

FUEL PROPERTIES AND SUITABILITY OF *EUCALYPTUS BENTHAMII* AND *EUCALYPTUS MACARTHURII* FOR TORREFIED WOOD AND PELLETS

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Torrefaction is the process of heating a material in the absence of oxygen, a pretreatment that represents a promising option for biofuels. Two eucalyptus species harvested in South Carolina, *E. benthamii* and *E. macarthurii*, were processed in a torrefier, and wood pellets were manufactured. Eucalyptus represents a promising biomass source in southern U.S. due to fast growth rates and the availability of cold-tolerant plantations. Analyses of moisture content, proximate and elemental composition, and net heating value of “light roasted” wood were assessed. The heating value of the eucalypts and pellets was enhanced by 19% (average), compared to the original material, while the moisture and volatiles content were drastically reduced. This reduction leads to an increase in the amount (w/w) of carbon, enhancing the energy content in the material. Thus, torrefaction is useful for improving the heating value of woody biomass, consuming little external energy due to recirculation and burning of gases for the process. The pellets showed increased energy density, providing improved properties for transportation and handling.

Keywords: Eucalyptus; Torrefied wood pellets; Proximate analysis; Ultimate analysis; Heating value

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INTRODUCTION

Torrefaction of wood is one of the most readily available bio-technologies (Sklar 2009), being an attractive pretreatment option for biomass intended for conversion and combustion processes (Zwart et al. 2006), with results more financially attractive than traditional pelletization of biomass, when comparing their economic feasibilities (Bergman and Kiel 2005). Torrefaction can be defined as a “mild pyrolysis” (Sklar 2009) in which the heating process produces degradation of hydrophilic polysaccharides (hemicellulose), reducing the hydroxyl radicals where water molecules typically attach or “bind” to the wood, along with a relative increase in lignin content in relation to the initial mass (which further increases the hydrophobic changes in the material) (Hakkou et al. 2006; Mburu et al. 2007). Wood typically begins to degrade at 180°C, releasing CO₂, acetic acid, and phenolic compounds (Girard and Shah. 1991), while hemicellulose begins degrading at 225°C (Rowell et al. 2005). Fonseca et al. (1998) described torrefaction as a slow heating of biomass in an oxygen-free atmosphere with a maximum temperature of 300 °C. The VOCs and hemicellulose fractions are combusted to generate heat, leaving cellulose and lignin to constitute the torrefied wood, which has a charcoal-

like appearance. Depending on the residence time of the wood in the torrefier, the torrefied wood yield can be high, varying between 66% and 75% (Sklar 2009).

The process of torrefaction applied to woody biomass is not a new concept, being evaluated as early as 1930, when Basore (1930) proposed a method for profitable disposal of large amounts of southern pine sawdust generated from sawmills in the southern U.S. The method proposed is very similar in concept to the actual parameters utilized in a typical modern torrefaction process, and the inventor even proposed the “densification” as briquettes, of the charcoal-like material obtained, in order to optimize handling, transportation, and utilization of the biomass. Furthermore, Basore and Moore (1942) proposed an additional refined method for the production of lump charcoal from pine sawdust. Despite these early efforts, torrefaction was not widely implemented as a process for biomass conversion or properties optimization. Instead, the torrefaction of biomass was overlooked and is still at an early stage in industrial applications, with limited commercial availability (Uslu et al. 2008).

In the last two decades, more research has been performed regarding the characteristics of the material produced in the torrefaction process (Bourgeois and Guyonnet 1988; Gevers et al. 2002), as well as the gases obtained in the heating chamber (Pach et al. 2002; Uslu et al. 2008); however, very little literature can be found regarding large-scale (industrial) manufacturing of torrefied wood. Few species have been tested, with most of the attention being focused on birch and southern pine (Bourgeois and Guyonnet 1988; Pach et al. 2002). In addition, the development of pretreatment options such as torrefied pellets has been deemed necessary by several large-scale biomass users, scientists, and members of the IEA Task 40 on sustainable international bioenergy trade (Junginger and Sikkema 2008). Among the types of biomass with promising properties for bioenergy production in the U.S. are some *Eucalyptus* species (Gonzalez et al. 2008, 2011b) and its utilization in pretreatments such as torrefaction should be evaluated in detail (Gonzalez et al. 2011c,d). Countries such as Brazil have already shown the potential that such short-rotation crops may have for the bioenergy industry (Couto et al. 2004; Rosillo-Calle 2004; Wright, L. 2006; Gonzalez et al. 2008). Recent advances in characteristics such as coppicing, cold tolerance, and growing rates make eucalypts an ideal biomass source for energy production in southern U.S. (Gonzalez et al. 2008; Wright, J. 2009, Gonzalez et al. 2011a,b). Typical plantations of eucalyptus can yield around 450 GJ ha⁻¹ year⁻¹ of energy, while traditional wood sources from commercial forests in the U.S. yield around 100 GJ ha⁻¹ year⁻¹ (Moreira 2006). Eucalypts have a rotation period (for bioenergy purposes) of less than 7 years (Carvalho et al. 2009). Despite these characteristics, very few species have been assessed for their properties for the bioenergy industry, especially in combination with pretreatments such as pelletization and torrefaction, and only Brazil has been consistently converting eucalyptus to charcoal for the pig iron and steel industry (Wright, L. 2006). With many Eucalyptus plantations established in the last 10 years in southern U.S. states such as South Carolina, Florida, Georgia, Alabama, and Louisiana (Wright, J. 2010; Gonzalez et al. 2011c,d; Pirraglia et al. 2011), and an increasing need to produce bioenergy from sustainable sources, it is necessary to perform further research in a timely manner evaluating the properties and suitability of eucalyptus species for bioenergy production. In view of these facts, two eucalyptus species, *E. benthamii* and *E. macarthurii* from a plantation in South Carolina

were assessed for its basic properties for biofuels production (Pirraglia et al. 2011), obtaining promising results for its suitability as a solid biofuel for power and heat generation; thus additional research needs to be performed in order to fully assess the properties of this material when subject to torrefaction and further pelletization pretreatments.

The main objective of this research was to assess the characteristics of torrefaction as biomass pretreatment in two cold-resistant eucalyptus species (*E. benthamii*, and *E. macarthurii*), measuring and comparing physico-chemical characteristics of the torrefied wood material. An additional objective of the research was to manufacture and evaluate the characteristics of torrefied wood pellets from the eucalyptus samples, torrefied in a large-scale torrefaction unit, and pelletized in a laboratory-scale pellet mill with the addition of binders, assessing the main fuel characteristics of these pellets.

MATERIALS AND METHODS

In previous work performed by the authors (Pirraglia et al. 2011), *E. benthamii* and *E. macarthurii* were evaluated for their raw material properties, and pellets were manufactured with the addition of binders. In the present work, it is of particular interest to further evaluate properties of the species when subjected to a torrefaction process, and pelletization with binders. In order to compare the original raw material and the processes applied to them, the same properties of the samples must be evaluated, utilizing the same procedures and standards. The following properties were evaluated for both the torrefied material, and the torrefied pellets with the addition of binders for the two species: moisture content of the torrefied material after cooling, proximate analysis (volatiles content, ash content, and fixed carbon content), ultimate analysis (C-H-O-N content), and high heating value (HHV). The procedures and standards used for the determination of these properties are fully described below. In order to achieve the proposed objectives, several ASTM standards were used to determine several properties of torrefied wood from *E. benthamii* and *E. macarthurii*, and for the determination of properties of wood pellets produced with the torrefied wood of *E. benthamii*. Such standards are briefly described below for each test performed:

- **Moisture Content Determination:** Determination of Moisture content is based on the ASTM Standard D 4442 (ASTM International. 2002a), Test Methods for Direct Moisture Content Measurement of Wood and Wood-based Materials. Samples were taken from torrefied wood produced with each species; and measures of its weight before and after drying were taken to a precision of ± 0.1 grams. Samples were dried in an oven at 103 ± 2 °C until there was no appreciable change in weight. Three replicates were completed for each species of torrefied *E. benthamii* and *E. macarthurii*.
- **Volatile Matter:** Determination of volatiles was performed following the ASTM D 3175-07, Standard Test Method for Volatile Matter in the Analysis Sample of Coal and Coke (ASTM International. 2009a). Three replicates were performed for each torrefied wood produced from *E. benthamii* and *E. macarthurii*. A Omegalux LMF-

3550 muffle furnace was utilized for this test, as well as for ash content and fixed carbon determination (Omega. 2011)

- **Ash Content:** Ash Content determination was performed following ASTM D 3174-04, Standard Test Method for Ash in the Analysis Sample of Coal and Coke from Coal (ASTM International. 2009b). Three replicates were used for each experiment in each species.
- **C-H-O-N Analysis:** The analysis for Carbon, Hydrogen, Oxygen, and Nitrogen of the torrefied wood produced was performed according to the standard ASTM D 3176, Standard Practice for Ultimate analysis of Coal and Coke (ASTM International. 2009c). Oxygen is back-calculated, assuming that the entire composition of the samples is reduced to these four elements (sulfur content is assumed negligible). Three replicates were analyzed for the torrefied wood of each species. A Perkin-Elmer Corporation's CHN Elemental Analyzer, model 2400 was utilized in performing this test (PerkinElmer. 2011; Hodge et al. 1991)
- **High Heating Value (HHV):** The High Heating Value of the torrefied wood produced was calculated following the ASTM Standard D 2015, Standard Test Method for Gross Calorific Value of Coal and Coke by the Adiabatic Bomb Calorimeter. A Parr Instruments 1108P Oxygen Combustion Bomb, and a 1341 Oxygen Bomb Calorimeter were used for determining this property (Parr Instruments. 2011)

The original biomass was collected during winter of 2010 at a plantation in Summerville, South Carolina, and present specimens of 8 years of age, with average basic densities of 0.55g/cc (± 0.04 g/cc) for *E. benthamii*, and 0.54g/cc (± 0.04 g/cc) for *E. macarthurii*, and an average of 10-12 inches in diameter at breast height (DBH). These specimens are part of a fast-growing, short rotation trials for bioenergy production, and are also freeze-tolerant specimens, being resistant to temperatures as low as 16°F (-8° to -9° C) (Arborgen 2010). Upon arrival, the eucalyptus biomass of both species was prepared by grinding the received (debarked) logs utilizing a WEIMA Horizontal Beaver 300 grinder with a ¾" screen size (WEIMA America, Inc., 2011), making it possible to obtain wood chips of size between 20 and 50 mm. In addition, the biomass was air-dried from its initial moisture content of ca. 54% to a moisture content of 18 to 20%. This particle size and moisture content allows for better flow of material in the feeding system of the torrefier unit. Additional characteristics of the original biomass, such as elemental composition, proximate analysis and heating value are detailed reported by Pirraglia et al. (2011).

Below, a brief description of the torrefied process applied to the eucalyptus biomass is given in order to adequately interpret the results of the experiments performed for determining the properties of torrefied eucalyptus. The torrefier unit utilized for the experiments was designed and developed at NC State University. The unit operates by heating the wood in a low-oxygen environment, producing irreversible changes in the wood. According to Brito et al. (2008), these changes occur in five phases. The first phase occurs at 100°C and involves the evaporation of free and bound water in the wood; the second phase occurs between 100°C and 250°C, producing changes in the OH groups of the wood constituents, and causing irreversible wood degradation. The third phase

occurs between 250°C and 330°C, and involves the total degradation of hemicelluloses, and significant modifications of lignin, with reduction of the β -O-4 linkage between units and the formation of resistant linkages (Nakamura et al. 2008; Rousset et al. 2009). In the fourth phase, it is assumed that the destruction of cellulose occurs between 330°C and 370°C; and the fifth phase (temperatures above 350°C) produces material with charcoal characteristics, and involves the most intense degradation of lignin (Guedira 1988). In the torrefactor utilized at North Carolina State University, only a partial 3rd phase is reached, by achieving an average temperature of 280°C, partially degrading the Hemicellulose (HC) and releasing around 30% of the original volatile matter (VOC's). The VOC's and HC are combusted to generate 80% of the torrefaction process heat. Figure 1 demonstrates a schematic view of the torrefaction process for the unit designed at NC State University (James 2009).

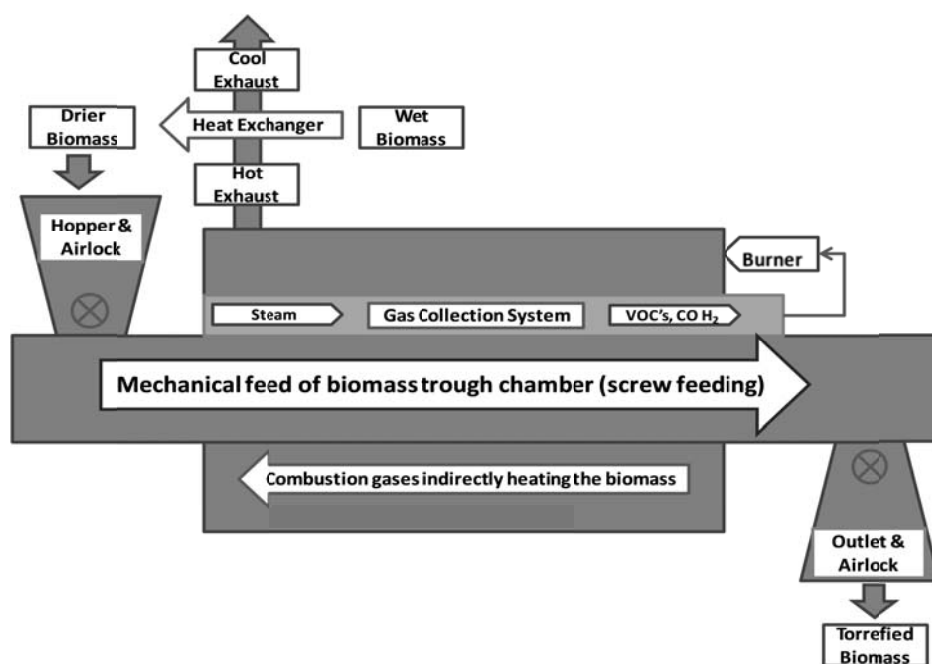


Figure 1. Schematic process for the production of torrefied eucalyptus

One of the most important variables affecting the torrefied wood that can be produced with this unit, along with the temperature of the chamber, is the residence time. The residence time determines the degree to which the wood is “roasted”, and its properties modified. The longer the residence time, the more coal-like the material is going to be obtained at the exit of the machine. This residence time also determines the yield. If the residence time is long (between 5 and 6 minutes), then most of the VOCs from the wood are removed, leaving a charcoal material with highly increased heating value, and extremely low moisture content, which is regained at the exit and posterior cooling of the torrefied wood, even when the material becomes hydrophobic (Bergman et al. 2005). It is advisable that characteristics from the torrefaction process, such as mass and energy balance for different torrefaction levels, must be further studied; variables such as residence time, temperature, and characteristics of the torrefied product are of

extreme importance to obtain a uniform product, and may provide improved results and more consistent comparisons between both species.

In order to establish the most similar conditions for an industrial-level process with continuous and high-volume flow, the machine was set up for low residence times, being three minutes from the instant in which the material enters the unit through the hopper, to the moment in which it exits the machine. These three minutes are the lowest residence time utilized in the machine in order to obtain a torrefied material. The result of a material with this residence time is described as “light roast” or “slightly torrefied”. In order to adequately define the level or “degree” of torrefaction, and make the results of this project reproducible, three variables are considered: Residence time, torrefaction temperature, and mass yield. The residence time was set at three minutes, as previously described, while the torrefaction temperature was set at 280°C, resulting in a mass yield between 80 and 85%. According to Almeida et al. (2010), the mass loss, or mass yield in a torrefaction process is an excellent indicator of the intensity of the treatment, making it possible to predict and calculate most of the energy properties of the biomass as a function of the overall mass loss; and it has been previously tested in eucalyptus species. In addition, the research team is attempting to establish a color scale in order to determine the “roast” degree by observing the change of color on the wood at the exit of the unit, changing from brown to black in different scales and depending on the particle size of the wood (James 2009). Some other properties that can be measured are applied to the torrefied wood in order to define the “roast” degree; a “lightly roast” is usually defined as torrefied wood that contains around 60 to 70% of the original VOCs content of the wood, and a carbon content 20 to 30% higher than the original carbon content, thus enhancing the heating value of the wood. Also, when considering a mass balance, or mass yield, the light torrefaction process has a yield of 80 to 85% compared to the original mass (W/W). In order to facilitate the understanding and discussion of the results, a code is used to define the species of torrefied wood. Table 1 describes the codes used and their meanings.

Table 1. Codes used for Identification of each Torrefied Wood According to the Species

Code	Torrefied wood description
EMT1	1 st replica of <i>E. macarthurii</i> tests, 3 samples per replica
EMT2	2 nd replica of <i>E. macarthurii</i> tests, 3 samples per replica
EMT3	3 rd replica of <i>E. macarthurii</i> tests, 3 samples per replica
EBT1	1 st replica of <i>E. benthamii</i> tests, 3 samples per replica
EBT2	2 nd replica of <i>E. benthamii</i> tests, 3 samples per replica
EBT3	3 rd replica of <i>E. benthamii</i> tests, 3 samples per replica

To achieve the additional objective of pelletizing the torrefied biomass, two different binders were added to the torrefied wood of *E. benthamii*, resulting in two pellet trials. To prepare the torrefied material for pelletization, further particle size reduction was necessary, and was performed with a C.S. Bell No. 20 Cast iron hammer mill with blower discharge (The C.S. Bell Milling & Grinding Co., 2011). This hammer mill was equipped with a 3 mm screen in order to obtain this particle size in the torrefied material.

After particle size reduction, the material was mixed with the binders in a tumbler mixer, and fed into a flat-die PP220 pellet mill (Pellet Pros. 2010), obtaining pellets short in length (10-15 mm), with 6 mm diameter, solid, and with few fines observed.

Due to the characteristics of the torrefied material, in terms of being abrasive and difficult to pelletize in a laboratory scale unit, this trial was only performed with *E. benthamii*. These pellets were studied using the same tests and procedures as the ones previously described for the evaluation of the raw material, gaining understanding of the differences produced when pelletizing torrefied wood with the addition of binders.

The binders utilized for the pellets were Dried Distiller Grain (DDG), and soybeans, with the addition of water to increase the moisture content of the wood prior to pelletizing. Dried Distillers Grains present good properties for gluing the wood particles together in pellets, due to its content of condensed distiller's solubles, including fats and starch (Weiss et al, 2007; Kingsly et al, 2010). Soybeans have also been successfully used as binder in the wood industry, especially wood composites (United Soybean Board, 2006), and present good characteristics for pelletizing wood (PelletPros, 2007). Details on the formulations for the pellets are provided below.

Previous work has been performed by the authors regarding pellets manufacturing from *E. benthamii* and *E. macarthurii* with the addition of binders, and pelletized in a flat-die mill from Pelletpros Inc., model PP220 (PelletPros, 2010; Pirraglia et al. 2011). Based on this previous experience, two different formulations of materials and binders were selected in order to produce pellets trials with the torrefied material. Table 2 describes these formulations.

Table 2. Formulations with Binders for Each Pellet Trial

Code	Binders and quantities added to Pellets		
EB6	3000 cc <i>E. benthamii</i>	750 cc soybeans	83.33 cc Water
EB7	3000 cc <i>E. benthamii</i>	750 cc DDG	83.33 cc Water

The formulations provided and tested have a ratio of 36:9:1 of *E benthamii*, Binder, and Water. This ratio provides a good approach for a scale-up production with the formula, and provides the right amount of moisture and binder to allow for better densification and flow of the material through the rollers and dies, improving production rate and durability of the pellets.

RESULTS AND DISCUSSION

Biomass properties for energy in *Eucalyptus* species has not been widely studied, especially properties of the biomass after being pretreated for solid biofuels, such as pelletizing and torrefaction. This characteristic, along with the fact that few data pertaining to the properties of *E. benthamii* and *E. macarthurii* are readily available, makes comparisons with previous literature difficult. To address this limitation, the authors compare the results obtained after the pretreatment and after the pelletization process, with results previously obtained for the original wood material of both species (Pirraglia et al. 2011).

Physic-chemical Tests of Torrefied Wood of *E. benthamii* and *E. macarthurii*

Moisture content

The moisture content for the torrefied wood of both eucalyptus species was calculated by comparing the weight of the wood after torrefaction and cooling vs. the weight after drying the torrefied samples in an oven at $103^{\circ}\pm 2^{\circ}$, being the moisture content reported in a dry-basis.

Three replicates for moisture content were performed using *E. macarthurii* (EMT) and *E. benthamii* (EBT), obtaining 5.55% (\pm) and 5.05% (\pm) average moisture content (dry basis) respectively. No technical published data were available describing moisture content of torrefied wood for the eucalyptus species evaluated in this research. The average moisture content of the samples is significantly lower than that of the received material for both species, both of them around 5% compared to the original moisture content around 54 to 57% in an oven-dry basis. Previous work has been pursued in determining the wood-water relations in torrefied eucalyptus species (Almeida et al. 2009; Rodrigues and Rousset 2009; Almeida et al. 2010), and deal with common species, such as *E. grandis*; the importance of this work is that allows for comparison with the eucalypt species utilized in the present work. In a study by Rodrigues and Rousset (2009), it is indicated that the chemical changes occurring in the wood of *E. grandis* due to torrefaction treatment at temperatures higher than 220°C are responsible for the hydrophobic properties and low moisture of the material. In this study, the authors also determined that the lack of moisture regain in the torrefied material heavily influences the usable heating value of the eucalyptus, making it an extremely important parameter.

Volatile matter, ash content, and fixed carbon content

Experiments for the determination of volatile matter, ash content, and fixed carbon content were performed for each torrefied wood species. These experiments were performed using the same methodology previously used by the authors in the analysis of wood samples from *E. benthamii* and *E. macarthurii* (Standards ASTM D 3174-04 and ASTM D 3175-07). Three replicates of each torrefied wood species were performed, and the results (average of the three replications) are presented in Fig. 2.

Results of the proximate analysis for the torrefied material indicated a higher content of ash ($4.02\% \pm 0.35\%$ for *E. macarthurii*, and $1.83\% \pm 0.14\%$ for *E. benthamii*) and fixed carbon ($22.15\% \pm 0.54\%$ for *E. macarthurii*, and $36.61\% \pm 0.58\%$ for *E. benthamii*) as compared to the original material. This characteristic is due to the displacement of volatile matter ($73.84\% \pm 0.49\%$ for *E. macarthurii*, and $61.60\% \pm 0.47\%$ for *E. benthamii*) from the samples, caused by the heat treatment. Figure 2 demonstrates the change in Volatiles, Fixed Carbon, and Ash content from the original material to the torrefied one. In this sense, the ash content obtained from *E. macarthurii* torrefied (4.0%) is notably higher than the one obtained from *E. benthamii* (1.8%). Furthermore, the fixed carbon content of *E. benthamii* torrefied (36.6%) was considerably higher than that obtained with *E. macarthurii* torrefied (22.1%). These results indicate that *E. benthamii* provided better fixed carbon content with lower ash content as compared to *E. macarthurii*, when exposed to the torrefaction process. However, both species showed an increase in their fixed carbon content in weight percentage.

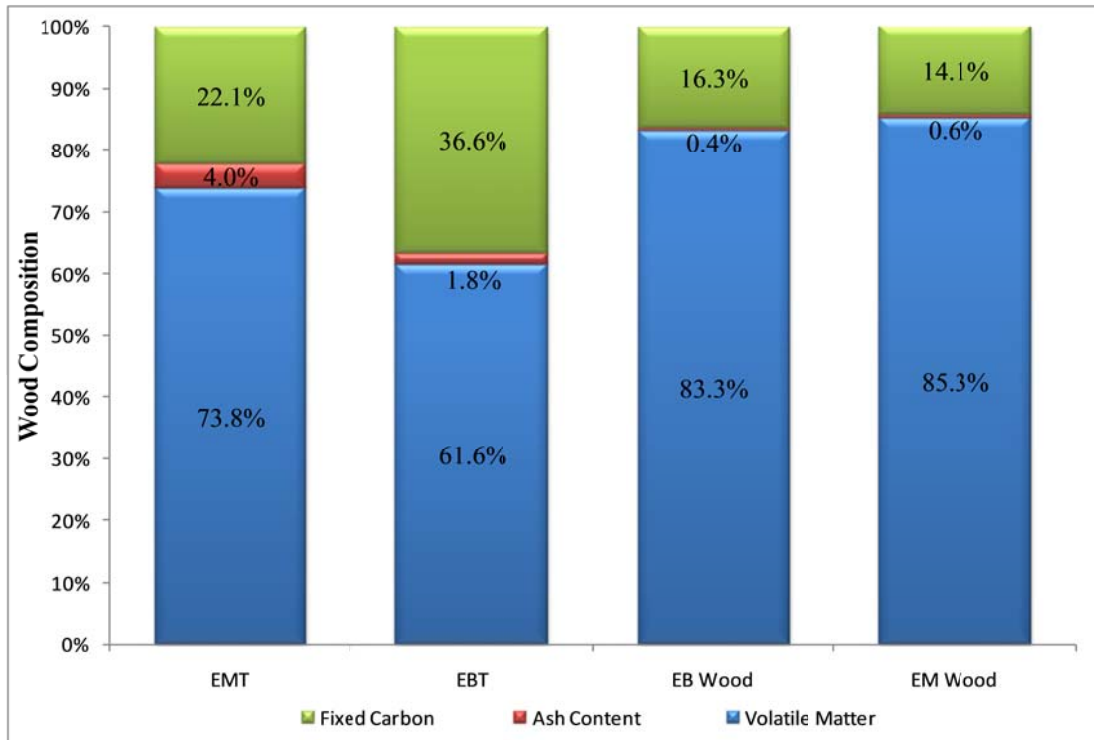


Figure 2. Proximate analysis for the torrefied wood compared to non-treated eucalyptus

The results demonstrated the suitability of both the torrefaction process and the utilization of its product as a biofuel, either as raw material for further processing, or for direct burning and co-firing, since the fixed carbon content is directly related to the heating value of a given biomass (Demirbas 1997; Parikh et al. 2005).

C-H-O-N analysis

The determination of carbon, hydrogen, oxygen, and nitrogen composition for each torrefied wood sample was determined assuming the composition of the torrefied wood is based only on these four elements, and the sulfur content is negligible. Figure 3 shows the average of C, H, O, and N, based on the average of three samples analyzed in each replica.

The ultimate analysis of the torrefied material showed lower oxygen content in the samples ($33.5\% \pm 0.24\%$ for *E. macarthurii*, and $29.39\% \pm 1.15\%$ for *E. benthamii*), when compared to the original material (Fig. 3). The nitrogen content did not show a difference as substantial as the one found for oxygen ($0.22\% \pm 0.01\%$ for *E. macarthurii*, and $0.16\% \pm 0.01\%$ for *E. benthamii*), while the samples also displayed a high increase in the elemental carbon content ($61.91\% \pm 0.30\%$ for *E. macarthurii*, and $66.36\% \pm 1.31\%$ for *E. benthamii*), due to the displacement of oxygen out of the samples. The increase in elemental carbon content is an indication of an enhanced heating value of the material being torrefied (Uslu et al. 2008), as it is demonstrated later for the HHV tests of the samples.

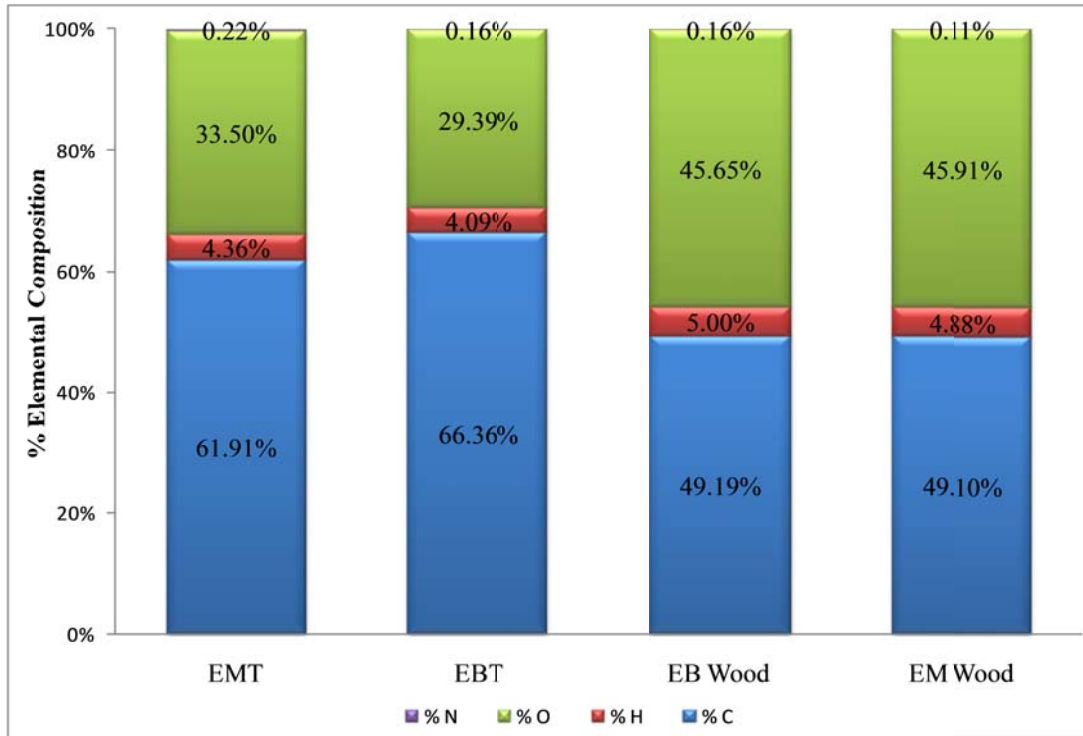


Figure 3. Ultimate analysis for torrefied vs. non-treated wood

High Heating Value (HHV)

Analysis of the heating value for the torrefied wood was performed in order to evaluate the effect of this treatment in the heating value delivered per unit of mass. Figure 4 shows the average heating values obtained for the torrefied wood of each species, in MJ/Kg.

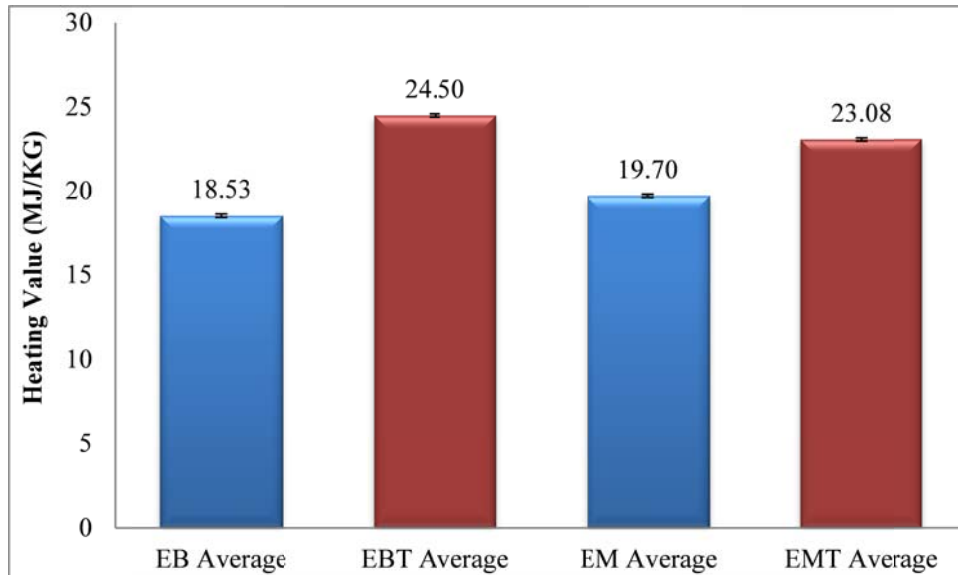


Figure 4. HHV analysis from the torrefied wood tests, with comparison of the average of 3 replicates in each category

Results from the net heating value tests of the torrefied wood (Fig. 4) indicate a high increase in the energy that can be obtained from the samples. An average increase of 19.7% could be obtained with the “light roast” torrefaction process for *E.macarthurii* (Temp: 280°C, Res. Time: 3 min., and Mass yield:80-85%), while an average of 19.6% increase in heating value could be obtained from “light roast” torrefaction of *E. benthamii*; both HHV calculations show a low standard deviation ($\pm 0.12\%$).

This enhanced heating value is related to the low moisture content of the product, and the partial elimination of volatiles content from the wood, leaving more carbon (as a percentage weight) for use as burning fuel.

An important concept to consider for the analysis of heating values in torrefied woody biomass is the Usable Heating Value (UHV), which determines the amount of energy effectively used (per unit of mass) based on the moisture content (dry basis) of the biomass (Rodrigues and Rousset, 2009). This UHV can be expressed as,

$$UHV = LHV*(1-MC) - 2.51*(MC) \quad (1)$$

where LHV represents the Higher Heating Value and the subtraction of the amount of energy required for condensation of water formed in complete combustion, and it can be expressed as:

$$LHV = HHV - 1.36 \quad (2)$$

With these equations, it is possible to calculate the usable energy from the torrefied biomass. Results from these calculations are presented in Table 3.

Table 3. Usable Heating Value of the torrefied Eucalyptus species

Species	MC (Dry basis, W/W)	HHV (MJ/Kg)	LHV (MJ/Kg)	UHV (MJ/Kg)
EB torrefied	0.0505	24.50	23.14	21.84
EM torrefied	0.0550	23.08	21.72	20.39

Results from the UHV indicate a usable energy content that is still higher than the High Heating Value of the original biomass (18.53 MJ/Kg for *E.benthamii* and 19.70 for *E.macarthurii*), indicating that even with the presence of moisture, and the loss of some of the energy content of the original material due to hemicellulose breakdown, the heat treatment is efficient in producing a high energy content product per unit of mass.

It is important to consider that the process of torrefaction requires little energy input, since the process is 80% self-sustained by the recirculation (heat source) of gases, while the remaining 20% is obtained from a propane flame stream. A detailed analysis of energy (energy balance) for the torrefactor and the product is highly recommended, since this process is noticeably increasing the HHV of the material, while using very little external energy. Such analysis would demonstrate the advantages and economical feasibility of this process with a biomass source such as eucalyptus. At the present time, authors report an estimated torrefied wood production cost of around 58 € ton⁻¹ (Uslu et

al. 2008), without considering feedstock costs, and in a small-scale factory. Further analysis is needed to assess production costs of torrefied wood considering economy of scale, and U.S. market conditions.

Physic-chemical Tests of Torrefied Wood Pellets Made from *E. benthamii*

As Zwart et al. (2006) concluded, a combination of torrefaction and pelletization may be one of the more feasible pretreatments of biomass in terms of transportation and suitability for bulk handling. In addition, the denominated “TOP Pellets” carry the advantages of an increased energy and material density (Bergman 2005), a hydrophobic property of the material, and a lower electrical demand required for further grinding/pulverization (Zwart et al. 2006). In this section, comparisons are made between previous pellets trials of *E. benthamii* and *E. macarthurii* performed by the authors (Pirraglia et al. 2011). Below, Table 3 summarizes the average properties obtained for the pellets produced.

Table 3. Summary of Properties for *E. benthamii* Torrefied Pellets and Comparison with Original Biomass Properties

Code	Pellets length	MC %	Volatile Matter	Ash Content	Fixed Carbon	C%	H%	O%	N%	HHV (MJ/Kg)
EB6	10 - 15 mm	7.54	68.95%	3.67%	27.38%	58.41	5.96	32.92	2.72	24.38
			(±0.22%)	(±0.07%)	(±0.27%)	(±0.15)	(±0.11)	(±0.16)	(±0.01)	(±0.03)
EB7		8.29	68.46%	3.66%	27.88%	55.55	5.99	36.26	2.21	24.30
			(±0.44%)	(±0.09%)	(±0.51%)	(±0.09)	(±0.22)	(±0.22)	(±0.03)	(±0.06)
EB	-	~54	83.26%	0.44%	16.30%	49.19	5.00	45.65	0.16	24.50*
			(±0.85%)	(±0.08%)	(±0.77%)					(±0.12)

* HHV of *E. benthamii* after torrefaction process, in order to compare how binders may affect HHV of torrefied material when pelletized

Moisture content

Moisture Content (MC) levels of the pellets trials were assessed while being stored at 70°F, and 50% Relative Humidity, being the equilibrium moisture content of the storage area EMC = 9.2%; showing a MC of 7.54%, and 8.29% for EB6 and EB7, respectively (oven-dry basis). These pellets remain at a MC lower than that of equilibrium (EMC), demonstrating the resilience of the material against regaining moisture after the heat treatment. The pellets manufactured from torrefied wood showed lower moisture content when compared to levels in conventional standard (10%) and utility quality pellets (10%), and a similar moisture content as compared to premium quality pellets (8%) (Pellets Fuel Institute. 2010). These results are in concordance with observations obtained by Cremers (2009) regarding the lower MC in torrefied wood pellets as compared to regular wood pellets. MC levels are important for transportation costs of pellets; a lower MC allows transporting and delivering more energy per unit of weight and volume.

Volatile matter, ash content, and fixed carbon content

The determination of volatile matter, ash content, and fixed carbon content in the pellets produced from *E. benthamii* wood were assessed utilizing 7 samples in 3

replicates. These experiments were performed using the same methodology employed previously for the analysis of *E. benthamii* and *E. macarthurii* wood samples (Pirraglia et al. 2011). Comparisons of the torrefied wood pellets were performed with the characteristics of the original wood as described in previous research by Pirraglia et al. (2011). The original wood of *E. benthamii* contains around 16% of fixed carbon, 0.44% of ash content, and 83% of volatile matter. In comparison, the torrefied and pelletized material contains 11% more fixed carbon (27% fixed carbon content in the torrefied pellets), and 14% less volatile matter (69% volatile matter in the torrefied pellets). These results are similar to those presented by several authors when comparing average results on torrefied wood samples (69.24% volatile matter, 29.74% fixed carbon, and 1.04% ash content, Pentananunt et al. 1990), and better than values obtained for beechwood by Couhert et al. (2009) in a recent study, with 75.7% of volatile matter, 0.4% of ash content, and 24.2% of fixed carbon. It is well known that fixed carbon content and volatile matter are directly related to the heating value (energy content) of a wood sample (Jimenez and Gonzalez 1991; Kucukbayrak et al. 1991; Raveendran and Ganesh 1996; Demirbas 1997; Cordero et al. 2001). A combination of higher fixed carbon and lower volatile matter content enhances the heating value of the wood, characteristics that are achieved through the torrefaction process, thus improving the properties of the wood for biofuels purposes. The results from Fig. 5 are also comparable to those obtained by Almeida et al. (2010) on the wood of *E. grandis* and *E. saligna*, for both, the untreated wood and the torrefied material, in which they obtained very similar values in fixed carbon, volatile matter, and ash content.

C-H-O-N analysis

The elemental composition (carbon, hydrogen, nitrogen, and oxygen content) was determined for the torrefied pellets trials, and the results were compared to the original wood material of *E. benthamii*. It is important to note that the composition of the pellets is based on these four elements, by assuming the sulfur content of the material is negligible (it is typically lower than 0.1% for wood materials). As compared to the original biomass, the torrefied pellets showed a considerable increase in the carbon and nitrogen content, a slight increase in hydrogen content, and presented a noticeable decrease in the oxygen content. According to a formula for estimating the gross heating value developed by Boie (1952), which utilized 16 biomass-based fuels, 66 coal, char and coke fuels, and 67 oil fuels (including alcohols), it was determined that an increase in carbon, nitrogen, and hydrogen content positively influences the gross heating value of a given biomass, while a decrease in the oxygen content also carries an increased heating value. As mentioned by Annamalai et al. (1987), this characteristic is due to the fact that if the amount of oxygen is greater, there will be less available percentages of carbon and hydrogen for combustion (due to combinations of CO, H₂O, OH groups, etc.). In the pellet trials developed (EB6 and EB7), there was less oxygen present, and thus, having more carbon and hydrogen available, which contributes to achieve a highly energy-dense fuel.

High Heating Value (HHV)

Previous results from the proximate and ultimate (CHON) analyses indicate an increase in the heating value of the torrefied and pelletized wood tested. Direct measure-

ments of the heating value by an adiabatic bomb were performed in order to verify the previous indications. The tests performed to determine the HHV of the torrefied pellet trials indicate similar heating values when compared to the original torrefied material of *E. benthamii* (24.28 MJ/Kg for EB6 and 24.30 MJ/Kg for EB7, versus an average of 24.5 MJ/Kg for torrefied EB). These results indicate that the binders have little effect in changing the energy properties of the torrefied material, while they allow for a better packing and densification of the torrefied wood, providing a more energy-dense product, and making it more suitable for transportation. This characteristic further enhances the desirable properties for handling and transportation in a solid biofuel produced from biomass sources. Further analysis of the cost/benefit of the binder is under development in order to determine the economic feasibility of using the binders.

As a final thought, pelletization of torrefied woody biomass is in the early stages of development and industrial application. In laboratory-scale mills, pelletization is difficult to achieve, due to low horsepower transmitted to the rollers and dies, and being a flat-die mill. Some authors report that successful production of torrefied pellets on an industrial scale has been already achieved (Boerrigter et al. 2006; Mitchell et al. 2007) by means of a TOP process, and ring-die pellet mills (Bergman 2005). This indicates that many technical difficulties of pelletizing torrefied biomass may be encountered in laboratory scale units only. In addition, it is stated that at torrefaction temperatures, the lignin in wood becomes plastic and serves as a natural binder for the individual wood particles (James 2010), improving the flowability of the material through the dies, which may be further improved with the addition of other binders. These properties will help overcome the abrasive nature of the torrefied material, allowing producing quality pellets with durability about 2 times more than regular wood pellets, and significantly lowering moisture uptake (James 2010).

Another benefit that favors the torrefaction and pelletization process is the fact that torrefaction reduces the fibrous nature of the wood, making it easily grindable, significantly reducing power consumption in the grinding operation prior to pelletization (Bridgeman et al. 2010). A major problem present in co-firing coal with biomass is the differences in energy density, burning range, and poor flowability of biomass, reducing the thermal efficiency and capacity of boilers (Hughes and Tillman 1998; Tillman 2000; Phanphanich and Mani 2011). Many of these problems can be addressed by utilizing torrefied wood pellets, which are easily grindable, improve combustion characteristics in co-firing, and are suitable for storage.

CONCLUSIONS

1. Torrefaction pre-treatment shows great potential for eucalyptus, eliminating water and volatiles, significantly enhancing its heating value by an average of 19% compared to the original material. More aggressive torrefaction temperatures and residence times may increase the heating value of the biomass significantly more.
2. Further enhancement can be achieved by changing residence time and operation temperature, these being critical variables for the product's uniformity. However, the utilized torrefaction process provides a good usable heating value for the biomass

evaluated, being higher than that of the original biomass, despite the mass and energy loss typical of this type of heat treatment.

3. Torrefied pellets have lower moisture and volatile matter, and higher fixed carbon content compared to traditional premium quality pellets, delivering higher energy content per bulk, with improved handling, and transportation.
4. In addition, binders do not affect the energy content of the pellets. However, further economic analysis of binders is necessary to assess the pre-treatment's impact on profitability.

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