Utilisation d'arbres de faible vigueur provenant de forêts feuillues tempérées comme matière première pour la fabrication de granules de bois

Thèse

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Thèse

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Sous la direction de:

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Résumé

La présente étude avait pour objectif de contribuer à une meilleure compréhension de la granulation des bois feuillus par l'évaluation de la possibilité technique de transformer des bois feuillus de faible vigueur (l'érable à sucre et le bouleau jaune) en granules conventionnels et granules de haute qualité, un type prometteur de transport énergétique. Trois études individuelles ont été réalisées et présentées dans cette thèse. La première étude visait à déterminer s'il y a des changements de teneur en extractibles, cendres, et lignine du bois entre les classes de vigueur des arbres. Les teneurs plus élevées en extractibles et en lignine dans les arbres peu vigoureux ont suggéré que ces derniers sont plus appropriés par rapport aux arbres vigoureux pour la conversion en biocombustibles solides. La deuxième étude visait à optimiser des procédés de granulation des bois feuillus. L'étude a porté sur l'influence des paramètres du procédé (la température et la force de compression) et des caractéristiques de la matière première (la taille des particules et la teneur en humidité) sur les propriétés physiques et mécaniques des granules de bois. Le procédé de granulation doit être effectué à une température d'environ 100 °C pour minimiser la force de friction dans le granulateur et à une teneur en humidité d'environ 11,2% pour maximiser la masse volumique et la résistance mécanique des granules produites. Cette étude a également confirmé que les arbres de faible qualité sont plus appropriés pour la fabrication de granules de bois que les arbres vigoureux. La troisième étude visait l'élaboration de granules de haute qualité. L'eau chaude à température élevée a été utilisée pour modifier les propriétés de la matière première avant granulation. Les caractéristiques de granulation du matériau traité ont été significativement améliorées. Les granules produites ont montré des propriétés améliorées incluant une plus faible teneur en cendres, une plus haute densité énergétique, une meilleure résistance à l'eau, et une meilleure résistance mécanique. Les résultats obtenus de toutes ces études ont démontré la nécessité de bien connaître les fondements de la granulation des bois feuillus et les solutions pratiques pour l'utilisation d'arbres feuillus de faible qualité, le premier peut être applicable pour le développement de procédés de granulation et le dernier peut contribuer à long terme à la restauration des forêts feuillues dégradées en termes de santé des forêts et de leur valeur.
Abstract

The objective of this study was to contribute to a deeper understanding of hardwood pelletization by investigating the technical possibility of converting low quality hardwoods (sugar maple and yellow birch) into conventional and advanced fuel pellets, a promising type of future energy carrier. Three individual studies were conducted and reported in this thesis. First study aimed at the investigation of the changes in extractives, ash, and lignin contents of wood among tree vigor classes. The higher contents of extractives and lignin in low vigor trees suggested that the low vigor trees are more suitable for conversion into solid biofuels. Second study aimed at the optimization of hardwood pelletizing process. The investigation was focused on the influence of process parameters such as temperature and compression force, and raw material characteristics such as particles size and moisture content on the physical and mechanical properties of wood pellets. The pelletizing process should be performed at a temperature of about 100 °C to minimize friction force in the press channel and at a moisture content of about 11.2% to maximize density and mechanical durability of pellets produced. This study also confirmed that low quality trees are more suitable for making wood pellets than vigorous trees. Third study aimed at the development of advanced fuel pellets. Hot water at elevated temperature was used to change the properties of raw material before pelletizing. Pelletization characteristics of treated material were significantly improved. The pellets produced represented a number of enhanced properties including ash content, energy content, water resistance and mechanical durability. The findings obtained through these studies emphasized the necessity of a good knowledge of the fundamentals of hardwood pelletization and proper solutions for the use of low quality hardwood trees in which the former can be applicable for pelletizing process development, and the latter in long term can contribute to the restoration of the degraded hardwood forests in terms of forests health and value.
Table des matières

Résumé ............................................................................................................................... iii
Abstract.............................................................................................................................. iv
Table des matières ........................................................................................................... v
Liste des tableaux............................................................................................................... vii
Liste des figures................................................................................................................ viii
Abréviations ..................................................................................................................... xi
Remerciements................................................................................................................ xii
Avant-propos .................................................................................................................... xiv
Chapitre 1. Introduction générale .................................................................................. 1
Chapitre 2. Literature review ......................................................................................... 3
  2.1. Biomass and bioenergy ......................................................................................... 3
  2.2. Characteristics of wood ....................................................................................... 5
  2.3. Fundamental of wood densification ..................................................................... 11
  2.4. Pelletizing process ............................................................................................... 17
  2.5. Quality parameters/characteristics ..................................................................... 21
  2.6. Recent trends and developments/ Pellets quality improvements ...................... 26
Chapitre 3. Objectives of this study ............................................................................. 30
  3.1. General objective ................................................................................................. 30
  3.2. Specific objective ................................................................................................. 30
  3.3. Hypotheses .......................................................................................................... 30
Chapitre 4. Material and methods .............................................................................. 31
  4.1. Material sampling and preparation .................................................................... 31
  4.2. Pelletizing experiment ....................................................................................... 35
Chapitre 5. Fuel properties of sugar maple and yellow birch wood in relation with tree vigor .......................................................................................... 37
  Résumé ........................................................................................................................... 38
  Abstract .......................................................................................................................... 39
  5.1. Introduction .......................................................................................................... 40
  5.2. Experimental ........................................................................................................ 42
  5.3. Results and discussion ....................................................................................... 46
  5.4. Conclusions ......................................................................................................... 51
Chapitre 6. Effect of process parameters and raw material characteristics on physical and mechanical properties of wood pellets made from sugar maple particles .......................................................... 52
  Résumé ........................................................................................................................... 53
  Abstract .......................................................................................................................... 54
  6.1. Introduction .......................................................................................................... 55
  6.2. Experimental ........................................................................................................ 58
  6.3. Results and discussion ....................................................................................... 65
  6.4. Conclusions ......................................................................................................... 76
Chapitre 7. Pressurized hot water treatment of sugar maple and yellow birch wood particles for high quality fuel pellet production ........................................................................... 77
  Résumé ........................................................................................................................... 78
  Abstract .......................................................................................................................... 79
7.1. Introduction........................................................................................................................................... 80
7.2. Experimental........................................................................................................................................ 81
7.3. Results and discussion......................................................................................................................... 85
7.4. Conclusions.......................................................................................................................................... 95
Chapitre 8. Conclusion générale et recommandations ............................................................................. 96
  8.1. Conclusion générale............................................................................................................................. 96
  8.2. Recommandations............................................................................................................................... 97
Bibliographie .................................................................................................................................................. 99
Annexe 1....................................................................................................................................................... 112
Liste des tableaux

Table 2-1. Specification of properties for pellets (EN 14961-1) ................................................................. 22
Table 5-1. Description of the 18 sample trees used for chemical analysis .............................................. 43
Table 5-2. Properties of sugar maple and yellow birch wood from different tree vigor class .................. 47
Table 5-3. Properties of extracted sugar maple and yellow birch wood ....................................................... 50
Table 6-1. Particle size distribution by tree vigor classes ............................................................................ 59
Table 6-2. Experimental ranges and levels of independent variables ........................................................ 60
Table 6-3. Experiment order ......................................................................................................................... 61
Table 6-4. Analysis of variance (ANOVA) for friction, density and compression strength of pellets pressed from particles of the most (R) and the least (M) vigorous trees .................................................................................. 66
Table 6-5. Summary of the regression modelling statistics (coded variables) ........................................... 67
Table 6-6. Properties of experimental pellets and commercial pellets ....................................................... 69
Table 7-1. Physical and chemical characterization of liquid and solid fractions resulting from the hot water treatment process ........................................................................................................ 86
Table 7-2. Physical and mechanical properties of pellets pressed from hot water treated and untreated sugar maple and yellow birch wood particles ................................................................................. 89
Liste des figures

Figure 2-1. Global biomass volumes required to achieve a 50% reduction in greenhouse gas emissions by 2050..................................................................................................................................................3

Figure 2-2. Share of bioenergy in the world primary energy mix ..............................................................................................5

Figure 2-3. Chemical structure of cellulose .................................................................................................................................5

Figure 2-4. Main components of hemicellulose ...........................................................................................................................6

Figure 2-5. Chemical structure of the principal components of lignin ..........................................................................................7

Figure 2-6. Distribution in percentages of cellulose, lignin and hemicelluloses within the cell-wall layers ..................9

Figure 2-7. Adsorption isotherms of hemicellulose, holocellulose, lignin and wood at 25 °C .............................................10

Figure 2-8. Diagram of the microfibril arrangements within the cell-wall layers .................................................................10

Figure 2-9. Models of a cross section and a longitudinal section of a microfibril ...............................................................11

Figure 2-10. Modulus versus temperature for an amorphous polymer ..................................................................................12

Figure 2-11. Glass-transition temperature of lignin and hemicellulose as a function of moisture content .................13

Figure 2-12. Schematic view of a transverse compression stress-strain curve for wood ..............................................14

Figure 2-13. Classification of agglomeration processes .........................................................................................................15

Figure 2-14. Deformation mechanisms of powder particles under compression ...............................................................16

Figure 2-15. Illustration of the pelletizing process ................................................................................................................17

Figure 2-16. Interactions between physical-mechanical and combustion properties of solid biofuels ...................23

Figure 2-17. Difference on molar H:C to O:C ratio between biomass and coal .................................................................24

Figure 2-18. Difference between gross and net calorific value at variable moisture content of fuel ..........................24

Figure 2-19. Selected future bioenergy carriers for supporting the flexible use of biomass ........................................26

Figure 2-20. Temperature regimes for the thermal decomposition of cellulose, hemicellulose and lignin .......27

Figure 2-21. Thermal decomposition of wood and its cell wall components .................................................................28

Figure 2-22. Location of selected trees in the forest stands ..............................................................................................31

Figure 4-1. Location of selected trees in the forest stands ..............................................................................................31

Figure 4-2. Raw material preparation ...................................................................................................................................32
Figure 7-4. Water absorption of pellet samples after immersion for 2, 24 and 168 hours in distilled water at room temperature ...........................................................................................................92

Figure 7-5. Volumetric swelling of pellet samples after immersion for 2, 24 and 168 hours in distilled water at room temperature ...........................................................................................................93

Figure 7-6. Typical load - deformation curves of pellets in transverse compression. From top to down are curves under compression for pellets pressed from 200 °C HWT, 175 °C HWT, 150 °C HWT and untreated (control) wood particles of moribund sugar maple trees ...........................................................................................................94

Figure 7-7. Appearance of pellets after compressive strength tests. From left to right are fracture patterns of broken pellets made of untreated (control) and hot water treated (150 °C, 175 °C, 200 °C) sugar maple wood particles ...........................................................................................................95

Figure A.1-1. Interaction plots of friction in N mm⁻¹ for pellets pressed from particles of the most vigorous sugar maple trees ...........................................................................................................112

Figure A.1-2. Interaction plots of density in g cm⁻³ for pellets pressed from particles of the most vigorous sugar maple trees ...........................................................................................................113

Figure A.1-3. Interaction plots of compressive strength in N mm⁻¹ or pellets pressed from particles of the most vigorous sugar maple trees ...........................................................................................................114

Figure A.1-4. Band of experimental load versus time curves obtained during pelletization of 729 pellets under controlled conditions ...........................................................................................................115

Figure A.1-5. Band of experimental load - deformation curves in transverse compression of pellets produced from various pelletizing parameters ...........................................................................................................115
**Abbréviations**

<table>
<thead>
<tr>
<th>Acronym</th>
<th>Full Form</th>
</tr>
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<tbody>
<tr>
<td>ASTM</td>
<td>American society for testing and materials</td>
</tr>
<tr>
<td>ANOVA</td>
<td>Analysis of variance</td>
</tr>
<tr>
<td>CEN</td>
<td>European committee for standardization</td>
</tr>
<tr>
<td>CHP</td>
<td>Combined heat and power</td>
</tr>
<tr>
<td>DBH</td>
<td>Diameter at breast height</td>
</tr>
<tr>
<td>EN</td>
<td>European standard</td>
</tr>
<tr>
<td>EU</td>
<td>European union</td>
</tr>
<tr>
<td>GCV</td>
<td>Gross calorific value</td>
</tr>
<tr>
<td>GHG</td>
<td>Greenhouse gas</td>
</tr>
<tr>
<td>HHV</td>
<td>Higher heating value</td>
</tr>
<tr>
<td>HWE</td>
<td>Hot water extraction</td>
</tr>
<tr>
<td>HWT</td>
<td>Hot water treatment</td>
</tr>
<tr>
<td>IEA</td>
<td>International energy agency</td>
</tr>
<tr>
<td>LHV</td>
<td>Lower heating value</td>
</tr>
<tr>
<td>MC</td>
<td>Moisture content</td>
</tr>
<tr>
<td>MRNF</td>
<td>Ministry of natural resources and wildlife</td>
</tr>
<tr>
<td>T₉</td>
<td>Glass transition temperature</td>
</tr>
</tbody>
</table>
For those People who on a Mission - Developing Solid Biofuels
I would say: "Power from the Woods - Our Sustainable Future"!
Remerciements

J'ai eu une excellente opportunité de compléter cette étude au Centre de Recherche sur les Matériaux Renouvelables (CRMR), Département des sciences du bois et de la forêt de l'Université Laval, Québec, Canada.

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Avant-propos

Cette thèse est le résultat d'un projet de recherche mené sous la supervision des professeurs Alain Cloutier, Alexis Achim et Tatjana Stevanovic du Département des sciences du bois et de la forêt de l'Université Laval. Le projet était axé sur l'étude des propriétés du carburant, les caractéristiques de granulation de bois feuillus avec l'objectif d'examiner le potentiel d'utilisation des bois feuillus de faible qualité pour la production de granules de bois à des fins bioénergétiques. Ce document est présenté sous forme d'une thèse de publications. On retrouve dans cet ouvrage une acte de conférence et trois articles scientifiques écrits en anglais présentés dans l'annexe 1 et les chapitres 5, 6, 7. Ceux-ci sont respectivement:


L'auteur de cette thèse, Quy Nguyen Nam, était responsable des travaux expérimentaux et la rédaction la première version de ces articles. Les co-auteurs, le professeur Alain Cloutier, professeur Alexis Achim et professeure Tatjana Stevanovic, ont supervisé les expériences et révisé les articles.
Les résultats de ce travail ont également été présentés dans les conférences suivantes:


Cette thèse se compose de huit chapitres basés sur des travaux rapportés dans trois articles scientifiques. Ces articles sont présentés dans les chapitres 5, 6 et 7. Le chapitre 1 fournit des informations générales sur cette recherche. Le chapitre 2 présente une revue de littérature. Le chapitre 3 présente les buts et les objectifs du projet. Le chapitre 4 décrit brièvement le matériel et les méthodes utilisés dans cette thèse. Le chapitre 5 traite avec les propriétés du combustible bois en relation avec la vigueur de l'arbre. Le chapitre 6 vise à discuter de l'effet des caractéristiques des matières premières et des paramètres de traitement sur les caractéristiques de granulation et les propriétés des granules de bois. Le chapitre 7 porte sur le traitement hydrothermique de particules de bois pour la fabrication de granules de haute qualité. Le chapitre 8 résume les résultats obtenus et présente les conclusions générales.
Chapitre 1. Introduction générale

Au cours des dernières décennies, les tiges des arbres les plus précieux dans les forêts feuillues de l'Est de l'Amérique du Nord, l'éralbe à sucre (*Acer saccharum* Marsh.) et le bouleau jaune (*Betula alleghaniensis* Britton), ont été récoltés pour la production de bois d'oeuvre de feuillus (Nyland 1992; Bédard et Majcen 2003; Pothier et al. 2013). Ainsi, les forêts actuelles contiennent une forte proportion d'arbres non-vigoureux de très peu ou pas de valeur commerciale (Havreljuk et al. 2014). Du point de vue de la gestion des forêts, la présence de ces arbres de faible vigueur et la faible qualité des tiges ont un effet négatif sur la santé des forêts et sur leur valeur. Par conséquent, dans les coupes partielles généralement appliquées à ces peuplements, la priorité de récolte est maintenant donnée aux arbres non-vigoureux tout en laissant croître les arbres plus vigoureux pour la récolte future (Delisle - Boulianne et al. 2014).

La bioénergie, l'énergie dérivée de la biomasse, est une des sources les plus importantes d'énergie renouvelable aujourd'hui. Plus de 80% de la biomasse utilisée pour l'énergie provient de forêts, sous la forme de billes, de copeaux de bois, granules de bois ou/et d'autres sous-produits. Les granules de bois utilisées d'une manière moderne et efficace pour la production de chaleur et d'électricité semblent être le plus polyvalent parmi toutes sortes de biocarburants solides (Jenkins 2010). Selon un rapport publié par Flach et al. (2014), la consommation mondiale de granules de bois est actuellement d'environ 20 millions de tonnes métriques, et les prévisions montrent une augmentation entre 50 et 80 millions de tonnes d'ici 2020. En 2014, 28 pays de l'Union européenne (EU) ont produit 13,3 millions de tonnes et importé plus de 6,5 millions de tonnes de granules de bois pour leurs besoins domestiques. Les plus grands exportateurs de granules de bois vers l'EU sont les États-Unis et le Canada. Les exportations canadiennes de granules de bois ont atteint 1,6 million de tonnes en 2014 et représentaient 84% de la production totale du pays (Aguilar et al. 2015).

Des recherches ont été menées dans le monde pour développer de nouvelles technologies pour la production de biocarburants de haute qualité à partir de ressources renouvelables (Ragauskas et al. 2006; Hunt 2009; Liu et al. 2013). Un certain nombre d'études ont été réalisées dans le but d'améliorer la qualité du combustible. Le traitement à l'eau chaude (HWT) est une étape importante dans le prétraitement de la biomasse ligneuse et la fabrication de produits à base de bois. La fraction solide après le traitement HWT a une teneur plus élevée en énergie, une stabilité dimensionnelle plus élevée, une plus grande résistance à la biodégradation et présente une réduction des émissions polluantes lors de la combustion (Navi et Sandberg 2011; Pu et al. 2011; Bach et al. 2013; Hartmann 2013; Pelaez - Samaniego et al. 2013; Hoekman et al. 2014). En outre, l'hydrophobicité du

L'utilisation croissante des granules combustibles et l'expansion des types de matières premières se traduisent par la nécessité d'une meilleure compréhension du procédé de granulation. En outre, le développement de granules de bois de haute qualité est toujours un défi. Ceci a été la motivation pour la présente étude. Elle avait pour objectifs de: (i) développer une compréhension plus profonde des difficultés de granulation des bois feuillus, (ii) améliorer le procédé de granulation des bois feuillus et la qualité des granules, et (iii) identifier une solution appropriée pour l'utilisation des arbres feuillus de faible qualité, qui contribue à l'amélioration de la santé des forêts feuillues dégradées.
Chapitre 2. Literature review

2.1. Biomass and bioenergy

2.1.1. Biomass resource and bioenergy potential

Plants annually convert 4500 exajoules (4500 x 10^18 joules) of solar energy and 125 gigatonnes of carbon from the atmosphere into biomass. Atmospheric carbon is captured by living plants from the atmosphere through photosynthesis process and released back into the atmosphere by decay or other processes of use. Biomass is therefore a carbon neutral source of energy (Ohlrogge et al. 2009; Kopetz 2013; Junginger et al. 2014).

The international energy agency (IEA) estimated that a 50% reduction in greenhouse gas (GHG) emissions by 2050 will require the contribution of the bioenergy production of 150 exajoules per year. It is more than 20% of the world primary energy. To reach the IEA 2050 target of 150 exajoules per year, primary energy from biomass would require 15 billion metric tonnes of biomass annually. At baled grass and wood chip densities of 150 and 225 kg m^-3, the transport volume calculated for 15 billion metric tonnes would be 100 or 60 billion cubic meters, respectively (Fig. 2-1). Using reported energy densities of conventional pellets, pyrolysis oil and torrified pellets, these densified products would require 28, 17, and 15 billion cubic meters, respectively (Richard 2010).

![Fig. 2-1. Global biomass volumes required to achieve a 50% reduction in greenhouse gas emissions by 2050 (Richard 2010).]
Biomass can be used to produce different kinds of bioenergy, biomaterials, and chemicals required in a modern society. As compared to other resources available, biomass is one of the most common and widespread resources in the world. Bioenergy, the energy derived from biomass, is one of the most important renewable energy sources today and expected to play a major role in medium to longer term in replacing fossil fuels and reducing greenhouse gas (GHG) emissions. By 2035, biomass could deliver 120 exajoules (50% of the world's energy needs) for heat, 15% exajoules for transport, and 18 exajoules (7%) for electricity (Faaij 2013; Kopetz 2013; Majer and Thrän 2015).

2.1.2. Present and future use of forest biomass for energy

Forest biomass is considered to be a sustainable source of energy, because as a part of its biological cycle, carbon dioxide is taken up by living trees and stored as forest biomass. When trees die and/or decay naturally, they release carbon dioxide and energy over a long period of time, and only carbon dioxide is in turn taken back up by renewed growth. However, converting the biomass into bioenergy can effectively capture the stored energy quickly and mostly in a constructive way. Although biomass-based bioenergy has a lower energy content compared to fossil fuels, in the short term it can actually generate higher carbon dioxide emissions than fossil fuels. But over time there is a net benefit to the atmosphere because the forest biomass is renewable. Therefore, the proper utilization of forest energy resources can be an environmentally friendly option (Faaij 2013).

More than 80% of the biomass used for energy comes from forests, in the form of logs, wood chips, wood pellets, and other by-products (Fig. 2-2). Logs are the traditional way to use wood as a fuel. Wood chips are the main alternative tree-derived fuel to logs. Medium to large boiler systems tend to use wood chips rather than logs, mainly because this form of wood fuel is easier to use in fully automated systems. Fuel pellets are far superior to both wood chips and logs. Today, wood pellets appear to be the most versatile of all wood-based fuels. They are appropriate to almost all requirements, regardless of ownership type and building size. They are also the most convenient to handle and offer the greatest number of advantages when used in automated systems (Jenkins 2010; Sultana and Kumar 2012; Lamers et al. 2014).
2.2. Characteristics of wood

2.2.1. Chemical composition

2.2.1.1. Cellulose

Cellulose ($\text{C}_6\text{H}_{10}\text{O}_5)_n$ is a crystalline thermoplastic natural polymer acting as reinforcing microfibers in the laminae of cell walls. Cellulose is formed by a condensation reaction of anhydroglucose units. These anhydroglucose units are linked together in the 1,4 ($\beta$-D-glucopyranose) positions (Fig. 2-3). This makes the cellulose molecule in the form of a long, straight, and very slender chain (Dinwoodie 2000; Kettunen 2009). The degree of polymerization in the molecule chain varies for different tree species and is on the average of 8000 - 10000 glucose units. Cellulose accounts for 40 – 50% of wood substance by weight, and has a density of about 1550 kg m$^{-3}$ (Dinwoodie 2000; Smith et al. 2003). Thermogravimetric analysis shows that cellulose is more thermally stable than other wood constituents (e.g lignin, hemicelluloses) and starts degrading at temperatures above 200 °C (Kim et al. 2001; Amidon et al. 2008).

Fig. 2-3. Chemical structure of cellulose (Mohan et al. 2006).
2.2.1.2. Hemicellulose

Hemicelluloses are heteropolysaccharides composed of sugar monomers other than glucose (Fig. 2-4). Common monomers are mannose, galactose, xylose, arabinose, galacturonic acid, and 4-O-methylglucuronic acid. These precursoric sugar units can also be categorized into pentoses, hexoses, hexuronic acids, and deoxyhexoses. The backbone of hemicellulose molecule can be a homopolymer, consisting only of one type of sugar units (e.g. xylans), or a heteropolymer, consisting of two or more sugar units (e.g. glucomannans). The degree of polymerization of hemicelluloses is lower than that of cellulose, only in the order of 150 - 200. They have a semi-crystalline structure, similarly as cellulose, but with the dominance of an amorphous structure. Hemicelluloses account for 25 - 35% of wood substance by weight, and has a density of about 1500 kg m\(^{-3}\) (Smith et al. 2003; Kettunen 2009).

The hemicellulose in hardwoods and softwoods are different in the proportion and type. The common/main hemicelluloses in softwoods are galactoglucomannan of about 15 - 20%, and arabinoglucomannan of about 10% of the total dry wood mass. Hardwood hemicelluloses consist mainly of glucuronoxylan accounting a percentage of 20 - 30% of the total dry wood mass and a small amount (less than 5% of the total dry wood mass) of glucomannans (Barnett and Jeronimidis 2003; Kettunen 2009). Chemical and enzymatic reactivity of hemicelluloses is high due to the availability of numerous hydroxyl groups and glycosidic bonds as well as of ester bonds in xylan acetyl groups. The hemicelluloses are alkali soluble and the glucomannans with a high galactose content are water soluble. The highly branched arabinogalactans are also an example of water soluble hemicelluloses (Barnett and Jeronimidis 2003). Hemicelluloses are less thermally stable than cellulose and lignin. Degradation temperature of hemicelluloses is low at about 160 °C (Amidon et al. 2008).

![Chemical structures of sugar monomers](https://example.com/chemical_structures.png)

Fig. 2-4. Main components of hemicellulose (Mohan et al. 2006).
2.2.1.3. Lignin

Lignin is a highly complex non-crystalline molecule comprising a large number of phenyl-propane units. Hardwood lignin is made up mainly of coniferyl and sinapyl alcohols and softwood lignin of coniferyl alcohols (Fig. 2-5). Different functional groups are present in the macromolecule of lignin include aromatic and aliphatic hydroxyls, benzyl alcohol and ether groups, carbonyl and methoxyl groups. Methoxyl groups represent about 12 - 18% and 15 - 22% of lignin proportions in softwoods and hardwoods, respectively (Barnett and Jeronimidis 2003).

Lignin contributes to natural durability, antimicrobial resistance, and mechanical strength of wood. Lignin is the most hydrophobic component in the cell walls with a density of about 1400 kg m\(^{-3}\) (Smith et al. 2003; Kettunen 2009). The carbon content in lignin is significantly higher (64%) than for cellulose (42%). Differences in the macromolecular composition are therefore directly influencing the total carbon content of the dry matter. Therefore, lignin is a main contributor to the heating value of a biomass fuel (Hartmann 2013). Lignin decomposition in wood was proposed to begin at 280 °C and continues to 450 - 500 °C.

Fig. 2-5. Chemical structure of the principal components of lignin (Mohan et al. 2006).

2.2.1.4. Extractives

Besides the cell structural components there are numerous kinds of organics compounds which are called wood extractives. The extractive compounds in wood can be grouped in two main types: lipophilic (soluble in organic solvents) and hydrophilic (water-soluble). The constituents of lipophilic extractives include terpenoids and steroids, fats, waxes, and phenolic compounds that consist of stilbenes, lignans, tannins, and different classes of flavonoids (Sjöström 1993; Sjöström and Alén 1999). Though with very small proportion of extractives in wood,
they may have a great influence on the properties and processing qualities of wood (Fengel and Wegener 1983; Dinwoodie 2000).

2.2.1.5. Mineral components

Mineral components are present in wood either as an integral part of substances manufactured by the living cells, or as extraneous substances brought up in suspension and deposited in the wood. Elements such as calcium, sodium, potassium, phosphorous, and magnesium are all components of wood tissue. Total inorganic constituents represents a small quantity of less than 1% (d.b.) in temperate woods or higher in tropical woods (Dinwoodie 2000).

2.2.2. Variation in wood characteristics

Wood is a highly variable material because it is produced in a biological environment and the tree is subjected to varying growth conditions (Barnett and Jeronimidis 2003). Wood characteristics can be categorized into three groups. The first group covers wood characteristics that form as a tree grows naturally. The second group of wood characteristics comprises biotically induced characteristics. These include microorganisms and animals that use tree parts as a food source or for nesting. Human influences include injuries due to forestry or logging operations. The third group includes abiotically induced wood characteristics of inanimate nature such as temperature, precipitation, wind, and snow (Richter 2015).

2.2.2.1. Variation in chemical composition

Chemical composition of wood varies with geographical, genus and species. In general, softwoods have higher lignin content (25 - 35%) and their hemicelluloses contain galactoglucomannan (15 - 23%) and arabinoglucuronoxylan (7 - 10%), while hardwoods have less lignin (18 - 30%) and their hemicelluloses contain acetylglucuronoxylan (15 - 30%) and glucomannan (2 - 5%). Regarding the chemical composition of the different cell wall layers (Fig. 2-6), the primary wall has the highest concentration of lignin and the lowest of cellulose, with the middle lamella being made up mostly of lignin (Barnett and Jeronimidis 2003).

The components showing the highest variation are the extractives, with the quantity and types of extractives varying widely between species, within different sites for the same species and within the same tree. Variation in chemical composition relates to the occurrence of different types of wood within tree stem such as juvenile and mature wood, sapwood and heartwood, reaction and normal wood (Barnett and Jeronimidis 2003).
2.2.2.1. Variation in physical and mechanical properties

Wood is an anisotropic material in its cellular organization resulting in anisotropic physical and mechanical properties. Properties parallel to the grain differ significantly from those in the transverse direction. In addition, transverse properties also differ radially and tangentially (Dinwoodie 2000; Barnett and Jeronimidis 2003). The adsorption isotherms of wood and its chemical constituents shown in Fig. 2-7 indicate that hemicelluloses are the most hygroscopic, followed by holocellulose (a combination of the hemicellulose and cellulose) and lignin.

Mechanical properties of wood relate directly to the microfibrillar organization of the cell wall (Fig. 2-8). The highest strength values of wood are obtained in longitudinal tension resulting from the high density of covalent bonds of cellulose in that direction (Fig. 2-9). The lowest strength values are obtained in tangential and radial tension due to the much weaker horizontal bonding between the cellulose molecules. In longitudinal compression, the microfibrillar structure is dislocated and the cellulose crystallite orientation is altered, resulting in the formation of cross running kinks (Dinwoodie 2000).
Fig. 2-7. Adsorption isotherms of hemicellulose, holocellulose, lignin and wood at 25 °C (adapted from Skaar 1984).

Fig. 2-8. Diagram of the microfibril arrangements within the cell-wall layers (Dinwoodie 2000).
2.3. Fundamental of wood densification

2.3.1 Glass transition temperature of wood

The natural organic structural polymers comprising wood or other lignocellulosic materials may be understood through polymer theory. In particular, the concept of glass transition has direct application to the material properties of the polymers comprising plant cell walls. As a practical matter, lignocellulosic raw materials are routinely subjected to elevated temperatures and pressures during the manufacturing processes. Thus, the glass transition ($T_g$) of wood or lignocellulosic polymers is of fundamental importance for the understanding of material behavior during composite consolidation.

Amorphous polymers (lignin and hemicelluloses) of wood, known as viscoelastic materials, can behave as viscous fluids and as linear elastic solids, depending on the time, temperature, and diluent concentration (Wolcott et al. 1990). Wood exhibits glassy and rubbery states as a function of its temperature. The glass transition temperature ($T_g$) of a material is the temperature at which there is an abrupt change in stiffness, and it mediates between glassy and rubbery regimes of polymers (Hillis and Rozsa 1985). Many properties of amorphous polymers, such as the elastic modulus, change dramatically when the material passes this softening point (Fig. 2-10). The apparent rubbery transition of wood reflects the $T_g$ values of hemicellulose and lignin that are about 80 °C and 90 °C, respectively (Hillis and Rozsa 1985).
Hillis and Rozsa (1978) studied the influence of wood components on the softening of wood and suggested that moisture lowered the softening point of hemicelluloses and lignin in wood, which is above 160 °C when these two components are isolated from the wood. Hemicelluloses in the cell wall softened first (at 54 – 56 °C), which decreased the wood stiffness. This enabled wood fibres to adapt their cross-sectional shape to the applied forces. The softening of lignin (at 72 – 128 °C) in the cell wall and middle lamella allows for further cross-sectional movement within and between the fibres (Kutnar and Šernek 2007).

Kelley et al. (1987) investigated the viscoelastic properties of wood by using dynamic mechanical thermal analysis and differential scanning calorimetry. They found that at 0% moisture content, the glass transition temperature of both lignin and hemicellulose is assumed to be 200 °C, a temperature near which degradation takes place. The glass transition temperature of lignin decreases with increasing moisture content but starts to reach a plateau at 70 °C near 10 to 15% moisture content. The $T_g$ of hemicellulose, however, continues to decrease with increasing moisture content with a value of -20 °C near 30% moisture content (Fig. 2-11).
Literature values for the $T_g$ of cellulose ranged from 200 °C to 250 °C, with an assumed value of 220 °C used for theoretical calculation (Back and Salmén 1982). The $T_g$ of hemicelluloses was summarized in the range of 150 - 220 °C, with an assumed value of 180 °C as the apparent mean $T_g$ of hemicelluloses containing side groups. The dry $T_g$ of native lignin may be of 205 °C.

Hardwood lignins have fewer phenolic hydroxyl groups, and substantially more methoxyl groups, resulting in a significantly lower softening temperature than softwood lignin. This helps to explain that different kinds of wood require different pelletizing temperature to produce high quality pellets.

2.3.2. Viscoelastic response of wood under compression

During transverse compressive loading, a typical stress-strain curve of wood has three distinct regions (Bodig 1965, Nairn 2006), as shown in Fig. 2-12. The initial part of the stress-strain curve for wood is a linear elastic region, in which the stress is directly proportional to strain. The second part, sometimes known as the plateau, is a plastic region in which strain increases rapidly with a small or no change in stress. After the plastic region, the stress increases sharply with strain. This region is termed the densification region (Tabarsa and Chui 2000).
As the proportional limit is exceeded, the stress–strain relationship is no longer linear. Stresses are now great enough to induce covalent bond rupture and permanent distortion at all three structural levels (Winandy and Rowell 2005). At the molecular level, the limit of reversible or recoverable hydrogen bonding has been exceeded. Covalent C–C and C–O bonds are breaking, thus reducing larger molecules to smaller ones. This reduction in degree of polymerization by covalent bond scission is nonrecoverable. At the microscopic level, stresses develop within the crystalline region of the carbohydrate microfibrils. Failure of the microfibril from stress overload causes actual covalent bond rupture and excessive microfibril disorientation. Additionally, the cell wall layers distort such that permanent micro cracks occur between the various cell wall layers. Separation of the cell wall layers is soon noticeable. At the macroscopic level, entire fibers actually distort in relation to one another, such that recovery of the original position is now impossible. The wood cells or wood fibers are actually failing either by scission of the cell, in which the cell actually fails by tearing into two parts to give a brash type of failure, or by cell-to-cell withdrawal (middle lamella failure), where the cells actually pull away from one another to give a splintering type of failure.

A yield point is exhibited at the beginning of cells collapse. When a majority of the cells have collapsed, densification begins (Wolcott et al. 1994). During densification, the stress rapidly increases as a result of the elimination of air voids resulting from the compression of the solid wood structure and consolidation of the collapsed cell walls. Cellular collapse occurs by elastic buckling, plastic yielding, or brittle crushing, depending on the test conditions and the nature of the cell wall material (Wolcott et al. 1989).
The relationships between compressive forces applied to wood and deformation are strongly influenced by the moisture content. Temperature also has effect on mechanical properties, especially in combination with moisture. Ellis and Steiner (2002) compressed five species in axial, tangential, and radial directions at moisture contents: 5%, 10%, 20%, and 30%. They found that higher initial moisture contents increased the plasticity of the wood leading to a lower Young’s modulus and lower levels of stress during compression, greater springback after release of stress, and greater recovery after swelling in water. Tabarsa and Chui (1997) compressed white spruce (*Picea glauca*) in radial direction at 20, 100, 150, and 200 °C and found that the plastic strain increased with increasing temperature.

### 2.3.3. Binding mechanism of wood densification

Depending on the feedstock and the agglomeration process, different binding mechanisms and forces are relevant for the bonding of individual particles. The pressure agglomeration is achieved by applying external forces to particles in different shaped dies to form enlarged agglomerates (e.g. briquettes and pellets). Fig. 2-13 provides an overview of established technologies (Klemm et al. 2013).

![Classification of agglomeration processes: (a) - punch and die press, (b) - screw extrusion press, (c) - ram extrusion press, (d) - pellet mill with flat die, and (e) - pellet mill with ring die (after Klemm et al. 2013).](image-url)
According to Rumpf (1962), the possible binding mechanisms can be divided into five major groups: (i) attraction forces between solid particles, (ii) interfacial forces and capillary pressure, (iii) adhesion and cohesion forces, (iv) solid bridges, and (v) mechanical interlocking. Solid bridges, due to inorganic bonding, chemical reaction, crystallization, melting at points of contact, due to frictional pressure, and sintering. Solid bridges can also be formed after hardening of the binder. Mechanical (particle) interlocking can be very important in the pelletizing process because of the different shapes (crystals, rounded or elongated) of the raw material and the high shear forces acting on the particles during pelletizing. Fig. 2-14 shows the deformation mechanism of the powder particles during compression (Çomoğlu 2007).

Mani et al. (2004) indicated in their studies that the compression curve was slowly increased during the initial stages of loading. During this period, particles moved and rearranged resulting from compression and the empty spaces were eliminated. This mechanism is called particle rearrangement, which occurs at low pressures. The initial stage was different between the types of material. It was short for barley straw grinds but was long for corn stover, switchgrass, and wheat straw grinds. During particle rearrangement, the slope of the compression curve was constant and as the compressive force progressed, the slope increased indicating densification by elastic, plastic deformation, and perhaps interlocking of particles. The time at which the transition of slope occurred was taken as particle rearrangement time.

According to Kaliyan and Morey (2009) the application of high pressures and temperatures, solid bridges may be developed by diffusion of molecules from one particle to another at the points of contact. Solid bridges may also be formed between particles due to crystallization of some ingredients, chemical reaction, hardening of binders, and solidification of melted components. Solid bridges are mainly formed after cooling of densified products.
2.4. Pelletizing process

The first patented biomass densification/compaction process was registered in 1880 by William H. S. in Chicago, Illinois, USA. Various variables (e.g. pressure, temperature, compression and die material, die shaping, pressing rolls rotating speed and gap between the roll and die) which affects the pelletizing process. Optimization of pelletizing process must be based on raw materials. Different raw materials need different parameters. Pelletizing process is presented in Fig. 2-15 (Nielsen et al. 2009a).

![Fig. 2-15. Illustration of the pelletizing process (Nielsen et al. 2009a).](image)

In the pelletizing process, material is fed continuously into the feed layer, but each die channel receives material intermittently, including short time intervals when no material is received. Consequently, both static and kinematic wall friction has to be overcome in the die channels. Friction between raw material particles and machinery (wall friction), and internal friction between raw material particles are highly relevant in the pelletizing process (Holm et al. 2006). The raw material is continuously added in the front of the rolls, and a flow of compressed particle thus passes through the channels. At the end of the channels, the compressed particle breaks off and form the pellets. The pellets maintain their shape and size due to bonding that occurs between the particles at the high pressure in press channels (Nielsen et al. 2009b). The pelletizing pressure is increased as the length of die channel increases. In contrast, the pressure is decreased as the diameter of the die hole increases. Hence, the dimensions of the press channels in the matrix have a great influence on the pressure needed to press the pellets though the matrix (Holm et al. 2006). During compression, raw material temperature is rising and binding agents (lignin, resin or starch) start to soften. The pelletizing process involves many
variables which affect the pelletizing process, namely pressure, temperature, compression and die material, die shaping, pressing rolls, rotating speed, and gap between the roll and die (Nielsen et al. 2009a).

### 2.4.1. Raw material

Almost all types of biomass can be pelletized. However, many factors need to be considered before biomass qualifies as an appropriate feedstock for pelletizing. The main characteristics of an appropriate biomass for pelletizing include moisture content, chemical composition, and flow characteristics (Obernberger and Thek 2010; Döring 2013).

### 2.4.2. Pelletizing equipment

The pellet press consists of a matrix (die) and a roller. There are two types of pellet press: press with a flat (disk) die (Fig. 2-13d) and press with a cylindrical (ring) die (Fig. 2-13e). Ring die pellet mills consist of a die ring that runs around fixed rollers. The material is fed to the rollers sideways and pressed through the holes of the die. The rollers of flat die pellet mills rotate on top of a horizontal die. The material in the pellet mill is squeezed between roller and die and forced to flow into the press channels. The extruded pellets are cut off at a specified length by a knife blade.

### 2.4.3. Processing parameters

#### 2.4.3.1. Moisture content

The role of moisture in pelletization is very important. Moisture acts as one of the binding agents in the pelletizing process. There is a critical limit of moisture that can produce good quality pellets. Too much moisture makes the particles slippery and they slide through the die holes easily resulting in reduced pellet quality. Too dry materials may plug the die channels if the resistance from the channel exceeds the compresive force of the roller (Bhattacharya 1989; Coronella et al. 2012). According to Hansen et al. (2009), the moisture content in the raw material should be about 10% before the pelletizing process begins. Raw material with a moisture content higher than 15% is difficult to pelletize.

Tabil (1996) studied the compaction behaviour of alfalfa pellets and concluded that grind moisture content in the range of 8.5 and 10% (wet base) was optimal for pelletizing. Li and Liu (2000) used oak wood, oak bark, pine wood, and cottonwood in the forms of sawdust and chips for producing densified fuel logs. They found that the
moisture ranges between 5 and 12% are necessary for all woody materials, and optimum moisture content is about 8%. Mani et al. (2006) studied the effect of raw material moisture content and particle size on the mechanical properties of wheat, barley straw, corn stover and switchgrass pellets. They reported that a decrease in moisture content from 15 to 12% (wet base) and a reduction in hammer mill screen size from 3.2 to 0.8 mm increased the density of pellets.

Moisture lowers the friction occurring when the biomaterial is compacted and pressed through the channels of the die and consequently also the back pressure. Therefore, at higher moisture content the friction and back pressure is low which results in a low density of densified products (Samuelsson et al. 2009).

**2.4.3.2. Temperature**

The die temperature is determined by the frictional heat generated during the pelletization. It is not a controlled parameter in the industrial process. However, it may be indirectly controlled through several factors such as species, particle size, moisture content, and feeding rate. The die temperature is required to be higher than the glass transition temperature of raw materials to produce strong pellets. However, the die temperature should not be too high because it can cause frequent blocking of materials in the die (Kaliyan and Morey 2009; Kaliyan and Morey 2010; Lam et al. 2013).

Rhén et al. (2005) studied the effect of raw material moisture content (6.3 - 14.7%), densification pressure (46 – 114 MPa) and temperature (26 – 114 °C) on the properties of pellets made from Norway spruce sawdust. They concluded that high temperature and low moisture content are the most important variables to increase the strength and density of pellets.

Zaini et al. (2008) investigated the density, heating value, and composition of pellets made from lodgepole pine infested with mountain pine beetle. They stated that the glass transition temperature of lignin and hemicelluloses is greatly dependent upon moisture content. Therefore, increasing temperature to 110 °C may lead to softening of the amorphous components (e.g. lignin) leading to an increased pellet density.

Nielsen et al. (2009a) showed that increasing temperature and moisture content decreased the energy requirements for all components of the pelletizing process and that beech required more energy than pine in all components. He also reported that increasing temperature resulted in stronger pellets.
Stelte et al. (2011) used beech, spruce and straw for fuel pellet production. They found that the compression strengths of the pellets were higher for pellets produced at higher temperature, and much higher for wood pellets than for straw pellets.

### 2.4.3.3. Particle size

Particle size, along with temperature and moisture content of the raw material, is one of the most important variables affecting pellet mechanical properties. Smaller particles provide a greater surface area, thus facilitating heat conduction, which is beneficial to activate the binding properties of chemical components (Carone et al. 2011). Small particles tend to produce pellets with higher density and durability. A mixture of particle sizes result in less porous pellets (Kaliyan and Morey 2006).

Bergström et al. (2008) investigated the effects of raw material particle size distribution (< 1.0 mm, 1.0 – 2.0 mm, and 2.0 – 4.0 mm) on pellets density. The results showed that particle size distribution of raw material has a minor influence on the density of pellets produced. According to them, the raw material was broken down during the pelletizing process resulting in a narrower particle size distribution.

### 2.4.3.4. Pelletizing pressure

Li and Liu (2000) studied the densification behaviour of oak sawdust, oak mulch, oak bark, oak chips, pine sawdust, cottonwood sawdust, and cottonwood mulch in the pressure range of 34 and 138 MPa. They found that increasing pressure increased the abrasive resistance, impact resistance and compressive resistance of logs made from these biomass materials. Mani et al. (2004) studied the effect of pressures in the range of 31.08 and 136.77 MPa on the density of pellets made from wheat, barley straw, corn stover, and switchgrass using a single pelletizer. They found that higher applied pressure resulted in higher density of pellets, except for the the corn stover. Rhén et al. (2005) reported that compression strength was significantly correlated with pellets density. The pressure in the tested range of 46 – 114 MPa had a small effect on pellet density.

### 2.4.3.5. Raw material composition and additives

The chemical composition of the biomass, which includes compounds like cellulose, hemicelluloses, protein, starch, lignin, crude fiber, fat, and ash also affect the densification process. During compression at high temperatures, the protein and starch plasticizes and acts as a binder, which assists in increasing the strength of the pelletized product (Kaliyan and Morey 2009).
Lehtikangas (2001) reports a weak correlation between lignin content and pellet durability. Generally, longer storage in bulk leads to microbial degradation of the hemicelluloses and cellulose fractions of the feedstock, such that those that were stored longer have a larger percentage of lignin content. As noted, moisture content also plays a critical role in pellet formation. However, the reason for this effect is not well understood. Traditional thinking is that some moisture content is necessary to develop intermolecular (van der Waals forces and hydrogen bonds) and interfacial forces that serve to bring particles in closer contact with one another during the binding process.

Nielsen et al. (2010) studied the effect of extractives and storage on the pelletizing process of sawdust. They concluded that the differences in extractive content between raw materials of different wood species may explain clearly the differences in pelletizing properties. The extractives which act as agents in the pelletizing processes help lower the pelletizing energy requirements and thus increase the capacity of the pellet mills, but also have a negative effect on the pellet strength.

2.5. Quality parameters/characteristics

CEN/TC 335 of solid biofuels has published 27 technical specifications for solid biofuels and these are upgraded to EN-standards. The two most important are fuel specifications and classes (CEN/TS 14961) and fuel quality assurance (CEN/TS 15234). The quality of fuel pellets is determined by several key parameters including moisture content, calorific value, mechanical durability, particle density, ash content, and ash melting point. These parameter values are defined in the EN 14961-1 standards as summarized in Table 2-1.
Table 2-1. Specification of properties for pellets (EN 14961-1).

<table>
<thead>
<tr>
<th>Quality parameter</th>
<th>European standard specifications</th>
</tr>
</thead>
<tbody>
<tr>
<td>Moisture content (MC), percent (wt. %)</td>
<td>MC10: ≤ 10%, MC15: ≤ 15%</td>
</tr>
<tr>
<td>Diameter (D) and length (L)</td>
<td>D06: D = 6 mm ± 1.0 mm, and 3.15 ≤ L ≤ 40 mm,</td>
</tr>
<tr>
<td></td>
<td>D08: D = 8 mm ± 1.0 mm, and 3.15 ≤ L ≤ 40 mm,</td>
</tr>
<tr>
<td></td>
<td>D10: D = 10 mm ± 1.0 mm, and 3.15 ≤ L ≤ 40 mm,</td>
</tr>
<tr>
<td></td>
<td>D12: D = 12 mm ± 1.0 mm, and 3.15 ≤ L ≤ 50 mm,</td>
</tr>
<tr>
<td></td>
<td>D25: D = 25 mm ± 1.0 mm, and 3.15 ≤ L ≤ 50 mm</td>
</tr>
<tr>
<td>Mechanical durability (DU)</td>
<td>DU97.5: ≥ 97.5%, DU96.5: ≥ 96.5%, DU95.0: ≥ 95.0%,</td>
</tr>
<tr>
<td></td>
<td>DU95.0 + : &lt; 95.0% (minimum value to be stated)</td>
</tr>
<tr>
<td>Amount of fines (F) (wt. %, &lt; 3.15 mm)</td>
<td>F1.0: ≤ 1.0%, F2.0: ≤ 2.0%, F3.0: ≤ 3.0%, F5.0: ≤ 5.0%,</td>
</tr>
<tr>
<td></td>
<td>F5.0 + : &gt; 5.0% (maximum value to be stated)</td>
</tr>
<tr>
<td>Ash content (A), percent (wt. % d.b.)</td>
<td>A0.7: ≤ 0.7%, A1.0: ≤ 1.0%, A1.5: ≤ 1.5%,</td>
</tr>
<tr>
<td></td>
<td>A2.0: ≤ 2.0%, A3.0: ≤ 3.0%, A5.0: ≤ 5.0%,</td>
</tr>
<tr>
<td></td>
<td>A7.0: ≤ 7.0%, A10.0: ≤ 10.0%, A10.0 + : &gt; 10.0%</td>
</tr>
<tr>
<td>Bulk density (BD)</td>
<td>BD550: ≥ 550 kg m⁻³, BD600: ≥ 600 kg m⁻³,</td>
</tr>
<tr>
<td></td>
<td>BD650: ≥ 650 kg m⁻³, BD700: ≥ 700 kg m⁻³,</td>
</tr>
<tr>
<td></td>
<td>BD700 + : &gt; 700 kg m⁻³ (minimum value to be stated)</td>
</tr>
<tr>
<td>Calorific value as received (MJ kg⁻¹)</td>
<td>Minimum value to be stated</td>
</tr>
<tr>
<td>Sulfur (S), percent (wt.% of dry basis)</td>
<td>S0.02: ≤ 0.02%, S0.05: ≤ 0.05%, S0.08: ≤ 0.08%</td>
</tr>
<tr>
<td></td>
<td>S0.10: ≤ 0.10%, S0.20: ≤ 0.20%,</td>
</tr>
<tr>
<td></td>
<td>S0.20 + : &gt; 0.20% (maximum value to be stated)</td>
</tr>
<tr>
<td>Nitrogen (N), percent (wt.% of dry basis)</td>
<td>N0.3: ≤ 0.3%, N0.5: ≤ 0.5%, N1.0: ≤ 1.0%,</td>
</tr>
<tr>
<td></td>
<td>N2.0: ≤ 2.0%, N3.0: ≤ 3.0%,</td>
</tr>
<tr>
<td></td>
<td>N3.0 + : &gt; 3.0% (maximum value to be stated)</td>
</tr>
<tr>
<td>Chlorine (Cl), percent (wt.% of dry basis)</td>
<td>Cl0.02: ≤ 0.02%, Cl0.03: ≤ 0.03%,</td>
</tr>
<tr>
<td></td>
<td>Cl0.07: ≤ 0.07%, Cl0.10: ≤ 0.10%,</td>
</tr>
<tr>
<td></td>
<td>Cl0.10 + : &gt; 0.10% (maximum value to be stated)</td>
</tr>
<tr>
<td>Ash melting behavior, °C</td>
<td>Deformation temperature should be stated</td>
</tr>
</tbody>
</table>
2.5.1. Moisture content

Moisture content is highly variable in biomass and has a significant effect on the fuel quality in terms of combustion chemistry and energy balance. The moisture content is also playing a major role in the interactions between physical-mechanical properties and calorific value of solid biofuels (Fig. 2-16).

![Diagram of interactions between physical-mechanical and combustion properties of solid biofuels.](image)

Fig. 2-16. Interactions between physical-mechanical and combustion properties of solid biofuels (adapted from Kaltschmitt and Hartmann 2009).

2.5.2. Calorific value

Calorific value or heating value is the amount of energy released per unit mass or volume of a material upon complete combustion. It can be categorized as either the higher heating value (also known as the gross calorific value) or the lower heating value (also known as net calorific value). Higher heating value (HHV) considers moisture in liquid state after combustion. Lower heating value (LHV) considers that water formed by combustion of the fuel remains in vapour form. These energy measures can be reported on dry basis, as-received basis, or wet basis.

The heating value obtained by the bomb calorimeter, which measures the enthalpy change between reactants and products at 25 °C, is termed HHV. The net calorific value at constant volume and the net calorific value at constant pressure of the biofuel are obtained by calculation from the gross calorific value at constant volume determined on the analysis sample. The calculation of the net calorific value at constant volume requires information about the moisture and hydrogen contents of the analysis sample. In principle, the calculation of the net calorific value at constant pressure also requires information about the oxygen and nitrogen contents of the sample.
The heating value is generally correlated to the presence of C, H and O, with an increase attributed to higher percentage of C and H, while a decrease is observed with higher degrees of oxidation. The range of heating values for biomass fuels is lower than that of bituminous coals since biomass fuels have higher oxygen concentration (Fig. 2-17). However, biomass exhibits increased heating values compared to several lignite varieties (Lu and Baxter 2011). The heating value in biomass fuels is also correlated to the presence of cellulose, hemicellulose, and lignin. Lignin has the highest heating values, while hemicellulose has the lowest due to its high oxidation degree (Lu and Baxter 2011; Moya and Tenorio 2013).

Fig. 2 - 17. Difference on molar H:C to O:C ratio between biomass and coal (Lu and Baxter 2011).

The HHV of oven-dried wood fuel is about 7% higher than the LHV. For wet wood this relative difference between the HHV and the LHV increases (Fig. 2-18), because the possible heat yield through recondensation of the produced water vapor is respectively higher (Hartmann 2013).

Fig. 2-18. Difference between gross and net calorific value at variable moisture content of fuel (Kaltschmitt and Hartmann 2009).
2.5.3. Volatile matter

During combustion, volatile matter and bound water in the wood fuel are released in the pyrolysis zone forming vapours consisting of water, tar, oils, and gases. Quantitative analysis of volatiles thus helps in designing fuel combustors such as wood or pellets stoves and boilers (Lata and Mande 2009).

2.5.4. Ash content and its melting properties

Mineral content in the fuel that remains in oxidized form after the completion of combustion is generally called the ash. The amount and nature of ash and its behaviour at high/elevated temperature affect the design and type of ash handling system employed in biomass combustors (Obernberger and Thek 2010; Döring 2013; Hartmann 2013).

The ash melting behaviour is depending on the ash constituents and it is thus directly correlated with the fuel composition. Fuels with a low ash melting point represent a high risk of slag deposition and adhesion for combustion chamber and heat exchanger surfaces. Such coatings can lead to blockages, breakdowns and to an inhibited combustion air supply (Obernberger and Thek 2010; Hartmann 2013).

2.5.5. Shape and dimensions of pellets

The particle size is relevant for several technical reasons. The main impact of an unsuitable size distribution is given for mechanical systems in silo emptying, conveying, or charging systems. Too long particles can lead to blockages and damages of the conveyor. In addition, the tendency to form a bridge during transhipment such as at the bottom of a silo is determined by an unsuitable size distribution (Obernberger and Thek 2010; Hartmann 2013).

2.5.6. Particle and bulk density

Bulk density is defined as the weight per unit volume of loosely packed biomass. It includes the actual volume of biomass, pore volume, and void volume between particles. Bulk density of the wood particles is influenced by the size and shape of the particles. Biomass with high bulk density is advantageous for combustion systems. Particle density (also called as single density) is defined as the mass of an individual particle divided by its volume. The particle density of solid fuels influences their combustion behaviour. Fuels with a higher particle density have a longer burnout time (Obernberger and Thek 2010; Hartmann 2013).
2.5.7. Mechanical durability

Durability or abrasive resistance of pellets is an important property to minimize dust emissions during handling, transport and storage. Dust emissions can be a health hazard to people handling pellets, disturb boiler feeding systems, and increase the risk of fire and explosion during shipping, handling, and storage (Temmerman et al. 2006; Obernberger and Thek 2010).

2.6. Recent trends and developments/ Pellets quality improvements

The future bioenergy provision concept for power, heat and transport fuels are characterized by more complex demands. A future energy market is characterized by the need for a sustainable flexible energy carrier with homogeneous properties for application in the fields of combined heat and power (CHP), heat and fuel. To some extent these energy carriers are already available today. However, in many cases untreated biomass cannot fulfill the requirements of existing and future conversion processes or demands. Fig. 2-19 represents two promising types of future bioenergy carriers, including the advanced solid biofuels and biogenic substitutes for natural gas (Billig et al. 2015).

Fig. 2-19. Selected future bioenergy carriers for supporting the flexible use of biomass (adapted from Billig et al. 2015).
The growing importance of pelletization does not only apply to wood fuels but also to agricultural residues and non-woody energy crops. However, the scope of using herbaceous fuels is today still limited due to their unfavorable properties. The upcoming search for new resources therefore needs to be associated with developing and applying new methods for fuel improvements. This can, for example, be the applications of additives or applying other pretreatment measures before pelletizing. Hydrothermal pretreatment (also known as hot water extraction or sometimes named as wet torrefaction) and torrefaction (also known as thermal rectification or dry torrefaction) are two promising types of pretreatment processes. The thermal decomposition of the major components of a biomass depends mainly on temperature, particle size, and time of exposure (Klemm et al. 2013). The rate and extent of thermal decomposition of woody biomass can be distinguished in five main reaction regimes (Fig. 2-20) including: regime A represents the physical drying temperature of the biomass, regime B is defined as a temperature zone in which softening of lignin occurs, regime C is characterized by depolymerization and condensation of the shortened polymers, regime D represents temperature range which leads to a limited devolatization and carbonization of the intact polymers, and regime E represents temperature range which leads to extensive devolatization and carbonization of the polymers as well as of the solid products that were formed in regime D. Fig. 2-20 indicates that hemicellulose decomposition starts at lower temperatures and proceeds over a narrower temperature range as compared to cellulose and lignin.

Fig. 2-20. Temperature regimes for the thermal decomposition of cellulose, hemicellulose, and lignin: (A) - Drying, (B) - Glass transition, (C) - Depolymerization and recondensation, (D) - Limited devolatization and carbonization, and (E) - Extensive devolatization and carbonization (after Klemm et al. 2013).
The whole wood starts to thermally degrade at about 200 °C (Fig. 2-21). Between about 300 - 375 °C, the majority of the carbohydrate polymers have degraded and only lignin remains. The hemicellulose components start to decompose at about 225 °C and are almost completely degraded by 325 °C. The cellulose polymer is more stable to thermal degradation until about 370 °C, and then decomposes almost completely over a very narrow temperature range. Both acid lignin and milled wood lignin start to decompose at about 200 °C, but are much more stable to thermal degradation as compared to the carbohydrate polymers. The decomposition of cellulose leads mainly to volatile gases, while lignin decomposition leads mainly to tars and char (Shafizadeh 1984).

![Graph of thermal decomposition of wood and its cell wall components](image)

Fig. 2-21. Thermal decomposition of wood and its cell wall components (adapted from Kaltschmitt and Hartmann 2009).

### 2.6.1. Torrefaction

Torrefaction is a mild pyrolysis process for the production of solid biofuels. It is performed at temperatures between 200 °C and 300 °C within an inert atmosphere for a certain period of time depending on the particle size. In the range of torrefaction temperatures, degradation of hemicelluloses occurs (Hillis 1984). Lignin and cellulose are subjected to partial depolymerization, and the crystallinity index of cellulose is increased (Tumuluru et al. 2011). After this treatment, the biomass contains typically 60 - 70% of the initial mass and 90% of the initial energy content. The physico-chemical properties of torrefied biomass are superior to those of the original biomass. Key benefits related to fuel storage, distribution, and utilization include energy densification, homogenization, improved grindability, pronounced hydrophobic character, enhanced reactivity.
2.6.2. Hydrothermal treatment/ Hot water extraction

Water is highly polar and a weak solvent for most organic compound under ambient conditions, but raising the temperature significantly above ambient has a dramatic effect. Increasing temperature means water becomes less polar. The polarity of hot water is measured by the value of the dielectric constant. When water is heated above 100 °C its dielectric constant decreases and water becomes similar to organic solvents. At 214 °C the dielectric constant of water is the same as that of methanol at room temperature. At 295 °C water becomes similar to acetone.

The effect of thermal pretreatment on properties of wood composites have recently been reviewed by Pelaez-Samaniego et al. (2013). These authors reported that many terms can be used to describe the hot water extraction (HWE) process, including autohydrolysis, hydrothermolysis, hot compressed water treatment, water hydrolysis, wet torrefaction, hydrothermal carbonization, aqueous fractionation, aqueous liquefaction. HWE temperatures are generally between 100 °C and 374 °C. However, many thermolabile compounds such as those in essential oil can be degraded at high temperatures (more than 200 °C). Some compounds can even degrade at 175 °C. In general, HWE is a fast, clean, cheap, environmentally friendly and green sustainable technique. HWE can extract polar, mid-polar and even non-polar compounds from plant samples. Phenolics, essential oils, flavonoids, pectins and proteins have been extracted using HWE. Temperature plays an important role in extraction efficiency of target compounds. Increasing temperature higher than 175 °C may cause degradation problems with many samples. Pressure is important to keep water in the liquid state. Green and sustainable processes are becoming more popular. The cost of the HWE process may appear more expensive in the laboratory scale, however, on an industrial scale it is competitive with commercial methods (Özel and Göğüç 2014).

HWE is capable of dissolving and thus washing out the inorganic components from solid biofuels. This is an additional advantage of HWE over torrefaction since it can produce cleaner solid fuels in term of inorganic content, which are beneficial to the combustion process. In addition, HWE process can produce solid fuels with a higher calorific value, higher energy yield and a better hydrophobicity at much lower temperature and holding reaction time (Bach et al. 2013). On the other hand, many valuable organic compounds such as flavonoids, vitamine, antioxidants and antimicrobials can be extracted selectively using HWE process (Özel and Göğüç 2014). Besides, other organic compounds such as acetic acid, formic acid, lactic acid, phenol, furfural, etc. are also found in the liquid fraction following HWE process (Amidon and Liu 2009). The potential use of these water-soluble organic substances for production of valuable products may contribute to the cost of HWE process (Ragauskas et al. 2006; Cybulksa et al. 2013).
Chapitre 3. Objectives of this study

3.1. General objective
The overall objective of this study is to contribute to a deeper understanding of hardwood pelletization by investigating the technical possibility of converting low quality hardwoods into conventional and advanced fuel pellets.

3.2. Specific objective
The specific objective of this study is to investigate the potential use of low quality hardwoods for the production of conventional and advanced fuel pellets. To achieve this objective the following work needs to be done:

(i) Determine the changes in chemical composition and calorific value of wood related to tree vigor;
(ii) Determine the influence of process parameters and raw material characteristics on physical and mechanical properties of wood pellets made from trees of different vigor classes, and to determine which combinations of these independent variables provide the best pelletizing process in terms of friction force and pellets quality;
(iii) Determine the effect of hot water treatment parameters on the physical and chemical characteristics of treated material, and to determine the advanced pretreatment and pelletization conditions to improve pelletization performance and quality of pellets with a focus on reducing the ash content and improving the main fuel properties (e.g. energy content, water resistance, and mechanical durability) of the fuel pellets.

3.3. Hypotheses
Hypothesis 1: There exists a variation on the amount of chemical compositions (e.g. ash, lignin, and extractive contents) between hardwood trees of different vigors.

Hypothesis 2: There exists optimal pelletizing parameters (e.g. compressive force, temperature, moisture content, and particle sizes) to a pelletization process in terms of friction force and pellets quality.

Hypothesis 3: Application of hot water treatment to wood particles will enhance pelletization characteristics and fuel properties of pellets.
Chapitre 4. Material and methods

4.1. Material sampling and preparation

4.1.1 Tree classification and selection

Tree sampling was carried out in July 2010 at two degraded hardwood stands composed mainly of sugar maple (*Acer saccharum* Marsh.) and yellow birch (*Betula alleghaniensis* Britton). The stands were located in the vicinity of Mont-Laurier, Québec, Canada (46°39'40"N, 75°36'30"W and 46°39'05"N, 75°36'25"W) (Fig. 4-1).

![Location of selected trees in the forest stands by Filip Havreljuk.](image)

The trees were classified according to the tree vigor (MSCR) classification system proposed by Boulet (2007) and described as follows. Trees of reserve stock (class R) are free of any symptoms of disease or damage and are considered as healthy trees with the highest probability of survival. Growing trees (class C) have minor defects but are not biologically declining and are expected to survive until the next harvest without risk of imminent wood decay. Low quality or defective trees (class S) are considered to be declining in terms of vigor,
wood quality, and volume increment, and are not expected to survive until the next harvest. Finally, moribund trees (class M) show signs of either lethal pathological infection or severe damage with high risk of trunk breakage. Moribund trees are biologically declining and are assumed to have a high probability of mortality before the next scheduled harvest.

After classifying each tree according to its vigor class, a total of 9 sugar maple trees and 9 yellow birch trees (3 vigorous/healthy trees, 3 weakened/defective trees, and 3 moribund/dying trees of each species) were selected and felled. Three sample logs were extracted from each sample tree and used as raw material of this study. Details on sample logs are presented in chapters 5 and 6.

4.1.2 Preparation of wood particles for pellets production

A band saw was used to convert sample logs into sticks (Fig. 4-2). A chipper was used for size reduction of large sticks into wood chips (Fig. 4-2). The wood chips were air-dried in layers of about 5 cm thick to a moisture content between 10 and 15%.

The air-dried wood chips were sent into a hammer mill and Pallmann ring refiner for final size reduction in preparing raw material (wood particles) for pelletization. Samples were ground using a hammer mill with a 12.5 mm screen size. The samples were then ground using the Pallmann ring refiner with three different screen sizes of 4.0 mm, 2.5 mm, and 1.5 mm (Fig. 4-3).

The fraction size distribution of the ground particle samples was analyzed. For each test, a sample of 100 g was placed in a stack of sieves arranged from the largest to the smallest opening. A Ro-Tap sieve shaker (Fig. 4-4) was used to determine the geometric mean particle size using standard sieve series number 6, 7, 14, 18, 35...
and 60 (sieve opening size: 3.35, 2.80, 1.40, 1.00, 0.50 and 0.25 mm, respectively). The duration of sieving was 15 minutes.

Fig. 4-3. Final size reduction step: Wood chips (top left), Pallmann ring refiner (bottom left) and its grinding chamber (middle) and its ground particles (top left), and the screens of different sizes (bottom right).

Fig. 4-4. Particle size distribution analysis: laboratory samples (left) and a Ro-Tap sieve shaker with standard sieve series (right).

4.1.3 Hydrothermal treatment

Air-dried wood particles were subjected to HWT using a 2-liter batch reactor (Fig. 4-5). The HWT process was performed at three controlled temperature levels of 150 °C, 175 °C, and 200 °C for 30 min under its self-generated pressures. In each treatment, an amount of 250 g dried wood particles (size of 0.5 - 1.0 mm screens) and 1000 ml deionized water (the equivalent ratio of 4:1 w/w) was loaded into the reactor. The reactor was heated up and maintained at the desired temperature for 30 min. Self-generated pressure values of about 0.47 MPa, 0.89 MPa, and 1.55 MPa were observed corresponding to the set temperatures of 150 °C, 175 °C, and 200 °C, respectively. The reactor was cooled rapidly by immersing it in a cold water bath. The gas was released to the atmosphere. The liquid fractions were filtered from the solids and collected for further analysis. The solid
fractions were then washed thoroughly with deionized water and exposed to ambient conditions for air drying. The air dried wood particles were packaged and stored for further analyses.

Fig. 4-5. Equipment and products of hydrothermal treatment of wood particles: a batch reactor (left), the solid fractions/treated wood particles (top right) and the liquid fractions (bottom right).

For analysis of ash content, a test specimen of about 2 g was weighed at an accuracy of 0.1 mg in an incineration crucible. The crucible plus specimen (Fig. 4-6) was placed in a furnace at room temperature. The temperature of the furnace was raised progressively (5 °C min⁻¹) to 250 °C and maintained at this temperature for 60 minutes. The furnace temperature was then raised progressively (5 °C min⁻¹) to 550 °C, and kept at this temperature for 180 minutes. After incineration the crucible with contents was cooled in a desiccator and weighed. The percentage of ash (A), based on the weight of the oven-dry sample, was calculated from Equation 4-1.

\[
A(\%) = \frac{W_1}{W_2} \times 100
\]

(4-1)

where \( W_1 \) is mass of ash, and \( W_2 \) is mass of oven-dry sample.

Fig. 4-6. Wood samples before combustion (left) and their corresponding ash (right) of untreated and hydrothermally treated (HWT) wood particles.
For HHV measurement, the samples were combusted in a Parr 6400 Automated Isoperibol calorimeter (Fig. 4-7). The calorimeter was calibrated with a benzoic acid standard (heat of combustion 26.454 MJ kg$^{-1}$). The ground material sample (approximately 0.5 g) was compacted into a tablet and burnt in an oxygen bomb calorimeter to determine the calorific value. A cotton thread was attached to the platinum ignition wire and placed in contact with the tablet (Fig. 4-7). The HHV was calculated from Equation (4-2).

$$HHV_d = HHV_w \times \frac{100}{(100 - MC)}$$

(4-2)

where $HHV_d$ is the higher heating value at constant volume of the dry (moisture-free) fuel, $HHV_w$ is the higher heating value at constant volume of the fuel as received, and $MC$ is moisture content of the sample (as percentage by mass on an air-dried basis).

![Image](image1.png)

Fig. 4-7. Measurement of higher heating value: Calorimeter (left), sample preparation (bottom right), and result display (top right).

### 4.2. Pelletizing experiment

#### 4.2.1 Single pelletizer development and setup

A schematic diagram of the single pelletizer used in this study is shown in Fig. 4-8. The pelletizer has a plunger and cylinder. The cylinder has an internal diameter of 6.25 mm and a length of 150 mm. The cylinder is wrapped with a heating element covered by insulation material. Two type-T thermocouples are placed close to the inside of the cylinder wall at each of the cylinder ends. The thermocouple close to the base was connected to a temperature controller (Fig. 4-9). Loading was made possible by a plunger attached to the MTS Model RT/50 testing machine which is equipped with a 50 kN load cell (Fig. 4-9).
4.2.2 Pellets making procedure

Initially the raw material (untreated and treated wood particles) was conditioned to the required moisture content. A known amount of raw material was prepared to compact in the cylinder of the single pelletizer. The cylinder was first heated to the desired temperature then the raw material was loaded into the cylinder and compressed by the piston to the desired pressure. The pellet formed was pressed out of the cylinder by removing the bottom plate.
Chapitre 5. Fuel properties of sugar maple and yellow birch wood in relation with tree vigor

by

Quy Nam Nguyen, Alain Cloutier, Alexis Achim, and Tatjana Stevanovic

in:

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Résumé

Les propriétés énergétiques du bois obtenues à partir de l’érable à sucre et du bouleau jaune de peuplements de feuillus tempérés situés dans la Province de Québec, Canada ont été étudiées pour savoir comment la vigueur des arbres affecte la composition chimique et la valeur calorifique du bois. Cette étude a porté sur les propriétés physiques et chimiques du bois dans le but d’utiliser le matériel pour la production de biocombustibles solides. Les éléments spécifiques mesurés comprenaient les valeurs calorifiques du bois, et les teneurs en substances extractibles, en cendres et en lignine. Des changements dans la composition chimique ont été trouvés entre les classes de vigueur des arbres. Les arbres peu vigoureux avaient des teneurs en extractibles, en cendres et en lignine plus élevées que les arbres vigoureux. Les extractibles totaux se situaient entre 4,8 et 7,3% pour l’érable à sucre, et entre 3,3 et 5,1% pour le bouleau jaune. La lignine de Klason se situait entre 21,4 et 23,5% pour l’érable à sucre, et entre 18,6 et 21,5% pour le bouleau jaune. La teneur en cendres se situait entre 0,3 et 0,9% pour l’érable à sucre, et entre 0,2 et 0,4% pour le bouleau jaune. Les effets combinés de la teneur en lignine élevée qui pourrait contribuer à une meilleure auto-liaison des particules et de la teneur en extractibles élevée qui pourrait faciliter le processus de granulation rendent les arbres peu vigoureux plus appropriés pour la conversion en biocombustibles solides. Les plus grandes quantités d'extractibles et de lignine présentes dans l’érable à sucre et le bouleau jaune de faible vigueur pourraient aussi avoir un rôle positif dans le maintien des valeurs calorifiques élevées de ce bois, malgré une plus forte teneur en cendres.
Abstract

The fuel properties of wood obtained from sugar maple (SM) and yellow birch (YB) of temperate hardwood stands located in the Province of Québec, Canada were studied to see how tree vigor affects the chemical composition and calorific value of the wood. This study focused on the physical and chemical properties of wood with the aim of using the material for the production of solid biofuels. Specific items measured included the wood’s calorific values, and the levels of extractives, ash, and lignin. Changes in chemical composition were found among tree vigor classes. The low vigor trees had higher extractives, ash, and lignin contents than the vigorous trees. Total extractives ranged between 4.88 and 7.32% in SM, and between 3.35 and 5.12% in YB. Klason lignin ranged between 21.46 and 23.53% in SM, and between 18.60 and 21.51% in YB. Ash content ranged between 0.38 and 0.97% in SM, and between 0.26 and 0.47% in YB. The combined effects of higher lignin content that could contribute to a better self-bonding of particles and of higher extractives content that could facilitate the pelletization process makes the low vigor trees more suitable for conversion into solid biofuels. The higher amounts of extractives and lignin present in the low vigor sugar maple and yellow birch trees could also have a positive role in maintaining the high calorific values of this wood despite higher ash content.
5.1. Introduction

In recent years, a clear interest has arisen for the use of forest biomass as a biofuel. This is mainly motivated by the low environmental impact and low carbon emissions resulting from the use of forest biomass as a source of energy. Also, it may not be achievable to meet the global increase in energy consumption without the contribution of biofuels (Hunt 2009; Demirbas 2010; Masia et al. 2010).

The most important characteristic of a biofuel is its energy content (Lestander 2013). The standard measure of the energy content of a fuel is its higher heating value (HHV), which is sometimes called the calorific value or the heat of combustion (Jenkins et al. 1998). It may be expressed as either the higher heating value (also known as the gross calorific value) or the lower heating value (also known as the net calorific value). The CEN/TS 14588 (2003) standard defines calorific or heating value as the quantity of energy released per unit mass or volume of a material upon complete combustion. Heating value is the most fundamental property in connection with fuel quality, and it is therefore widely used as a mean to compare different types of fuels or feedstocks.

The heating value of woody materials is directly related to its chemical composition. The calorific value of moisture-free wood is approximately 18 to 21 MJ kg$^{-1}$, depending on the tree species (Schobert 2013). Because lignin has a higher amount of energy per unit mass than cellulose (26 and 18 MJ kg$^{-1}$, respectively), the calorific value varies with the lignin content, and to a much greater extent, with the resin content (Wang et al. 1982; White 1987; Moya and Tenorio 2013). Studies were performed to determine the relationships between the chemical composition (hemicellulose, cellulose, and lignin) of woody biomass and its HHV. No direct relation between the hemicellulose and cellulose content of a biomass and its HHV was reported (Demirbas 2005). However, a highly significant linear correlation was found between the HHV and lignin content within a biomass (White 1987; Demirbas 2001; Demirbas 2005; Telmo and Lousada 2011).

Inorganic minerals (ashes) are noncombustible, and their presence in wood has an adverse effect on the heating value (Wang et al. 1982; Kataki and Konwer 2001; Demirbas 2002; Moya and Tenorio 2013; Schobert 2013). The ash content of biomass varies in a broad range from around 0.5% (dry basis (d.b.)) for some clean woody biomasses to 12% (d.b.) for straws and cereals; these ash values can be even higher if the fuel is contaminated with mineral impurities (van Loo and Koppejan 2008).

The chemical composition in the tree stems differs from that in the branches, knots, tops, and roots. Marked variations were also observed within a stem, especially in the radial direction from juvenile to mature wood, and from heartwood to sapwood (Barnett and Jeronimidis 2003). Differences between reaction wood and normal wood were also reported (Sjöström and Alén 1999). Generally, the lignin content of hardwoods varies in the
range of 16 to 24%. These estimates are based on Klason lignin content (acid-insoluble lignin) determined without accounting for the acid-soluble lignin.

Acid-soluble lignin is low molecular weight lignin that is solubilized in the acidic hydrolysis solution. Inclusion of acid soluble lignin concentration in the total lignin value is necessary, as acid soluble lignin can represent an important portion of lignin, especially in hardwoods (Sluiter and Sluiter 2011). The available data on within tree variation suggest that the lignin content of sapwood is equal to or slightly higher than that of the heartwood, higher in juvenile wood than in mature wood, and lower in tension wood than in normal wood. In many species, lignification of the wood tissue is nearly completed during the year of formation in the cambial zone (Sarkanen and Ludwig 1971).

Woody biomass could be considered as a potential renewable energy source with supplementary value-added applications depending on its chemical composition variation (Lindsey et al. 2013; Sengupta and Pike 2013; St-Pierre et al. 2013). Wood extractives can have many uses as natural dyes for fabrics, foods and cosmetics, as well as potential medicines. These materials are receiving increased attention as low-volume but high-value products (Lavoie and Stevanovic 2005; Rossi et al. 2013). These extractives could be recovered before using wood as a fuel. Thus, it is of interest to study the effect of extractive removal on the calorific value of wood. The extractive compounds in wood can be grouped in two main types: lipophilic (soluble in organic solvents) and hydrophilic (water-soluble).

The constituents of lipophilic extractives include terpenoids and steroids, fats, waxes, and phenolic compounds that consist of stilbenes, lignans, tannins, and different classes of flavonoids. Fats are known as an energy source for parenchyma cells, whereas terpenoids and phenolics protect the tree against microorganism attacks (Sjöström 1993; Sjöström and Alén 1999).

Over the past decades, the most valuable tree stems within the Eastern hardwood forests in North America, sugar maple (Acer saccharum Marsh.) and yellow birch (Betula alleghaniensis Britton), have been harvested for the production of hardwood lumber (Nyland 1992; Bédard and Majcen 2003; Pothier et al. 2013). Thus, the current forests contain a large proportion of non-vigorous trees of very little or no commercial value (Havreljuk et al. 2014).

From the forest management standpoint, the presence of these low vigor and quality stems has a negative effect on forest health and value. Therefore, in the partial cuts typically applied to such stands, harvesting priority is
now given to the non-vigorous trees while leaving the more vigorous trees to grow for future harvest (Delisle-Boulianne et al. 2014).

To determine tree vigor in the field prior to harvesting, the tree vigor classification system proposed by Boulet (2007) has been applied to hardwood stands in the Province of Québec, Canada. This evaluation is based on pathological symptoms (e.g., presence of cankers and fungi), mechanical defects (e.g., cracks, leaning) and other visible features (e.g., trunk splits, defoliation).

Healthy trees have several defense mechanisms against fungal infection. However when a tree is wounded or otherwise weakened by living organisms, especially fungi, its chemical composition may change (Jane et al. 1970; Stokland et al. 2012). According to Barnett and Jeronimidis (2003), the occurrence of physical wounds, mechanical stress, or attack by biotic organisms induces many trees to produce traumatic tissues, and to synthesize and deposit protecting compounds in the affected areas. Most of these compounds are extractives and appear in high amounts in the affected parts of the wood.

Fungal attack has been found to be associated with the increase in lignin content because of its resistance to degradation by pathogens, as reported by Hawkin and Boudet (2003) for cider gum (Eucalyptus gunnii). Increases in extractive content following fungal attack was also reported by Barry et al. (2001) for shining gum (Eucalyptus nitens).

To the best of the authors’ knowledge, there are no reports in the literature on the effect of tree vigor on the calorific value of wood. In addition, the assessment of chemical composition and fuel characteristics of the biomass is important for the efficient utilization and conversion of the feedstock into biofuels and chemicals. Therefore, the aim of the present work was to determine the changes in chemical composition and calorific value of wood related to tree vigor for both sugar maple and yellow birch.

Because the current work was conducted with the intention of using the material for the production of solid biofuels with supplementary value-added applications, the investigation was focused on the determination of extractives, ash, and lignin content of wood, and the measurement of calorific values of both extracted and non-extracted wood samples.

5.2. Experimental

5.2.1. Materials and sample preparation
Tree sampling was carried out in July 2010 at two degraded hardwood stands composed mainly of sugar maple and yellow birch. The stands were located in the vicinity of Mont-Laurier, Québec, Canada (46°39'40"N, 75°36'30"W and 46°39'05"N, 75°36'25"W). The trees were classified according to the tree vigor (MSCR) classification system proposed by Boulet (2007) and described as follows. Trees of reserve stock (class R) are free of any symptoms of disease or damage and are considered as healthy trees with the highest probability of survival. Growing trees (class C) have minor defects but are not biologically declining and are expected to survive until the next harvest without risk of imminent wood decay. Low quality or defective trees (class S) are considered to be declining in terms of vigor, wood quality, and volume increment, and are not expected to survive until the next harvest. Finally, moribund trees (class M) show signs of either lethal pathological infection or severe damage with high risk of trunk breakage. Moribund trees are biologically declining and are assumed to have a high probability of mortality before the next scheduled harvest.

After classifying each tree according to its vigor class, 18 sample trees (9 sugar maple trees and 9 yellow birch trees) were selected and felled (Table 5-1). The selected trees were assigned to one of the 3 following categories: R - vigorous (healthy) tree, S - weakened (defective) tree, and M - moribund (dying) tree. Figure 5-1 presents the visual appearance of representative sample trees and their wood showing advanced biodeterioration (sap-staining fungi and/or wood rotting fungi) of wood of the less vigorous trees.

Table 5-1. Description of the 18 sample trees used for chemical analysis

<table>
<thead>
<tr>
<th>Tree vigor *</th>
<th>Type of defect/ Characteristics/ Description</th>
<th>DBH (cm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Healthy tree</td>
<td></td>
<td></td>
</tr>
<tr>
<td>(R class)</td>
<td>Green foliage and without wounds</td>
<td>Sugar maple: 30.0</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Yellow birch: 33.5</td>
</tr>
<tr>
<td></td>
<td></td>
<td>29.3</td>
</tr>
<tr>
<td></td>
<td></td>
<td>35.0</td>
</tr>
<tr>
<td>Weakened tree</td>
<td>Wounds, poor foliage (defoliation), presence of sap-staining fungi</td>
<td>Sugar maple: 39.5</td>
</tr>
<tr>
<td>(S class)</td>
<td></td>
<td>Yellow birch: 36.8</td>
</tr>
<tr>
<td></td>
<td></td>
<td>31.3</td>
</tr>
<tr>
<td></td>
<td></td>
<td>36.3</td>
</tr>
<tr>
<td>Dying tree</td>
<td>Partial bark lose or fallen off, presence of decay/ wood rotting fungi (fungal infection)</td>
<td>Sugar maple: 37.9</td>
</tr>
<tr>
<td>(M class)</td>
<td></td>
<td>Yellow birch: 33.7</td>
</tr>
<tr>
<td></td>
<td></td>
<td>27.2</td>
</tr>
<tr>
<td></td>
<td></td>
<td>31.3</td>
</tr>
</tbody>
</table>

DBH= Diameter at breast height of tree stem measured under bark at 1.3 m from the root collar, *The assignment of each tree to a vigor class in the field was based on a visual assessment of tree morphological characteristics. Trees were classified according to MSCR classification system proposed by Boulet (2007).
After the trees were felled, measured and marked, a sample log (of about 60 cm long) was extracted from each sample tree at a distance of approximately 70 cm from the root collar, wrapped in plastic bags, transported to the Renewable Materials Research Center, Laval University, Québec, Canada, and then stored at -5 °C in a laboratory freezer to avoid enzyme-induced changes and oxidation of the extractives (Sjöström and Alén 1999; St-Pierre et al. 2013).

Analyses in the present study were conducted only for the disks sectioned from sample logs. The bark was then removed from the disks. The wood material was converted into chips, dried at 30 °C for approximately 48 h, and milled. The coarse material was ground in a centrifugal mill (Pulverisette 19 grinder, Fritsch, Germany). The wood flour was sieved through 40 mesh (425 μm) and 60 mesh (250 μm) screens. After grinding and sieving,
the samples were kept in closed polyethylene bags and stored at -5 °C in a laboratory freezer until further analyses.

5.2.2. Determination of chemical properties

Ash content, extractive content in an ethanol-toluene mixture, and extractive content in hot water were determined in accordance with ASTM D1102-84, ASTM D1107-96 and ASTM D1110-84 (method B - hot water solubility) standards, respectively. When wood is treated with strong acids, the carbohydrates are hydrolyzed and an insoluble residue is obtained; this insoluble residue is the acid-insoluble lignin (Klason lignin). Because some of the wood extractives would remain with the insoluble lignin during the hydrolysis, these extractives were first removed from the wood by solvent extraction. In the present study, acid-insoluble lignin was determined in accordance to the ASTM E1721-01 standard. The chemicals used include ethanol (anhydrous grade) from Commercial Alcohols Inc. (Canada), toluene and 72% sulfuric acid (A.C.S grade) from VWR International (Canada). Acid-soluble lignin was analyzed within 4 h of the hydrolysis process using UV-Vis absorption spectroscopy (Varian, Cary 50 UV-Vis, Varian Inc., Australia) at 205 nm. Three replicates were performed for each test sample and the mean values were used for statistical analyses.

5.2.3. Determination of the calorific value

The gross calorific value (GCV), or higher heating value (HHV), represents the absolute value of the specific energy of combustion per unit mass of a solid biofuel burned in oxygen in a calorimetric bomb under specified conditions. In the present study, the HHV was determined in accordance to the CEN/TS 14918:2005 standard. The samples were combusted in a Parr 6400 Automated Isoperibol calorimeter (Parr Instrument Company, Illinois, USA). The calorimeter was calibrated with a benzoic acid standard (heat of combustion 26.454 MJ/kg). The ground material sample (approximately 0.5 g) was compacted into a tablet and burnt in an oxygen bomb calorimeter to determine the calorific value. A cotton thread was attached to the platinum ignition wire and placed in contact with the tablet. The HHV, based on the oven-dry weight of the sample, was calculated from Equation (5-1).

\[ HHV_d = HHV_w \times \frac{100}{(100 - MC)} \]  

(5-1)

where \( HHV_d \) is the higher heating value at constant volume of the dry (moisture-free) fuel, \( HHV_w \) is the higher heating value at constant volume of the fuel as received, and \( MC \) is moisture content of the sample (as percentage by mass on an air-dried basis).
5.2.4. Data analysis

Analyses of variance (ANOVA) using SAS/STAT software version 9.2 (SAS Institute Inc., NC, USA) of the SAS system were performed. Differences between individual levels were compared through Tukey's multiple comparison tests at a 5% probability level.

5.3. Results and discussion

5.3.1. Chemical composition

The chemical composition of sugar maple and yellow birch wood is presented in Table 5-2. Ash content is an important parameter to monitor since it can cause problems in the combustion and conversion processes (Obernberger and Thek 2010; Meincken and Tyhoda 2013). In the current study, the mean ash content of the sugar maple wood varied from 0.38% in the R vigor class to 0.73% in the S vigor class and 0.97% in the M vigor class \( (p < 0.0001) \). The mean ash content of yellow birch wood varied from 0.27% in the R vigor class to 0.26% in the S vigor class and 0.47% in the M vigor class \( (p = 0.0036) \). The M class in both sugar maple and yellow birch contained more ash than the other two higher quality classes (S and R). This finding is in agreement with the results obtained by Good et al. (1955), who also found that the amount of ash is higher in stained wood as compared to sound sugar maple sapwood. According to Shigo and Sharon (1970), the increase in mineral content is associated with the processes of discoloration and decay in the sugar maple. Tattar et al. (1971) studied changes in wood constituents associated with the discoloration and decay of sugar maple. These authors also found that the amounts of total, insoluble, and soluble ash content increased appreciably in the order of clear tissue, discolored tissue, and decayed tissue. Wood from the less vigorous trees has shown biodeterioration in the form of sapstain fungi and wood rotting fungi (Fig. 5-1). Therefore, another explanation for the elevated ash contents in M class wood could be that the original mineral components of wood were not efficiently removed during biodeterioration, which then became concentrated in the remaining material.

The ash contents of the extracted wood samples were also determined. The results shown in Tables 5-2 and 5-3 indicated that the ash content of all extracted wood samples was appreciably lower than for the non-extracted wood samples. A larger amount of ash could be removed from the wood samples of the least vigorous trees. Ash removals of 46% and 40% were obtained for the wood samples from the least vigorous sugar maple and yellow birch trees, respectively.
Table 5-2. Properties of sugar maple and yellow birch wood from different tree vigor class

<table>
<thead>
<tr>
<th>Properties</th>
<th>Sugar maple</th>
<th>Yellow birch</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Vigorous tree (R class)</td>
<td>Weakened tree (S class)</td>
</tr>
<tr>
<td>Ash content (%)</td>
<td>0.38(^c) (0.03)</td>
<td>0.73(^b) (0.07)</td>
</tr>
<tr>
<td>Ethanol-toluene extract (%)</td>
<td>3.17(^a) (0.22)</td>
<td>2.39(^b) (0.24)</td>
</tr>
<tr>
<td>Hot water extract (%)</td>
<td>2.27(^b) (0.11)</td>
<td>2.48(^b) (0.11)</td>
</tr>
<tr>
<td>Total extracts (%)</td>
<td>5.44(^b) (0.20)</td>
<td>4.88(^c) (0.31)</td>
</tr>
<tr>
<td>Klason lignin (%)</td>
<td>21.46(^c) (0.33)</td>
<td>22.89(^b) (0.76)</td>
</tr>
<tr>
<td>Acid soluble lignin (%)</td>
<td>3.59(^a) (0.49)</td>
<td>3.53(^a) (0.40)</td>
</tr>
<tr>
<td>Total lignin (%)</td>
<td>25.06(^c) (0.35)</td>
<td>26.40(^b) (0.52)</td>
</tr>
<tr>
<td>HHV (MJ kg(^{-1}))</td>
<td>19.59(^a) (0.08)</td>
<td>19.68(^a) (0.10)</td>
</tr>
</tbody>
</table>

Standard deviations are in parentheses. All values are based on the oven-dry weight of the original (non-extracted) sample. For each property of a tree species, the comparison is based on means. Values with different letters (a, b, c) indicate significant differences at the α = 0.05 level, for each species separately.

The ash contents of fuels should be as low as possible since ashes in biomasses have a number of negative impacts on the combustion process. Low ash content in wood fuels could benefit the end users in terms of
energy content and time for emptying ash (van Loo and Koppejen 2008). As reported by Alakangas et al. (2006), a raw material ash content of less than 0.7% is a requirement for the production of high quality solid biofuels (e.g., wood briquettes and wood pellets). From these fuel requirements, the primary limitation for the use of low quality trees as raw materials for the production of solid biofuels is the higher ash content levels as compared to healthy trees. However, the present results showed that extraction can reduce the ash content significantly. In addition, hot water extraction could increase the dimensional stability and compressibility of woody materials (Paredes et al. 2008; Pelaez-Samaniego et al. 2014). Therefore, the results suggested that hot water extraction should be applied to the wood from low vigor trees in order to produce a superior solid biofuel by the removal of undesirable inorganic substances with water prior to the wood's pelletization.

The extractive content is an important parameter that directly affects the heating value (Kataki and Konwer 2001). A high extractive content makes a lignocellulosic material more desirable as a fuel since the extractives increase the heating value of the material (Demirbas 2002; Telmo and Lousada 2011). In the current study, the mean extractives content was 5.44%, 4.88%, and 7.32% for wood samples from sugar maple trees of class R, S, and M (p < 0.0001), respectively. The mean extractive content was 3.35%, 3.47%, and 5.12% for wood samples from yellow birch trees of class R, S, and M (p = 0.0027), respectively. Wood from the M class trees for sugar maple and yellow birch contained more extractives than wood from the R class trees. This finding is in agreement with the studies of Pearce and Woodward (1986) and Pearce (1991), who reported higher amounts of extractives in the reaction zones of the wood around wounds than in healthy sapwood, and around infections caused by wood-decay fungi. Similarly, St-Pierre et al. (2013) found a higher amount of extractives in wood from low vigor yellow birch trees than from fungi infected trees. In addition, Barnett and Jeronimidis (2003) have shown that the occurrence of physical wounds, mechanical stresses, or attacks by biotic organisms induced the production of traumatic tissues in many trees, which involved the accumulation of protecting compounds in the affected areas. Most of these compounds were extractives and appeared in higher concentrations in the corresponding wood.

The ANOVA indicated that there were significant effects of tree vigor classes on the lignin content of sugar maple (p < 0.03) and yellow birch (p < 0.05) trees. The total and Klason lignin contents in the sugar maple ranged from 25.06% and 21.46%, respectively, in wood from the R class trees, to 26.40% and 22.89% in the S class, and to 26.83% and 23.53% in the M class trees. A similar pattern was observed for yellow birch, in which total and Klason lignin contents increased significantly from 23.20% and 18.60%, respectively, in the wood from the R class trees, to 24.23% and 20.18% in the S class trees, and to 25.70% and 21.51% in the M class trees. A possible explanation for the appreciable increase in lignin content is that the lignin actually increases during the
discoloration and decay processes due to its resistance to degradation by pathogens (Hawkin and Boudet 2003; Craft and Audia 1962). Jane et al. (1970) also mentioned that some wood rotting fungi, termed 'brown rot', break down the cellulose and the related materials and leave a brown residue that consists mostly of lignin. Therefore, the higher lignin contents obtained in this work could be due to the loss of cellulose and other polysaccharides, leaving behind the discolored lignin residual.

Prior research (Nguyen et al. 2015) has shown that wood from low vigor trees was more suitable for making wood pellets due to a lower friction in the pelletizer die and a higher mechanical strength of the pellets. It is known that wood extractives could act as lubricant in the pellets die and lignin could act as adhesive between the wood particles (Kaliyan and Morey 2009). Therefore, the higher extractive content and the higher lignin content found in wood from low vigor trees in the current study could explain the lower friction in the pellets die and the higher mechanical strength of pellets made from this material as reported by Nguyen et al. (2015).

5.3.2. Calorific value

The calorific values of sugar maple and yellow birch are presented in Tables 5-2 and 5-3. The results showed that variations in the HHV between the tree vigor classes and the wood species were very small, ranging from 19.57 to 19.82 MJ kg⁻¹ for the non-extracted wood samples and from 19.28 to 19.64 MJ kg⁻¹ for the extracted wood samples. The ANOVA indicated that there were no significant effects of tree vigor classes on the HHV of non-extracted wood samples from sugar maple (p = 0.55) and yellow birch trees (p = 0.07). Only a small increase in the HHV of extracted wood samples was recorded in the M class of yellow birch (p = 0.03). This finding could be explained by the variations in amounts of extractives, lignin and ash among the tree vigor classes. The higher amounts of extractive and lignin contents in wood from the low vigor trees had a positive impact on the calorific values even when considering the negative effect of their higher ash content on this property.

The results of the present study also indicated that the removal of extractives did not alter the HHV of sugar maple wood, while the removal of extractives showed a small reduction in the HHV of yellow birch wood. These findings are supported by the results reported by Rossi et al. (2013), in which the removal of extractives did not affect, led to a decrease, and caused an increase of the HHV in jatoba (*Hymenaea courbaril*), cedro arana (*Cedrelinga catenaeformis*), and ipe (*Tabebuia* spp.) woods, respectively. These authors suggested that the HHV of wood after extractives removal depends on their chemical composition. Also, this finding may be explained by the nature of the components and the relative quantities present in the fuel. Some extractives contain terpenoid hydrocarbons and lipids, while others contain phenolic compounds with higher oxygen
contents than the former compounds. Since combustion of an organic compound is related to its chemical composition, organic compounds containing only carbon and hydrogen produce more heat energy when burned than those containing also oxygen (Senelwa and Sims 1999; Moya and Tenorio 2013).

Table 5-3. Properties of extracted sugar maple and yellow birch wood

<table>
<thead>
<tr>
<th>Properties</th>
<th>Sugar maple</th>
<th></th>
<th>Yellow birch</th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Vigorous tree (R class)</td>
<td>Weakened tree (S class)</td>
<td>Dying tree (M class)</td>
<td>Vigorous tree (R class)</td>
</tr>
<tr>
<td>Ash (%)</td>
<td>0.29c</td>
<td>0.41b</td>
<td>0.52a</td>
<td>0.17b</td>
</tr>
<tr>
<td></td>
<td>(0.06)</td>
<td>(0.03)</td>
<td>(0.02)</td>
<td>(0.02)</td>
</tr>
<tr>
<td>Total lignin (%)</td>
<td>26.46b</td>
<td>27.76ab</td>
<td>28.93a</td>
<td>24.00b</td>
</tr>
<tr>
<td></td>
<td>(0.35)</td>
<td>(0.64)</td>
<td>(0.85)</td>
<td>(0.55)</td>
</tr>
<tr>
<td>HHV (MJ kg⁻¹)</td>
<td>19.57a</td>
<td>19.64a</td>
<td>19.56a</td>
<td>19.28b</td>
</tr>
<tr>
<td></td>
<td>(0.07)</td>
<td>(0.07)</td>
<td>(0.05)</td>
<td>(0.01)</td>
</tr>
<tr>
<td>Mass yield (%)</td>
<td>94.55b</td>
<td>95.12a</td>
<td>92.67c</td>
<td>96.65a</td>
</tr>
<tr>
<td></td>
<td>(0.20)</td>
<td>(0.31)</td>
<td>(0.03)</td>
<td>(0.11)</td>
</tr>
<tr>
<td>Energy yield (%)</td>
<td>94.45a</td>
<td>94.93a</td>
<td>92.39b</td>
<td>95.21a</td>
</tr>
<tr>
<td></td>
<td>(0.31)</td>
<td>(0.52)</td>
<td>(0.67)</td>
<td>(0.35)</td>
</tr>
</tbody>
</table>

Standard deviations are in parentheses. All values are based on the oven-dry weight of extracted sample. For each property of a tree species, the comparison is based on means. Values with different letters (a, b, c) indicate significant differences at the $\alpha = 0.05$ level. Note: * determined after wood sample were extracted sequentially with ethanol-toluene mixture and hot water in accordance with ASTM D1107-96 and ASTM D1110-84 standards, respectively; † obtained by dividing the weight of extracted solid fraction by weight of its original (non-extracted) sample; and ‡ obtained by multiplying the mass yield by HHV of the extracted sample and dividing by HHV of the original (non-extracted) sample.
One can observe from Table 5-3 that the extraction had a negligible effect upon the HHV of sugar maple and yellow birch, this effect being somewhat more significant ($p = 0.0003$) in the case of yellow birch wood which contains triterpene extractives rich in carbon, which are extracted with ethanol (St-Pierre et al. 2013). While HHV represents the absolute value of the specific energy of combustion per unit mass of a solid biofuel, energy yield represents the percentage of energy conserved in the solid fraction following the extraction process. It is an important parameter for the evaluation of the biofuel conversion process. Results indicated that energy yield following the extraction of sugar maple and yellow birch wood ranged between 92.39 and 95.31% (Table 5-3). It corresponds to about 5 to 8% of energy loss through the extraction. The energy yield calculated for wood sample of the least vigorous (M vigor class) trees is significantly lower than that of the most vigorous (R vigor class) trees for both sugar maple ($p = 0.0024$) and yellow birch ($p = 0.02$). This could be explained by the greater amounts of extractives removed from wood of the least vigorous trees through the extraction. Because of the high value by-products obtained through the extraction process, the higher amounts of extractives in the wood of less vigorous trees could compensate for the energy loss in the solid fraction through the extraction.

5.4. Conclusions

Tree vigor for sugar maple and yellow birch had significant effects on the chemical composition of the wood, which included extractives, ash, and lignin content. The less vigorous trees had higher wood extractives, ash, and lignin content than healthy trees.

There were no significant differences in the higher heating value of wood between tree vigor classes and between the non-extracted and the extracted wood samples. This can be considered as a value-added advantage because the greater amounts of high-value extractives in the less vigorous trees could be a supplementary benefit that compensates for the energy loss in the solid fraction through the extraction.

The higher extractive content and higher lignin content of wood from less vigorous trees found in this study support prior research which suggested that wood particles from less vigorous trees were more suitable than those from vigorous trees to make wood pellets.
Chapitre 6. Effect of process parameters and raw material characteristics on physical and mechanical properties of wood pellets made from sugar maple particles

by

Quy Nam Nguyen, Alain Cloutier, Alexis Achim, and Tatjana Stevanovic

in:

Biomass and Bioenergy 80: 338-349
Résumé

Le but de la présente étude était d'étudier l'influence des paramètres du procédé et les caractéristiques des matières premières sur les propriétés physiques et mécaniques de granulés de bois fabriqués à partir de particules de bois d’érable à sucre provenant d’arbres de différentes vigueurs. Les granules de bois ont été fabriqués dans un granulateur tout en contrôlant la température (75, 100 et 125 °C), la teneur en humidité (8,1, 11,2 et 17,2%), la force de compression (1500, 2000 et 2500 N) et la taille des particules (<0,25, 0,25 - 0,5 et 0,5 - 1,0 mm). La taille des particules a été le facteur le plus influant sur la friction dans le granulateur, suivie par la teneur en humidité, la force de compression et la température. La teneur en humidité était le facteur le plus important affectant la densité des granules, suivie par la température, la force de compression et la taille des particules. La température était le facteur le plus important pour la résistance mécanique en compression des granules, suivie par la force de compression, la taille des particules et la teneur en humidité. La friction dans le granulateur diminue avec l'augmentation de la taille des particules et la teneur en humidité du matériau, et augmente avec l'augmentation de la force de compression. La friction a diminué d'abord lorsque la température a augmenté de 75 °C à 100 °C, et ensuite a augmenté avec la température. La densité et la résistance mécanique en compression des granules a augmenté avec l'augmentation la température et la force de compression, et a diminué avec l'augmentation de la taille des particules et de la teneur en humidité. Le procédé de granulation doit être effectué à 100 °C pour minimiser la friction et une teneur en humidité de 11,2% pour maximiser la densité et la résistance mécanique en compression de granules. Les particules de bois provenant des arbres de faible vigueur étaient plus appropriées pour la fabrication de granules de bois en termes de friction dans le granulateur et de résistance mécanique en compression que celles des arbres vigoureux.
Abstract

The aim of the present study was to investigate the influence of process parameters and raw material characteristics on physical and mechanical properties of wood pellets made from particles of sugar maple wood from trees of different vigor. Pellets were made in a single pelletizer while controlling temperature (75, 100 and 125 °C), raw material moisture content (8.1, 11.2 and 17.2%), compression force (1500, 2000 and 2500 N) and particle size (<0.25, 0.25 - 0.5 and 0.5 - 1.0 mm). Particle size was the most important factor influencing friction in the die, followed by moisture content, compression force and temperature. Moisture content was the most important factor affecting pellet density, followed by temperature, compression force and raw material particle size. Temperature was the most important factor for pellet compression strength, followed by compression force, raw material particle size and moisture content. Friction in the die decreased with increasing particle size and moisture content of the material and increased with increasing compression force. It decreased initially with increasing temperature from 75 °C to 100 °C, and then increased with increasing temperature. Density and strength of pellets increased with temperature and compression force, decreased with increasing particle size, and decreased with increasing moisture content. Pelletizing should be performed at 100 °C to minimize friction and a moisture content of 11.2% to maximize density and compression strength of the pellets. Wood particles from sugar maple trees of low vigor were more suitable for making wood pellets in terms of friction in the pelletizer and compression strength than those from vigorous trees.
6.1. Introduction

The Northern hardwood forests of Québec, Canada, are composed largely of sugar maple (*Acer saccharum* Marsh.) and yellow birch (*Betula alleghaniensis* Britton). Several decades of subjecting these hardwood forests to selective harvesting where the most valuable high quality trees were removed, left the productive hardwood stands with low commercial value (Bédard and Majcen 2003). The restoration of more productive and ecologically desirable conditions through appropriate silvicultural practices requires the ability to gradually remove low vigor trees, with retention of vigorous growing stock of the desired species. However, the cost of harvesting low vigor trees rapidly becomes an obstacle because they often have low commercial value for traditional products such as lumber or pulp (Pothier et al. 2007). In such situations, wood energy markets can provide the economic incentive for removal of low value trees from forests.

Provided the existence of a suitable market, harvesting priority can be given to low vigorous trees in partial cuts as a means to restore depleted hardwood forests. Among tree classification systems that have been developed to assess tree vigor prior to harvesting, the MSCR classification system proposed by Boulet (2007) is used as tree marking guide for selection cuttings in the province of Québec, Canada. Based on pathological symptoms (e.g. presence of cankers and fungi), mechanical damage (e.g. cracks, leaning) and other visible features (e.g. improper branch shedding, crown openness), it defines more than 200 vigor codes, which are then grouped into four vigor classes: trees of reserve stock (class R) are free of any symptoms of disease or damage and are considered as healthy trees with highest probability of survival; growing trees (class C) have minor defects but are not biologically declining and are expected to survive until the next harvest without risk of imminent wood decay; low quality or defective trees (class S) are considered to be declining in terms of vigor, wood quality and volume increment, and are not expected to survive until the next harvest; and moribund trees (class M) show signs of either lethal pathological infection or severe damage with high risk of trunk breakage. Moribund trees are biologically declining and are assumed to have a high probability of mortality before the next harvest entry. According to Hartmann et al. (2008), typical fungal infections for sugar maple involve the northern tooth fungus (*Climacodon septentrionalis* P. Karst.), which causes severe crown dieback. Sugar maple trees showing signs of this fungal infection are classified as moribund. The unused available annual forest biomass in the Province of Québec, Canada, is estimated at 6.4 million anhydrous metric tons including 3.5 million tons in the form of tree trunks and 2.9 million tons in the form of crowns and branches. More than half (55%) of this volume is coming from low vigor trees not suitable for lumber production (MRNF). The availability of these unused low quality trees could thus represent a significant source of feedstock for the production of biofuel.
In recent years, a clear interest arose for the use of forest biomass as biofuels. This was motivated not only by its low environmental impacts and carbon emissions, but also because meeting the global increase in energy consumption may not be achievable without the contribution of biofuels (Hunt 2009; Masia et al. 2010). It has been argued recently that a better use of biomass would be to burn it in the form of solid fuel for the generation of electricity to power electric vehicles (Campbell et al. 2009). Particularly, modern solid fuel combustion technology has been developed with much cleaner and efficient combustion than in the past. Solid biomass fuel, e.g. in the form of pellets, could thus become a more important energy commodity (SFU). The CEN/TS 14588 standard (CEN/TS 14588 (2003)) defines wood pellets as densified biofuel made from pulverized woody biomass with or without additives, usually with a cylindrical form in diameter of 6 to 10 mm and a random length of typically 5 to 40 mm with broken ends. The homogeneity in size, shape and quality of pellets makes them well-suited for fully automated feed systems (Lai 2012). Pellets could be used as fuel directly in several applications from residential heating stoves to central heating boilers (Obernberger and Thek 2010). They can even be used as a fuel source in large-scale power plants (Döring 2013), or as part of the process to produce liquid fuels (Xie and Gathergood 2013). Wood pellets are standardized and traded at both national and international levels. These features combined with their advantages such as environmental benefits, high energy content due to their high density and low moisture content, and relative convenience of transportation and storage explain the rapid growth of the global wood pellets market, both in terms of production and consumption.

In Canada, the wood pellets industry is well developed. It produced more than 1.5 million tons in 2011 and continues to be one of the world leaders in wood pellet production. In the beginning of 2012, Canada had 39 operational wood pellets plants with a capacity of 3.2 million tons per year (Bradley 2012). The production capacity has grown significantly in recent years. However, wood pellets producers are facing a shortage of the traditional raw material supply (i.e. sawdust and wood chips) due to the recent downturn in the commodity wood products industry. This has increased the interest in finding new raw material sources from forests in sufficient volumes and in a sustainable manner. With the increase in fossil energy prices and environmentally efficient technologies such as those used by St-Pierre et al. (2013) and Ngueho Yemele et al. (2008) for the extraction of wood and bark, the harvesting of low quality sugar maple and/or yellow birch trees from high-graded hardwood forests as new sources of raw material for biofuel pellets production is likely to become a viable option.

Hardwood particles are difficult to use for pellets production because the frictional forces in the compression channels of the die are high compare to softwoods and other biomass. Consequently, the pellet mill is prone to blockage when hardwoods are used (Holm et al. 2006; Bradfield and Levi 1984). According to Obernberger and Thek (2010), the process of densification in the pellet mill depends on the friction between the compression channel and the raw material, which is largely determined by the moisture content of the raw material. This is
why the optimal moisture content has to be determined according to the pelletizing technology and the raw material used. Moisture contents in the range of 5 to 10% are usually optimal for woody raw material. Moisture contents of 6 to 10% for beech wood (Stelte et al. 2011), of 5% for olive tree wood (Carone et al. 2011), and of 6 to 8% for pine wood (Nielsen et al. 2009b) have been reported as appropriate. Tumuluru (2014) reported that it is possible to produce corn stove pellets from feedstock at high moisture content in the range of 28 to 38% (w.b.) by varying other process parameters. Gunnerman (1977) found that the moisture content of organic fibrous material in pelletization can be within the range of 16 to 28% but the best results were obtained for moisture content between 20% and 24%. Both Tulumuru (2014) and Gunnerman (1977) suggested that high moisture content pellets can be dried to safe storage moisture content by using cost and energy efficient drying systems.

Another problem occurring during pelletization is overheating. Heat is generated during pelletization due to friction between the ground material and the walls of the pellet mill. Serrano et al. (2011) studied the heat distribution in a pellet press and found that under operation at stable conditions, the temperature of the die is about 90 °C, while the temperature of the biomass is about 70 °C. Nielsen et al. (2009b) reported that the pelletizing process generates heat that maintains the temperature of the operating die at 110 - 130 °C. The results conducted by Holm et al. (2006) showed that the pellet mill was blocked after a few minutes of pelletizing a mixture of 40% (by dry weight, wt) pine shavings and 60% (wt) beech dust. The temperature of the pellets in the matrix was recorded at approximately 120 °C in this case. However, a mixture of 80% (wt) pine shavings and 20% (wt) beech dust could be pelletized without problems with pellets temperature of approximately 105 °C while the pellets temperature of pure pine shavings was approximately 90 °C.

A single pelletizer cannot simulate all aspects of a large-scale pelletizing process; for example, the flow of feedstock into the openings of the press channels (Mišljenovic et al. 2014; Stelte et al. 2013). However, using a large-scale pellet mill to study the effects of various parameters on pelletizing characteristics and pellets quality is time and material consuming, and some process parameters such as temperature and friction in the channels of the die cannot be easily controlled because of their interaction effects (Shang et al. 2014). A single pelletizer is therefore widely used method for testing the pelletizing characteristics of a new type of biomass. The effect of die temperature, pressure, raw material moisture content, and other parameters can be quickly tested and screened with the single pelletizer before scaling up to an industrial pellet mill.

The objectives of this study were (i) to investigate the effect of compacting temperature, moisture content of the particles, compression force, and particles size on the physical and mechanical properties of pellets pressed from raw material obtained from low and high vigor sugar maple trees; (ii) to determine which combinations of these independent variables provide the best pelletizing process in terms of friction force and pellets quality.
6.2. Experimental

6.2.1 Raw material

Pelletizing experiments were conducted only from material obtained from the sugar maple trees sampled in this study. The trees were harvested in July 2010 from two mixed hardwood stands composed mainly of sugar maple and yellow birch located in the vicinity of Mont-Laurier, Québec, Canada (46°39'40"N, 75°36'30"W and 46°39'05"N, 75°36'25"W). Sugar maple trees in this area were classified according to the tree vigor (MSCR) classification system proposed by Boulet (2007). Based on external traits, each tree of the stand was classified into one out of the four vigor classes as follows: class M - moribund tree, class S - tree of surviving stock (low quality or defective tree), class C - tree of conservation stock (growing tree), or class R - tree of reserve stock. After classifying each tree according to its corresponding tree vigor class, a total of 9 sugar maple trees (3 trees classified as R, 3 trees classified as S, and 3 trees classified as M and suffering from a major fungal attack) were felled and sampled. The sample trees classified as S and M had part of their crown dead and part of the bole visibly hollow. Two 1.2 m long logs were cut from each sample tree. The first log was cut at a distance of 1.3 m from the root collar, and the second at a distance of one-third of the trunk length down from the crown base. The 18 sample logs were wrapped in green condition and transported to the Renewable Materials Research Center at Université Laval, Québec, Canada for analysis.

Various types of equipment were used to convert the sample logs into wood particles with suitable size for pellets production. A band saw was used to convert the logs into sticks. Then, a chipper was used to convert the sticks into wood chips, which were air-dried in layers of about 50 mm thick to a moisture content of about 15% (dry basis). After air seasoning, the wood chips were fragmented with a hammer mill and finally processed into a Pallmann ring refiner (Type PSKM 8, Ludwig Pallmann K.G., Zweibrücken, Germany) for final size reduction. The refiner screen sizes used in the present study were 4.0 mm, 2.5 mm and 1.5 mm.

Coning and quartering method was used to produce laboratory samples from a combined sample in accordance with CEN/TS 14780 (2006). The fraction size distribution of the ground particles samples was analyzed from laboratory samples. For each test, a sample of 100 g was placed in a stack of sieves arranged from the largest to the smallest opening. A Ro-Tap sieve shaker (Model RX-29, The W.S Tyler Company Ltd., Ontario, Canada) was used to determine the geometric mean particle size using standard sieve series number 6, 7, 14, 18, 35 and 60 (sieve opening size: 3.35, 2.80, 1.40, 1.00, 0.50 and 0.25 mm, respectively). The duration of sieving was
15 minutes. After sieving, the mass retained on each sieve was weighed. The sieve test was replicated three times for each ground sample. The results of particle size distribution analysis are presented in Table 6-1.

The results presented in Table 6-1 show that most ground material obtained with the 1.5 mm refiner screen size was retained on the sieves with openings of 0.25, 0.5, and 1.0 mm. The fine grind fraction for trees of class M was significantly higher than that of trees of the more vigorous classes S and R at any specific refiner screen size, but especially for 1.5 mm. This could be explained by the reduction in resistance to fragmentation of wood from the less vigorous sugar maple trees.

The sieved ground material was placed inside conditioning chambers. Each of the sieved samples was divided into 3 parts. Each part was placed inside conditioning chambers under the following air temperature and relative humidity conditions: 20 °C and 40%, 20 °C and 60%, and 20 °C and 80%, corresponding to equilibrium moisture contents in the wood grinds of 8.1, 11.2, and 17.2% on a dry basis, respectively.

Table 6-1. Particle size distribution by tree vigor classes.

<table>
<thead>
<tr>
<th>Sizes of refiner screen</th>
<th>Tree vigor classes</th>
<th>Fraction (%)</th>
<th>Sieve opening size (mm)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td>&lt; 0.25</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Mean</td>
<td>Std</td>
</tr>
<tr>
<td>1.5 mm R</td>
<td>22.9</td>
<td>0.88</td>
<td>37.4</td>
</tr>
<tr>
<td>S</td>
<td>29.0</td>
<td>0.26</td>
<td>37.0</td>
</tr>
<tr>
<td>M</td>
<td>31.9</td>
<td>0.74</td>
<td>38.0</td>
</tr>
<tr>
<td>2.5 mm R</td>
<td>10.7</td>
<td>0.31</td>
<td>21.7</td>
</tr>
<tr>
<td>S</td>
<td>11.9</td>
<td>0.63</td>
<td>22.5</td>
</tr>
<tr>
<td>M</td>
<td>13.1</td>
<td>1.05</td>
<td>25.3</td>
</tr>
<tr>
<td>4.0 mm R</td>
<td>5.8</td>
<td>0.67</td>
<td>15.1</td>
</tr>
<tr>
<td>S</td>
<td>6.3</td>
<td>0.27</td>
<td>17.2</td>
</tr>
<tr>
<td>M</td>
<td>7.3</td>
<td>0.68</td>
<td>17.2</td>
</tr>
</tbody>
</table>

6.2.2 Experimental design

A full split-plot factorial design with four independent variables was used to study the influence of temperature, moisture content, compressive force, and particle size on the physical and mechanical properties of pellets. The factor temperature is denoted $X_1$ (for whole-plot factor), and the other three factors (sub-plot factors): moisture
content, compressive force and particle size are denoted $X_2$, $X_3$, and $X_4$ respectively. The experimental plan is a three-level factorial design in 81 independent experiments (or runs). Each run was replicated three times, and three dependent variables: friction, density and strength were measured. A total of 729 pellets were produced from particles of the most vigorous (R class) trees and 729 pellets were produced from particles of the least vigorous (M class) trees (Table 6-3). The three levels of each independent variable are coded by 0, 1, and 2 numbers for the low, middle, and high levels, respectively, as shown in Table 6-2.

6.2.3 Pelletization

Pellets were individually produced in a single pelletizer under controlled conditions as indicated in Table 6-2. The single pelletizer (Fig. 6-1) used in the present study is based on the designs proposed by Mani et al. (2006) and Carone et al. (2011), with a minor modification by setting up a control program to assist compression and automatic ejection of the pellet while protecting the piston from getting jammed and to monitor the applied load and displacement during the compression process. This addition enabled a better analysis of the behavior of frictional forces at the wall of the press channel (Fig. 6-1). Teflon rods were installed above and below the pellet to prevent the piston from getting jammed in the cylinder. Fig. 6-2 shows the load application program.

Table 6-2. Experimental ranges and levels of independent variables.

<table>
<thead>
<tr>
<th>Independent variables</th>
<th>Code</th>
<th>Coded variable levels</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Code</td>
<td>0</td>
</tr>
<tr>
<td>Temperature (°C)</td>
<td>$X_1$</td>
<td>75</td>
</tr>
<tr>
<td>Moisture content (% d.b.)</td>
<td>$X_2$</td>
<td>8.1</td>
</tr>
<tr>
<td>Compressive force (N)</td>
<td>$X_3$</td>
<td>1500</td>
</tr>
<tr>
<td>Particle size (mm)</td>
<td>$X_4$</td>
<td>&lt; 0.25</td>
</tr>
</tbody>
</table>
The cylinder of 6.25 mm inner diameter and 150 mm long was first heated to the desired temperature. After equilibration, a 0.5 g sample of ground wood (Fig. 6-3) was put into the cylinder and compressed by the piston at a speed of 12 mm min\(^{-1}\) to the desired pressure. After a retention time of 10 seconds at full pressure, the pellet was pressed out of the cylinder by removing the bottom plate. The force-displacement data were recorded for the whole cycle of compaction and ejection of pellets. Data for adhesive friction and sliding friction were measured as illustrated in Fig. 6-2. Each pellet produced was stored in a sealed vial (Fig. 6-4) for further measurements.
Fig. 6-1. Set up and schematic diagram of the single pelletizer: (A) stainless steel plunger of 6.0 mm in diameter; (B) cylindrical die with 6.25 mm inner diameter and 150 mm long; (C) heating element; (D) material used or wood pellet; (E) top and (F) bottom teflon compression assist rod of 6.25 mm in diameter and 10 mm long; and (G) removable backstop of 20 mm wide, 45 mm high and 120 mm long without handle.

Fig. 6-2. Schematics of the compression force and ejection force as a function of piston movement during a pelletizing cycle. Dark yellow (top), grey (middle) and pink (bottom) curves show the pelletizing cycles applied at compression forces of 2500 N, 2000 N and 1500 N, respectively.
Fig. 6-3. Materials (left side), pelletized products (right side). The illustrated pellets were produced from wood particles of 11.2 % (d.b.) moisture content at three preset temperatures under a compression force of 2000 N.

6.2.4 Determination of physical and mechanical properties

6.2.4.1 Moisture content
Samples of preconditioned and conditioned particles were tested. Moisture was determined in accordance with CEN/TS 14774 standard (2004). The oven was set at 105 ± 2 °C and samples were dried for 24 hours.

6.2.4.2 Pellet density
The density of a single pellet was calculated by dividing its mass by its volume. Mass was measured by using an analytical balance with an accuracy of ±0.0001 g. Volume was calculated from the diameter and length of the pellet, assuming a perfectly cylindrical shape. The diameter and length of individual pellets were measured using an electronic caliper with an accuracy of ±0.01 mm. Pellet density was determined immediately following the ejection of the pellets. The values were reported as an average of 3 measurements.

6.2.4.3 Pellet strength
The compression strength of pellets was measured following the method proposed by Nielsen et al. (2009b) and Bergström et al. (2010). A universal MTS testing machine was used to measure the peak load in diametrical compression of the pellets. The sample was compressed radially between two parallel horizontal plates. The
Compression speed was set to 6 mm min⁻¹. The ratio between the peak load in compression and the length of the pellet was defined as the compression strength. The pellets were kept in vials and tested for compression strength about two hours after production. Compression strength values were reported as an average of 3 measurements.

Fig. 6-4. Storage and dimensional measurements of pellets (left side) and representation of the diametrical compression of a wood pellet (right side), including the typical load-deformation curve.

6.2.5 Statistical analyses

A second order quadratic model was used to describe the effect of independent variables in terms of linear and quadratic effects, and their interactions. The proposed model for the responses is shown in Equation 1.

\[ y = b_o + \sum_{i=1}^{n} b_i x_i + \sum_{i=1}^{n} b_{ii} x_i^2 + \sum_{i=1}^{n} \sum_{j=1}^{n} b_{ij} x_i x_j + \varepsilon \]  (6-1)

where \( y \) is the dependent variable, \( x_i \) and \( x_j \) are the coded independent variables, \( b_o \) is the intercept coefficient, \( b_i, b_{ii}, b_{ij} \) are the linear, quadratic, and interaction terms, respectively, \( n \) is the number of independent variables and \( \varepsilon \) is the random error.

The experimental data was analyzed using the proc glm procedure of the SAS/STAT software, version 9.2 of the SAS system for Windows.
6.3. Results and discussion

Tables 6-4 and 6-5 summarize the results obtained from the ANOVA. The results show that linear terms of all main effects (temperature, moisture content, compression force and raw material particle size) were significant at the $P < 0.001$ level for all three dependent variables (friction, pellet density and pellet strength) indicating their strong effect on these properties. The interactions were also all significant at the $P < 0.02$ level or less.

The main effects and interaction plots are displayed in Figures 6-5, 6-6, 6-7 and 6-8. By visualizing the main effect plots and comparing the F-value obtained for each main effect, the factor raw material particle size ($X_4$) was the most important for friction followed by factors moisture content ($X_2$), compression force ($X_3$) and temperature ($X_1$). Moisture content was the most important factor for pellet density followed by temperature, compression force and raw material particle size. Temperature was the most important factor for pellet compression strength, followed by raw material particle size, moisture content, and compression force. Factor temperature appeared to have a greater effect on pellet strength, with a steeper slope.

By examining the main effects plot and interaction plots in Figures 6-5, 6-6, 6-7 and 6-8 that correspond to the significant effects identified in the ANOVA (Table 6-4), the most obvious findings are that level 1 of the moisture content factor should be chosen to maximize pellet density and pellet strength, while level 1 of the temperature factor should be chosen to minimize friction force.
Table 6-4. Analysis of variance (ANOVA) for friction, density, and compression strength of pellets pressed from particles of the most (R) and the least (M) vigorous trees.

<table>
<thead>
<tr>
<th>Source of variation</th>
<th>Degrees of freedom</th>
<th>Sum of squares</th>
<th>Mean squares</th>
<th>F value</th>
<th>Pr &gt; F</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>R</td>
<td>M</td>
<td>R</td>
<td>M</td>
<td>R</td>
</tr>
<tr>
<td>Friction</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Model</td>
<td>80</td>
<td>80</td>
<td>6746.9</td>
<td>4713.1</td>
<td>84.3</td>
</tr>
<tr>
<td>Error</td>
<td>162</td>
<td>162</td>
<td>115.5</td>
<td>114.0</td>
<td>0.7</td>
</tr>
<tr>
<td>Corrected total</td>
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<td>242</td>
<td>6682.5</td>
<td>4267.2</td>
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<td></td>
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<td></td>
<td></td>
</tr>
<tr>
<td>Density</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Model</td>
<td>80</td>
<td>80</td>
<td>1150388</td>
<td>1020863</td>
<td>14379</td>
</tr>
<tr>
<td>Error</td>
<td>162</td>
<td>162</td>
<td>15768</td>
<td>11492</td>
<td>97</td>
</tr>
<tr>
<td>Corrected total</td>
<td>242</td>
<td>242</td>
<td>1166156</td>
<td>1032356</td>
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</tr>
<tr>
<td></td>
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<tr>
<td>Model</td>
<td>80</td>
<td>80</td>
<td>48724.0</td>
<td>51674.0</td>
<td>609.0</td>
</tr>
<tr>
<td>Error</td>
<td>162</td>
<td>162</td>
<td>12619</td>
<td>544.4</td>
<td>7.7</td>
</tr>
<tr>
<td>Corrected total</td>
<td>242</td>
<td>242</td>
<td>49895.9</td>
<td>52218.4</td>
<td></td>
</tr>
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<td></td>
<td></td>
</tr>
</tbody>
</table>
Table 6-5. Summary of the regression modelling statistics (coded variables).

<table>
<thead>
<tr>
<th>Components</th>
<th>Parameter</th>
<th>Pellets pressed from grinds of the most vigorous (class R) trees</th>
<th>Pellets pressed from grinds of the least vigorous (class M) trees</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Friction (R² = 0.87)</td>
<td>Density (R² = 0.90)</td>
<td>Strength (R² = 0.90)</td>
</tr>
<tr>
<td>Intercept</td>
<td>b₀</td>
<td>29.64***</td>
<td>958***</td>
</tr>
<tr>
<td>Temperature (X₁)</td>
<td>b₁</td>
<td>-7.79***</td>
<td>40***</td>
</tr>
<tr>
<td>Moisture content (X₂)</td>
<td>b₂</td>
<td>-3.65***</td>
<td>93***</td>
</tr>
<tr>
<td>Compression force (X₃)</td>
<td>b₃</td>
<td>3.47***</td>
<td>70***</td>
</tr>
<tr>
<td>Particle size (X₄)</td>
<td>b₄</td>
<td>-7.54***</td>
<td>-36***</td>
</tr>
<tr>
<td>X₁²</td>
<td>b₁₁</td>
<td>2.03***</td>
<td>10”</td>
</tr>
<tr>
<td>X₂²</td>
<td>b₂₂</td>
<td>-1.29***</td>
<td>-53***</td>
</tr>
<tr>
<td>X₃²</td>
<td>b₃₃</td>
<td>ns</td>
<td>-7”</td>
</tr>
<tr>
<td>X₄²</td>
<td>b₄₄</td>
<td>0.73”</td>
<td>ns</td>
</tr>
<tr>
<td>X₁X₂</td>
<td>b₁₂</td>
<td>1.51***</td>
<td>-16***</td>
</tr>
<tr>
<td>X₁X₃</td>
<td>b₁₃</td>
<td>ns</td>
<td>-5”</td>
</tr>
<tr>
<td>X₁X₄</td>
<td>b₁₄</td>
<td>1.38***</td>
<td>5”</td>
</tr>
<tr>
<td>X₂X₃</td>
<td>b₂₃</td>
<td>-0.37”</td>
<td>-10”</td>
</tr>
<tr>
<td>X₂X₄</td>
<td>b₂₄</td>
<td>1.78***</td>
<td>ns</td>
</tr>
<tr>
<td>X₃X₄</td>
<td>b₃₄</td>
<td>-0.77***</td>
<td>ns</td>
</tr>
</tbody>
</table>

Note: ns = non-significant, *** significant at P < 0.0001; ** significant at P < 0.01; * significant at P < 0.05.

6.3.1. Friction

The friction ranged from 11.7 to 41.4 N mm⁻¹ with the overall mean of 19.6 N mm⁻¹ and from 10.8 to 34.1 N mm⁻¹ with the overall mean of 17.9 N mm⁻¹ for pellets pressed from particles of the most and the least vigorous trees, respectively. Temperature had a greater effect on friction at low moisture content than at high moisture content. At the highest moisture content used in this study (17.2%), the effect of temperature on friction became negligible. At moisture contents of 8.1% and 11.2%, a minimum in friction was found at 100 °C. This is supported by results obtained by Nielsen et al. (2009b), in which a minimum in friction was reached at approximately 105 °C for both a hardwood species (European beech) and a softwood species (Scots pine). These findings could explain partially why a mixture of 80% (wt) pine shavings and 20% (wt) beech dust could be pelletized without
problems at a temperature of approximately 105 °C in the die, while a mixture of 40% (wt) pine shavings and 60% (wt) beech dust could not be pelletized at a temperature of approximately 120 °C in the die because of blockage as reported by Holm et al. (2006).

As illustrated in Figure 5-5a, the friction force in the die of the single pelletizer decreased with increasing particle size and moisture content of the material and increased with increasing compression force. The interaction effect shows that the effect of temperature was more important at lower moisture contents. Generally, friction decreased when temperature increased from 75 °C to 100 °C, and then tends to increase. The higher friction force in the die was observed for pellets pressed from wood particles of the most vigorous trees in comparison to those of the least vigorous trees (Figure 6-5a). A lower friction during pelletizing with a single pelletizer implies a lower energy consumption in pelletizing with a large-scale pellet mill. It is significant to reduce production costs and risks of die blockage.

A regression analysis based on the friction data was performed. Quadratic models in terms of coded variables with a coefficient of determination $R^2 = 0.87$ for pellets pressed from particles of vigorous trees ($F_R$) and $R^2 = 0.86$ for those pressed from particles of the low vigor trees ($F_M$) are given in Equations 2 and 3, respectively.

\[
F_R = 29.6 - 7.79X_1 - 3.65X_2 + 3.47X_3 - 7.54X_4 + 2.03X_1^2 - 1.29X_2^2 + 0.73X_4^2 \\
\quad + 1.51X_1X_2 + 1.38X_1X_4 - 0.37X_2X_3 + 1.78X_2X_4 - 0.77X_3X_4
\]  \quad (6-2)

\[
F_M = 23.8 - 4.86X_1 + 3.58X_3 - 5.96X_4 + 1.2X_1^2 - 1.61X_2^2 + 0.72X_4^2 \\
\quad + 1.01X_1X_2 + 1.03X_1X_4 - 0.41X_2X_3 + 0.89X_2X_4 - 0.53X_3X_4
\]  \quad (6-3)

where $X_1$ is coded independent variable for temperature, $X_2$ is coded independent variable for moisture content, $X_3$ is coded independent variable for compression force, and $X_4$ is coded independent variable for particle size.

### 6.3.2. Properties of pellets

#### 6.3.2.1. Moisture content

The experimental results presented in Table 6-6 show that the moisture content of the pellets was dependent on the moisture content of the raw material used and the preset temperature of the die. In general, the moisture content of the pellets produced was within the moisture content range of commercial pellets (Table 6-6), except for pellets pressed from high moisture content material of 17.2% at temperature levels of 75 and 100 °C. In these conditions, the pellet moisture contents were 14.3% and 11.9%, respectively.
Table 6-6. Properties of experimental pellets and commercial pellets

<table>
<thead>
<tr>
<th>Process parameters</th>
<th>Properties of pellets</th>
<th>Type of material</th>
</tr>
</thead>
<tbody>
<tr>
<td>Temp (°C)</td>
<td>MC (% d.b.)</td>
<td>UD (kg m⁻³)</td>
</tr>
<tr>
<td>75</td>
<td>8.1</td>
<td>977</td>
</tr>
<tr>
<td></td>
<td>11.2</td>
<td>1029</td>
</tr>
<tr>
<td></td>
<td>17.2</td>
<td>952</td>
</tr>
<tr>
<td>100</td>
<td>8.1</td>
<td>1054</td>
</tr>
<tr>
<td></td>
<td>11.2</td>
<td>1059</td>
</tr>
<tr>
<td></td>
<td>17.2</td>
<td>947</td>
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<tr>
<td>125</td>
<td>8.1</td>
<td>1107</td>
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<tr>
<td></td>
<td>11.2</td>
<td>1099</td>
</tr>
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<tr>
<td>75</td>
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<td>1008</td>
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<td>1035</td>
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<td>17.2</td>
<td>966</td>
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<td>100</td>
<td>8.1</td>
<td>1065</td>
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<td>11.2</td>
<td>1062</td>
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<td>17.2</td>
<td>964</td>
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<tr>
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<td>8.1</td>
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<tr>
<td></td>
<td>11.2</td>
<td>1101</td>
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<tr>
<td></td>
<td>17.2</td>
<td>1024</td>
</tr>
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</table>

Commercial pellets

<p>| | |</p>
<table>
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<tbody>
<tr>
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<td>1267</td>
<td>-</td>
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<td>1162</td>
<td>-</td>
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</tbody>
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Industrial scale process

<p>| | |</p>
<table>
<thead>
<tr>
<th></th>
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</thead>
<tbody>
<tr>
<td>Fine partice (&lt; 1.0 mm)</td>
<td>1263</td>
</tr>
<tr>
<td>Coarse particle (2.0 - 4.0 mm)</td>
<td>1276</td>
</tr>
</tbody>
</table>

Temp. = temperature, UD = unit density, CS = compression strength, MC = moisture content.
6.3.2.2. Unit density

Density ranged from 879 to 1188 kg m$^{-3}$ with an overall mean of 1026 kg m$^{-3}$ and from 887 to 1193 kg m$^{-3}$ with an overall mean of 1038 kg m$^{-3}$ for pellets pressed from particles of both the most and the least vigorous sugar maple trees, respectively. As illustrated in Figure 6-5b, density increased with increasing temperature and compression force and decreased with increasing particle size. The interaction effect shows that the effect of temperature was more important at lower moisture content. The data indicate that at low temperature (75 °C), there was a moisture content threshold point of about 11.2% beyond which further increases led to a lower pellet density (Figures 6-5b and 6-7).

Quadratic models were developed with a coefficient of determination $R^2 = 0.90$ for density of pellets pressed from particles of the most vigorous trees ($D_{R}$) and $R^2 = 0.91$ for those pressed from particles of the least vigorous trees ($D_{M}$) are given in Equations 4 and 5, respectively.

$$D_R = 958 + 40X_1 + 93X_2 + 70X_3 - 36X_4 + 10X_1^2 - 53X_2^2 - 7X_3^2 - 16X_1X_2 - 5X_1X_3 + 5X_2X_3 - 10X_2X_3$$  \hspace{1cm} (6-4)

$$D_M = 992 + 24X_1 + 67X_2 + 67X_3 - 42X_4 + 11X_1^2 - 42X_2^2 - 8X_3^2 + 6X_1X_2 - 12X_1X_3 + 6X_2X_4 - 10X_2X_3$$  \hspace{1cm} (6-5)

where $X_1$ is coded independent variable for temperature, $X_2$ is coded independent variable for moisture content, $X_3$ is coded independent variable for compression force, and $X_4$ is coded independent variable for particle size.

6.3.2.3. Compression strength

Compression strength ranged from 19.8 to 91.7 N mm$^{-1}$ with an overall mean of 43.0 N mm$^{-1}$ for pellets pressed from particles of the most vigorous trees and from 22.8 to 93.6 N mm$^{-1}$ with an overall mean of 46.6 N mm$^{-1}$ for pellets pressed from particles of the least vigorous trees. The plotted results reveal that the strength of pellets increased along with temperature increases for most treatments, except for the moisture content of 17.2%, for which no significant differences in pellet compression strength were found between 75 °C and 100 °C (Figures 6-5c and 6-8).

In general, the compression strength of pellets pressed from particles of the least vigorous trees was higher than that of pellets pressed from particles of the most vigorous trees (Figure 6-8). The difference in hot water extractives and lignin content between particles from the most and the least vigorous sugar maple trees seems to have an impact on pellet compression strength. According to Stokland et al. (2012), the chemical composition of wood may change when a tree is weakened by living organisms, especially fungi. Increases in the amount of lignin have been reported due to its resistance to degradation by pathogens (Hawkin and Boudet 2003), while
Jane et al. (1970) mentioned that some wood rotting fungi, more specifically brown rots, break down the cellulose and related materials and leave a brown residue consisting largely of lignin. Increases in the amount of extractives were determined in wood from low vigor sugar maple trees, typically that from the fungus infected trees (Bradley 2012). The increase in amount of extractives around infections caused by wood decaying fungi in silver maple (Acer saccharinum) was also reported by Pearce and Woodwards (1986).

Natural binders such as lignin and proteins in biomass have been softened during pelletization, and form a strong bonding (solid bridge formation) between particles. Thus, lower amount of lignin in the biomass results in poor adhesion and low compression strength of pellets, both at low and high levels of pelletizing temperature (Kylian and Morey 2010; Stelle et al. 2010). Hot water extractives determined from ASTM D 1110-84 standard usually include tannins, gums, simple sugars, various coloring matter and starches. According to Wood (1987), starch acts as a binder. In the presence of heat and moisture, starch plasticizes, which has been shown to increase pellet compressive strength. In addition, the lower hardness due to fungal attack of a certain amount of particles from the less vigorous sugar maple trees could contribute to pellet compressive strength.

The developed quadratic models for compression strength with a coefficient of determination $R^2 = 0.90$ for pellets pressed from particles of the most vigorous trees ($S_R$) and $R^2 = 0.94$ for those pressed from particles of the least vigorous trees ($S_M$) are given in Equations 6 and 7, respectively.

$$S_R = 25.8 + 12.7X_1 + 17.4X_2 + 10X_3 - 8.1X_4 + 2.2X_1^2 - 8.2X_2^2 + 3.3X_4^2$$
$$- 3.5X_1X_2 - 2.7X_1X_4 - 1.9X_2X_3 - 1.3X_3X_4$$

(6-6)

$$S_M = 28.3 + 15.6X_1 + 13.8X_2 + 10.4X_3 - 7.2X_4 + 2.2X_1^2 - 5.8X_2^2 - 0.9X_3^2 + 2.6X_4^2$$
$$- 4.9X_1X_2 + 0.9X_1X_3 - 3.6X_2X_4 - 1.8X_2X_3 - 1.3X_3X_4$$

(6-7)

where $X_1$ is coded independent variable for temperature, $X_2$ is coded independent variable for moisture content, $X_3$ is coded independent variable for compression force, and $X_4$ is coded independent variable for particle size.
Fig. 6-5. Main effects plots of friction (a), density (b), and compression strength (c) for pellets pressed from particles of the most vigorous trees (class R) and the least vigorous trees (class M). Temperature (X1) at three levels (0, 1 and 2) of 75 °C, 100 °C and 125 °C. Moisture content (X2) at three levels (0, 1 and 2) of 8.1, 11.2 and 17.2%. Compression force (X3) at three levels (0, 1 and 2) of 1500, 2000 and 2500 N. Particle size (X4) at three levels/ranges (0, 1 and 2) of smaller than 0.25 mm, within 0.25 to 0.5 mm and within 0.5 to 1.0 mm.
Fig. 6-6. Interaction plots of friction for pellets pressed from particles of the most vigorous trees (R₁ - R₆), and from the least vigorous trees (M₁ - M₆).
Fig. 6-7. Interaction plots of density for pellets pressed from particles of the most vigorous trees (R - R₆), and from particles of the least vigorous trees (M - M₆).
Fig. 6-8. Interaction plots of compression strength for pellets pressed from particles of the most vigorous trees (R1 - R6), and from particles of the least vigorous trees (M1 - M6).
6.4. Conclusions

Wood particles produced from sample logs of vigorous and non-vigorous sugar maple trees were pressed into pellets. Compared to particles from vigorous trees, the use of wood particles from the non-vigorous trees reduced friction, and increased the compression strength of the pellets produced. The pelletizing process should be performed at a temperature of about 100 °C to minimize friction and at a moisture content of about 11.2% to maximize density and compression strength of the pellets produced. There are two reasons for using roundwood from low quality hardwood trees for pellet production. First, the use of low quality trees in the form of roundwood as a new raw material source for pellet production could reduce the pellet industry's dependence on uncertain deliveries of wood residues from sawmills. Second, the use of low vigor trees, which are often of low quality for traditional end-uses, could be beneficial to the pelletizing process in terms of friction and compression strength of the pellets produced from that material. This, however, would require the use of an appropriate milling screen size. In the present study, a 1.5 mm milling screen size could be inappropriate since it produced a large proportion of fine grinds (31.9% for the non-vigorous trees and 22.9% for the vigorous trees), while a 2.5 mm milling screen size produced a much lower proportion of fine grinds (13.1% for the non-vigorous trees and 10.7% for the vigorous trees). Additionally, the reduction in friction of pellets pressed from wood particles of the non-vigorous trees resulted in reduced energy consumption and caused less risk of die blockage.
Chapitre 7. Pressurized hot water treatment of sugar maple and yellow birch wood particles for high quality fuel pellet production

by

Quy Nam Nguyen, Alain Cloutier, Tatjana Stevanovic, and Alexis Achim

Submitted to:

Biomass and Bioenergy
Résumé

Cette étude a été menée dans le but de transformer des bois feuillus de faible qualité en granules de bois de haute qualité en utilisant l'eau chaude comme procédé de prétraitement. Les particules de bois provenant de l'érable à sucre et du bouleau jaune ont d'abord été prétraitées avec de l'eau chaude à 150 °C, 175 °C et 200 °C pendant 30 minutes dans un réacteur sous pression. Les fractions solides après l'extraction à l'eau chaude ont ensuite été comprimées en granules avec un granulateur. Les granules produits ont montré plusieurs propriétés améliorées par rapport à celles obtenues à partir de particules de bois non-traitées. L'augmentation de masse volumique et de la densité énergétique des granules a atteint environ 30% et 40%, respectivement. La résistance mécanique en compression a été augmentée jusqu'à trois fois ou plus. Les résultats ont également indiqué que les granules de haute résistance à l'eau ont été obtenus en utilisant un matériau traité avec de l'eau chaude à une température d'environ 200 °C pour l'érable à sucre et le bouleau jaune. En outre, une réduction considérable en friction dans le granulateur a été observée lorsque des particules extraites ont été utilisées.
Abstract

This study was conducted with the aim of converting low quality hardwoods into high quality fuel pellets by using hot water as a pretreatment method. Particles from sugar maple and yellow birch trees were first pretreated with hot water at 150 °C, 175 °C and 200 °C for 30 minutes in a batch reactor. The solid fractions following hot water treatment were then compacted into pellets using a single pelletizer. The produced pellets exhibited a number of enhanced quality properties as compared to those obtained from untreated wood particles. The increases in density and energy content of pellets reached approximately 30% and 40% respectively. Compressive strength was increased up to three times or even higher. Results also indicated that pellets of high water resistance were obtained using material treated with hot water at a temperature of about 200 °C for both sugar maple and yellow birch wood. In addition, considerable reduction in friction in the die was observed when treated wood particles were used.
7.1. Introduction

Wood pellets used for the production of heat and power are standardized and traded at both national and international levels (Obernberger and Thek 2010; Döring 2013; Malik et al. 2015). According to a report by Flach et al. (2014) the global wood pellet consumption currently is around 20 million metric tonnes, and forecasts show a rise between 50 and 80 million tonnes by 2020. In 2014, 28 countries of the European Union (EU) produced 13.3 million tonnes and imported about 6.5 million tonnes of wood pellets for their domestic needs. The largest wood pellet exporters to the EU are the USA and Canada. As part of this, Canadian wood pellet exports reached 1.6 million tonnes in 2014 and represented 84% of total wood pellet production in Canada (Aguilar et al. 2015).

The reasons explaining such a strong and dynamic development of the global wood pellet market in recent years, both in terms of production and consumption, includes the advantages of using wood pellets such as environmental benefits, high energy content, and relative convenience of transportation and storage. Furthermore, solid biofuel combustion technology has been developed resulting in much cleaner and efficient combustion. Solid biofuel in the form of pellets, could thus become a more important energy commodity in the near future. The need to improve the quality of the fuel pellets has become increasingly important. However, the current major challenges in the development of high quality commercial wood pellets are their low mechanical properties and high hygroscopicity (Obernberger and Thek 2010, Lam et al. 2013). The affinity of wood pellets to water is a serious concern (Tooyserkani et al. 2013; Craven et al. 2015), especially for exports for which wood pellets travel long distances often across oceans (Tooyserkani et al. 2013). Therefore, minimizing the break-up of pellets upon moisture adsorption and improving their mechanical strength are as important as increasing their energy content.

Research has been conducted worldwide to develop new technologies for the generation of high quality biofuels from renewable resources (Ragauskas et al. 2006; Hunt 2009; Liu et al. 2013). In the literature, a number of studies were performed with the purpose of improving the mechanical durability and water resistance of fuel pellets. A recent research by Craven et al. (2015) reported that the reduction of moisture absorption of wood pellets could be obtained by adding a coating layer of hydrophobic substances (parafin oil, mineral oil and linseed oil). These authors also reported that the treatment of wood pellets with oil did not change the initial pellet strength and it may have the drawbacks of oil treatments such as the risks of contamination to both humans and environment. Other studies reported that hydrothermal treatment and torrefaction are two promising pretreatments that can be employed for converting biomass into solid biofuels (Coronella et al. 2012; Klemm et al. 2013; Billig et al. 2015). Torrefaction, a thermal treatment process performed within a temperature range from 200 °C to 300 °C in an inert atmosphere has been reported as a potential approach to upgrade many fuel properties of both torrefied wood material and its densified pellets. More specifically, several properties of wood
change following torrefaction. As the wood becomes more fragile, it is easier to grind, its heating value increases, moisture absorption is reduced and dimensional stability is improved. The main drawbacks of torrefaction include its negative impact on density and mechanical durability of wood pellets (Peng et al. 2013; Ghiasi et al. 2014; Shang et al. 2014; Peng et al. 2015). Hydrothermal treatment, also known as hot water extraction or hot water treatment (HWT) is an important step in the pretreatment of woody biomass and in wood-based products manufacturing. The solid fraction after HWT process has a higher energy content, higher dimensional stability, higher compressibility and lower springback, higher resistance to biodegradation, lower pollutant emissions during combustion (Navi and Sandberg 2011; Pu et al. 2011; Bach et al. 2013; Hartmann 2013; Pelaez-Samaniego et al. 2013; Hoekman et al. 2014). Moreover, the hydrophobicity of wood material was remarkably improved after HWT (Pelaez-Samaniego et al. 2014; Bach et al. 2016), which implies that water be removed much easier from the material using mechanical processes such as pressing, centrifugation and filtration compared to water in untreated material (Zhao et al. 2014). Therefore, HWT woody biomass could be used as a bio-derived material and fuel (Ragauskas et al. 2006; Amidon and Liu 2009; Pu et al. 2011; Zhao et al. 2014; Billig et al. 2015). In addition, the HWT process could be used to extract valuable chemicals from the biomass which can be converted into high-value products (Ragauskas et al. 2006, Cybulska et al. 2013).

The objectives of this study were (i) to investigate the effect of hot water treatment parameters on the physical and chemical characteristics of treated material, and (ii) to determine the pretreatment and pelletization conditions required to improve pelletization performance and quality of pellets with a focus on reducing the ash content and improving the main fuel properties (e.g. energy content, water resistance and mechanical durability) of the fuel pellets.

### 7.2. Experimental

#### 7.2.1 Raw material

The experiments in this present study were performed using the material from six sugar maple (*Acer saccharum* Marsh.) trees and six yellow birch (*Betula alleghaniensis* Britton) trees (three vigorous trees and three moribund trees of each species). Trees were sampled in July 2010 in two degraded hardwood stands composed mainly of sugar maple and yellow birch. The two stands were located in the vicinity of Mont-Laurier, Québec, Canada (46°39’40”N, 75°36’30”W and 46°39’05”N, 75°36’25”W). Sample trees were classified according to the tree vigor (MSCR) classification system proposed by Boulet (2007) and described as follows. Trees of reserve stock (class R) are those free of any symptoms of disease or damage and are considered as vigorous/healthy trees with the highest probability of survival. Growing trees (class C) have minor defects but are not biologically
declining and are expected to survive until the next harvest without risk of imminent wood decay. Low quality or
defective trees (class S) are considered to be declining in terms of vigor, wood quality, and volume increment,
and are not expected to survive until the next harvest. Finally, moribund trees (class M) show signs of either
lethal pathological infection or severe damage with high risk of trunk breakage. Moribund trees are biologically
declining and are assumed to have a high probability of mortality before the next scheduled harvest.

7.2.2. Hot water extraction process

Dried wood particles were subjected to HWT using a 2-liter batch reactor (Model 4522, Parr Instrument
Company, Moline, Illinois, USA) controlled by a 4842 Parr temperature controller. The HWT process was
performed at three controlled temperature levels of 150 °C, 175 °C, and 200 °C for 30 min under its self-
generated pressures. In each treatment, an amount of 250 g dried wood particles (size of 0.5 - 1.0 mm mesh)
and 1000 ml deionized water (the equivalent ratio of 4:1 w/w) was loaded into the reactor. The reactor was
heated up and maintained at the desired temperature for 30 min. Self-generated pressure values of about 0.47
MPa, 0.89 MPa, and 1.55 MPa were observed corresponding to the set temperatures of 150 °C, 175 °C, and
200 °C, respectively. The reactor was cooled rapidly by immersing it in a cold water bath. The gas was released
to the atmosphere. The liquid fractions were filtered from the solids and collected for further analysis. The solid
fractions were then washed thoroughly with deionized water and exposed to ambient conditions for air drying.
The air dried wood particles were packaged and stored for further analyses.

7.2.2.1. Total dissolved solids and pH value

The amount of residue remaining from a filtered liquor sample after drying the sample at 105 °C to constant
weight is defined as total dissolved solids [Sluiter et al. 2008]. In the present study, the total dissolved solid
content (TDS) was calculated from Equation (7-1).

\[
TDS \, (\%) = \frac{W_r - W_d}{W_l} \times 100
\]  

(7-1)

where \(W_r\) is the total weight of dish and extraction liquor after drying at 105 °C to constant weight; \(W_d\) is the
weight of the dish, \(W_l\) is the weight of the extraction liquor sample before drying.
The pH of the HWT liquor sample was measured after cooling to room temperature using a pH meter (Accumet AB15 Basic, Fisher Scientific, Singapore). All the experiments were performed in duplicate, with average values reported.

7.2.2.2. Higher heating value and ash content

The higher heating value (HHV) represents the absolute value of the specific energy of combustion per unit mass of a solid biofuel burned in oxygen in a calorimetric bomb under specified conditions. In the present study, the HHV and ash content were determined according to CEN/TS 14918:2005 and ASTM D1102-84 standards, respectively. The HHV based on the oven-dry weight of the sample, was calculated from Equation (7-2).

\[
HHV_d = HHV_w \times \frac{100}{(100 - MC)}
\]  

(7-2)

where \(HHV_d\) is the higher heating value at constant volume of the anhdrous fuel, in MJ kg\(^{-1}\); \(HHV_w\) is the higher heating value at constant volume of the fuel as received, in MJ kg\(^{-1}\); and \(MC\) is moisture content of the sample (as percentage by mass on an air-dried basis).

7.2.3. Pelletization process

7.2.3.1 Pellet production

Pellets were individually produced in a single pelletizer under controlled temperature and compaction force of 100 °C and 2000 N, respectively. Details about the single pelletizer used and the pellet production process can be found in Nguyen et al. (2015). For the present study, the HWT and untreated wood particles of both sugar maple and yellow birch were conditioned at 20 °C and 60% RH to equilibrium, and subsequently processed in the single pelletizer. The single pelletizer with a cylinder of 6.25 mm inner diameter and 150 mm long was first heated to the desired temperature of 100 °C. After equilibration, a sample of wood material of about 0.5 g was put into the cylinder and compressed by a piston at a speed of 12 mm min\(^{-1}\) to the desired pressure. After a retention time of 10 s at full pressure, the pellet was pressed out of the cylinder by removing the bottom plate. The force - displacement data were recorded for the whole cycle of compaction and ejection of pellets. Data for adhesive and sliding friction were measured as illustrated in Fig. 7-1. Each pellet produced was stored in a sealed vial for further measurements.

7.2.3.2 Physical and mechanical properties
The density of a single pellet was calculated by dividing its mass by its volume. Mass was measured by using an analytical balance with an accuracy of ±0.0001 g. Volume was calculated from the diameter and length of the pellet, assuming a perfectly cylindrical shape. The diameter and length of individual pellets were measured using an electronic caliper with an accuracy of ±0.01 mm. Pellet density was determined immediately following the ejection of the pellets from the pelletizer. Three replications were performed for each treatment and the mean values were used for statistical analyses.

The water resistance of pellets (water absorption and volumetric swelling) was qualitatively assessed by immersing each oven dried pellet in an aluminum weighing dish containing 25 ml of deionized and distilled water at room temperature (Fig. 7-3). Weight, diameter and length of each pellet were recorded after 2 h, 24 h and 168 h of immersion. Three replications were performed for each treatment and the mean values were used for statistical analyses. The percentage of water absorption (WA) and volumetric swelling (VS) were calculated from Equations (7-3) and (7-4) respectively.

\[
WA(\%) = \left(\frac{W_i - W_o}{W_o}\right) \times 100 \quad (7-3)
\]

where \(W_i\) is the mass of a pellet measured after water immersion for 2h, 24h, and 168h, and \(W_o\) is the initial weight of the oven-dry sample.

\[
VS(\%) = \left(\frac{V_i - V_o}{V_o}\right) \times 100 \quad (7-4)
\]

where \(V_i\) is the volume of the pellet measured after water immersion for 2h, 24h, and 168h, and \(V_o\) is the initial volume of the oven-dry sample.

The compressive strength of pellets was determined following the procedure described by Nguyen et al. (2015). The pellets produced were kept in vials and tested for compressive strength about two hours after production. A universal MTS testing machine was used to measure the peak load in transverse compression of the pellets. The pellet specimen was compressed radially between two parallel horizontal plates until it broke or showed signs of fracture. The compression speed was set to 6 mm min\(^{-1}\). The ratio between the peak load in compression
and the pellet length was defined as the compressive strength. Three replications were performed for each treatment and the mean values were used for statistical analyses.

7.2.4 Data Analysis

The experimental data was analyzed using the proc glm procedure of the SAS/STAT software, version 9.2 (SAS Institute Inc., NC, USA) of the SAS system.

7.3. Results and discussion

7.3.1. Hot water treatment process

In the hot water treatment (HWT) process also known as hot water extraction or autohydrolysis, wood was subjected to a water treatment at temperatures between 150 °C and 200 °C resulting in products in three different states (gas, liquid and solid fractions). The total dissolved solid and the pH value of the liquid fraction are two important extraction indicators for this process. Analysis of liquid fractions showed that the pH decreased as the extraction temperature increased (Table 7-1). During hot water extraction, acetyl groups in wood hydrate and dissociate, creating acetic acid and thus lowering the pH of the liquid fraction (Liu et al. 2013).

Data in Table 7-1 show that extraction temperatures of 150 °C, 175 °C and 200 °C resulted in different amounts of dissolved solid (TDS), which can be arranged in rank order as the lowest, the highest and intermediate respectively. In general, TDS in the liquid fractions increased with increasing temperature from 150 °C to 175 °C, and then decreased to an intermediate value for an extraction temperature of 200 °C. The elevated temperature could promote the formation of a larger amount of gases during hot water extraction (Yang et al. 2007). Since the reactor with contents (e.g. gases, liquor and solid residual) after hot water treatment was cooled down, a part of the gases (e.g. volatile condensable components) then could condense and mix into the liquid phase used for further analysis of total dissolved solid contents, while the other part of the gases (non-condensable components) was released to the atmosphere. Beside this loss, a part of the volatile matters in the liquid phase could be removed during oven drying. It suggested that given the elevated extraction temperature of 200 °C, the loss of volatile matters would be more important in the gaseous phase. This also contributed to the lower dissolved solid contents of the liquid fractions as indicated in Table 7-1.

Ash content is an important fuel property that needs to be monitored because it can have a negative impact on the combustion and conversion processes (Obernberger and Thek 2010; Meincken and Tyhoda 2014). As reported by Alakangas et al. (2006), a raw material ash content of less than 0.7% is a requirement for the
production of high quality solid biofuels such as wood briquettes and wood pellets. Furthermore, lower ash content requires less frequent ash cleaning, which is important for the automatic operation of pellet boilers and stoves. In the present study, the ash content of untreated and HWT wood particles are presented in Table 7-1.

Table 7-1. Physical and chemical characterization of liquid and solid fractions/products resulting from/following the hot water treatment process.

<table>
<thead>
<tr>
<th>Characteristics of products from HWT process</th>
<th>Raw materials and pretreatment conditions</th>
</tr>
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<tbody>
<tr>
<td>Tree species/ Tree vigor</td>
<td>Hot water treatment</td>
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<tr>
<td></td>
<td>Before treatment</td>
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<tr>
<td></td>
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<tr>
<td>pH value of the extraction liquor</td>
<td>Sugar maple/ R class</td>
</tr>
<tr>
<td></td>
<td>Sugar maple/ M class</td>
</tr>
<tr>
<td></td>
<td>Yellow birch/ R class</td>
</tr>
<tr>
<td></td>
<td>Yellow birch/ M class</td>
</tr>
<tr>
<td>Total dissolved solid content (TDS), %</td>
<td>Sugar maple/ R class</td>
</tr>
<tr>
<td></td>
<td>Sugar maple/ M class</td>
</tr>
<tr>
<td></td>
<td>Yellow birch/ R class</td>
</tr>
<tr>
<td></td>
<td>Yellow birch/ M class</td>
</tr>
<tr>
<td>Ash content, %</td>
<td>Sugar maple/ R class</td>
</tr>
<tr>
<td></td>
<td>Sugar maple/ M class</td>
</tr>
<tr>
<td></td>
<td>Yellow birch/ R class</td>
</tr>
<tr>
<td></td>
<td>Yellow birch/ M class</td>
</tr>
<tr>
<td>Higher heating value (HHV), MJ kg⁻¹</td>
<td>Sugar maple/ R class</td>
</tr>
<tr>
<td></td>
<td>Sugar maple/ M class</td>
</tr>
<tr>
<td></td>
<td>Yellow birch/ R class</td>
</tr>
<tr>
<td></td>
<td>Yellow birch/ M class</td>
</tr>
</tbody>
</table>

Note: † self-generated pressure; ‡ determined following the test for pH of wood at room temperature; R class is designated as vigorous trees; M class is designated as moribund trees; N/A - Not applicable. For each property, the comparison is based on means. Numbers in parenthesis represent standard deviation. Values with different letters indicate significant differences at the α = 0.05 level.

The results indicated that the ash content of all HWT wood samples was appreciably lower than for untreated wood samples. The results also revealed that the ash content of HWT wood samples decreased when the extraction temperature increased. Data in Table 7-1 suggest that HWT could remove a significant proportion of
the ash present in wood samples. The ash content of moribund sugar maple and yellow birch wood samples reduced from 0.97% and 0.47%, respectively for untreated wood samples to 0.20% and 0.03% after HWT at 200 °C. This corresponds to ash removals of 79% and 93% from moribund sugar maple and yellow birch trees, respectively.

The HHV based on the oven-dry weight of all HWT wood samples were significantly higher than those of the untreated wood samples (Table 7-1). Results also indicated that the HHV of the HWT wood samples increased with the extraction temperature. This finding could be explained by variations in the amounts of lignin and ash between the untreated and HWT wood samples. First, the lower ash content in HWT wood samples could have a positive impact on the HHV. Second, the HWT process dissolved greater proportions of the lower heating value materials such as hemicelluloses and related materials, leaving behind a lignin-rich residue. Because lignin has a higher amount of energy per unit mass than the other two main structural components of wood (White 1987; Moya and Tenorio 2013), its increased mass proportion could contribute to increase the HHV of HWT wood samples.

7.3.2. Pelletization process

The pelletization process depends on the friction between the compression channel and the raw material (Oberberger and Thek 2010). A lower friction during pelletizing with a single pelletizer implies a lower energy consumption when pelletizing with a large-scale pellet mill (Shang et al. 2014). These authors reported that the pelletizing of torrefied wood chips involves high energy consumption and heat generation in the press channel which can cause pellet self-ignition. Similarly, a study by Mišlenovic et al. (2014) indicated that torrefied material required higher force for pellet discharge because of the higher friction generated on the surface of the press channel wall. These authors also confirmed that more energy is required for pelletizing torrefied wood. Conversely, the results of the current study (Fig. 7-1) reveal that pelletizing HWT wood particles could result in lower friction force in the press channel.
7.3.2.1. Pellet density and energy content

The pellets produced from HWT wood particles of both sugar maple and yellow birch had a higher density compared to those produced from untreated wood particles. In the case of pellets produced from HWT wood particles, the results in Table 7-2 show that pellet density increases with increasing HWT temperature. Pellets produced from untreated moribund (M class) sugar maple wood particles had a density of 1046 kg m$^{-3}$, whereas density of pellets produced from treated wood particles obtained from the same tree vigor class reached 1361 kg m$^{-3}$ following HWT at 200 °C. It is about 30% higher than for pellets produced from untreated wood particles.

One of the most important characteristics of a biofuel is its energy content, which is a measure of how much energy is contained per unit volume of a fuel (Lestander 2013). Higher energy content is associated with higher
heating value (HHV) and higher pellet density. The energy content is obtained by multiplying the HHV by the density of pellets. The energy contents obtained in the current study are shown in Fig. 7-2. Since both the HHV and density of pellets increased, the energy content of pellets made from HWT wood particles rapidly increased with extraction temperature. The average results on energy content calculated for pellets made from 200 °C HWT sugar maple and yellow birch particles were 29.1 and 28.4 GJ m⁻³ respectively. This is about 43% and 41% higher respectively, than for the control pellets. This enhancement in energy content would result in substantial decreases in the costs of handling and transportation.

Table 7-2. Physical and mechanical properties of pellets pressed from hot water treated and untreated sugar maple and yellow birch wood particles.

<table>
<thead>
<tr>
<th>Quality parameters/ properties of fuel pellets</th>
<th>Raw materials and pretreatment conditions</th>
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<tr>
<td></td>
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<td>Tree species/</td>
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<td>Tree vigor</td>
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<td>After treatment at temperature of</td>
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<td>150 °C</td>
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<td>175 °C</td>
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<td>200 °C</td>
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<tr>
<td>Moisture content, %</td>
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<tr>
<td>Sugar maple/ R class</td>
<td>6.4</td>
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<td>5.6</td>
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<td>6.6</td>
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<tr>
<td>Sugar maple/ M class</td>
<td>6.6</td>
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<td></td>
<td>5.8</td>
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<td>6.3</td>
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<tr>
<td>Density, kg m⁻³</td>
<td></td>
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<tr>
<td>Sugar maple/ R class</td>
<td>1027H (1.70)</td>
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<tr>
<td></td>
<td>1070G (4.67)</td>
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<tr>
<td></td>
<td>1269C (4.61)</td>
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<tr>
<td></td>
<td>1350A (7.05)</td>
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<tr>
<td>Sugar maple/ M class</td>
<td>1046D (3.74)</td>
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<tr>
<td></td>
<td>1102C (8.47)</td>
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<td></td>
<td>1284C (8.44)</td>
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<tr>
<td></td>
<td>1361A (10.02)</td>
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<tr>
<td>Yellow birch/ R class</td>
<td>1011J (3.44)</td>
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<tr>
<td></td>
<td>1116F (4.50)</td>
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<td></td>
<td>1245D (7.05)</td>
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<td></td>
<td>1321A (9.65)</td>
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<tr>
<td>Yellow birch/ M class</td>
<td>1037H (6.40)</td>
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<tr>
<td></td>
<td>1138F (6.61)</td>
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<td>1288C (5.06)</td>
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<td></td>
<td>1358A (7.83)</td>
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<td>Compressive strength, N mm⁻¹</td>
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<tr>
<td>Sugar maple/ R class</td>
<td>44.0H (1.70)</td>
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<tr>
<td></td>
<td>59.6E (2.38)</td>
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<td></td>
<td>130.8D (1.16)</td>
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<td></td>
<td>164.9B (3.50)</td>
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<tr>
<td>Sugar maple/ M class</td>
<td>50.8G (1.05)</td>
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<td></td>
<td>69.1E (1.16)</td>
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<td>143.3C (1.00)</td>
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<td></td>
<td>176.4A (1.60)</td>
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<td>Yellow birch/ R class</td>
<td>28.3J (0.82)</td>
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<td>51.3G (1.39)</td>
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<tr>
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<td>133.2D (2.72)</td>
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<tr>
<td></td>
<td>169.6B (3.24)</td>
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<tr>
<td>Yellow birch/ M class</td>
<td>37.6H (1.57)</td>
</tr>
<tr>
<td></td>
<td>57.9F (1.70)</td>
</tr>
<tr>
<td></td>
<td>139.2C (1.35)</td>
</tr>
<tr>
<td></td>
<td>177.0A (1.41)</td>
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</table>

Note: For each property, the comparison is based on means. Numbers in parentheses represent standard deviations. Values with different letters indicate significant differences at the α = 0.05 level. R class is designated as vigorous trees, and M class is designated as moribund trees.
7.3.2.2. Water resistance properties

Visual observations showed that the pellets produced from treated wood particles were stable and could hold their integrity up to one week or longer in water, whereas the control pellets disintegrated right after immersion and lost their entire integrity no longer than 5 min in water (Fig. 7-3). These observations provided a first direct evidence on the water resistance improvement of pellets made from HWT wood particles.

It should be taken into consideration that pellets produced from untreated (control) and 150 °C HWT wood particles did not provide a measurable result of water absorption or volumetric swelling because they disintegrated in water as shown in Fig. 7-3. Therefore, the results of water absorption and volumetric swelling were calculated for the pellets made from 175 °C HWT and 200 °C HWT particles only and are presented in Figs. 7-4 and 7-5, respectively. It also should be noted in Figs. 7-4 and 7-5 that water absorption and volumetric swelling results are the average values for each tree species (sugar maple and yellow birch) calculated from the individual pellets of both vigorous and moribund trees.
Fig. 7-3. Water immersion tests: Appearance of the pellets before being immersed in water (a) and after immersion for 5 min (b), 24 h (c) and 1 week (d) in distilled water at room temperature (23 °C). From left to right: pellets made from hot water treated (150 °C, 175 °C and 200 °C) and untreated (control) wood particles of moribund sugar maple trees.
The results presented in Fig. 7-4 show that pellets made from 175 °C HWT wood particles absorbed much more water than pellets made from 200 °C HWT wood particles. The latter had a moisture content of only about 7% (d.b.) after immersing in water for 2 hours, whereas pellets made from 175 °C HWT wood particles had a moisture content of more than 30% (d.b.). The results also showed that water absorption made from sugar maple wood was higher than those from yellow birch wood, except for pellets made from HWT material at 200 °C and immersed in water for 2 hours (Fig. 7-4). These results could be explained by the high content of lipophilic extractive constituents of yellow birch wood which are not removable by hot water treatment (Lavoie and Stevanovic 2006).

Fig. 7-4. Water absorption of pellet samples (sugar maple wood pellets in white and yellow birch wood pellets in black) after immersion for 2, 24 and 168 hours in distilled water at room temperature (23 °C). Pellets produced from 175 °C and 200 °C HWT wood particles were indicated by the numbers 1 and 2, respectively. No results are presented for pellets produced from untreated (control) and 150 °C HWT wood particles because they disintegrated in water. Data are represented as means of three replications for each treatment. Bars accompanied by different letters (a, b, c, d, e, f, g) are significantly different at p < 0.05 (Tukey’s test).

The calculated results of volumetric swelling for the pellets made from 175 °C HWT and 200 °C HWT sugar maple and yellow birch wood particles are presented in Fig. 7-5. Pellets produced from untreated (control) and 150 °C HWT wood particles did not provide a measurable result because they disintegrated in water as shown
Results showed that volumetric swelling of pellets decreased from 175 °C to the 200 °C temperature. At the lower HWT temperature of 175 °C the volumetric swelling of pellets made from yellow birch wood was lower than for sugar maple. However, at the higher HWT temperature of 200 °C, a significant difference of volumetric swelling between sugar maple and yellow birch wood pellets was observed only after 24 hours of immersion or more.

Volumetric swelling varied in the range from 34.7% to 70.1% and from 4.7% to 24.4% for the pellets produced from 175 °C and 200 °C HWT wood particles, respectively. Obernberger and Thek (2010) reported that the swelling of untreated commercial wood pellets may exceed 400% in volume. The present research results on water resistance confirm that HWT could be used as a pretreatment method to improve the water resistance property of fuel pellets in terms of reducing both water absorption and swelling.

![Fig. 7-5. Volumetric swelling of pellet samples (sugar maple wood pellets in white and yellow birch wood pellets in black) after immersion for 2, 24 and 168 hours in distilled water at room temperature (23 °C). Pellets produced from 175 °C and 200 °C HWT wood particles were indicated by the numbers 1 and 2, respectively. No results are presented for pellets produced from untreated (control) and 150 °C HWT wood particles because they were disintegrated in water. Data are represented as means of three replications for each treatment. Bars accompanied by different letters (a, b, c, d, e, f, g) are significantly different at p < 0.05 (Tukey's test).](image-url)
7.3.2.3. Compressive strength

The compressive strength of pellets pressed from HWT wood particles was significantly higher compared to that of pellets made from untreated wood particles (Table 7-2). The results also indicated that compressive strength of pellets produced from HWT wood particles increased with treatment temperature. Pellets produced from moribund sugar maple (M class) wood particles had a mean compressive strength of 50.8 N mm$^{-1}$, whereas the mean compressive strength of pellets produced from wood particles HWT at 200 °C reached 176.4 N mm$^{-1}$. This represents an increase of about 300% as compared to that of pellets produced from untreated wood particles. According to Pu et al. (2011), lignin content of the HWT pretreated wood material increased with the increase of extraction temperature. Moreover, during HWT lignin can migrate from the inner to the outermost layer of the cell walls and leave a deposit of molten lignin on their surface (Pelaez-Samaniego et al. 2015). As a result, stronger pellets may have results from HWT because the greater lignin content provided an evidence for the better adhesion of the particles during pelletization (Kaliyan and Morey 2009). The observations on pellet strength tests (Figs. 7-6 and 7-7) provided additional visual evidence for the enhancement in compressive strength of pellets produced from treated wood particles.

![Typical load - deformation curves of pellets in transverse compression. From top to down are curves under compression for pellets pressed from 200 °C HWT, 175 °C HWT, 150 °C HWT and untreated (control) wood particles of moribund sugar maple trees.](image)

Fig. 7-6. Typical load - deformation curves of pellets in transverse compression. From top to down are curves under compression for pellets pressed from 200 °C HWT, 175 °C HWT, 150 °C HWT and untreated (control) wood particles of moribund sugar maple trees.
7.4. Conclusions

Hot water treatment of wood particles obtained from vigorous and moribund trees of two hardwood species, sugar maple and yellow birch, resulted in improved properties of the pellets manufactured from this material. The main findings of the present study can be summarized as follows: (i) Hot water treatment performed at 175 °C and 200 °C improved the properties of pellets produced from treated wood materials including: the removal of up to 93% of the ashes, the increase in density and energy content of the pellets up to approximately 30% and 40% respectively, the increase in mechanical strength of the pellets up to 300%, and a remarkable improvement of water resistance properties of the pellets in terms of reduced water absorption and volumetric swelling; (ii) The differentiation in quality of pellets made of wood particles obtained from different tree vigor and species including: the lower ash content and higher water resistance of yellow birch pellets as compared to those of sugar maple pellets and the higher compressive strength of pellets produced from moribund trees compared to those produced from vigorous trees; and (iii) The lower friction forces obtained during pelletization of hot water treated wood material associated with its higher density provide evidence for the enhancement of the pelletization process in terms of risk of die blockage and energy consumption. In addition, hot water treatment offers a significant advantage in term of conversion efficiency by avoiding an energy-extensive drying process in comparison with other treatments such as heat treatment which requires dry material. This conversion efficiency is more significant in the context of the current study in which green wood from moribund trees was used as a new type of raw material for the production of fuel pellets. This provides another evidence of energy efficiency. More work is also needed to address other aspects regarding the hot water treatment process such as the unused liquid fractions and the energy consumption, which play an important role in cost and energy balance of the whole process.
Chapitre 8. Conclusion générale et recommandations

8.1. Conclusion générale
Ce projet avait pour objectif d'étudier le potentiel d'utilisation des bois feuillus de faible qualité pour la production de granules énergétiques. Trois aspects différents du procédé de granulation de bois feuillus incluant le nouveau type de matière première, l'optimisation des paramètres, et le développement de granules de haute qualité ont été étudiés afin de contribuer au développement d'une compréhension plus profonde de la granulation des bois feuillus et une meilleure utilisation des bois feuillus de faible qualité. Les résultats de ces recherches ont été rapportés en détail dans les chapitres 5, 6 et 7. Les principales conclusions sont les suivantes:

La revue de la littérature (chapitre 2) a révélé plusieurs tendances comme la demande croissante en biomasse forestière pour la bioénergie, l'utilisation croissante de la biomasse forestière sous forme de biocombustibles solides densifiés pour la production de chaleur et d'électricité, les exigences techniques et environnementales croissantes dans les domaines de la production et de l'utilisation de biocombustibles. La revue de la littérature a également montré que le prélèvement des arbres de faible qualité a été considéré comme l'option la plus prometteuse pour réhabiliter les forêts feuillées dégradées. En conclusion, le scénario de la bioénergie a montré une opportunité potentielle à fournir le marché pour les bois feuillus de faible qualité. Cependant, les difficultés techniques dans la transformation de ce nouveau type de matière première en biocombustibles solides avec une meilleure forme et la meilleure qualité posent de nombreux défis.

Le premier travail expérimental a été effectué pour déterminer les changements dans la composition chimique et la valeur calorifique du bois liés à la vigueur des arbres (chapitre 5). Cette étude a révélé que les arbres peu vigoureux avaient des teneurs en extraits et en lignine plus élevées que les arbres vigoureux. Les résultats ont également indiqué qu'il n'y avait aucune différence dans la valeur calorifique supérieure du bois entre les classes de vigueur des arbres, et entre les échantillons de bois non-extraites et extraits. En conclusion, les caractéristiques techniques et la composition des combustibles de bois feuillus de faible qualité sont intéressantes en comparaison avec des arbres vigoureux.

Les difficultés de granulation des bois feuillus et les paramètres appropriés pour la granulation du nouveau type de matières premières, par exemple l'érable à sucre, ont été étudiés en détail dans la deuxième étude (chapitre
Il est évident que les résultats présentés dans ce chapitre ont contribué à une meilleure compréhension de la granulation des bois feuillus incluant l'optimisation des conditions de granulation (paramètres du procédé et caractéristiques de la matière première) et les avantages de la granulation des bois feuillus de faible qualité. Cette étude a confirmé que les particules de bois provenant d'arbres peu vigoureux sont plus appropriées pour la fabrication de granules de bois en comparaison avec celles des arbres vigoureux.

La troisième étude (chapitre 7) a permis d'appliquer les connaissances obtenues à partir des études précédentes et de les utiliser pour étudier les caractéristiques de granulation des particules de bois (l'érable à sucre et le bouleau jaune) traitées avec l'eau chaude. Les résultats sur l'évolution des granules de haute qualité rapportés au chapitre 7 permettent de conclure que l'application du traitement hydrothermique aux particules de bois avant granulation a produit des granules avec de nombreuses propriétés avancées telles que la résistance à l'eau, la haute densité énergétique, et une très faible teneur en cendres. En outre, la conclusion sur les forces de friction lors de la granulation de bois traité avec l'eau chaude fourni une preuve de l'amélioration du processus de granulation en termes de risque de blocage de la matrice et de consommation d'énergie, qui sont mentionnés comme d'importants facteurs limitant la granulation des bois feuillus non traités.

Rassemblant les résultats et conclusions obtenus à partir de toutes les études mentionnées ci-dessus, il est évident que des granules de qualité conventionnelle et de haute qualité peuvent être produites à partir de bois feuillus de faible qualité. Les paramètres du procédé obtenus à partir des expériences avec le granulateur peuvent être utilisés pour prédir les paramètres des procédés aux échelles commerciale et pilote.

**8.2. Recommandations**

Pour la production de granules traditionnelles, les résultats de la deuxième étude (chapitre 6) indiquent qu'une teneur en humidité de la matière première d'environ 11,2% est optimale pour le procédé de granulation. La haute teneur en humidité du bois vert provenant des arbres sur pied est un problème pour le procédé de granulation en termes de technologies et d'efficacité énergétique. Par conséquent, des mélanges de particules de bois humides provenant d'arbres debouts et de résidus de bois sec (par exemple la sciure et les copeaux de bois) provenant de l'industrie du bois d'œuvre de feuillus pourrait être une solution potentielle à considérer. Plus de travail est nécessaire pour identifier les approches les plus prometteuses afin d'avoir un mélange approprié.
Pour la production de granules de haute qualité utilisant l’eau chaude comme milieu de prétraitement, les résultats de la troisième étude (chapitre 7) sur les forces de friction lors de la granulation de matériau traité à l’eau chaude fourni une preuve de l’efficacité énergétique. En outre, le traitement à l’eau chaude peut offrir un avantage significatif en terme d’efficacité de conversion en évitant un procédé de séchage énergivore comme c’est le cas avec d’autres procédés tels que la torréfaction qui nécessite un matériau séché. Cette efficacité de conversion est plus significative dans le contexte de la présente étude, dans laquelle le bois vert des arbres de faible qualité a été utilisé comme nouveau type de matière première pour la production de granules énergétiques. Cela donne une autre preuve de l’efficacité énergétique. Plus de travail est également nécessaire pour répondre en détail à un certain nombre de questions concernant le procédé de traitement à l’eau chaude tel que les fractions liquides non utilisées et la consommation d’énergie, qui jouent un rôle important dans l’équilibre des coûts et de l’énergie de l’ensemble du procédé.
Bibliographie


Kettunen P. O (2009). Background to the topics concerned in the present papers. Materials Science Forum, 599:1-78.


Fig. A.1-1. Interaction plots of friction in N mm\(^{-1}\) for pellets pressed from particles of the most vigorous sugar maple trees. X1: temperature, X2: moisture content, X3: compressive force, and X4: particle size.
Fig. A.1-2. Interaction plots of density in g cm\(^{-3}\) for pellets pressed from particles of the most vigorous sugar maple trees. X1: temperature, X2: moisture content, X3: compressive force, and X4: particle size.
Fig. A.1-3. Interaction plots of compressive strength in N mm$^{-1}$ or pellets pressed from particles of the most vigorous sugar maple trees. X1: temperature, X2: moisture content, X3: compressive force, and X4: particle size.
Fig. A.1-4. Band of experimental load versus time curves obtained during pelletization of 729 pellets under controlled conditions.

Fig. A.1-5. Band of experimental load - deformation curves in transverse compression of pellets produced from various pelletizing parameters.