

Original article

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Hardness of chemically densified Yellow birch in relation to wood density, polymer content and polymer properties

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Abstract: Density of wood can be increased by filling its porous structure with polymers. Such densification processes aim to increase hardness of wood and are particularly interesting for flooring applications. This study aims to evaluate efficiency of different polymers for chemical densification based on the polymer properties. Yellow birch (*Betula alleghaniensis* Britt.) was chemically densified with seven monomer mixtures through acrylate monomer impregnation and electron beam *in-situ* polymerization. Chemical retention and polymer content of densified woods were recorded. Hardness of treated and untreated Yellow birch was measured and compared to hardness of Jatoba (*Hymenaea courbaril* L.). All densified woods showed higher or comparable hardness to Jatoba. Hardness of densified wood was analyzed in relation to initial density of wood and polymer content of the material using multivariable linear mixed models. Efficiency of polymers for chemical densification was evaluated through effect of polymer content on hardness with interaction coefficients. Polymer films corresponding to monomer impregnating mixtures were prepared through low

energy electron beam and characterized by their glass transition temperature, micro hardness, indentation modulus and crosslinking density. Polymers showed statistically significantly different efficiencies and were separated in two main groups. Overall, polymer efficiency increased with increasing glass transition temperature of polyacrylates.

Keywords: acrylate; densification; electron beam; hardness; impregnation; linear mixed models.

1 Introduction

Wood is a versatile natural material that can be used in various applications such as structural and appearance products, or converted to fibers, chemicals and energy. With regards to all these applications, wood quality can be defined in various ways depending on its end use. For instance, in the flooring industry the most important characteristics are hardness as well as aesthetic appearance and visual grade for both industry standards and consumer perceptions (Jonsson 2008; Lutz 1977). Hardness of wood remains an obstacle to its use in non-residential buildings with heavy traffic areas (Drouin et al. 2013).

Chemical densification is a process increasing density and hardness of wood through impregnation with monomers, oligomers or resins followed by their *in-situ* polymerization or curing. Many authors have addressed this challenge (Cai and Blanchet 2015; Ellis and O'Dell 1999; Goldstein and Dreher 1960; Moore et al. 1983; Schneider 1995; Trey et al. 2010). Such products, known as polymer impregnated wood (PIW), have been produced for decades (Schneider and Witt 2004) and are very attractive as they preserve wood appearance and present increased performances. Chemical densification can be used to add value to low density wood that can then access markets where high density and hardness are required or improve strength and hardness of high-density species. Although PIWs are a combination of wood and polymer, there is no analysis of the material based on properties of both wood and polymer.

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PIWs have been made with both softwood and hardwood, using a variety of monomer systems and polymerization mechanisms (Hill 2006). Vinyl monomers are the preferred chemistry for chemical densification as they can polymerize easily through free-radical mechanism which can be initiated by heat or radiations. Electron beam induced polymerization is attractive as it runs with very fast rates, polymerization occurs within a very short time and is less dangerous than X or γ -rays. Amongst monomer systems, methyl methacrylate (MMA) is the most commonly used but presents risks due to high vapor pressure and flash point. Many other vinyl monomers and monomer mixtures such as styrene, acrylonitrile, hexanediol diacrylate (HDDA), glycidyl methacrylate (GMA), 2-hydroxyethyl methacrylate (HEMA), trimethylolpropane triacrylate (TMPTA) or ethylene glycol dimethacrylate (EGDMA) have been successfully used in wood densification (Cai and Blanchet 2010; Chao and Lee 2003; Devi and Maji 2012; Dong et al. 2015; Ellis and O'Dell 1999; Yong Feng et al. 2013; Zhang et al. 2006).

Several characteristics of monomers were identified as favorable for PIW's hardness. Ellis and O'Dell (1999) showed that combination of HDDA with monomers presenting the ability to enter the cell wall or react with the cell wall, such as polar HEMA or reactive isocyanates, led to increased hardness of PIW. On the other hand, cell wall penetration of the resin was found to be detrimental for hardness at low chemical retention because filled lumens and reduction of the void volume are the primary cause of increased hardness of chemically densified wood (Klüppel 2017). Mixtures of monomers are preferred to the use of individual monomers and lead to greater improvement of mechanical properties and hardness (Devi and Maji 2007; Ellis and O'Dell 1999). However, Zhang et al. (2006) showed that adding only 10 wt% of diacrylate to crosslink MMA did not affect significantly hardness compared to MMA alone. Densification of wood with polyethylene glycol dimethacrylate led to higher hardness than using the corresponding acrylate monomer (Trey et al. 2010). Trey suggested that higher glass transition temperature (T_g) was responsible for improved hardness of polymethacrylates PIWs over the corresponding polyacrylates. Despite the availability of a wide range of acrylate monomers presenting a variety of structures, many acrylate monomers remain unexplored in chemical densification.

As wood hardness increases with wood density and hardness of PIW increases with polymer loading (Heräjärvi 2004; Rowell 2012), both should be considered in order to analyze performances of polymers in chemical densification. Furthermore, Ding et al. (2013) concluded that the increase of density alone was not the single factor affecting properties of densified wood (Ding et al. 2013). High chemical retention also leads to increased cost of the manufactured products, which

can be disadvantageous in a cost driven market as in North America (Morrell 2018). High performance system providing high hardness with lower polymer loading could be of major interest to produce PIWs that are economically viable. Carefully choosing monomer systems for high performances with lower polymer loading could be a wise route in PIW development.

Blomberg et al. (2005) evaluated the efficiency of different mechanical densification processes by comparing the effect of densified wood density on hardness (Blomberg et al. 2005). In order to establish a relationship between PIW performances with wood and polymer properties, a similar approach is proposed in this study to compare the efficiency of different polymers in chemical densification. This study aims to analyze hardness of PIWs made with different monomer mixtures using a multivariable linear mixed model with effect of initial wood density and polymer content of PIW. Efficiency of polymers was evaluated through comparison of the effect of polymer content on hardness.

2 Materials and methods

2.1 Materials

Yellow birch (YB) (*Betula alleghaniensis* Britt.) and Jatoba (*Hymenaea courbaril* L.) hardwood lamellas, 4.2 mm thick, were provided by Boa-Franc S.E.N.C. (Saint-Georges, Canada). Raw sawn planks, free of knots and cracks, of size 8 cm \times 110 cm \times 4.2 mm were sanded to P150 grit and cut to specimen final size of 4 cm \times 4 cm \times 3.8 mm. Wood material was stored in a conditioning room at 23 °C and 42% RH until constant mass. These conditions lead to an average equilibrium moisture content of 8%. Specimen cutting is reported in Figure 1A.

Seven monomer mixtures were impregnated into nineteen planks with three repetitions to minimize plank to plank variations. In total 57 samples per treatment and control specimen were analyzed. Acrylate monomers hexanediol diacrylate (HDDA), trimethylolpropane triacrylate (TMPTA), tripropylene glycol diacrylate (TPGDA), glycerol propoxy triacrylate (GPTA), pentaerythritol tetraacrylate (PPTTA), dipentaerythritol hexaacrylate (DPHA) and polyethylene glycol 400 diacrylate (PEG400DA) were provided by EMCO-Inortech (Terrebonne, Canada).

Seven monomer mixtures were selected from preliminary work in order to obtain polymers over a wide range of T_g , indentation modulus and micro hardness. Mixtures with a viscosity lower than 150 cP at 25 °C were also preferred. All seven mixtures impregnated in this study are mixtures of one di- and one multi-functional (>2) acrylate monomers by weight. Viscosity of monomer mixtures was measured using a piston-type viscometer Viscolab 4100 (Cambridge Applied Systems Inc., Boston, USA) operated at 25 °C. Composition and viscosity of monomer mixtures are detailed in Table 1.

2.2 Polymer film preparation and characterization

Two layers of electrical tape were placed on glass plates in order to create 2.5 \times 2.5 cm square cases. A volume of 230 μ L of monomer

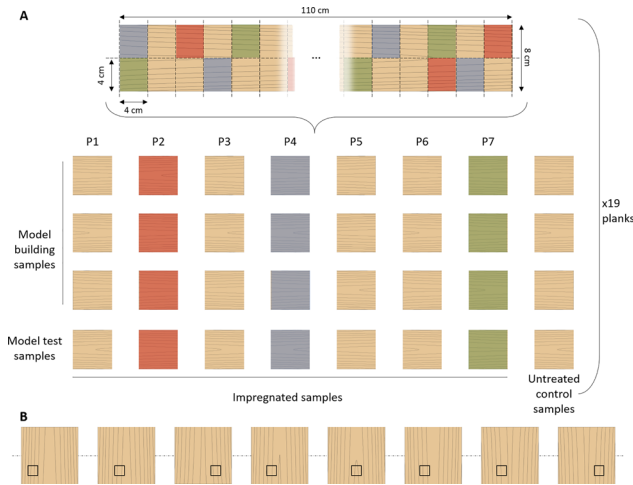


Figure 1: Specimen cutting into a randomized complete block design (A) and laser cutting into the sample for SEM imaging (B).

mixture was placed in the middle of the case and spread onto the surface. This procedure allowed to produce polymer films of $308 \pm 4 \mu\text{m}$ thickness measured with a caliper. Samples were polymerized using a low energy electron beam accelerator EBLab 200 (Comet, Flamatt, Switzerland). A dose of 100 kGy was delivered with an accelerating voltage of 200 kV, heating current of 9.297 mA and 9 m s^{-1} conveyor speed.

Mechanical properties of polymers were measured using dynamic mechanical analyzer DMA Q 800 (TA Instruments, New Castle, USA) equipped with tension clamps. Polymer films were laser cut into 5 mm wide and 10 mm long stripes for DMA analysis. T_g was measured by performing a temperature sweep test. Temperature ramped from -10 to $200 \text{ }^\circ\text{C}$ at a $3 \text{ }^\circ\text{C min}^{-1}$ rate. Oscillating frequency and sinusoidal strain were 1 Hz and 0.05%. For each polymer, three analyses were performed. T_g was defined as the peak of $\text{Tan } \delta$ curve. Crosslink density was calculated from E' , storage modulus in the rubbery plateau, R the gas constant ($R = 8.314 \text{ J K}^{-1} \text{ mol}^{-1}$) and T the

Table 1: Monomer mixture names, composition and viscosity at $25 \text{ }^\circ\text{C}$.

Mixture	Composition	Viscosity (cP at $25 \text{ }^\circ\text{C}$)
P1	25 wt% TPGDA 75 wt% PPTTA	73
P2	75 wt% TPGDA 25 wt% GPTA	18
P3	75 wt% TPGDA 25 wt% DPHA	31
P4	75 wt% HDDA 25 wt% TMPTA	9
P5	25 wt% HDDA 75 wt% PPTTA	50
P6	50 wt% PEG400DA 50 wt% PPTTA	81
P7	50 wt% PEG400DA 50 wt% PETA	143

temperature in Kelvin corresponding to the E' value (Hill 1997; Kron-gauz 2010) with Equation (1):

$$\text{Crosslink density (mol m}^{-3}\text{)} = \frac{E'}{3RT} \quad (1)$$

Microindentations were performed on polymer films using a Tritrec SA microindentation Tester MHT3 (Anton-Paar, Graz, Austria). A Berkovich type diamond tip was employed, a three-sided pyramid with an equivalent cone semi-angle θ of 70.3° . The elastic modulus and the Poisson's ratio of the diamond tip are equal to 1141 GPa and 0.07. The micro-indenter tip was calibrated. A force P and the penetration depth h were measured simultaneously as the indenter penetrates into the film. Loading and unloading curves were recorded. Typical depth penetration curves allowed to calculate polymers indentation modulus and indentation micro hardness. The elastic property was assessed by the physical principle that initial unloading indentation is pure elastic response (Oliver and Pharr 1992). For each polymer specimen, 10 microindentation tests were repeated. The maximum load was reached in a short period of 5 s. Then the load was kept as constant for 300 s. Finally, it was quickly retracted in a time of 5 s to reduce the creep effect on indentation modulus measurement (Feng and Ngan 2002).

2.3 Preparation of densified wood

From each plank, four samples were randomly assigned to each polymer and control series as showed in Figure 1A. Before impregnation, mass and dimensions of YB specimens were recorded. YB samples were placed in a container, immersed in monomer mixtures and weighed down by metallic wire mesh to keep them immersed. The container was placed in a reactor, vacuum was pulled to 3.3 kPa and held for 90 s. Upon the release of the vacuum, samples were wiped off, weighed and wrapped in aluminum foil. Samples were irradiated with 3.8 MeV high energy electrons delivered by a Dynamitron generator (Radiation Dynamics, Inc., Brentwood, USA) with heating current of 42 mA and conveyor speed of 14.6 m min^{-1} . Doses were recorded with Kodac BioMax alanine dosimeters films placed under the foil at the top and bottom of impregnated samples. Mean dose deposited at the top and bottom of specimens were 80 and 110 kGy respectively. Recorded doses are averages of four samples with 4 dosimeter strips at the top and bottom. A total of 532 samples were densified, 399 of which were used to build the model and 133 were used to test the model.

2.4 Chemical retention and polymer content

Chemical retention (CR%) into the wood specimens and polymer content (PC%) in final PIW were calculated from mass before (m_i) and after (m_f) impregnation according to Equations (2) and (3) respectively.

$$\text{CR}\% = \frac{m_f - m_i}{m_i} \times 100 \quad (2)$$

$$\text{PC}\% = \frac{m_f - m_i}{m_f} \times 100 \quad (3)$$

2.5 Hardness measurements

Hardness measurement protocol of treated and untreated specimens was adapted from the DIN EN 1534-11 standard and ASTM D1037-12 test

method. According to EN 1534, hardness is calculated from the mean of along the grain and across-grain indentation diameters. However, due to manual measurement and the collapse of fibers during indentation, the measurement of indentations was found indicative rather than accurate and induces operator bias. Heräjävi (2004) did not find significant differences between hardness measured from indentation diameter and depth, and calculating hardness from indentation depth provide values better correlated to density (Lykidis et al. 2016). On the other hand, from ASTM D1037-12, hardness is measured from the load required to indent wood 5.64 mm deep. This test method is somewhat more accurate but very severe for the material and not adapted for samples with a cross section less than 50 mm by 50 mm (Green et al. 2006). Because the indentation depth and the maximum load can be measured accurately by the testing machine, test method used herein was adapted. The hardness was measured by the maximum force required to penetrate 1 mm deep inside the wood. A universal testing machine QTest/5 Elite Controller (MTS, Eden Prairie, USA) with a 5 kN load cell and 10 mm diameter steel ball indenter were used. Load was applied and recorded to indent wood specimens at a rate of 3.9 mm min⁻¹. A total of 57 indentations (one on each sample) were recorded for each group of treated and untreated YB specimens and 64 samples for Jatoba. Jatoba was used as a target reference.

2.6 Scanning electron microscopy (SEM)

Monomer penetration and morphology of PIWs was observed using a FEI Quanta 250 microscope (FEI Company Inc. Thermo-Fisher Scientific, OR, USA) operated with an acceleration voltage of 15 kV. Morphology samples were laser cut into the densified samples. Samples were all placed at the same vertical position on the laser table and the laser was driven across the samples vertically in order to cut out 7 mm squares at the same distance from the original sample's cross-section as showed in Figure 1B. To improve image quality, samples were coated with a gold-platinum alloy before imaging. Images were recorded at $\times 100$ magnification.

2.7 Statistics and data analysis

Statistical analysis of data was performed using R software (R Foundation for Statistical Computing, Vienna, Austria). Because samples came from various planks with repetitions for each plank, a random

effect was applied to the plank identity for all analyses. One-way ANOVA were executed to compare CR and PC values between treatments and hardness between densified and natural wood. A log transformation on hardness was used to ensure homogeneity of variance. Pairwise Tukey comparisons of least-squares mean was used to identify significant differences at 0.05 level. Variations of natural YB hardness with density were analyzed using a linear model. For densified wood, hardness data were analyzed with linear mixed models using initial density, polymer content and polymer type as explanatory variables. β coefficients associated to polymer content for each polymer were used as indicators of polymer efficiency for chemical densification. Normal distribution of residuals was examined and residuals as a function of predicted values were inspected to ensure validity of the model. Coefficient of determination were calculated by squaring the correlation coefficient between measured values and predicted values.

3 Results and discussion

3.1 Polymer properties

All seven monomer mixtures undergo free-radical polymerization due to collision of monomers with low energy electrons (Coqueret 2017) under electron beam irradiation. Mixtures differ by the structure of monomers varying with the number of acrylate functionalities as well as different backbone structures and lengths. These differences will lead to polymers exhibiting different T_g and crosslink densities (Van Krevelen and Te Nijenhuis 2009). Properties of polymers are summarized in Table 2. It can be seen that all seven mixtures display different T_g , crosslink densities, micro-hardness and indentation modulus. The higher T_g was observed with mixture P4 at 137 °C. This is coherent with its composition being 75 wt% of diacrylate HDDA with a short and stiff alkyl backbone, and 25 wt% of short triacrylate TMPTA.

When comparing polymers, it can be seen that replacing triacrylate GPTA in mixture P2 by hexacrylate

Table 2: Polymer properties: T_g , crosslink density, micro hardness and indentation modulus.

Polymer	Polymer properties			
	T_g (C)	Crosslink density (10^3 mol m ⁻³)	Micro hardness (MPa)	Indentation modulus (GPa)
P1	114 (± 5)	26 (± 4)	85 (± 5)	1.18 (± 0.07)
P2	96 (± 2)	8 (± 2)	17 (± 2)	0.38 (± 0.04)
P3	107 (± 4)	26 (± 1)	40 (± 7)	0.12 (± 0.01)
P4	130 (± 10)	39 (± 2)	61 (± 8)	0.45 (± 0.05)
P5	77 (± 6)	NA	113 (± 8)	1.30 (± 0.1)
P6	71 (± 1)	17 (± 2)	24 (± 5)	0.50 (± 0.1)
P7	71 (± 1)	43 (± 2)	43 (± 5)	0.31 (± 0.03)

Standard deviations are in parentheses. NA, not applicable. Crosslink density could not be calculated for P5

DPHA in mixture P3 led to higher T_g and higher crosslink density. Analogously, replacing PPTTA in mixture P6 by PETA in mixture P7 did not affect T_g but led to a higher crosslink density. A higher crosslink density is coherent with crosslinker structure since PETA is a small crosslinker compared to PPTTA that contains long ethoxy chains.

3.2 Wood impregnation

Chemical retention and polymer content were calculated using Equations (2) and (3) respectively. Figure 2A and B presents chemical retention and polymer content for all seven mixtures. Chemical retention describes extent of impregnation and is representative of the impregnation process while polymer content is representative of the impregnated material composition. Chemical retention and polymer content were different between mixtures and higher chemical retention led to higher polymer content of PIW. Analysis of variance on chemical retention and polymer content values (Table 3) indicated that differences between mixtures were statistically significantly different ($\alpha = 0.05$).

Highest mean chemical retention was observed for mixture P4 with 49% and lowest retention was obtained with mixtures P3, P5, P6 and P7 with 42%. Many factors can influence penetration of monomers into the wood such as molecular weight, polarity and viscosity (Cai and Blanchet 2010; Rowell 2012) as well as wood characteristics such as porosity pattern, vessel size as well as the amount of inaccessible pores and closed lumens (Ding et al. 2008; Wu et al. 2017; Zhang et al. 2005). Chemical retention overall decreased with increasing viscosity of mixture. Mixture P1 with TPGDA was retained to 46% while the use of HDDA in mixture P5 led to 42% chemical retention despite a lower viscosity. This could be attributed to the tripropylene backbone of TPGDA that exhibits higher polarity and thus higher affinity with wood than HDDA and its alkyl backbone. Mixture P1 also presents lower surface tension than P5 allowing higher penetration. Impregnation of wood overall produced materials with polymer content ranging from 20 to 42%.

3.3 Hardness of polymer impregnated wood

Hardness of PIWs was evaluated using a modified version of Brinell hardness and results are displayed in Figure 2C. The hardness of the control group shows expected natural variability of wood. Naturally, wood hardness varies with density, wood structure as well as early/late wood ratio

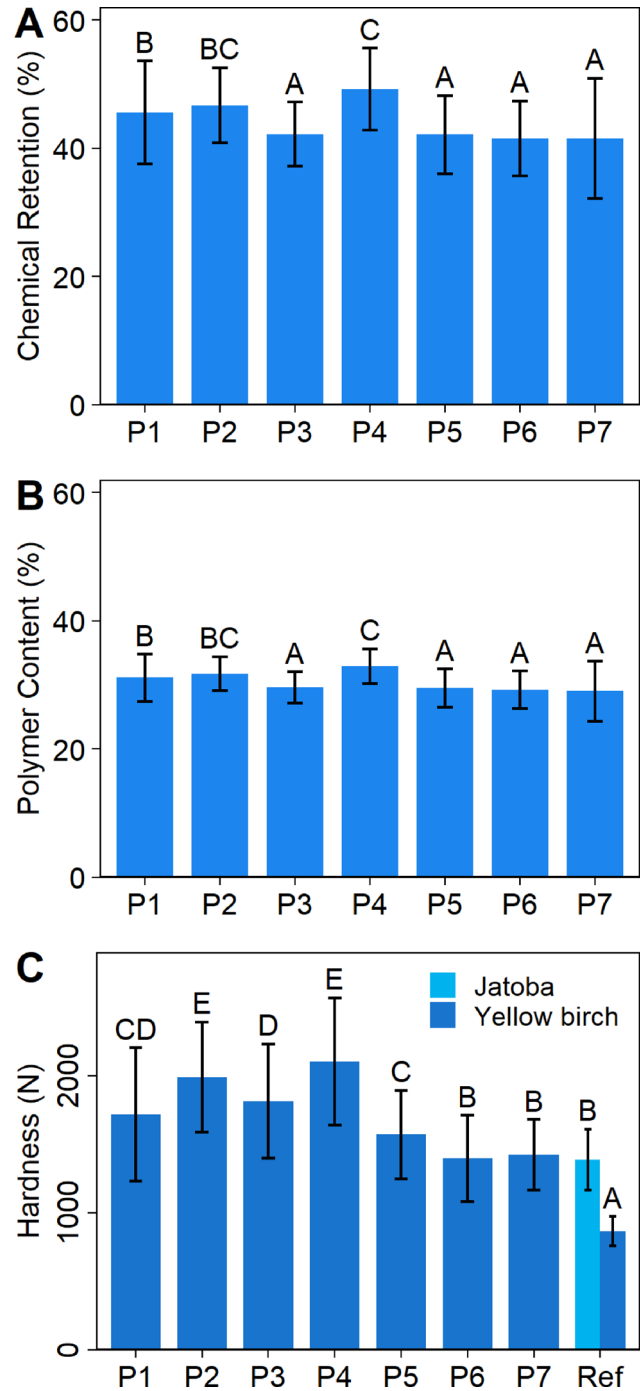


Figure 2: Mean values and standard deviation of chemical retention (A) polymer content (B) and hardness (C) of densified and natural YB (dark blue) and Jatoba (light blue). Letters above bars represent groups revealed by Tukey comparison test after ANOVA analysis ($\alpha = 0.05$). Groups sharing a letter are not significantly different.

(Dinwoodie 1975; Hirata et al. 2001). These variations were minimized by testing several samples from different planks. Treated samples showed an increased variability compared to natural wood. This may be due to the

Table 3: ANOVA results for chemical retention (CR), polymer content (PC) and hardness of impregnated YB.

Source	Degree of freedom	Type III sum of squares	Mean square	F value	Pr > F
<i>ANOVA for chemical retention of impregnated YB</i>					
Mixture	6	3208.00	534.60	20.34	<0.0001***
Planks	18	8298.00	461.00	17.54	<0.0001***
<i>ANOVA for polymer content of impregnated YB</i>					
Mixture	6	746.74	124.46	20.97	<0.0001***
Planks	18	18.81	102.32	17.24	<0.0001***
<i>ANOVA for hardness of impregnated and natural YB</i>					
Mixture	7	1.79E-05	2.56E-06	312.46	<0.0001***
Planks	18	4.87E-06	2.70E-07	32.99	<0.0001***

Note: ***p < 0.001.

impregnated polymer and the differences in polymer content of PIWs. Increased variability of treated samples over natural wood samples was also found for Sugar maple (*Acer saccharum*, Marshall), Scots pine (*Pinus sylvestris*, L.) and hybrid poplar acrylate or methacrylate densified woods (Cai and Blanchet 2015; Koubaa et al. 2012; Trey et al. 2010).

Hardness of Jatoba was found to be greater than natural YB samples as expected and reported in the literature (Ross 2010). Jatoba is known for its high density and hardness and is frequently used for flooring products. Densification of YB allowed to achieve hardness comparable or higher than Jatoba (Figure 2C). The fast vacuum process used herein and 42% mean chemical retention were enough to successfully reach exotic wood hardness performance using local feedstock.

ANOVA on hardness of natural and densified samples is presented in Table 3. Hardness values of all densified YB were significantly greater than hardness of reference natural YB (R). Average hardness increased from 64% with mixture P7 up to 143% with mixture P4. Amongst mixtures, four groups were identified by Tukey multiple comparisons ($\alpha = 0.05$).

While looking at the raw data presented in Figure 2, one may conclude that mixture P4, leading to the highest densified wood hardness, was the best performing mixture proposed here. Similarly, mixtures P6 and P7 being the least effective. These observations were confirmed by performing an ANOVA analysis on hardness (Table 3). This conclusion would be insufficient as it does not consider the materials composition. In fact, mixture P4 is also the system with the highest chemical retention and as hardness of chemically densified wood increases with chemical retention, its higher hardness is

most likely due to higher chemical retention and polymer content.

3.4 Polymer impregnated wood morphology

Figure 3 shows morphology of PIW and penetration of mixtures P3, P5 and P7. These systems, showed same mean chemical retention of 42% despite having different viscosity (31, 50 and 143 cP respectively). For all systems, polymer was found in both vessel and fiber lumens. However overall location of the polymer varied. For P3, polymer was located in both fiber and vessel lumens, while for P5 and P7, polymer was located predominantly in the vessels. This suggests that lower viscosity mixtures are capable of penetrating the small diameter cells as well as vessels while for higher viscosity, vessels seem to be the main infiltration path and fiber lumens remain empty. Despite the same chemical retention, densified wood with mixture P3 showed statistically significantly higher hardness than mixtures P5 and P7 (Figure 2C) indicating that filling the fiber lumens rather than the vessels is preferred to achieve higher hardness at the same CR values.

3.5 Hardness of wood in relation to density

Most strength properties of wood, such as hardness, are reported to increase with density following a power law $f = ap^b$ (Dinwoodie 2000; Kollmann and Côté 1968). As shown in Figure 4, hardness of reference untreated samples increases with density of YB. Both linear and power models were able to describe 53% of hardness variations and were very similar in the range of densities studied here.

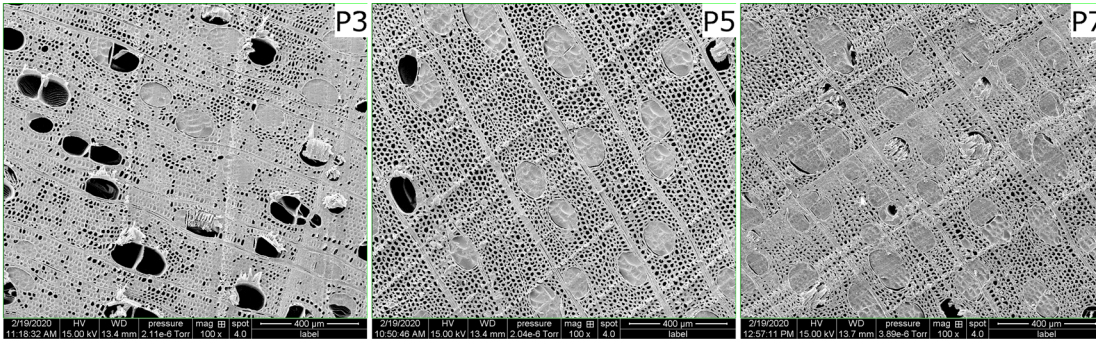


Figure 3: SEM micrographs of densified YB with mixtures P3, P5 and P7. Viscosities of mixtures are 31, 50 and 143 cP respectively.

The same similarities were found between linear and power models for Scots pine (*P. sylvestris* L.) (Grekin and Verkasalo 2013). Hardness of wood is also dependent on many other variables such as early/latewood ratio, wood being sapwood or heartwood, drying conditions, position within the tree and growing conditions (Grekin and Verkasalo 2013; Hansson and Antti 2006; Hirata et al. 2001; Wang et al. 2009). As these details are unknown here, adding a random effect on the board was used to account for them and allows to increase R^2 value to 0.67 for the linear model and to 0.63 for the power model. Similar coefficients of correlation were found for linear analysis of Brinell hardness variations of Silver birch (*Betula pendula* Roth.) and European white birch (*Betula pubescens* Ehrh.) with density (Heräjärvi 2004). For these reasons, the use of a linear mixed model is judicious since good linear correlation was found in this data set.

3.6 Multivariable linear mixed model analysis

This study aims to evaluate hardness of PIWs with different monomer mixtures and discuss their performances based on properties of polymers. PIWs are materials composed of wood filled with polymers. Hence, initial density of wood and proportion of polymer in the material must be considered. Mean polymer content of wood after impregnation being between 29 and 33% (Figure 2B), materials are still mostly composed of wood. It is then expected that PIW hardness would be affected by initial density of the sample as well as chemical retention of impregnation mixture. However, because chemical retention is more representative of the impregnation process rather than the material composition, evaluating PIW hardness with respect to polymer content seems to be more appropriate.

Multivariable linear regression models, are a powerful data analysis that can help answer specific scientific questions when more than one predictor variables are involved. More specifically, they assume a linear relationship between predictors and a continuous response. In this study, the objective is to investigate hardness of PIW, the continuous response, in relation to initial density of wood (ρ_i) and polymer content of material (PC), both predictor variables. Linear mixed models are a type of linear regression model that allow addition of random effects to take into account the sampling strategy. Here, because the 19 planks received all seven monomer mixtures, a random effect was added on the plank in order to account for similarities between samples from the same plank.

The use of multivariable linear regression is supported by Figure 5 presenting hardness of densified woods over polymer content (PC). The color gradient displays initial

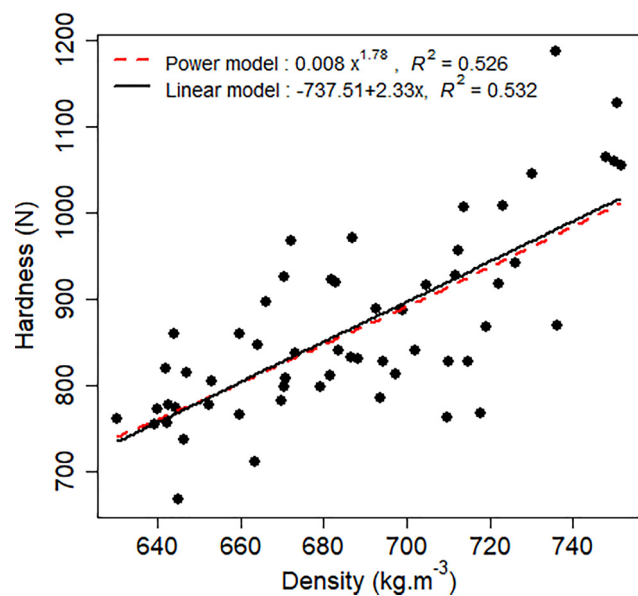


Figure 4: Hardness variations of natural YB with density.

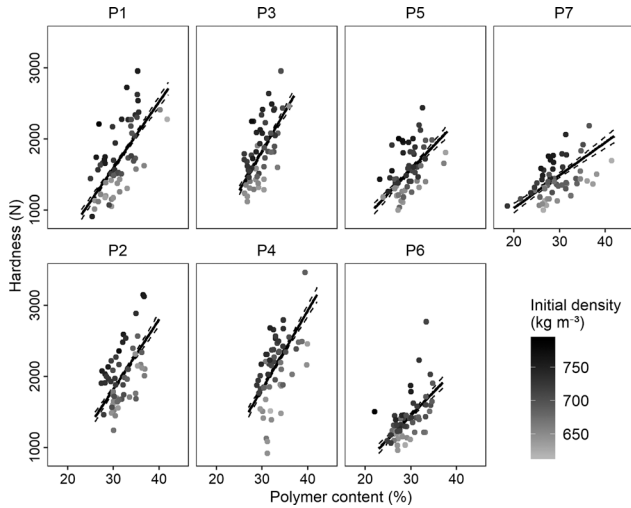


Figure 5: Correlation between hardness and polymer content of PIW for each polymer system. Color gradient represents initial wood density of the sample.

density (ρ_i) of the sample. Figure 5 shows that while hardness of PIW increased with respect to PC, samples with higher ρ_i remained harder than samples with lower ρ_i . Consequently, both PC and ρ_i of the sample must be considered when comparing hardness of PIW and the use of multivariable linear regression is appropriate.

First, in order to perform multivariable linear mixed models, variables must be independent. For each series, Pearson's correlation between ρ_i and PC was checked and was between -0.12 and -0.17 , low enough to ensure that explanatory variables were independent. Then, a linear mixed model with interactions was used to analyze effect of ρ_i , PC and polymer type (Polymer_i) with the following model structure:

$$\text{Hardness} = \beta_0 + \beta_1 \text{PC} + \beta_2 \rho_i + \beta_{3i} \text{Polymer}_i + \beta_{4i} \text{PC} \times \text{Polymer}_i + \beta_{5i} \rho_i \times \text{Polymer}_i + \varepsilon \quad (4)$$

where β_0 is the intercept, β_1 , β_2 and β_{3i} are estimates of the main effect of the polymer content, initial density and polymer for the reference level P1. β_{4i} and β_{5i} are the estimates for the interaction between polymer content and initial density with the polymer type. Polymer content and initial density were covariates and polymer type was treated as a factor variable.

The model detailed in Equation (4) was constructed in order to explore a few hypotheses. First, polymer content, initial density of wood and polymer type were added in order to evaluate their main effect on hardness of densified wood. Interaction element $\beta_{4i} \text{PC} \times \text{Polymer}_i$ was added to investigate influence of polymer type on polymer content

effect. Finally, interaction element $\beta_{5i} \rho_i \times \text{Polymer}_i$ was added in order to verify effect of initial density of wood in each densified wood series.

Table 4 displays F and p value of the model of the three variables as well as interactions. Initial density of wood, polymer content, polymer type as well as the interaction elements had a very significant effect ($p < 0.0001$) on hardness of densified wood. The F value for the interaction coefficient was much lower than the main effects F value. This suggests that hardness of PIWs increased mainly with polymer content and was different between polymers only on a lower level.

Table 5 shows a summary of variables effects from model in Equation (4). PC showed a strong positive effect. Hardness of densified wood increases of 93.7 N with a 1% increase of polymer content for the model with reference level mixture P1. This is consistent with the assumption that increased polymer content leads to increased hardness of PIW and is supported by data presented by Deka et al. (2007) and Hazarika et al. (2015). Hardness of resin impregnated Norway spruce (*Picea abies* (L.) Karst) and Fig wood (*Ficus hispida*) was higher for samples with higher mean chemical retention and thus higher polymer content. Furfurylated Scots pine (*P. sylvestris* L.) also presented the same feature of increased hardness with chemical retention (Lande et al. 2004). Balsa (*Ochroma pyramidale* (Cav. ex Lam.) Urb.) impregnated with vinyl monomers, also showed the same trend (Wright and Mathias 1993).

Initial density of wood also showed strong positive effect supporting the assumption that hardness of densified wood increased with increasing initial density of wood. Thus, as shown by the color gradient in Figure 5, wood presenting high initial density remained harder than lower initial density samples with the same polymer content.

Effect of interaction between initial density and polymer content with polymer type were also greatly significant ($p < 0.0001$). From results displayed in Table 4, it can be concluded that effects of PC and ρ_i content on densified

Table 4: Results of F -tests for each fixed-effects term in the densified wood hardness model according to Equation (3).

	df	F -value	p -Value
Polymer content (PC)	1	419.024	<0.0001 ***
Initial density (ρ_i)	1	250.514	<0.0001 ***
Polymer	6	63.513	<0.0001 ***
<i>Interactions</i>			
Polymer \times PC	6	7.639	<0.0001 ***
Polymer \times ρ_i	6	5.806	<0.0001 ***

Note: *** $p < 0.001$.

Table 5: Summary of multivariable regression analysis (Equation (4)) predicting hardness of densified wood (N = 395).

Variable	Hardness	
	β	SE β
PC	93.7***	7.7
ρ_i	7.8***	0.7
Polymer		
E2	-74.2	818.3
E3	216.1	773.7
E4	-1842.8**	829.1
E5	1960.5**	762.1
E6	2545.8***	783.0
E7	3035.8***	769.2
PC \times polymer		
PC \times E2	4.7	12.3
PC \times E3	17.9	13.1
PC \times E4	16.9	12.1
PC \times E5	-25.8**	11.5
PC \times E6	-28.3**	11.9
PC \times E7	-47.8***	9.5
$\rho_i \times$ polymer		
$\rho_i \times$ E2	0.2	0.9
$\rho_i \times$ E3	-0.7	0.9
$\rho_i \times$ E4	2.2**	0.9
$\rho_i \times$ E5	-1.7*	0.9
$\rho_i \times$ E6	-2.6***	0.9
$\rho_i \times$ E7	-2.4***	0.9
Constant	-6679.5***	589.3

Note: a random effect was added on planks (19 groups, 18 degrees of freedom). * $p < 0.05$; ** $p < 0.01$; *** $p < 0.001$. β coefficient estimates and standard error (SE).

wood were different for at least one polymer. Overall, hardness of densified wood increased with PC and ρ_i , but their effect depended on the mixture impregnated. Effect of PC for mixtures P2, P3 and P4 were not statistically significantly different from effect of P1 ($\alpha = 0.05$). However, effect of PC for mixtures P5, P6 and P7 were significantly different from reference level P1.

Interaction coefficient $\beta_{5i} \rho_i \times \text{Polymer}_i$ was added to verify effect of initial density of wood for each mixture series. Effect of initial density of wood was significant but varied between the different treatments (interaction coefficient $p < 0.0001$) suggesting that effect of initial density of wood for each mixture series was different between series. This could be due to the relatively large sample size on which density was calculated from and the high variability of wood density on a board. The lack of information about early/late wood ratio, grain angle, moisture content, wood processing parameters, height position in the tree or region of provenance can account for the variability amongst treatments group (Dinwoodie 1975; Grekin and Verkasalo 2013; Hansson and Antti 2006; Holmberg 2000).

Significance of interaction coefficient between initial density and polymer could also be due to electron beam irradiation and *in-situ* polymerization of monomers. Ionized radiation of wood causes chemical changes on cellulose and hemicelluloses that were detrimental to materials mechanical properties and hardness (Schnabel et al. 2015; Starr et al. 2014). Electron beam irradiation was used to graft methacrylate monomers to cellulose (Alberti et al. 2005). Irradiation of wood resulted in formation of radicals on cellulose fibers which further reacted with GMA leading to modified cellulose fibers. Additionally, some monomers could have penetrated the cell walls which was showed to be favorable to PIW hardness by Ellis and O'del (1999) or detrimental when CR was low (Kluppel 2017). With this in mind, perhaps the significance of the interaction between wood initial density and polymer results from different degree of polymer grafting onto the wood components or monomer penetration into the cell walls between monomer mixtures.

The model described in Equation (4) was tested using 133 PIW samples. Hardness was predicted on the basis of sample's initial density, polymer content and coefficients described in Table 5. Hardness was measured according to the same modified protocol. The scatter plot of observed and predicted hardness is illustrated in Figure 6. Good correlation was obtained and demonstrates robustness of the model as it was able to predict hardness of the test data set with 81% accuracy.

From the linear mixed model in Equation (4) and the summary of coefficients in Table 5, β coefficients can be extracted leading to Equation (5) for each polymer (i):

$$\text{Hardness}_i = \beta_{6i} + \beta_{7i}\text{PC} + \beta_{8i}\rho_i + \varepsilon \quad (5)$$

with β_{6i} , the intercept and β_{7i} and β_{8i} are the estimates of the effect of polymer content and initial density for each polymer. The β_{7i} coefficients associated to each polymer were extracted and represent polymer densification efficiency: a higher coefficient suggesting a greater ability to increase hardness with polymer content. Coefficients are presented in Figure 7. Lines presented in Figure 5 illustrate predicted values of the model for an average density value of 698 kg m^{-3} (mean value of samples initial density from the data set).

Tukey comparison test allowed to separate the β_{7i} coefficients in three statistically significantly different groups ($\alpha = 0.05$) indicating that effect of PC on hardness was different amongst mixtures. Mixtures P3 and P4 were the most effective at increasing hardness with polymer content and their efficiency was significantly different from mixtures P5, P6 and P7.

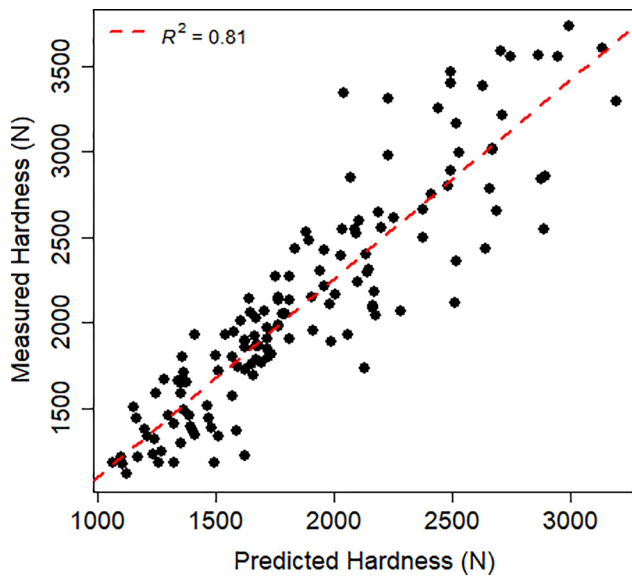


Figure 6: Measured hardness against predicted values of densified YB.

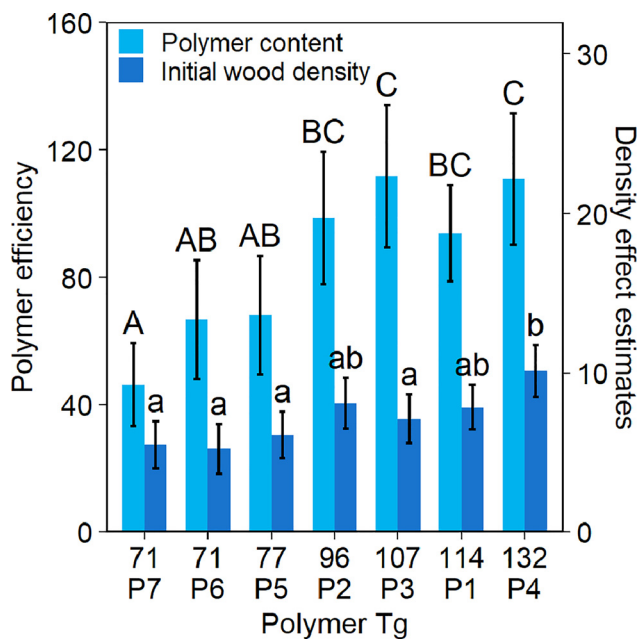


Figure 7: Estimate effect coefficients of initial wood density and PIW polymer content describing hardness of PIW (Equation (5)) against glass transition temperature of polymer systems. Letters above bars describe groups revealed by Tukey test ($\alpha = 0.05$). Groups sharing a letter are not statistically significantly different.

From the grouping established in Figure 7, efficiency of mixtures can be compared and discussed based on the properties of polymer films prepared outside of the wood using electron beam irradiation. Overall, efficiency of polymers for chemical densification increased with T_g of

polymers as suggested by Trey et al. (2010). Yet, efficiencies were significantly different only between high and low T_g s polymers below and above the median value 96 °C. Within these groups, efficiencies were not statistically significantly different ($\alpha = 0.05$).

No clear correlation appeared between microhardness or Indentation modulus of polymer films with polymer efficiency for chemical densification of wood. Mechanical characterisation of polymer films using micro/nano-indentations is very much dependent on the polymer-substrate system (Wen et al. 2017). Moreover, experiments on hardness of PMMA showed that nano hardness values were different than bulk hardness values (Hamada and Kaneko 1992). Adding the fact that PIW are wood-polymer composite materials and the possible chemical wood-polymer interaction, micro indentations of polymer films to predict their efficiency does not seem to be appropriate.

Properties of polyacrylates prepared by electron beam irradiation are affected by electron beam processing parameters such as beam energy, dose or carrier speed and such phenomenon is known as dose rate affect. Polymerization rate and conversion are very much increased with dose rate due to an increased number of initiating species (Burth et al. 2010; Coqueret 2017; Defoort et al. 2001). T_g slightly increased with increasing dose rate while impact toughness showed a declining trend (Schissel et al. 2017; Xiancong et al. 2008). In this study, polymerization of acrylate films and PIWs were not carried under the exact same conditions. While dose was kept constant (100 kGy), beam energy and dose rate were different. Considering this, polymer generated in wood might be slightly different from polymer films generated under low energy electrons explaining why mechanical properties of polymer films were not able to predict efficiency of polymers for chemical densification.

4 Conclusion

This study investigated the effect of different acrylate monomer mixtures on hardness of polymer impregnated yellow birch. Hardness of chemically densified Yellow birch achieved comparable or higher hardness than exotic wood Jatoba. Mean chemical retention of 42% was enough to achieve hardness comparable to exotic wood using local feedstock. Hardness of polymer impregnated yellow birch overall was analyzed in relation to initial wood density and polymer content of the sample through a multivariable linear mixed model. Hardness of densified wood increased with initial wood density and polymer content. Hardness of both high- and low-density woods was improved though chemical densification. Densified

wood that showed higher initial density remained harder than densified wood of lower initial density and effect of polymer content was different amongst polymer systems. Coefficient effects of polymer content for each polymer was used as an efficiency index of polymers in chemical densification. The multivariable mixed model proposed herein is an appropriate and effective approach to compare efficiency of different polymer systems that can be used in chemically densified wood products development to select the most efficient polymer system. Polymer efficiency was discussed based on polymer glass transition temperature, micro hardness and indentation modulus. Overall, efficiency increased with polymer glass transition temperature but was significantly different only between the lowest and highest glass transition temperatures. On the other hand, micro hardness and indentation modulus were not correlated to polymer efficiency measured herein. This is most likely due to the difference of polymerization parameters used for polymers and polymer impregnated wood. Hence, glass transition temperature of polymer can be used as predictor of polymer efficiency and polymers with high glass transition temperature should be preferred in order to maximize hardness of polymer impregnated wood with a given polymer content. Finally, it is important to consider that beam parameters have an impact on polymer properties when performing radiation polymerization. Testing polymer properties directly on polymer located in the lumens should give more accurate information about polymer properties.

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