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# Combined effects of torrefaction and pelletization parameters on the quality of pellets produced from torrefied biomass



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#### HIGHLIGHTS

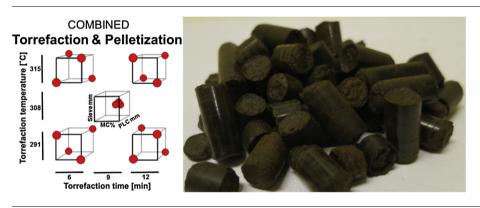
- Pilot scale torrefaction × pelletization experiments combined for the first time.
- To emphasize industrial applicability, a continuous torrefaction process was used.
- Torrefaction was done at high temperatures and short times favoring high throughput.
- Effect of torrefaction, moisture, and die channel length on pellet quality analyzed.
- Production windows for torrefied wood and its pelletization are identified.

#### ARTICLE INFO

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#### G R A P H I C A L A B S T R A C T



#### ABSTRACT

A combined torrefaction and pelletization study was performed at industrially relevant settings using a factorial design. First, wood chips of Scots pine were torrefied at high temperatures (291–315 °C) and short residence times (6–12 min), facilitating high throughput in a continuous pilot-scale torrefaction process. Then the torrefied materials were pelletized, also in pilot-scale, using varying moisture contents (MCs) (10–14%), sieve sizes (4–6 mm), and press channel lengths (PCLs) (25 and 30 mm), in all 19 batches, each of 400 kg. The resulting so called black pellets exhibited bulk densities of 558–725 kg m<sup>-3</sup>, durabilities of 46.3–86.5%, and fines contents of 3.8–85.8%.

Through multiple linear regression modelling of all 11 responses, it was found that the parameter with the greatest influence on the responses was the torrefaction temperature, followed by torrefaction time, MC, and PCL. Longer PCL and higher MC resulted in higher pellet quality, with less fines and greater bulk density and durability. Furthermore, a low torrefaction degree decreased the amount of power required for pelletization. The energy required to grind pellets into a powder (<0.5 mm) decreased with increasing torrefaction degree as expected, but also with decreasing MC before pelletizing. Pyrolysis-GC/MS analysis of thermal degradation products from the pellets revealed correlations with the torrefaction temperature and time, but no correlations with the pelletization process.

These results are useful for mapping chemical changes in torrefied materials and identifying complementary torrefaction and pelletization settings. Specifically of interest is adjustment of PCLs at low intervals to better match friction properties of torrefied materials.

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Torrefaction is a process that upgrades lignocellulosic biomass and can be combined with densification (pelletization or briquetting) to produce energy carriers with significantly improved fuel characteristics compared to raw biomass [1-4]. The process involves thermal treatment of biomass at 200-350 °C in an inert or oxygen-reduced atmosphere for a defined residence time [5-7]. This increases the fuel's gross calorific value and hydrophobicity [5,8], while reducing the required grinding energy [9-12] as well as the fuel's susceptibility to fungal and microbial degradation [13,14], all of which are beneficial with respect to storage, transport, and end-use. Although torrefaction increases the energy content of the biomass per unit weight, a subsequent densification step markedly improves the energy density per unit volume, thereby facilitating logistics throughout the supply chain. Pelletization of torrefied materials produces what we herein refer to as "black pellets" and has a positive economic impact at multiple points in the supply chain [2,4]. Moreover, under certain transportation conditions, black pellets have lower CO<sub>2</sub>-equivalent emissions than conventional (white) pellets according to life cycle assessments [3]. As the "torrefaction degree" (a parameter whose value increases with both the torrefaction temperature and the residence time) increases, the black pellets become more similar to bituminous coal with respect to key fuel gualities such as heating value, fragility, powder flowability, and bulk density [9]. These improved properties allow black pellets to be co-fired with coal at high ratios making them a promising fuel substitute in coal-fired power plants [15,16], as well as a potential feedstock for entrained flow gasification [17–19].

Although torrefaction enhances the feedstock's fuel properties, the changes it causes present challenges when fine tuning the densification process, e.g. to find the optimum settings of moisture content and press channel length. Pelletization of white pellets has been suggested to depend on several factors including: (1) the feedstock's chemical and physical properties, e.g. its contents and quality of extractives, lignin, and fibers [20] and particle size distribution [21], (2) the feedstock's pre-treatment e.g. initial moisture content [22,23], the use of additives [24] or pre-heating [20], and (3) the pelletization equipment employed e.g. pellet press type, die temperature, and press channel dimensions [1,20,23,25,26]. The agglomeration of the lignocellulosic materials to form pellets is suggested to result from several different interparticle bonding mechanisms favored by the softening of different components at high pressures and temperatures [27,28]. The known bonding forces in pellets include solid bridges, attractive forces between solid particles, particle interlocking, and forces of adhesive, cohesive, and interfacial forces [20].

It is also well recognized that torrefaction significantly influences the molecular composition of biomass: some of its components are volatilized, some are thermally disintegrated and others undergo major changes in structure and composition [28– 31]. Extractives start degrading and volatilizing at 100–250 °C, hemicelluloses at 200-230 °C with complete degradation at around 245 °C, and cellulose at 350-500 °C. Lignin degradation begins at 350 °C and continues beyond 500 °C, although it may undergo small changes starting at 200-270 °C [6,29-31]. Therefore, extractives, which have plasticizing and lubricating effects during pelletization, may be largely lost during torrefaction. In addition, hemicelluloses are disintegrated, which has been reported to remove vital hydroxyl groups that may function as moisture binding agents [32,33]. Lignin is recognized to be crucial for pellet strength [1] and is used as an additive to enhance binding and increase pellet quality [24,28,34-36]. Because lignin is the last component to degrade, its abundance relative to the hemicellulose

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[28] but it undergoes several chemical rearrangements including dehydration, bond cleavages, demethoxylations, and condensation reactions [30,37]. Pelaez-Samaniego et al. [28] showed that the migration of lignin liquid intermediates (LLI) and their deposition on cell surfaces depended on the torrefaction degree, suggesting that the torrefaction conditions could be optimized to produce LLI in quantities favorable for downstream pelletization. However, it is currently not completely clear how the material changes during torrefaction or how specific bonding mechanisms affect densification.

A few single-pellet press studies of black pellet production from torrefied biomass have highlighted factors that make it difficult to consistently produce high quality pellets [21,32,33,38-42]. However, Kiel [43] and van der Stelt et al. [44] claim that torrefaction improves the mechanical properties of the resulting pellets because of their higher lignin content. Stelte et al. [32] and Li et al. [33] showed that pelletization of torrefied biomass required more energy than that of non-torrefied material due to the increased friction in the press channel. Misljenovic et al. [41] also reported a greater energy requirement during pelletization, but found that torrefaction increased pellet strength for some raw materials while reducing it for others. Peng et al. [40] found it more difficult to compress torrefied samples into strong pellets than the raw material, and suggested either a higher die temperature or adding moisture into torrefied particles to improve the compression process. Another approach was taken by Duncan et al. [45] and Nicksy et al. [46] who investigated the potential of making spherical torrefied pellets to reduce abrasion and thus fines production during transport.

While maintaining process control of many pelletization parameters is relatively straightforward in a single pellet press, larger continuously fed systems such as pilot scale ring die systems and conventional industrial pellet presses offer only a few controllable parameters, such as the press channel geometry. Many key parameters such as the die temperature cannot easily be directly controlled, which creates various challenges when scaling up black pellet production [42]. In addition, pilot scale pelletization is laborious, which makes it essential to maximize the knowledge gained from each experimental campaign. In order to determine causeeffect relations and identify optimal production conditions, there is a need for more dedicated parametric studies on black pellet production on pilot, demo, and industrial scales.

Parametric studies using factorial experimental designs [47], can be used to identify important parameters, optimize process settings based on quality responses, and assess process stability [48]. For example Duncan et al., [46] used a factorial experimental design to simultaneously study several different quality responses during the production of spherical pellets of torrefied material, revealing that the torrefaction temperature was by far the parameter that most strongly influenced pellet quality.

Similarly, Rudolfsson et al. [49] demonstrated in a parametric study on single press pellets of torrefied spruce that high quality pellets could only be produced within a narrow design space. They emphasized the need for industrial optimization, particularly with respect to the challenge of maintaining a consistent moisture content throughout the production process. Larsson et al. [50] were among the first to perform a pilot-scale parametric pelletization study using torrefied materials. They used a full factorial design and a ring die press to study the influence of torrefaction temperature and the material's moisture content on the resulting pellet quality. However, the number of parameters included in the study was limited by the low capacity of the pilot torrefaction facility ( $20 \text{ kg h}^{-1}$ ); consequently, a decision was taken to expand the number of influential parameters and adjust the design space.

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