings to date, suggest that such energy densities combined with good pellet quality ($DU \geq 97.5\%$) are simply not obtainable. This statement is supported by the fact that, as indicated by the IEA [41], dozens of companies have not been able to demonstrate otherwise.

Is this epiphany a setback for the green energy revolution? Certainly not. The observed energy densities (a.r) in this study are much higher than for common renewable solid fuels such as stem wood chips (40% MC) (6.7 GJ m^{-3}) [59], seasoned (20% MC) birch fuel wood (6.1 GJ m^{-3}) [59] and commercial grilling charcoal (3.6 GJ m^{-3}) [60]. For comparison, consider the energy densities of fossil-based milled peat (3.2 GJ m^{-3}), lignite (12.8 GJ m^{-3}) and bituminous coal (17.8 GJ m^{-3}) [24]. As coal is replaced with renewable fuels in pulverised-fuel power plants, energy densities will influence on-site supply. For example, replacing bituminous coal with torrefied beech pellets would require an increase in storage volume by a factor of 1.46 (i.e. 17.8 GJ m^{-3}/12.2 GJ m^{-3}) in order to ensure the same period of operation between fuel deliveries. These differences will also affect fuel throughput of coal mills. 100% coal substitution using the pellets and determined energy densities herein (e.g. replacing bituminous coal with beech 250), the energy density differences translate to a needed 50% extra capacity of pulverisers in order to supply the same fuel energy throughput to the boiler.

Returning to the question posed by the IEA report: What needs to be understood in order to achieve a good quality product when pelleting torrefied lignocellulosic biomass? The results have shown that moderate degrees of torrefaction (250–280 °C, 20–75 min) had a large (mostly negative) influence on pellet durability yet a large (mostly positive) effect on bulk density. Based on these findings and in comparison with previous pilot-scale studies, this degree of torrefaction verges on optimal if product standards (i.e. DU and BD) are to be fulfilled (ENplus deficiencies are another matter). The results cast new light on previous studies, in which the pelleting process was not described [36,38]. Could better pellets have been produced if greater attention was given to pelleting? The inability of companies to produce good quality torrefied pellets, those fulfilling earlier claims [58] is understandable based on the results presented here. With a high torrefaction degree there is a trade-off at the expense of pellet durability and increased pelleting energy. But durability is also a function of feedstock. Fine-tuning of the pelleting process could likely improve pellet quality results (e.g. through use of additives [38] or process enhancements [61]). The pelleting energy of torrefied feedstock is clearly more energy intensive compared to untreated pellet production. This will have operational implications, for example, accelerated die wear, higher production (electricity) costs and be reflected in the emission footprint of pellets.

4. Conclusions

Torrefaction and pelleting technologies are vital links in the solid biofuel utilisation chain and enable a fast-track means of mitigating CO_2-equivalent emissions in society. This study generated specific knowledge on the densification of torrefied feedstock and how pellet production differs from that using untreated lignocellulosic biomass. A moderate degree of torrefaction strongly influenced the pelleting behaviour of beech, poplar, wheat straw and corn cob compared to the pelleting of these feedstock in their untreated form. The optimal moisture content for pelleting and suitable press channel lengths were substantially reduced with torrefied feedstock. Significant increases in the pelleting energy, extruded pellet temperature and the amount of production fines were observed in pelleting torrefied feedstock. Pellet quality showed feedstock-specific variations but was generally characterised by a decrease in mechanical durability and an increase in bulk density. The lower heating values (as received) of the feedstock improved through torrefaction (increases from 2.0 to 5.9 MJ kg^{-1}). These improvements, resulting from both torrefaction itself and the lower product moisture content, combined with bulk density improvements (increases from 43 to 120 kg m^{-3}) enabled improved energy densities for all feedstock. The energy density range (11.9–14.1 GJ m^{-3} as received) was comparable to that of lignite (12.8 GJ m^{-3}). Only pellets made of torrefied beech fulfilled the durability requirements and all torrefied pellets had an ash content exceeding the ENplus A1 limit. The results from this systematic pilot-scale study will help clarify earlier claims regarding properties of pellets made from torrefied lignocellulosic biomass.

CRediT authorship contribution statement

David A. Agar: Conceptualization, Methodology, Validation, Investigation, Data curation, Formal analysis, Writing – original draft, Writing – review & editing, Visualization. **Magnus Rudolfsson:** Methodology, Formal analysis, Investigation, Data curation, Writing – review & editing. **Simon Lavergne:** Investigation. **Thierry Melkior:** Resources. **Denilson Da Silva Perez:** Resources. **Capucine Dupont:** Resources, Writing – review & editing. **Matthieu Campargue:** Resources. **Gunnar Kalén:** Methodology, Investigation, Data curation. **Sylvia H. Larsson:** Writing – review & editing, Supervision, Project administration.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgements

This collaborative study was made possible through the EU Horizon 2020 research and innovation programme MOBILE FLIP 637020 and the Swedish Bio4Energy Strategic Research Environment. The authors warmly thank Markus Segerström for assistance with pelleting and characterisation at the Biomass Technology Centre. The French National Research Agency (ANR) is acknowledged for partial funding of the GENEPI platform within the framework Investments for the Future (Ref: ANR-11-EQPX-0019).

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.renene.2021.06.094.

Nomenclature

BD	bulk density (kg m^{-3})
C	carbon content (%)
Cl	chlorine content (%)
DU	mechanical durability (%)
H	hydrogen content (%)
HHV	higher heating value, also known as gross calorific value (MJ kg^{-1})
LHV	lower heating value, also known as net calorific value (MJ kg^{-1})
MC	moisture content, wet basis (%)
MHF	multiple-hearth furnace
N	nitrogen content (%)
O	oxygen content (%)
PCL	press channel length
S	sulphur content (%)
ρ_{ar}	bulk density, as received (kg m^{-3})
σ_{ar}	energy density, as received (GJ m^{-3})

References

[1] M.R. Allen, O.P. Dube, W. Solecki, F. Aragón-Durand, W. Cramer, S. Humphreys, M. Kainuma, J. Kala, N. Mahowald, Y. Mulugetta, R. Perez, M. Wairiu, K. Zickfeld, Global Warming of 1.5°C. An IPCC Special Report on the Impacts of Global Warming of 1.5°C above Pre-industrial Levels and Related Global Greenhouse Gas Emission Pathways, in the Context of Strengthening the Global Response to the Threat of Climate Change, Sustainable Development, and Efforts to Eradicate Poverty, 2018.

[2] A. EC, Policy framework for climate and energy in the period from 2020 to 2030, in: E. Commission (Ed.), 2014.

[3] BBC, Climate Change: China Aims for 'carbon Neutrality, 2020, p. 2060.

[4] J. Doat, Un noveau produit énergétique pour les pays en dévelopment: le bois torréfié, Rev. Bois et For. Trop. 208 (1985) 57–67.

[5] J. Kjarstad, F. Johnsson, The European power plant infrastructure - presentation of the Chalmers energy infrastructure database with applications, Energy Pol. 35 (2007) 3643–3664.

[6] G. Wang, J.G. Deng, Y. Zhang, Q. Zhang, L. Duan, J.K. Jiang, et al., Air pollutant emissions from coal-fired power plants in China over the past two decades, Science of the Total Environment, 2020, p. 741.

[7] D. Agar, J. Gil, D. Sanchez, I. Echeverria, M. Wihersaari, Torrefied versus conventional pellet production - a comparative study on energy and emission balance based on pilot-plant data and EU sustainability criteria, Appl. Energy 138 (2015) 621–630.

[8] M.R. Kabir, A. Kumar, Comparison of the energy and environmental performances of nine biomass/coal co-firing pathways, Bioresour. Technol. 124 (2012) 394–405.

[9] IPCC. reportClimate Change 2014: Synthesis Report. Contribution of Working Groups I, II and III to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change Geneva, Switzerland2014. p. 151.

[10] V. Repellin, A. Govin, M. Rolland, R. Guyonnet, Energy requirement for fine grinding of torrefied wood, Biomass Bioenergy 34 (2010) 923–930.

[11] M.J. Prins, K.J. Ptasinski, F. Janssen, Torrefaction of wood - Part 2. Analysis of products, J. Anal. Appl. Pyrol. 77 (2006) 35–40.

[12] T.G. Bridgeman, J.M. Jones, I. Shield, P.T. Williams, Torrefaction of reed canary grass, wheat straw and willow to enhance solid fuel qualities and combustion properties, Fuel 87 (2008) 844–856.

[13] T.G. Bridgeman, J.M. Jones, A. Williams, D.J. Waldron, An investigation of the grindability of two torrefied energy crops, Fuel 89 (2010) 3911–3918.

[14] T.C. Acharjee, C.J. Coronella, V.R. Vasquez, Effect of thermal pretreatment on equilibrium moisture content of lignocellulosic biomass, Bioresour. Technol. 102 (2011) 4849–4854.

[15] M. Phanphanich, S. Mani, Impact of torrefaction on the grindability and fuel characteristics of forest biomass, Bioresour. Technol. 102 (2011) 1246–1253.

[16] M.J. Prins, K.J. Ptasinski, F. Janssen, More efficient biomass gasification via torrefaction, Energy 31 (2006) 3458–3470.

[17] J. Deng, G.J. Wang, J.H. Kuang, Y.L. Zhang, Y.H. Luo, Pretreatment of agricultural residues for co-gasification via torrefaction, J. Anal. Appl. Pyrol. 86 (2009) 331–337.

[18] D. Agar, M. Wihersaari, Bio-coal, torrefied lignocellulosic resources - key properties for its use in co-firing with fossil coal - their status, Biomass Bioenergy 44 (2012) 107–111.

[19] J.H. Peng, X.T. Bi, S. Sokhansanj, C.J. Lim, Torrefaction and densification of different species of softwood residues, Fuel 111 (2013) 411–421.

[20] M.M. El-Wakil, Powerplant Technology, McGraw-Hill Inc., Singapore, 1984.

[21] S. Mani, L.G. Tabil, S. Sokhansanj, Grinding performance and physical properties of wheat and barley straws, corn stover and switchgrass, Biomass Bioenergy 27 (2004) 339–352.

[22] L.S. Esteban, J.E. Carrasco, Evaluation of different strategies for pulverization of forest biomasses, Powder Technol. 166 (2006) 139–151.

[23] M. Tamura, S. Watanabe, N. Kotake, M. Hasegawa, Grinding and combustion characteristics of woody biomass for co-firing with coal in pulverised coal boilers, Fuel 134 (2014) 544–553.

[24] Phyllis2, Database for (treated) biomass, algae, feedstocks for biogas production and biochar, in: T.N.O. ECN (Ed.), 2020.

[25] K.J. Loo van Sjaak, Handbook of Biomass Combustion and Co-firing, Twente University Press, Enschede, The Netherlands, 2002.
[26] A. Uslu, A.P.C. Faaij, P.C.A. Bergman, Pre-treatment technologies, and their effect on international bioenergy supply chain logistics. Techno-economic evaluation of torrefaction, fast pyrolysis and pelletisation, Energy 33 (2008) 1206–1223.
[27] G. Thek, I. Obernberger, Wood pellet production costs under Austrian and in comparison to Swedish framework conditions, Biomass Bioenergy 27 (2004) 671–693.
[28] Z. Dobo, A. Fry, Investigation of co-milling Utah bituminous coal with prepared woody biomass materials in a Raymond Bowl Mill, Fuel 222 (2018) 343–349.
[29] P. McNamee, L.I. Darvell, J.M. Jones, A. Williams, The combustion characteristics of high-heating-rate chars from untreated and torrefied biomass fuels, Biomass Bioenergy 82 (2015) 63–72.
[30] T.G. Bridgeman, L.I. Darvell, J.M. Jones, P.T. Williams, R. Fahmi, A.V. Bridgwater, et al., Influence of particle size on the analytical and chemical properties of two energy crops, Fuel 86 (2007) 60–72.
[31] J. Saastamoinen, M. Aho, A. Moilanen, L.H. Sorensen, S. Clausen, M. Berg, Burnout of pulverized biomass particles in large scale boiler - single particle model approach, Biomass Bioenergy 34 (2010) 728–736.
[32] Anttila M. Biohiilipellettien Ja Hiilen Jauhatus- Ja Yhteispolttokokeet 0.5 MW: N Pölypolttolaitteistossa. Biohiiliseminaari. Espoo, Finland2012.
[33] J. Li, A. Brzdekiewicz, W.H. Yang, W. Blasiak, Co-firing based on biomass torrefaction in a pulverized coal boiler with aim of 100% fuel switching, Appl. Energy 99 (2012) 344–354.
[34] P.J. Carl Wilén, Timo Järvinen, Kai Sipilä, Fred Verhoeff, Jaap Kiel, Wood Torrefaction – Pilot Tests and Utilisation Prospects, VTT Technical Research Centre of Finland, Kuopio, Finland, 2013.
[35] S.H. Larsson, M. Rudolfsson, M. Nordwaeger, I. Olofsson, R. Samuelsson, Effects of moisture content, torrefaction temperature, and die temperature in pilot scale pelletizing of torrefied Norway spruce, Appl. Energy 102 (2013) 827–832.
[36] T. Jarvinen, D. Agar, Experimentally determined storage and handling properties of fuel pellets made from torrefied whole-tree pine chips, logging residues and beech stem wood, Fuel 129 (2014) 330–339.
[37] M. Rudolfsson, E. Boren, L. Pommer, A. Nordin, T.A. Lestander, Combined effects of torrefaction and pelletization parameters on the quality of pellets produced from torrefied biomass, Appl. Energy 191 (2017) 414–424.
[38] L.E. Arteaga-Perez, H. Grandon, M. Flores, C. Segura, S.S. Kelley, Steam torrefaction of Eucalyptus globulus for producing black pellets: a pilot-scale experience, Bioresour. Technol. 238 (2017) 194–204.
[39] M. Rudolfsson, S.H. Larsson, T.A. Lestander, New tool for improved control of sub-process interactions in rotating ring die pelletizing of torrefied biomass, Appl. Energy 190 (2017) 835–840.
[40] B. Batidzirai, A.P.R. Mignot, W.B. Schakel, H.M. Junginger, A.P.C. Faaij, Biomass torrefaction technology: techno-economic status and future prospects, Energy 62 (2013) 196–214.
[41] J.K. Marcel Cremers, Middelkamp Jan, Joop Witkamp, Shahab Sokhansanj, Staffan Melin, Sebnem Madrali Status Overview of Torrefaction Technologies - A Review of the Commercialisation Status of Biomass Torrefaction, 2015.
[42] D.A. Agar, M. Rudolfsson, G. Kalen, M. Campargue, D.D. Perez, S.H. Larsson, A systematic study of ring-die pellet production from forest and agricultural biomass, Fuel Process. Technol. 180 (2018) 47–55.
[43] ENplus® Handbook Version 3.0. Part III: Pellet Quality Requirements.
[44] S. Jacob, D.D. Perez, C. Dupont, J.M. Commandre, F. Broust, A. Carriau, et al., Short rotation forestry feedstock: influence of particle size segregation on biomass properties, Fuel 111 (2013) 820–828.
[45] N. Kaliyan, R.V. Morey, Factors affecting strength and durability of densified biomass products, Biomass Bioenergy 33 (2009) 337–359.
[46] H. Bjork, A. Rasmuson, Moisture equilibrium OF wood and bark CHIPS IN superheated steam, Fuel 74 (1995) 1887–1890.
[47] S. Mani, L.G. Tabil, S. Sokhansanj, Effects of compressive force, particle size and moisture content on mechanical properties of biomass pellets from grasses, Biomass Bioenergy 30 (2006) 648–654.
[48] T.K. Shoulaifar, N. DeMartini, A. Ivaska, P. Fardim, M. Hupa, Measuring the concentration of carboxylic acid groups in torrefied spruce wood, Bioresour. Technol. 123 (2012) 338–343.
[49] S. Frodeson, G. Henriksson, J. Berghel, Pelletizing pure biomass substances to investigate the mechanical properties and bonding mechanisms, Bioresources 13 (2018) 1202–1222.
[50] T.K. Shoulaifar, N. DeMartini, S. Willfor, A. Pranovich, A.I. Smeds, T.A.P. Virtanen, et al., Impact of torrefaction on the chemical structure of birch wood, Energy Fuel. 28 (2014) 3863–3872.
[51] D. Agar, N. DeMartini, M. Hupa, Influence of elevated pressure on the torrefaction of wood, Energy Fuel. 30 (2016) 2127–2136.
[52] M.J. Prins, K.J. Ptasinski, F. Janssen, Torrefaction of wood - Part 1. Weight loss kinetics, J. Anal. Appl. Pyrol. 77 (2006) 28–34.
[53] N.P.K. Nielsen, D.J. Gardner, C. Felby, Effect of extractives and storage on the pelletizing process of sawdust, Fuel 89 (2010) 94–98.
[54] E. Sjöström, Wood Chemistry – Fundamentals and Applications, Academic Press Inc., London, UK, 1993.
[55] M.T. Miranda, F.J. Sepulveda, J.I. Arranz, I. Montero, C.V. Rojas, Analysis of pelletizing from corn cob waste, J. Environ. Manag. 228 (2018) 303–311.
[56] N. Kaliyan, R.V. Morey, Densification characteristics of corn cobs, Fuel Process. Technol. 91 (2010) 559–565.
[57] J.C. Huang, Y. Qiao, Z.Q. Wang, H.P. Liu, B. Wang, Y. Yu, Valorization of food waste via torrefaction: effect of food waste type on the characteristics of torrefaction products, Energy Fuel. 34 (2020) 6041–6051.
[58] P. Bergman, Combined Torrefaction and Pelletisation – the TOP Process, Energy Research Centre of the Netherlands, Petten, The Netherlands, 2005.
[59] E. Alakangas, Properties of Wood Fuels Used in finland - BioSouth Project Report, 2005.
[60] Z. Jelonek, A. Drobniak, M. Mastalerz, I. Jelonek, Environmental implications of the quality of charcoal briquettes and lump charcoal used for grilling, Sci. Total Environ. 747 (2020) 141267.
[61] M. Rudolfsson, D.A. Agar, T.A. Lestander, S.H. Larsson, Energy savings through late-steam injection - a new technique for improving wood pellet production, J. Clean. Prod. (2020) 254.