

# Wood Impregnation in Relation to Its Mechanisms and Properties Enhancement

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The principle of wood impregnation entails treating wood with a monomer/impregnating agent that diffuses into the cell walls, often followed by polymerization to change desired properties. Numerous studies related to this matter have been reported and continue to attract more interest, as wood impregnation can significantly improve wood properties. These processes can be grouped into two approaches: active modification involves the chemical alteration of wood structure by cross-linking, and passive modification features filling of cell cavities and/or cell walls with impregnating agents without any chemical reaction taking place. Wood impregnations could have resulted in an increase in its weight gain due to impregnating agents filling its cavities. It will diminish the utilization of wood as an engineering material in selected application fields. Owing to the extensive literature available, this article summarizes the representative achievements of wood impregnation. The mechanisms, benefits, and drawbacks of various impregnating agents on wood properties, along with grouping the impregnating agents that cause greater or lesser weight gain of wood were analyzed, compared, and evaluated. Thus, according to the application state of wood impregnations, the problems existing in those processes and the developmental trends in the future are also discussed in this review.

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## INTRODUCTION

Wood is a heterogeneous biomaterial that is prone to undergoing dimensional changes in response to easily changing atmospheric conditions. In addition, wood is also susceptible to environmental and biological attacks, so its durability and mechanical strength are often insufficient for long-term end uses. This has motivated a great deal of effort from numerous researchers to find solutions to the issues raised by these drawbacks. Over the past decade, impregnation of wood has become one of the most discussed techniques and continued to attract more interest due to their good results.

The principle of many impregnation processes entails treating wood with a monomer/impregnating agent, which then diffuses into the cell walls, followed by polymerization to impart the desired properties. Both active modification by cross-linking

and passive modification by bulking are included in this process (Rowell and Young 1981, Hill 2006). Hardwood and softwood have been impregnated with a variety of impregnating agents such as waxes and oils (Lesar and Humar 2011; Esteves *et al.* 2014; Ahmed *et al.* 2017; Belchinskaya *et al.* 2021), thermosetting resins, including epoxy, phenol formaldehyde, urea formaldehyde, *etc.* (Furuno *et al.* 2004; Gabrielli *et al.* 2010; Zhao *et al.* 2016; Biziks *et al.* 2018; Schwarzkopf *et al.* 2020; Wang and Zhao 2022), monomer/polymer, including methacrylate, acrylate, styrene, unsaturated polyester, followed by radiation or catalyst-thermal treatment (Chao *et al.* 2003; Hadi *et al.* 2013), noncyclic and cyclic anhydride (Li *et al.* 2001; He *et al.* 2020; Augustina *et al.* 2021), as well as organic acid (Fang *et al.* 2000; L'Hostis *et al.* 2017; Grosse *et al.* 2019; Lee *et al.* 2020; Augustina *et al.* 2022). These types of impregnating agents have been classified as either nonbonded-leachable (NBL), nonbonded-nonleachable (NBNL), or bonded-nonleachable (BNL), depending on the bonding efficiency of the impregnating agent to and from the wood cells (Rowell and Young 1981). For the purpose of understanding the effectiveness of impregnation process on dimensional stability (long-term or short-term application of product services), these categories can serve as the underlying mechanism.

In order to achieve adequate results of physical properties, particularly in terms of dimensional stability, the wood's capillary system (void spaces/pores) must be impregnated with the impregnating agents prior to curing. The type and condition of wood, along with the properties of chemical, *i.e.*, polarity and nature of each chemical, and impregnation conditions, including the processes employed, and the parameters of those processes, *i.e.*, time, pressure, curing, *etc.*, are undoubtedly essential for impregnation treatability (Sint *et al.* 2013; Augustina 2021). The permeability and penetrability of wood species are related to impregnation treatability, since the wood has a capillary structure that offers the primary path for liquid penetration into wood, *i.e.*, cell wall and/or lumen penetration (Flynn 1995; Taghiyari 2012; Wen *et al.* 2014; Augustina 2019). The penetration depth of impregnating agents is also affected by the average molecular weight (MW) of impregnating agent. According to the findings of Imamura *et al.* (1998), the presence of impregnating agents in the wood cells was found to be affected by the MW, or size of the chemicals. Furthermore, cell wall-deposited impregnating agents would have a more profound effect on the wood's properties than those located in the cell lumen. For this reason, it is essential that the impregnating solution has the capacity to swell the cell wall, allowing the penetration of impregnating agents.

As a result of impregnating agents filling the wood cavities, an impregnation process may have led to an increase in its weight gain (Matsuda 1987; Hill 2006). Consequently, it will be diminishing the utilization of wood, per unit of service, as an engineering material in certain application fields. Lightweight engineering materials with greater dimensional stability are needed in respected fields, including those that produce boats, windmills, small wind turbine blades, thermal insulation, and aircraft (Pourrajabian *et al.* 2019; Jang and Kang 2022). According to Sørensen *et al.* (2010), weight savings are critical for wind turbine companies, since the materials must have high stiffness, be fatigue resistant, have high dimensional stability, be damage tolerant, and be lightweight in order for the blades to meet the required design life. However, there is no guarantee that a higher increase in weight gain of impregnated wood means better wood properties (Dong *et al.* 2015; Ahmed *et al.* 2017). According to Rowell *et al.* (2005), at extremely high weight percent gain (WPG), the anti-swelling efficiency (ASE) began to decline to some extent. In propylene, butylene-oxide, and methyl-isocyanate-treated wood, the ASE increases as the WPG increases up to 25% to 30%, while it begins to decrease at higher WPGs. This

may be due to the fact that at a WPG of 33%, small cracks begin to form in the tracheid walls, and at a WPG of 45%, a major crack occurs within cells.

Owing to the extensive literature available, this article summarizes the significant advances that have been made in the field of wood impregnation. The mechanisms, benefits, and drawbacks of various impregnating agents, along with grouping the impregnating agents that cause or less cause an increase in weight gain of wood, are analyzed, compared, and evaluated with respect to their effects on wood properties, especially dimensional stability. In light of this, the paper provides a review of the current state of wood impregnations, the problems that have arisen during these processes and the development trend in the future.

## **IMPREGNATION PROCESS**

Wood impregnation is any method of altering the desired properties of wood by introducing a chemical, a mixture of chemicals, or an inert material (impregnating agents) into the wood cell wall (Hill 2006). These processes can be grouped into two categories: active and passive modification. The former modification involves chemical alteration of wood structure by cross-linking, whereas the latter modification involves the filling of cell cavities and/or cell walls with impregnating agents without any chemical reaction taking place. According to Hill (2006), the impregnating agents can be fixed using two main mechanisms. Monomer or oligomer impregnation, with subsequent polymerization within the cell wall, is one mechanism for fixing the impregnating agents; the other is diffusion of soluble material into the cell wall, followed by treatment in order to render the material insoluble (immobile/locked). Moreover, wood impregnation can be further subcategorized into diffusion, capillary, and pressure treatment (Bi *et al.* 2021). These processes are typically carried out by initial vacuum and pressure cycles at high pressure in the industrial scale. While, in the laboratory scale, one or more processes are usually used.

The most common method for controlling the quality of impregnation process is the increased mass brought on by the impregnating agents. Chemical uptake (CU) measurement is used to characterize the impregnation, whilst WPG measurement is used to characterize the quantity of cured impregnating agents present in the wood (Behr *et al.* 2018). Therefore, this section will discuss wood impregnation, which is mainly done by vacuum, pressure, and a combination of both processes, as well as the relationship between factors that affected the process including type of impregnating agents, impregnation parameters, and wood permeability with WPG and/or CU, as well as the changes in the wood properties especially dimensional stability.

### **Type of Impregnating Agents**

As previously stated, the impregnation process can be divided into two categories: active modification by cross-linking and passive modification by bulking. Cross-linking, as described by Rowell and Young (1981), take place when two or more structural units of the wood cell wall are chemically bonded together (cross-linked), preventing the units from swelling/shrinking as a result of exposure to moisture. In the presence of suitable crosslinking agents, the hydroxyl groups on the same or different cellulose, hemicellulose, and lignin polymers are capable of forming cross-link with each other. Meanwhile, cell wall bulking is defined as an increase in wood volume after treatment that is proportional to the theoretical volume of chemical added. Bulked wood may experience negligible

additional swelling/shrinking of the wood cell wall upon contact with water.

Impregnating materials can be divided into several categories: wax and oil, thermosetting resin, monomer/polymer, cyclic and noncyclic anhydrides, as well as organic acids. Different impregnating materials will have distinct reactions and phenomena when applied to wood. Based on the bonding effectiveness, these types of impregnating agents can be divided into three broad categories. The first classes of impregnating agents are termed nonbonded-leachable (NBL), and they include wax and oil, as well as monomer/polymer impregnating agents. The second one is nonbonded-nonleachable (NBNL) which can be represented by thermosetting resin treatment. The third class is bonded-nonleachable (BNL), which includes anhydride and organic acid treatment. The additive impregnates and forms covalent bonds with the wood cell walls, imparting long-term moisture resistance and dimensional stability.

#### *Nonbonded Leachable (NBL)*

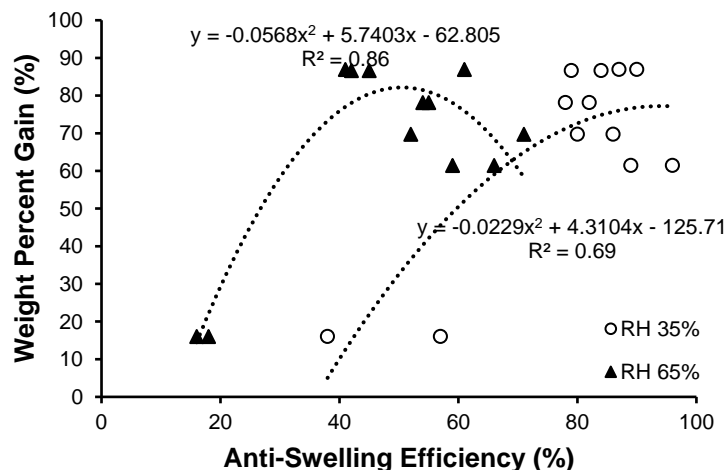
As mentioned before, NBL classes include wax and oil, as well as monomer/polymer as impregnating agents. Benefits of using waxes and oils include their good water repellency and wood protection, environmentally friendly materials derived from dried and/or extracted from plant parts, cost-effectiveness, abundance, and non-toxic nature (Chau *et al.* 2015; Chen *et al.* 2020). In terms of chemical structure, natural waxes can be described as esters of long-chain carboxylic acids and alcohols (Scholz *et al.* 2010a). It is possible to apply wax into wood either in a melted state while diluting it in organic solvents, in the form of emulsions, or as suspensions through impregnation or in conjunction with other treatments such as plasma, *etc.* The type of waxes can be divided into several categories, including natural waxes such as beeswax (Li *et al.* 2020) and carnauba wax (Chen *et al.* 2020); natural fossil waxes derived from petroleum or lignite such as paraffin (Esteves *et al.* 2014; Jiang *et al.* 2020) and montan wax; synthetic such as hydrocarbon or amide waxes (Scholz *et al.* 2010a). Meanwhile, the different kinds of oils that can be utilized as impregnating agents include linseed oil (Chen *et al.* 2020; Liu *et al.* 2020), tung oil (Ahmed *et al.* 2017; He *et al.* 2019), hemp oil (Baar *et al.* 2021), engine oil (Belchinskaya *et al.* 2021), becker oil (Ahmed *et al.* 2017), vegetable oil (Wei *et al.* 2022), and other plant oil-based substances (Vasiliauskiene *et al.* 2020).

Impregnation with waxes and oils has been studied extensively, with many different approaches taken by researchers. Methods range from the straightforward--dipping, soaking/immersing, vacuum-pressure--to the intricate ones by combining those processes with heat treatment, hot and cold soaking methods (physical impregnation), as well as mixing both of methods. For this specific sort of impregnating agent, three types of treatment exist: Type I, treatment decreases moisture uptake, but the swelling is relatively identical as untreated wood over time. This treatment enhances water repellency but not dimensional stability; as for Type II, treatment decreases swelling but not its moisture uptake. This treatment enhances dimensional stability but not water repellency; as for Type III, treatment decreases both moisture uptake and swelling. This treatment enhances water repellency and dimensional stability (Rowell and Bank 1985).

This sub-section focuses on waxes and oils impregnation with vacuum-pressure method. There are basically three major processes involved in wax impregnation: melting microcrystalline wax to a liquid state, impregnating hot wax fluid into wood cells; and then allowing the microcrystalline wax to solidify within the wood's cell cavities and the intercellular space (Zhang *et al.* 2020). It was reported that carnauba wax (CW) could provide WPG ranging from 3.7% to 9.9% with water absorption (WA) reduced by 53% in

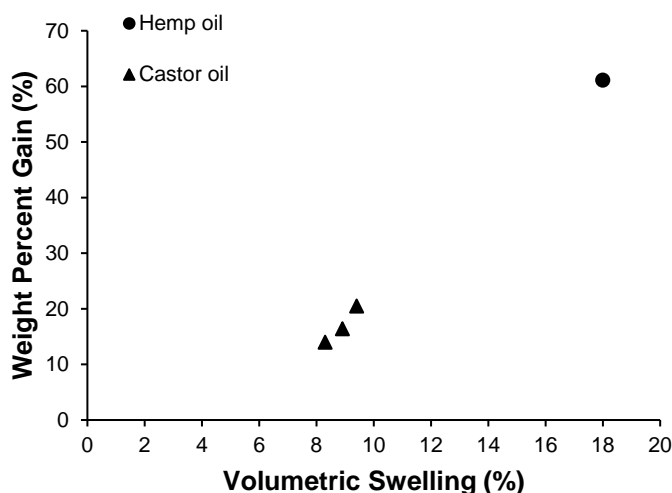
comparison to that of untreated wood after soaking for 1 h, but the values increased to 129% after 192 h. Wang *et al.* (2017) studied paraffin wax emulsion (PWE) in different solid contents (1, 2, 4, and 8%) and particle sizes (535, 400, 320, 232, and 171 nm). They discovered that WPG increased with elevating solid contents and decreased with elevating particle sizes. During this process, the WPGs produced were 2.0% and 11.5% for 1% and 8% solid content of PWE, whereas for particle size of 535 and 171 nm were <2.0% and 3.5% in loblolly pine, respectively. Jiang *et al.* (2020) investigated various PWE concentrations (2, 5, and 8%) and treatment duration. It was found that WPG increased with elevating PWE concentrations, which were 14.5% and 4.8% for 8% and 2% concentration throughout the same treatment time, respectively. As a result, WA decreased as the concentration increased, reaching 40.9% and 52.6% for 8% and 2% concentrations throughout the same treatment time, respectively.

Waxes do not react chemically with the hydroxyl groups of wood; however, they could form a hydrophobic film-barrier on wood surface. Scholz *et al.* (2010b) and Humar *et al.* (2016) were persuaded that there is no interaction between the hydrophobic wax and hydrophilic cell wall with its hydroxyl groups. Furthermore, this compound intends to fill cell lumens, thus achieving a hydrophobic treatment. This compound will be deposited in wood capillaries, reducing water penetration through capillary action and ultimately limiting the dimension swelling. Scholz *et al.* (2010c) found that pore volume decreased from 65-68% to 12-13% and 53-58% to 7-9% for pine and beech treated samples. Nonetheless, there are still significant issues that limit its widespread use. Incomplete and unequal penetration, temperature instability, and insufficient resistance to high temperatures are the common issues (Bi *et al.* 2021). According to Chen *et al.* (2020), wax impregnation using CW has been demonstrated to be effective at inhibiting water absorption in short-term applications. During periods of immersion, the hydrophobic film-barrier may be eroded, allowing water molecules to swell and open up the inaccessible region of wood cell walls. This phenomenon is supported by Fig. 1. Increasing the relative humidity (RH) from 35% to 65% reduced the ASE of PWE treated wood significantly. It implies that as RH increases, the slope of moisture adsorption and T/R ratios increases, thus it will eventually reduce ASE (Chau *et al.* 2015).



**Fig. 1.** Relationship between weight percent gain and anti-swelling efficiency in different relative humidity conditions using paraffin wax emulsion (Data obtained from Esteves *et al.* 2014)

In addition, oils impregnation using vacuum-pressure method have been reported by other researchers (Table 1). Chen *et al.* (2020) and Liu *et al.* (2020), used linseed oil (LO) derived from seed of *Linum usitatissimum* as impregnating agents. Once entering the wood structure, LO can block the lumen and generate a stable LO film on the pore surface, thus limiting water uptake. Liu *et al.* (2020) showed that LO-treated wood could achieve a WPG of around 21.7% and result in lower tangential and radial swelling values. These values were reduced by 19.3% and 15.2%, respectively, when compared to that of untreated. Other researchers reported that the utilization of hemp oil and castor oil could produce WPG of 61.1% and 60.6%, respectively with lower volumetric swelling (VS) than that of untreated (He *et al.* 2019; Baar *et al.* 2021). Due to its larger molecular size, hemp oil also exhibits higher VS and WPG than that of castor oil (Wei *et al.* 2022) (Fig. 2).



**Fig. 2.** Relationship between weight percent gain and volumetric swelling on hemp oil and castor oil (Data obtained from He *et al.* 2019 and Wei *et al.* 2022)

As there is no chemical bonding between wood and oil, it might only reduce the water adsorption rate but not the final moisture content. Furthermore, the oxidative polymerization of LO requires a prolonged time, often allowing the oil to exude from the wood. Therefore, it was discovered that epoxidized LO (ELO) could accelerate oxidative polymerization. ELO-treated wood has shown considerable improvements in ASE, water repellency, biodegradation and leaching resistance compared to LO-treated wood. Chen *et al.* (2020) reported that ELO-treated wood could produce WPG around 31.0% with ASE around 38%. The key improvement mechanism of ELO-modified wood was discovered to be the covalent bonded to the wood cell wall (new hydroxyl and carbonyl groups generated) following the opening of the epoxy ring.

In most cases, oils impregnation led to increased WPG, which is mainly accumulated in the surface area and/or stayed in the cell lumens. This phenomenon was supported by Hill (2006), who noted the inability of triglycerides in vegetable oils to penetrate the cell walls. It is also agreed by Baar *et al.* (2021) and Olsson *et al.* (2001b) that hemp oil and linseed oil have too large molecule size and hydrophobic character to enter the cell wall during impregnation, and consequently uptake occurs through pore cavities such as the lumen of tracheid and ray parenchyma cells.

**Table 1.** Oil Impregnation on Wood using Vacuum-Pressure Method

Author	Liu <i>et al.</i> 2020	Baar <i>et al.</i> 2021	He <i>et al.</i> 2019	Chen <i>et al.</i> 2020	Wei <i>et al.</i> 2022	Wei <i>et al.</i> 2022	Wei <i>et al.</i> 2022
Wood Species	Chinese ash ( <i>Fraxinus mandshurica</i> )	European beech ( <i>Fagus sylvatica</i> L.)	Chinese fir ( <i>Cunninghamia lanceolata</i> (Lamb.) Hook)	Poplar ( <i>Populus cathayana</i> Rehd)	Poplar ( <i>Populus euramericana</i> cv. I-214)	Poplar ( <i>Populus euramericana</i> cv. I-214)	Poplar ( <i>Populus euramericana</i> cv. I-214)
Sample Size (mm)	20 x 20 x 20	20 x 20 x 45	20 x 20 x 20	20 x 20 x 20	53 x 53 x 310	53 x 53 x 310	53 x 53 x 310
Method	Vacuum-pressure	Vacuum-pressure	Vacuum-pressure	Vacuum-pressure	Vacuum-pressure	Vacuum-pressure	Vacuum-pressure
Vacuum (kPa)	10	10	10	100	98	98	98
Vacuum time (min)	90	120	60	30	30	30	30
Imp. Pressure (kPa)	101.3 (1 atm)	NA	101.3 (1 atm)	500	1400	1600	1800
Imp. Time (min)	90	NA	NA	120	60	120	90
Concentration (%)	-	-	-	-	-	-	-
Curing temperature (°C)	103	103	100	80	Continuous curing 80 - 95 - 103 for 24 h	Continuous curing 80 - 95 - 103 for 24 h	Continuous curing 80 - 95 - 103 for 24 h
Impregnating Agent	Linseed oil	Hemp oil	Tung oil	Epoxidized Linseed oil	Castor oil	Castor oil	Castor oil
WPG (%)	21.7	61.1	60.6	30.9	16.4	14.0	20.5
ASE (%)	-	10	-	38	-	-	-
TS/RS (%)	2.7/1.7	-	1.6/1.5	-	-	-	-
MA/WA (%)	7.0	-	5.8	-	81.7	94.7	89.8
VS (%)	-	18	-	-	8.9	8.3	9.4
BC (%)	-	-	-	2.4	-	-	-

Note: WPG = weight percent gain; ASE = anti-swelling efficiency; TS/RS = tangential and radial swelling; MA/WA = moisture and/or water absorption; VS = volumetric swelling, BC = bulking coefficient, NA = not available.

The introduction of monomers and the subsequent *in situ* polymerization of these monomers into a polymer chain is one approach to permanently encapsulating novel materials in wood. These compounds undergo *in situ* polymerization after being exposed to radiation (Hadi *et al.* 2019) or by radical initiator at elevated temperatures (Che *et al.* 2018). A wide variety of vinyl monomers that are commercially available on the market, including acrylonitrile, glycidyl methacrylate, methyl methacrylate, hydroxyethylene, ethylene glycol, dimethacrylate, butyl acrylate, butyl methacrylate, styrene, acrylamide, or acrylonitrile, have been studied by several researchers.

This type of modification is partially effective to improve wood dimensional stability. According to Chao and Lee (2003), styrene (ST) treated wood could give WPG values around 35.9% and 48.3% for 1- and 20-min vacuum conditions, respectively. After 24 h soaking, WA may be lowered from 50% (untreated) to  $\approx$  20%. Hadi *et al.* (2019) investigated the effects of methyl methacrylate (MMA) impregnation on different wood species. They observed that the WPG of MMA-treated wood was around 14.62 to 23.75%; hence, it could achieve a WA of around 24% to 56.7%. According to Meints *et al.* (2018), polyethylene glycol (PEG) treated wood resulted in higher WPG as concentration increased. The WPG was around 10%, 25%, and 42% for 15, 30, and 45% of concentration, respectively. Ding *et al.* (2012) found that poly-MMA could result in an ASE of around 67% with lower WA. Despite the fact that this treatment might provide better dimensional stability, especially ASE, it can only be applied for a brief period of time (short-term application). According to He *et al.* (2011), ST treated wood could provide 88.4% of ASE for almost 2 h water soaking process, then the values subsequently decreased to 23.9% after 168 h water soaking process. This phenomenon was confirmed by Meints *et al.* (2018). After undergoing leaching treatment, the WPG of PEG treated wood was reduced significantly. Leaching of PEG 400 was nearly complete after 14 days of immersion in water (99% mean loss), whereas leaching of PEG 1000 resulted in less mass loss (75%) throughout the same period of time. Furthermore, PEG may liquefy as a result of the enormous amount of water absorbed and exuded from treated wood in high-humidity environment.

In summary, due to the penetration constraints, polymerization of hydrophobic monomers or co-polymer, such as MMA, styrene copolymer and PEG, respectively could not guarantee sufficient dimensional stability, especially when used in longer term service. The utilization of hydrophobic monomers or polymers was unable to enter or penetrate the wood cell wall; instead, these substances could only be filled in the lumen due to hydrophilic nature of wood cell walls. Consequently, pre-treatments have been introduced into this process by methacrylation, *a*-bromoisobutyryl bromide, and tosylation. These pre-treatments might involve modifying the samples with hydrophobic monomers or copolymers, which would result in an improvement of water repellence and dimensional stability (Ermeýdan *et al.* 2014).

#### *Nonbonded Nonleachable (NBNL)*

As mentioned before, NBNL classes include thermosetting resin as impregnating agents. The term “thermosetting resin” refers to a specific kind of synthetic resin with high or low molecular weight oligomers. These oligomers are able to penetrate the cell wall and undergo subsequent polymerization, which is mainly polycondensation, to form the infusible and insoluble high molecular weight polymers. According to Stefanowski *et al.* (2018), there are two main types of thermosetting resin used for wood impregnation: formaldehyde-based resin, such as urea-formaldehyde (UF), phenol-formaldehyde (PF),



melamine-formaldehyde (MF); and non-formaldehyde-based, such as methylene diphenyl di-isocyanate (MDI) and 1,3-methylol-4,5-dihydroxyethyleneurea (DMDHEU). In addition, tannin and furfuryl alcohol are included in this category but will not be discussed in this sub-section. A summary of thermosetting resin in both types based on Stefanowski *et al.* (2018) can be seen in Table 2. These resin types have led to a significant improvement on dimensional stability as an increase of WPG (Table 3). Pittman *et al.* (1994) investigated various types of MF resin. They discovered that the WPG and dimensional stability for MF-, plasticized MF- (PMF), methylated MF (MMF), and melamine-ammeline-formaldehyde (MAF)-treated wood increased significantly. The WPG ranged from 43 to 91%, whereas ASE was around 8% to 53%. According to Deka *et al.* (2000), high molecular weight (MW) of PF and MF resins resulted in WPG values around 12.1% to 40.8% with ASE around 29.3% to 70.6%. In addition, high MW PF yielded a greater increase in WPG and ASE than that of MF. This is likely because PF has a higher bulking coefficient than that of MF. According to Deka *et al.* (2000) the bulking coefficient (BC) ranged around 5.5% to 15.4% and 4.5% to 14.3% for PF and MF, respectively. UF impregnation could result in WPG above 110% and ASE less than 20%.

It is well known that thermosetting resin could be deposited in the lumen cells and function as bulking agent. Frihart (2004) proposed four different scenarios for resin penetration into the cell wall: first, resin simply occupies the free volume within the cell wall, thereby preventing shrinking and swelling; second, there is a mechanical interlocking effect due to cured resin extent from the lumen into the cell wall; third, the polymer network is composed of the cross-linked resin within the free volume of the cell wall; fourth, there is chemical crosslink formation with the cell wall polymeric components. This phenomenon is supported by the relationship between WPG and BC in various thermosetting resins (Fig. 3a). As can be seen in Fig. 3a, an increase in WPG may result in an increase in BC. Higher BC implies that a greater proportion of void cells covered by this compound, resulting in greater dimensional stability improvement. According to Fig. 3b, MUF may provide the highest WPG, followed by PF resins and PF oligomers. When compared to MUF and PF oligomers, the PF resin may provide better BC.

The use of high molecular weight (HMW) of resins could only provide gross penetration, indicating micrometer level penetration. According to Kamke and Lee (2007), gross penetration is caused by the passage of liquid resin into the porous structure of wood due to hydrodynamic flow and capillary action, mostly filling cell lumens. Hill (2006) stated that pre-polymerization of the resin before treatment led to a significant reduction in ASE. This was owing to the fact that bigger molecules had a restricted capacity to enter the cell wall. As seen in Fig. 3c, greater MW leads to a lower WPG and BC, while different phenomenon occurred for low molecular weight (LMW) resins.

Stamm and Seborg (1939) outlined three essential criteria for effective resin treatment of wood: 1. The size of the molecules needs to be sufficiently small enough to allow for penetration of the cell wall; 2. The resin molecules need to be soluble in polar solvents in order to allow for diffusion into microstructure of the cell wall; 3. The resin molecules need to possess sufficient polarity in order to exhibit high affinity with the cell wall. In light of these criteria, it is important to emphasize that the MW of resin needs to be small enough in order to accomplish cell wall penetration through the nanostructure of tissues. This was in agreement with Kamke and Lee (2007), who stated that the MW distribution, viscosity, solid content, and surface tension of the resin would directly affect its penetration. Thus, in turn, will contribute to the enhancement of wood properties, especially dimensional stability.

**Table 2.** Summary of Different Thermosetting Resin Type for Impregnation Process

Type of Thermosetting Resin	Advantages	Disadvantages	Manufacture	Reaction with Wood Cells	References
PF	Improve dimensional stability (ASE up to 50%), biodeterioration, high bonding strength, water resistance, and flame resistance	High formaldehyde emission, curing temperature relatively high, drying process hard to control	Polycondensation of phenol and formaldehyde in an alkaline medium containing excess formaldehyde	Cell wall bulking (gross penetration), thus forms rigid cross-linked network upon curing (cell wall penetration).	Laborie 2002; Klüppel and Mai 2013; Bi <i>et al.</i> 2021; Lang <i>et al.</i> 2022
UF	Low price, improve strength, short-term dimensional stability, durability, strength, faster initial drying velocity, fewer initial checks	High formaldehyde emission, poor water resistance, prone to polymerization during storage, short shortage period.	Combining urea (CH <sub>4</sub> N <sub>2</sub> O) and formaldehyde to produce branched and linear polymers with three-dimensional structure upon curing	Reaction of formaldehyde and ammonium chloride to produce hexamethylene-tetramine and hydrochloric acid, thus those acid reactive functional group forms three-dimensional network structure	Sernek <i>et al.</i> 1999; Zhao <i>et al.</i> 2020; Bi <i>et al.</i> 2021
MF	Improve dimensional stability and biodegradation resistance, high hardness and stiffness, as well as low flammability	High production cost, prone to cracking under dry and wet curing condition	Reaction of the primary amino groups of melamine with formaldehyde to form methylol melamines with up to six methylol groups	Methylol melamines react to macromolecules by forming methylene bridges or ether bond through self-condensation.	Gindl <i>et al.</i> 2003; Altgen <i>et al.</i> 2020; Bi <i>et al.</i> 2021
PUF	Improve dimensional stability, low formaldehyde emission	Leaching due to incomplete resin cure or alkaline degradation of hemicellulose	Mechanical blending of UF resin and alkaline-type of PF resins (resol).	Permanent swelling by cell wall bulking	Tomita and Hse 1998; Stefanowski <i>et al.</i> 2018; Kupfernagel <i>et al.</i> 2022
MUF	Good strength, good hydrolysis resistance, and lower cost than MF	High cost and slow curing	Copolymerization of urea and melamine during resin formation in order to give superior resin qualities	Melamine ring structure increases the crosslink extent of the cured resin to form polymers with high bond strength	Gindl <i>et al.</i> 2002; Stefanowski <i>et al.</i> 2018; Mohd Ali <i>et al.</i> 2020
MMF	No color change, improve hardness and thermal properties, as well as microbial resistance, and low cure temperature	High cost; partially methylated resins more sensitive to temperature; long shelf life	Synthesized from melamine, formaldehyde and methanol	Cell wall bulking and crosslinking with hydroxyl groups of the cellulose molecules in the wood	Klüppel and Mai 2013; Stefanowski <i>et al.</i> 2018
MDI	Moisture resistance	Higher cost than UF	Manufactured from aniline, formaldehyde and phosgene	Covalent bonded urethane bridges formed with hydroxyl groups of the cellulose molecules in the wood	Stefanowski <i>et al.</i> 2018

DMDHEU	Improve dimensional stability and decay resistance, reduce moisture uptake/FSP,	Brittleness, crack, and high formaldehyde emission	Synthesized from urea, glyoxal and formaldehyde	Reduces the pore size by occupying the void space present in the cell walls (cell-wall bulking), thus forms cross-linking of cellulose molecules and polymerization (auto-condensation) via the reactive hydroxymethyl groups.	Militz 1993; Dieste <i>et al.</i> 2009; Sanberg <i>et al.</i> 2017; Emmerich <i>et al.</i> 2020a and b.
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**Table 3.** Thermosetting Resin Impregnation on Wood using Vacuum-Pressure Method

Author	Lang <i>et al.</i> 2022							Girins <i>et al.</i> 2021							Klüppel and Mai (2013)		Zhang <i>et al.</i> 2022			
Wood Species	Beech ( <i>Fagus sylvatica</i> )							Silver birch ( <i>Betula pendula</i> )							Scotch pine ( <i>Pinus sylvestris</i> L)		Poplar wood ( <i>Populus tomentosa</i> )			
Sample Size	25 x 25 x 10							20 x 20 x 20							25 x 25 x 10		20 x 20 x 20			
Method	Vacuum-pressure							Vacuum-pressure							Vacuum-pressure		Vacuum-pressure			
Vacuum (kPa)	0.008							20							10		5			
Vacuum time (min)	45							60							30		30			
Imp. Pressure (kPa)	101.3							101.3							1200		1000			
Imp. Time (min)	60							120							120		240			
Curing temperature (°C)	NA							140							Temperature cycles: 20,40,75,40,103		140			
Impregnating Agent	Phenol formaldehyde (PF) resins							PF oligomers							LMW-PF	MMF	MUF			
Molecular Weight/Mn (g/mol)	237			305	405	520		220			338			467			400	NA	≤ 450	
Concentration (%)	5	10	15	20	20	20	20	10	15	20	10	15	20	10	15	20	30*	30*	10	20
WPG (%)	6.7	12.3	18.3	24.7	24.5	24.6	24.6	10.7	14.0	17.1	10.2	13.6	16.9	8.9	12.4	15.6	50	40	21	43
ASE (%)	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	72	82
TS/RS (%)	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	8.5/5.5	10/5.0	-	-
WA (%)	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
BC	3.8	6.8	9.6	12.2	12.8	11.0	7.9	7.0	9.5	10.7	5.6	7.8	8.5	3.5	5.0	6.2	-	-	6	11

Note: WPG = weight percent gain; ASE = anti-swelling efficiency; TR/SR = tangential and radial swelling; WA = water absorption; BC = bulking coefficient, NA = not available, PF = phenol formaldehyde; LMW-PF = low molecular weight PF; MMF = methylated melamine formaldehyde, MUF = methylated urea formaldehyde. (\*) marks representing solid content.

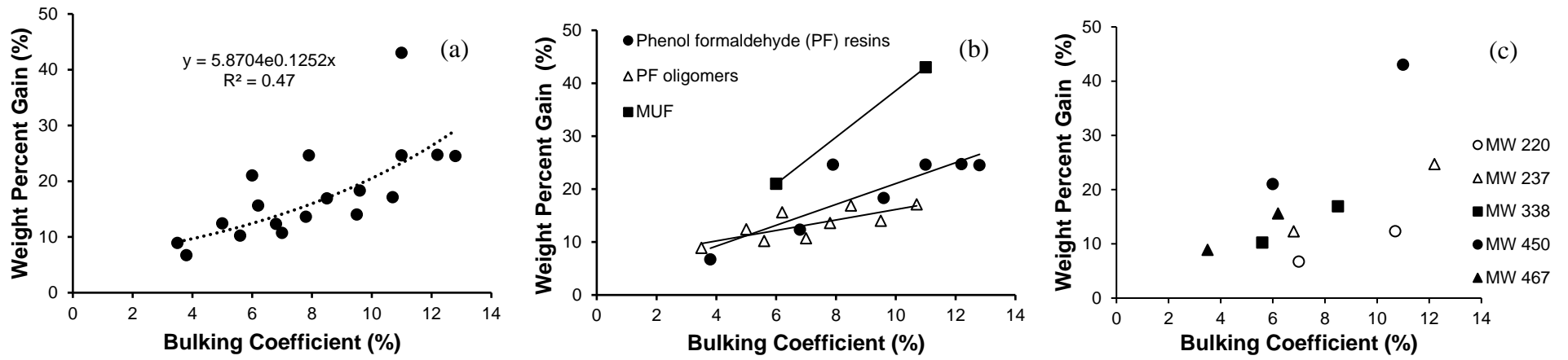


Fig. 3. Relationship between weight percent gain and bulking coefficient in different thermosetting resins (Table 3).

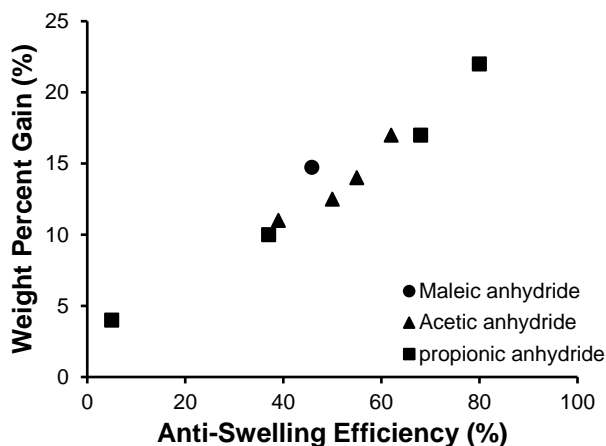
According to the findings of Klüppel and Mai (2013), LMW-PF treatment might offer a greater WPG (50%) than MMF treatment (40%) at a similar concentration of impregnating solution. Furuno *et al.* (2004) added that the amount of resin loading for LMW resin treated wood was found to be approximately 5.9% to 62.5% at a concentration of 1% to 15%, respectively. In addition, BC and WA were 12.6% and 37.1%, respectively, with ASE around 65% at 10% concentration. The lumina of tracheid and ray parenchyma cells contained very little or no phenolic resin when the concentration was between 1% and 5%. At 10% concentration, it was possible to observe a few dispersed cells that had phenolic filling their lumina. Even at concentration of 15%, the number of cell walls that had been filled with resin was extremely low and also scattered. This phenomenon can be observed in the photograph taken with a light microscope. The penetration mechanisms of LMW resins are also reported by Furuno *et al.* (2004). LMW resin is able to penetrate easily into the cell walls, and nearly all of it was found to be located in the wall polymer, with little or no presence resin in the cell lumina. When the concentration of resin is raised, any excess resin that is unable to enter the wall should remain in the lumina, coating and/or filling the space. The cells that had resin in their lumina were distributed sporadically throughout the tissue. As more resin penetrates the walls to form wall polymer, the walls will become bulkier and will absorb less water, leading to an increase in ASE as a direct consequence of this process. Girins *et al.* (2021) came to the conclusion that the MW of resin has a greater impact on enhancing dimensional stability than the solid content. Furthermore, it was also found that 30% WPG is necessary to penetrate wood cell wall and achieve ASE of 70%. Hill (2006) also mentioned that the improvement in dimensional stability has a significant correlation with the amount of mono-methylol phenol content that is present in the PF resin.

#### *Bonded Nonleachable (BNL)*

As mentioned before, BNL classes include anhydrides and organic acids as impregnating agents. Anhydride modification can be grouped into two categories, modification using cyclic anhydrides and non-cyclic anhydrides. Cyclic anhydride can be formed as succinic, alkenyl succinic, phthalic, maleic, glutaric, cyclohexane-1,2-dicarboxylic anhydride, *etc.* Meanwhile, non-cyclic anhydride, such as acetic, propionic, butyric, isobutyric, or hexanoic anhydride, may also be utilized (Hill 2006). These reactions can be carried out with or without catalyst at varying curing temperature (Li *et al.* 2001; He *et al.* 2020).

Among those anhydrides, cyclic anhydride can be represented by maleic anhydride (MAN). On the other hand, acetic anhydride (acetylation) has been the focus of substantial research for non-cyclic anhydride and is currently undergoing commercialization. Several studies have conclusively shown that the dimensional stability of anhydride-impregnated can significantly increase compared with untreated. He *et al.* (2020) investigated the impregnation of cyclic anhydride (maleic anhydride/MAN) at 110 °C for 8 h, which produced a high ASE with only 14.7% of WPG. Li *et al.* (2001) studied the impregnation of non-cyclic anhydride (acetic and propionic anhydride) with and without catalyst at a curing time 90 °C for 8 h in two different wood species. They discovered that acetic anhydride (acetylation) without catalyst can give higher WPG and ASE in hinoki, and yellow-poplar wood compared with propionic anhydride (propionylated). In the presence of catalyst (*i.e.*, potassium acetate, sodium acetate, and sodium propionate), propionylated as well as acetylated wood of both hinoki and yellow poplar were significantly accelerated. Compared to acetylated wood, the propionylated wood exhibited a considerably greater

increase in WPG and ASE. Research suggested that propionylation at 90 °C with catalysts could impart wood with a high dimensional stability. As shown in Fig. 4, WPGs of anhydride treated wood, particularly those treated with maleic, acetic, and propionic anhydride, did not exceed 25%, and the ASE values were around 80%. Hill (2006) concluded the amount of substitution for acetic or other linear chain anhydride rarely exceeds a WPG of 25% unless there has been cell wall damage. This demonstrates that anhydride can be an effective impregnating agent for less weight gain increases with higher dimensional stability.



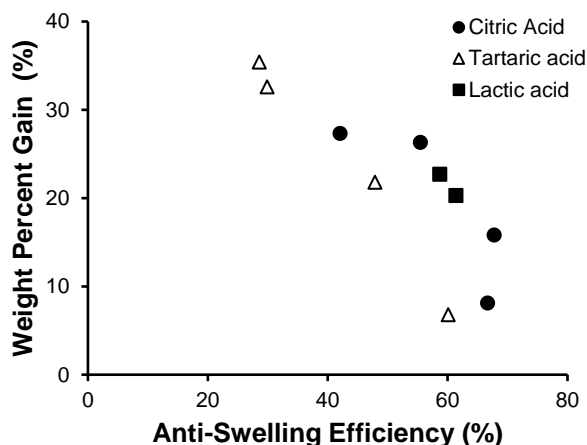
**Fig. 4.** Relationship between weight percent gain and anti-swelling efficiency in different anhydride compounds (Data obtained from Li *et al.* 2001 and He *et al.* 2020).

The reaction between wood and cyclic anhydrides does not produce a by-product, and it results in the modified wood polymers with a covalently bonded carboxylic group. When the temperature is raised above 100°C, there is some formation of diester, which leads to cross-linking within the cell wall matrix (Hill 2006). On the other hand, when the reaction involving non-cyclic anhydride is taking place with the cell wall to form an ester bond, the by-product of this reaction is acetic acid. This phenomenon is in agreement with the findings of He *et al.* (2020) who stated that MAN can be partially replaced by hydroxyl groups and that the remaining hydroxyl groups are somewhat altered. One possible explanation for these alterations is that the carboxyl group formed as a by-product of MAN's reaction with one single hydroxyl group. There is a different phenomenon reported by Li *et al.* (2001) with acetylation of wood. According to them, the swelling of the wood cell wall caused by the volume occupied by its compound and the blocking of the OH groups (bulking agents) is the primary mechanism of this process. Hill *et al.* (2005) investigated the cell wall micropore blocking for anhydride-modified wood. After the wood was treated, it was discovered that the degree of cell wall micropore accessibility was decreased significantly. This may be due to the chemically bonded adduct that occupy spaces in the cell wall. However, those mechanisms are still being debated between researchers. Hill (2006) stated that although cross-linking cannot be completely ruled out, hence bulking also contributed to the dimensional stabilization of wood.

Organic acids can be found in a variety of natural sources, including animals, plants, and microbes. They may be covalently bonded in groups such as amides, esters, and peptides and contain one or more carboxylic acid groups. Some commonly used organic acids include acetic, lactic, citric, malic, and ascorbic (Gurtler 2014). The effects of organic

acids in wood get more attention since these acids may also play a role for wood dimensional stability.

Many studies have been using these acids in order to enhance the dimensional stability of wood. L'Hostis *et al.* (2017) conducted research on the topic of improving the characteristic of beech through *in situ* formation of polyesters of citric acid (CA) and tartaric acid (TA) in combination with glycerol at varying curing temperatures. It was found that CA and TA treatment of wood results in WPG values around 8.1% to 27.3% and 6.8% to 35.4%, respectively. These values were reduced with the increase of curing temperature. On the other hand, a different occurrence was observed with ASE, which intensified when the curing temperature increased. The ASE of CA- and TA-treated wood were 42.1% to 66.7% and 29.9% to 60.1%, respectively. In the study on the effects of lactic acid (LA) on the dimensional stability of wood, Grosse *et al.* (2019) found that the results were very encouraging. They observed that LA-treated wood cured at a variety of temperatures exhibited a similar phenomenon as previous studies. WPG and ASE of LA treated wood were 22.7% and 58.7%, as well as 20.3% and 61.4% at 140°C and 160°C, respectively. This could be owing to higher curing temperature, which leads to better fixation in the wood cell walls *via* ester bonds both in cases of organic acids treated wood. As shown in Fig. 5, WPGs of organic acid treated wood, especially using CA, TA, and LA, did not exceed 40% with ASE of 30% to 60%. This shows that organic acid can be an effective impregnating agent for less weight gain increases with higher dimensional stability, especially when proper curing temperature is applied.



**Fig. 5.** Relationship between weight percent gain and anti-swelling efficiency in different organic acid compounds (Data obtained from L'Hostis *et al.* 2017 and Grosse *et al.* 2019)

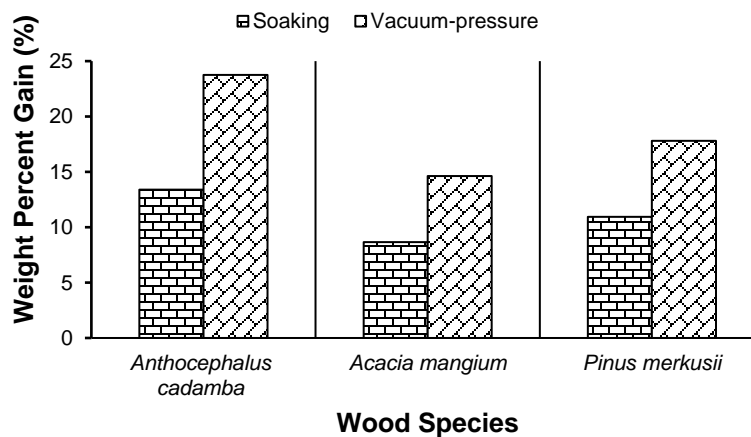
In general, the reaction mechanism that took place between wood and organic acid, in particular CA, involved a two-step esterification process. This began with the formation of anhydride, which was then followed by the reaction of cyclic anhydride with hydroxyl groups of wood; this sequence resulted in the formation of ester linkages (Lee *et al.* 2020). Fang *et al.* (2000) provided conclusive evidence that the cross-linking esterification reaction process takes place between polycarboxylic acid and wood. Instead of passing through a nucleophilic addition of the hydroxyl group and the carbonyl group, it was deduced that the crosslinking reaction went through cyclic anhydride intermediates.

As noted previously, the improved dimensional stability of modified wood has been found to be a function of WPG. From this section, it could be observed that impregnating agents in the first and second class (Fig. 1-3) could gain higher WPG than that of the third class (Fig. 4 and 5). However, the increasing of WPG in impregnated wood is not always correlated with an increase of wood properties, especially dimensional stability (Dong *et al.* 2015; Ahmed *et al.* 2017). According to Rowell (2005), when the WPG was exceptionally high, the ASE started to decrease, at least to some degree. In propylene, butylene-oxide, and methyl-isocyanate-treated wood, the ASE increases as the WPG increases up to 25% to 30%, while it begins to decrease at higher WPGs. This may be due to the fact that at a WPG of 33%, small cracks begin to form in the tracheid walls, and at a WPG of 45%, a major crack occurs within cells. A similar phenomenon also occurs in anhydride modification. Hill (2006) indicated that acetic or other linear chain anhydride rarely reaches a WPG of 25%, unless there is cell wall damage. In addition, oils impregnation increased WPG, and they predominantly accumulated on the surface area and/or remained in the cell lumens. Furthermore, an increase in WPG may cause microscopic cracks in the cell wall layers. According to Olsson *et al.* (2001a) a higher uptake between 75% and 105% could cause further damage due to the pressure gradient at the oil front. This process caused a change in the internal stress of the cell walls, which led to the formation of microcrack in the S1 layer. According to Rowell *et al.* (1978), succinic anhydride modified wood at 61% WPG exhibited extensive cell wall splitting. Hill and Jones (1996) concluded that dimensional stability does not increase much beyond a WPG of 30%.

## Impregnation Parameters

### Methods

Impregnation is accomplished by soaking or by vacuum-pressure to accelerate the impregnating agent into wood cell walls. The outcomes of these methods vary in terms of wood properties, especially dimensional stability. Hadi *et al.* (2019) compared the soaking and vacuum-pressure methods for treating three different kinds of wood species using MMA solution (Table 4). As shown in Fig. 6, the vacuum-pressure method resulted in higher WPG (14.6% to 23.7%) compared with the soaking method (8.7% to 13.4%).



**Fig. 6.** Comparison of weight percent gain produced from soaking and vacuum-pressure method in different wood species (Table 4)

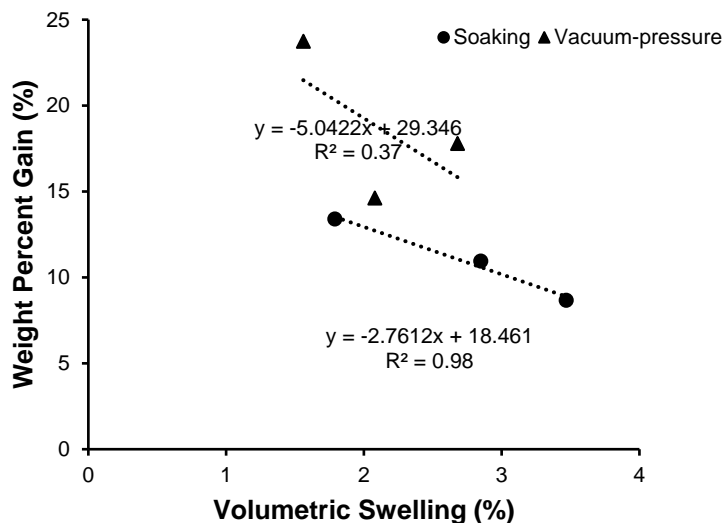


**Table 4.** Comparison between Soaking and Vacuum-Pressure Method in Impregnation Process

Author	Hadi <i>et al.</i> 2019					
Wood Species	Jabon ( <i>Anthocephalus cadamba</i> (Roxb.) Miq.)	Mangium ( <i>Acacia mangium</i> Wild.)	Pine ( <i>Pinus merkusii</i> Jungh. & de Vriese)	Jabon ( <i>Anthocephalus cadamba</i> (Roxb.) Miq.)	Mangium ( <i>Acacia mangium</i> Wild.)	Pine ( <i>Pinus merkusii</i> Jungh. & de Vriese)
Sample size (mm)	50 x 50 x 50	50 x 50 x 50	50 x 50 x 50	50 x 50 x 50	50 x 50 x 50	50 x 50 x 50
Method	Soaking	Soaking	Soaking	Vacuum-pressure	Vacuum-pressure	Vacuum-pressure
Vacuum (kPa)	-	-	-	506.6	506.6	506.6
Vacuum time (min)	-	-	-	30	30	30
Imp. Pressure (kPa)	-	-	-	506.6	506.6	506.6
Imp. Time (min)	-	-	-	30	30	30
Impregnation agents	MMA	MMA	MMA	MMA	MMA	MMA
Concentration (%)	-	-	-	-	-	-
WPG (%)	13.4	8.7	10.9	23.7	14.6	17.8
ASE (%)	-	-	-	-	-	-
TS (%)	-	-	-	-	-	-
WA (%)	80.9	32.8	46.4	56.7	24	41.5
VS (%)	1.8	3.5	2.8	1.6	2.1	2.7

Note: WPG = weight percent gain; ASE = anti-swelling efficiency; TS = thickness swelling; WA = water absorption; VS = volumetric swelling, MMA = methyl methacrylate.

Higher WPG led to an enhancement in dimensional stability, particularly in the reduction of volumetric swelling (VS). A negative correlation between WPG and VS in the soaking method ( $y = -2.7612x + 18.461$ ;  $R^2 = 0.98$ ) and the vacuum-pressure method ( $y = -5.0422x + 29.346$ ;  $R^2 = 0.37$ ) served as evidence for these findings (Fig. 7). According to Table 4, the vacuum-pressure method enhanced ASE values by 12.3% to 29.5% compared to the soaking method. Meanwhile, TS and WA were reduced by 7.3% to 27.8% and 13% to 18.3%, respectively compared with the soaking method.



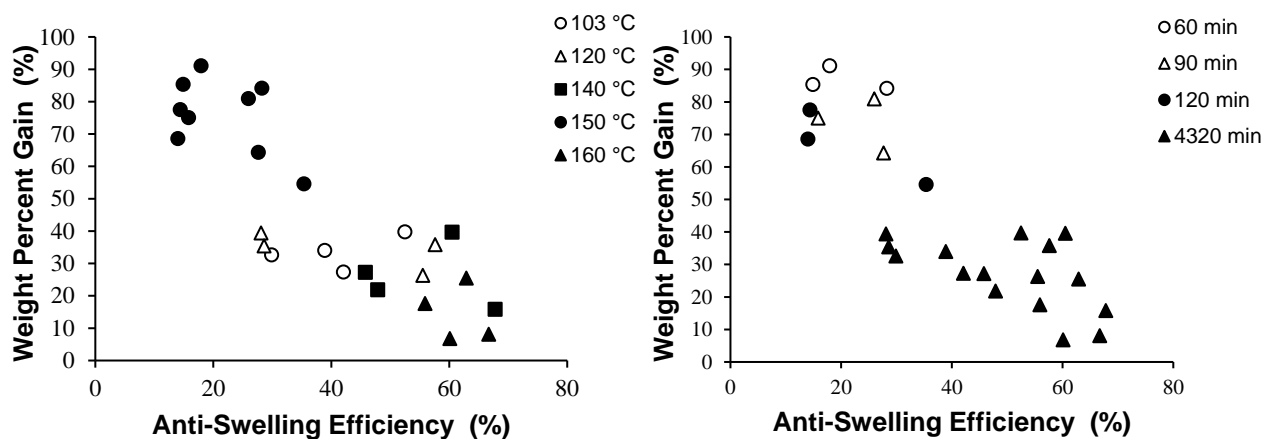
**Fig. 7.** Comparison between impregnation method on weight percent gain and volumetric swelling (Table 4)

Those results indicated that the vacuum-pressure method was more efficient and effective than the soaking method under similar impregnation conditions. The utilization of an initial vacuum prior to the process has the potential to draw out the air trapped inside the lumina cells, which will subsequently open the void cells and make them accessible for impregnation process. The application of pressure following those processes provides a deep and uniform penetration, which in turn fills the void cells more efficiently. This occurs because the pressure forces the impregnating agents into the wood structure. Yildiz *et al.* (2010) and Paril (2016) both came to the same conclusion on this phenomenon. In contrast, the soaking method may only just fill the wood surface due to insufficient diffusion time.

One of the most essential processes in wood industry is liquid impregnation, which can change desired properties by using related functional impregnating agents. However, the improvement is typically constrained by the impregnation process either in materials used or impregnation parameters, *i.e.*, curing condition, pressure, vacuum, *etc.* This can have an effect on the amounts of impregnating agents that are diffused into wood cells, thus resulting in inadequate amount for the improvement of wood properties. Thereby, numerous studies have been done in order to avert those drawbacks. Combining diverse methods prior and/or after impregnation process has been garnering greater interest, including the application of thermal treatments (Paril *et al.* 2016; Ahmed *et al.* 2017), and catalysts.

### Curing Conditions

In order to prevent the impregnating agent from evaporating and allow them to diffuse into the cell wall properly, impregnated wood is kept in non-drying environments, either wrapped in foil or submerged in the impregnation solution (Klüppel and Mai 2013). This process is known as using a curing condition, which requires heat in order to get satisfactory fixation. Impregnated wood is usually dried and cured in an oven in laboratory-scale research. The temperature is kept low at first so as not to cause drying failures, and then it is raised slowly to guarantee the complete fixation (Klüppel and Mai 2013). In an impregnation process, the most important parameter is the curing conditions, which include time and temperature. Different impregnating agents will have different curing conditions, as well as different impacts on wood properties (Table 5).



**Fig. 8.** Relation between weight percent gain and anti-swelling efficiency in different curing temperatures and times (Table 5)

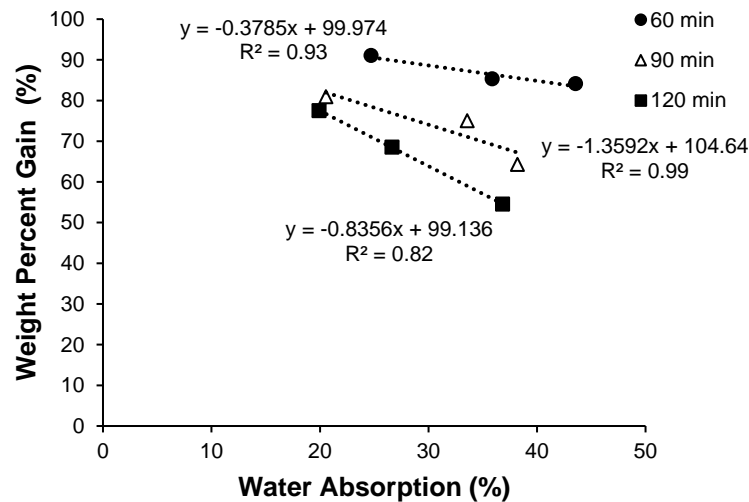
L'Hostis *et al.* (2017), reported that citric acid (CA) and tartaric acid (TA) alone, and in combination with glycerol (G+CA and G+TA) were found to reduce WPG at varying curing temperatures. A negative correlation between curing temperature and WPG ( $y = -1.47x + 170.55$ ;  $R^2 = 0.51$ ) served as evidence for these findings (Fig. 8). According to Fig. 8, curing temperature at 103 °C results in positive correlation between WPG and ASE, whereas an increase of curing temperature tends to give a negative correlation between those parameters.

Extending the curing time also tends to make a negative correlation between WPG and ASE. This could be due to wood degradation at higher curing temperature, which reduces WPG, but still results in higher ASE. This phenomenon excludes the curing temperature at 150 °C, which showed higher WPG than others due to the utilization of LMW-PF admixed urea. A similar phenomenon also was apparent in the relation between WPG and WA in different curing times at a curing temperature of 150 °C (Fig. 9). It was found that an increase of curing time results in the reduction of WPG and WA of treated wood. This phenomenon was confirmed by Behr *et al.* (2018), who stated that if the curing temperature and duration were increased, differential scanning calorimetry (DSC) results showed a higher degree of conversion.

**Table 5.** Comparison between Impregnation Process using Vacuum-Pressure Methods in Different Curing Conditions.

Author	Izreen <i>et al.</i> (2011)									L' Hostis <i>et al.</i> (2017)															
Wood Species	Jelutong ( <i>Dyera costulata</i> )									Beech ( <i>Fagus sylvatica</i> )															
Sample Size (mm)	150 x 50 x 5									20 x 20 x 20															
Impregnating Agent	LmwPF admixed urea									Glycerol + Citric Acid	Glycerol + Tartaric Acid	Citric Acid	Tartaric Acid	Glycerol + Citric Acid	Glycerol + Tartaric Acid	Citric Acid	Tartaric Acid	Glycerol + Citric Acid	Glycerol + Tartaric Acid	Citric Acid	Tartaric Acid	Glycerol + Citric Acid	Glycerol + Tartaric Acid	Citric Acid	Tartaric Acid
Vacuum (kPa)	85			85			85			10				10				10				10			
Vacuum time (min)	15			15			15			20				20				20				20			
Imp. Pressure (kPa)	340			340			340			1000				1000				1000				1000			
Imp. Time (min)	30			30			30			60				60				60				60			
Concentration (%)	20			30			40			-				-				-				-			
Curing Temperature (°C)	150			150			150			103				120				140				160			
Curing time (min)	60	90	120	60	90	120	60	90	120	4320				4320				4320				4320			
WPG (%)	84.1	64.3	54.6	85.3	75.0	68.6	91.1	80.9	77.5	39.7	27.3	34.0	32.6	35.8	26.3	39.4	35.4	39.6	15.8	27.2	21.8	25.5	8.1	17.6	6.8
ASE (%)	28.3	27.7	35.4	14.9	15.8	14.0	17.9	26.0	14.4	52.5	42.1	38.9	29.9	57.6	55.5	28.1	28.6	60.5	67.8	45.8	47.9	62.9	66.7	55.9	60.1
TS (%)	2.0	1.1	0.8	3.0	2.7	1.8	2.9	1.5	2.5	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
WA (%)	43.6	38.2	36.8	35.9	33.6	26.6	24.7	20.6	19.9	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
BE	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-

Note: WPG = weight percent gain; ASE = anti-swelling efficiency; TS = thickness swelling; WA = water absorption; BE = bulking efficiency.



**Fig. 9.** Relation between weight percent gain and water absorption in different curing times at curing temperature 150 °C (Data obtained from Izreen *et al.* 2011 in Table 5)

Several researchers have employed both dry and wet curing conditions in their work. During the first three steps of wet curing, the impregnating agent is cured in a wet state under non-drying conditions, and typically it is packed in polyethylene terephthalate (PET) bags before drying (Klüppel and Mai 2013). Under humid conditions, impregnating agents diffused from the cell wall into the lumens. Some researchers investigated dry and wet curing conditions in an effort to improve the impregnation process. Klüppel and Mai (2013), evaluated the wet and dry curing condition of wood treated with phenol formaldehyde (PF) and methylated melamine formaldehyde (MMF) resins. This research demonstrated that treated wood exhibited a greater density with no radial gradient when cured under wet conditions. Density profiles exhibited a similar pattern with no discernible peak on the wood surface. In the case of dry curing, MMF-treated wood exhibited a narrow radial gradient, whereas PF-treated wood exhibited no radial slope. Dry curing produced better results than wet curing in terms of WPG and tangential bulking, especially in PF-treated wood. In addition, wet curing causes more severe degradation than dry curing due to higher degree of hydrolysis (particularly due to high pH). Mechanisms of dry and wet curing in thermosetting resins are also reported by Klüppel and Mai (2013). During the drying process, the free water in the lumens is the first to evaporate, leading to an increase in the chemical concentration within the lumens. Hence, because of the concentration gradient that exists between lumen and cell wall, dissolved molecules are able to diffuse into the cell wall micropores. This process will carry on until the solution is completely saturated, at which point the resin will begin to precipitate in the cell wall. The solubility of the chemical has a significant bearing on how much of it is deposited. During wet curing, the molecules of resin go through poly-condensation as they are being solubilized. When the molecular size increases, its solubility decreases, and as a result, the resin eventually precipitates and does not migrate after further drying. It is possible to summarize the findings by saying that dry curing led to an unequal distribution of both resins, whereas wet curing appeared to immobilize the resin, preventing migration during drying.

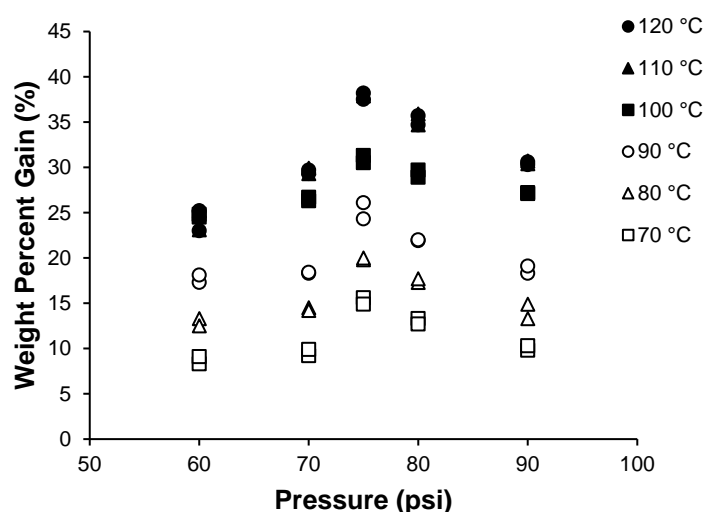
### *Catalysts*

Catalysts are commonly employed throughout the entire chemical modification, including impregnation process. Sodium acetate, zinc chloride, magnesium perchlorate, magnesium chloride hexahydrate, urea-ammonium sulphate, dimethylformamide, sodium acetate, magnesium persulfate, trifluoroacetic acid, boron trifluoride, and pyridine have been usually employed as catalysts in this process (Li *et al.* 2009; Rowell 2014; Hom *et al.* 2020). Catalysts are added during the process to shorten the reaction time while also enhancing the dimensional stability of wood. Li *et al.* (2009) reported that the application of various catalysts can significantly increase their WPG (15% to 27%) in comparison to uncatalyzed ones that were subjected to the same reaction conditions (5% to 16%). Because the catalysts were dissolved in acetic anhydride, they were able to produce a very effective catalytic effect on the acetylation process, which led to a higher rate of weight gain. In addition, by extending the reaction time during catalyzed acetylation, a WPG value of approximately 25% and an ASE value of more than 75% were attainable. In contrast, after an 8 h reaction at 75 °C, mechanical properties of acetylated wood catalyzed by zinc chloride and magnesium perchlorate decreased significantly, especially the bending strength by 23% to 29%. This could be due to zinc chloride, which is categorized as a strong Lewis acid and causes an extremely acidic reaction state. On the other hand, magnesium perchlorate is categorized as an oxidizer, which causes wood deterioration. Therefore, it could be affecting those mechanical properties compared with the untreated condition. Hom *et al.* (2020) reported that acetylation wood with pyridine as catalyst could obtain WPG value of about 27.4% to 32.9% and ASE around 55% to 64% after ten wetting cycles, as well as reduction in water absorption by 35%. Another case reported by Rowell and Elis (1978) involved an impregnation process using propylene oxide, butylene oxide, and acrylonitrile with triethylamine, sodium hydroxide and ammonium hydroxide as catalysts, which resulted in an increase of WPG (25.7% to 29.2%), but did not significantly attain dimensional stability to any great extent after seven wetting cycles. Thygesen *et al.* (2010), reported furfurylated wood produced with citric acid as the catalyst exhibited substantial fluorescence at a wavelength greater than ~ 450 nm. When the concentration of catalyst was at its highest level, there is an observed increase in the fluorescence intensity. Conjugation of poly-furfuryl acid (FA) is the primary sources fluorescence in furfurylated wood, and it exhibits a red-shift in landscapes as the degree of polymerization (DP) increases.

### *Pressure and Thermal Modification*

Thermal modification prior to impregnation process could accelerate its impregnation treatability. This could be due to the air within the cell cavities and voids being preheated during this process, causing it to become hot and swollen. The air within the cell cavities and voids rapidly contracts when the hot wood is immersed in room temperature impregnating agents, allowing the solution to be taken up into the wood void structure (Ahmed *et al.* 2017). Similar mechanisms known as physical impregnation, which are affected by different impregnating conditions (hot and cold), have also been reported by Paril *et al.* (2016). Impregnating agent may be able to penetrate wood cell as a result of a drastic change in temperature from hot to cold, which creates a partial vacuum (contraction). According to Ahmed *et al.* (2017), thermal modification prior to oil impregnation could give WPG around 67.6% to 267.1% for tung oil; 95.3% to 283.6% for pine tar; and 118.7% to 371.2% for beckers. Thermally modified wood along with oil

impregnation resulted in better dimensional stability with ASE around 10.8% to 69.5%. According to Ahmed *et al.* (2017) micro-pore blocking occurs when a hydrophobic compound is deposited in the cell lumens as a result of oil impregnation using water repellent compounds. The deposition occurs along the main capillary paths of the wood, thus limiting its ability to absorb water. Despite the fact that these mechanisms have been shown to improve its dimensional stability with higher WPG, it may cause morphological changes, such as vessel shrinkage, cell wall buckling, and disintegration of ray parenchyma that occur during thermal modification. In addition, mold growth may have been hastened due to the formation of soluble carbohydrates and the hydrolyzed hemicellulose into oligomeric and monomeric structure (soluble sugars) when subjected to extreme climates. Ahmed *et al.* (2017) reported that mold grades for thermally modified wood along with oil impregnation were around 3.0 to 4.5 for tung oil; 3.0 to 5.2 for pine tar; and 1.0 for beckers.



**Fig. 10.** Relation between weight percent gain and pressure in different thermal condition (Deka *et al.* 2000)

Thermal and/or heat modification after impregnation process also has been reported by several researchers. L'Hostis *et al.* (2017) reported that heat treatment after an impregnation process using citric acid (CA) and tartaric acid (TA) alone, and in combination with glycerol (G+CA and G+TA) could give WPG around 6.8% to 39.7% with ASE around 29.9% to 67.8%. A significant drop in WPG occurred as temperature was increased, which was pronounced at 160 °C. As can be seen, lower temperature applied results in lower WPG, whereas extend the temperature results in higher WPG (Fig. 10). L'Hostis *et al.* (2017) highlighted that increasing heat temperature improves fixation in wood cell walls, reduces mass loss against decay (*Coriolus versicolor*), and decreases leaching rates caused by grafting via ester linkage and/or the formation of an insoluble polyester. These phenomena were seen in both cases where CA and TA treatment was performed alone. However, the effect is accentuated in the scenario where glycerol is used in conjunction with other treatments (G+CA and G+TA). In addition, different amounts of pressure applied results in different loading of impregnating agents (WPG). As can be seen in Fig. 10, lower pressure applied results in lower WPG, then the values increase up to 75 psi of pressure, then starts to decrease at 80 psi of pressure.

### Compression Modification

The utilization of compression prior to impregnation process has received more attention due to the possibility for better wood treatability. This includes the possibility of more void cells being open or available, the movement of air or water out of the wood, and the creation of temporarily low pressure within the wood. All of these factors can result in a significant quantity of impregnating agents and high level of effectiveness in the impregnation process (Wang and Zhao 2022). In the early observation, studies on pre-compression combined with impregnation have been done with water (Iida *et al.* 1998, 2002; Zhao *et al.* 2018, 2019), as well as resin as impregnating agents (Wang and Zhao 2022). Research related to this modification have been carried out by several researchers, particularly regarding to effect of compression ratio, compression direction, compression speed, and compression-unloading places (Zhao *et al.* 2018), as well as different moisture content (Zhao *et al.* 2019) on impregnation process.

According to the findings of Zhao *et al.* (2018), pre-compression of woods followed by water impregnation leads to a remarkably increased of CU (impregnating uptake), as compression ratio (CR) is increased. The CU were 0.05 and 0.40 g/cm<sup>3</sup> for poplar, as well as 0.02 and 0.43 g/cm<sup>3</sup> for Chinese fir at CR 10% and 60%, respectively. A positive correlation was shown between CR and CU on poplar ( $y = 0.0065x - 0.0244$ ;  $R^2 = 0.98$ ) and Chinese fir ( $y = 0.0074x - 0.0446$ ;  $R^2 = 0.99$ ) wood species. As CR increases, there will be more space for possible impregnation, especially the void cavities (vessel and tracheid lumen) and more compression energy will be stored in wood. The process of the wood recovering itself in a very short time after being compressed and then submerged in water is referred to as spring-back. This phenomenon generates an increase in the wood cavity volume, which in turn leads to the transient drop in pressure within the wood cavity. Because of the pressure difference between outside and inside of the wood (atmosphere pressure and water pressure), the water was drawn into the wood cells (Zhao *et al.* 2018). In addition, impregnation of pre-compressed wood in radial directions at different speeds showed that CU at 0.5 and 1 mm/min of compression speed was nearly the same as those pre-compressed in 45° directions, but the values was higher at 3, 5, and 10 mm/min of compression speed (Zhao *et al.* 2018).

## WOOD PERMEABILITY

The permeability of wood is an essential factor, since it has a significant influence on its application, especially for its treatability which deals with impregnation process (Taghiyari 2012). Its permeability varies greatly due to their complex characteristics. A combination of factors, including position and morphology, respond to account for their variation. There is significant difference between families, genera, and species, even within the same species (Wen *et al.* 2014). Therefore, wood permeability between hardwood-softwood, sapwood-heartwood, and directions (*i.e.*, tangential, radial, and longitudinal), as well as the path of fluid/liquid flow during impregnation will be discussed in this section.

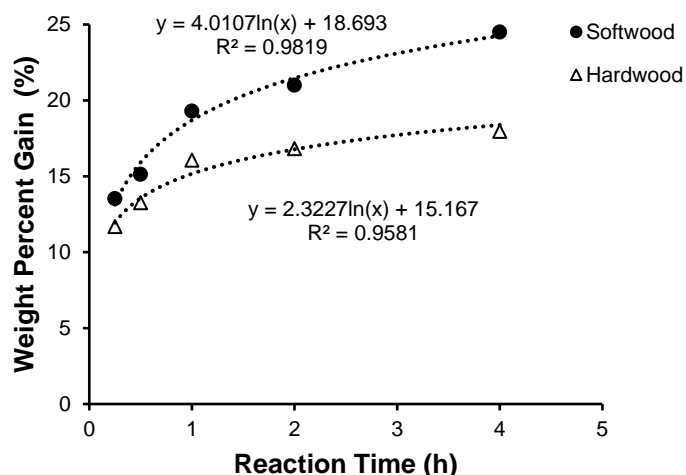
Many studies have been conducted on this matter, and the results of those investigations provide an overview of the liquid flow path during impregnation process. Rowell and Ibach (2018) compared the WPG of softwoods and hardwoods in terms of their respective reaction time while employing acetic anhydride (Table 6). According to the findings of the researchers, compared to hardwoods, softwood reacts more quickly and produces a higher level of bonded acetyl groups, which is observed as WPG (Fig. 11).



Softwood may have less pentosans in their hemicelluloses in comparison to hardwood. Pentosans may slow down the reaction time and reduce the amount of acetyl content (WPG) due to the lack of a primary hydroxyl group. Another reason that possibly causes the limitation of hardwood is due to their complex structure. Hardwood is composed of four principal cell types, including vessels, fibers, ray parenchyma, and axial parenchyma. These cell types are around 20% to 60%, 15% to 60%, 5% to 30%, and 1% to 24% of the volume, respectively. In contrast, softwood is composed of axial tracheid cells, ray parenchyma, and resin canals, with tracheid being the primary component and accounting for 90% to 95% of the total wood volume (Ding *et al.* 2008). Longer reaction time resulted in higher WPG for both softwood and hardwood. A positive correlation between WPG and reaction time in softwood and hardwood served as evidence for these findings. Wardrop and Davies (1961) stated that the path of penetration was essentially similar within hardwood and/or softwood species. Nevertheless, they found that there appeared to be another tissue system that assisted the liquid penetration.

**Table 6.** WPG of Softwood and Hardwoods Reacted with Acetic Anhydride

Parameter	Category	Wood Species	Reaction time of acetic anhydride					Authors
			0.25	0.5	1	2	4	
WPG	Softwood	<i>Pinus sylvestris</i>	13.7	15.4	19.5	20.7	24.9	Rowell and Ibach (2018)
		<i>Pinus radiata</i>	13.4	15.2	19.7	21.8	24.8	
		<i>Picea abies</i>	13.5	14.8	18.7	20.5	23.8	
	Hardwood	<i>Fagus sylvatica</i>	10.6	11.9	15.4	16.3	17.5	
		<i>Quercus rubra</i>	11.9	13.9	17.2	17.8	19.4	
		<i>Populus tremuloides</i>	12.6	14.0	15.6	16.4	17.0	



**Fig. 11.** Comparison between reaction time of softwood and hardwood on average of weight percent gain during impregnation process (Table 6)

Matsumura *et al.* (1999) studied sapwood and heartwood of radiata pine in relation to the impregnation process. They observed that liquid uptake of sapwood ( $0.6 \text{ g/cm}^3$ ) was higher than that of heartwood ( $0.1 \text{ g/cm}^3$ ). Taylor *et al.* (2002), Engelund *et al.* (2013), and Wen *et al.* (2014) reported that sapwood has a higher moisture content (MC) because it serves as the tree's main transpiration system, and generally has lower specific gravity indicates fewer cell wall materials, which is equivalent to a greater void volume and porosity. This phenomenon resulted in better treatability during impregnation process.

Higher MC can cause the wood cell cavity to enlarge, which facilitates the diffusion process of impregnating agent and speeds up the process for reaching the maximum impregnation level (Purba *et al.* 2014; Zhao *et al.* 2019). According to Uphill *et al.* (2014), the liquid pathways in the sapwood of Scots pine (*Pinus sylvestris*) move axially from one tracheid to the other through bordered pits and radially along the ray tracheid. Impregnation of heartwood is generally limited by pits clogged with extractives, as well as tyloses, various gummy, resinous and chalky exudates that also often formed in the vessel lumens within this portion (Wang and De Groot 1996, Lukmandaru and Takahashi 2009; Qiu *et al.* 2019).

Scholz *et al.* (2010b) investigated the effect of different anatomical directions on wax impregnation into the sapwood of European beech and pine. It has been found that the radial and tangential penetration of European beech has a shallower depth of penetration than that of pine sapwood. This could be due to the complex membrane structure of the parenchyma tissue that prevents liquids from passing through them. The radial direction of pine wood exhibited a higher penetration depth compared to that of tangential direction, but the reverse phenomenon occurred on European beech. Sernek *et al.* (1999), reported that the tangential direction of beech was shown to have a higher penetration than that in the radial direction. This could be due to beech containing a narrow latewood band with fewer and smaller vessels, as well as more pits, which prevents radial penetration. Moreover, air permeability of beech is almost 90 times higher in the tangential direction than that in the radial direction.

In addition, the presence of bordered pit aspiration on the tracheid walls, particularly in softwood species, will also affect their liquid permeability during the application. Bordered pits provide routes for communication between neighboring tracheid walls. As the wood's moisture content begins to fall, the pressure difference between tracheid starts to increase, leading to the development of bordered pits aspiration. This phenomenon occurred as an efficient response to counteract embolism of water-conducting in the sapwood portion (Lehringer *et al.* 2009). Matsumura *et al.* (2005) studied the profile of bordered pit aspiration in different portions of *Cryptomeria japonica*, particularly in green and air-dry condition. It was discovered that the amount of pit aspiration percentage (ASP) is initially lower in the sapwood portion, then it rises gradually in the intermediate portion, and then it increases significantly in the heartwood portion. This indicates that the aspiration of bordered pits starts to develop during heartwood formation in green condition. However, the value is higher in sapwood in an air-dry condition. This phenomenon was observed by Usta and Hale (2006). Wood samples in green condition were found to have higher longitudinal and tangential permeability than that of kiln drying condition. This happened due to higher open bordered pit and lower bordered pit aspiration percentage in green condition, while the inverse phenomenon occurred in kiln drying condition. Therefore, physical modification using cryogenic treatment *via* liquid nitrogen (Yorur and Kayahan 2018), mechanical opening using needles, drills, slit discs, laser and *etc.*, as well as biological modification using white rot fungus, bacterias, and enzymes have been done in order to limits the drawback and improve the capability of impregnation and penetration (Lehringer 2011).

## PROBLEMS AND FUTURE DEVELOPMENT TRENDS OF WOOD IMPREGNATION

Unfortunately, the majority of the methods for impregnation use hazardous or poisonous chemicals, which can raise environmental concerns either throughout the service life or at the end-life of wood products. Therefore, it would be important to mitigate the utilization of those chemicals by using environmentally friendly agents. In addition, the introduction of nanomaterials and the advancement of nanotechnology are also crucial for addressing the issues plaguing the conventional chemical modification process.

### Development of Environment-friendly Impregnating agents

The development of environmentally friendly and/or green technology has attracted increasing attention. Many studies have noted interest in investigating the use of renewable impregnating agents such as those produced from natural products, biomass-derived compounds, and biological-based chemical materials (Table 7). According to Dong *et al.* (2020), environmentally friendly processes, renewable modifying agents, non-toxic, and biodegradable resultant products are necessary for sustainable wood modification.

It is possible for environmentally friendly-impregnating agents made from renewable materials to replace synthetic compounds. Plant-based and biomass-based impregnating agents may play a significant role in green technologies due to their abundant sources. These compounds are mainly extracted from the vulnerable areas of a plant, *i.e.*, bark, heartwood, leaves, and seeds (Sommerauer *et al.* 2019), and sourced from agricultural/ biomass, *i.e.*, vegetables and fruits (Lee *et al.* 2020; Liu *et al.* 2020; Baar *et al.* 2021). Beside those compounds, lignin-based compounds could also become potential resources in terms of this matter. Lignin is the most abundant natural aromatic polymer in biomass cell walls. It is mainly produced through isolation of black liquor yielded from pulping processes (Mili *et al.* 2021). Most application attempts to use lignin as an impregnant involve substitution of petroleum-based raw materials by partially or completely changing the resin/adhesive formulation. Studies on lignin-based resin have been conducting recently, including lignin-phenol-formaldehyde (Pang *et al.* 2017; Rodrigues *et al.* 2023), lignin-polyurethane resin (Garcia 2018), and soy protein adhesive enhanced by biomass lignin (Pradyawong *et al.* 2017). The utilization of lignin is expected to ensure efficient reactivity by providing abundant phenolic hydroxyl groups and free aromatic C3 or C5 position, as well as the ability to undergo hydroxymethylation and polymerization by condensation with formaldehyde (Rodrigues *et al.* 2023). In addition, biological-based compounds used as impregnants have included by-products of biological activity, such as chitosan from crustacean shells (Larnøy *et al.* 2005; Huang *et al.* 2021), propolis extract produced by *Apis mellifera* (Woźniak *et al.* 2020), and beeswax. There is also a possible variation between those compounds due to differences in species of biological organisms, source of food especially for propolis and beeswax, and extraction method.

The development of impregnating agents using those compounds can be a good steppingstone for seizing a more environmentally friendly technology that can support long-term economic, ecological, and social growth. There is promising evidence for the use of renewable or ‘non-biocidal’ compounds in the impregnation process, but the related studies are still limited. To further assess the environmental behavior of treated wood itself and to better understand the mechanism and optimization of the process, more research is required to be done. In addition, issues related to the cost, processing conditions, and machinery also hinder efforts to bring the process for scaling up to industrial development.

**Table 7.** Summary of Environment Friendly-Impregnating Agents from Renewable Agents

Sources	Type of impregnating agents	Advantages	References
Plant-based compounds	Bio-oil	Substitution for petroleum-based phenolic compound; cost-competitive; environment friendly	Robinson <i>et al.</i> 2011; Liu <i>et al.</i> 2020; Baar <i>et al.</i> 2021
	Lignin-based resin	Bio-protection against biodegradation agents; higher proportion of hydroxy-methylation;	Mansouri <i>et al.</i> 2007; Chirkova <i>et al.</i> 2011;
	Extractives-based	Resistance of biodegradation agents; desirable change in vibrational properties (low $\tan \delta$ ), non-hazardous;	Matsunaga <i>et al.</i> 1999; Matsunaga <i>et al.</i> 2000; Malik <i>et al.</i> 2016; Balfas 2019; Malik and Ozarska 2019
	Tannin-based	High biological activity; phenolic rich biomolecules; water soluble; improve dimensional stability	Tondi <i>et al.</i> 2013; Sommerauer <i>et al.</i> 2019; Mubarak <i>et al.</i> 2022
Biomass-derived compounds	Suberin monomers	Natural aromatic-aliphatic crosslinked polyester; fully ‘green’ product; improve dimensional stability; reduce water vapor uptake	Gandini <i>et al.</i> 2006; Zhang and Ma 2021;
	Humin-based resin	Longer ignition time, slower heat release rate, lower; lower mass increase; enhance dimensional stability	Sangregorio <i>et al.</i> 2020
	Furfuryl alcohol	Environmentally friendly; improve physical-mechanical properties; enhance durability of wood	Lande <i>et al.</i> 2011; Li <i>et al.</i> 2016; Hadi <i>et al.</i> 2021; Mrtha <i>et al.</i> 2021; Sun <i>et al.</i> 2022
	Succinic acid	Renewable resources; improve dyeing ability, non-hazardous; substitution for petrochemical-succinic acid and maleic anhydride	Chang <i>et al.</i> 2016
	Sodium alginate	Biocompatibility, bioactive, cost-competitive; possesses a gelling ability	Zhang <i>et al.</i> 2022
	Itaconic acid	Improve UV resistance; substitution for maleic anhydride	Chang <i>et al.</i> 2016;
	Citric acid	Odourless, act as pH stabilizer, ‘green’ binder, cross-linking agent in improving dimensional stability as well as biological durability.	Lee <i>et al.</i> 2020
Biological-based compounds	Beeswax	Highly water repellent; low melting point, low density; enhance color stability, hydrophobicity and antibacterial properties	Li <i>et al.</i> 2020
	Propolis extract	Antifungal activity against mold and yeast; bio-friendly compound; substitution for petroleum-based phenolic compound	Woźniak <i>et al.</i> 2020
	Chitosan	Antifungal activity against forest pathogenic and decaying fungi; eco-friendly cross-linked	Larnøy <i>et al.</i> 2005; Huang <i>et al.</i> 2021

## Utilization of Nanomaterials Impregnating Agents

Since wood is considered a porous material, it can be easily impregnated with a wide variety of impregnating agents as long as those particle sizes fit or are even smaller than that of the wood pore's structure. The three distinct types of wood pores recognized by The International Union of Pure and Applied Chemistry (IUPAC) are macropores (pore size > 50 nm), mesopores (2 nm < pore size < 50 nm), and micropores (pore size < 2 nm). Yin *et al.* (2015) studied the comparison of changes in micropores and mesopores within wood cell walls of sapwood and heartwood. It was discovered that the liquid flow during impregnation process is mainly connected through vessels, tracheids, and rays (diameter range of 15 to 400 nm) in longitudinal direction, continued to flow in transversal direction by pits (diameter range of 0.4 to 30 nm), then finally reached the lumen of fibres through pores with nanometer scale diameter. In addition, the presence of bordered pit aspiration with different shapes (circular, slit-like, and blurred), and variation of pit chamber and aperture diameter added to the complexity of the process (Usta and Hale 2006). Those structures may act as barriers, since larger particles may give rise to clogging of those pathways during impregnation.

The introduction of nanomaterials and the advancement of nanotechnology are crucial for addressing the issues plaguing the conventional chemical modification process, such as insufficient impregnation and high side effect rate. Applying a nanotechnology-based treatment to the wood cell wall has the potential to enhance wood quality. Nanomaterials, a relatively new kind of modification, are ideally suited to the current trend in wood modification technology due to their strong functionality and effective impregnation effect.

Studies on the application of nanomaterials in wood impregnation are primarily concerned with the physical-mechanical properties, as well as biodegradation resistance (Papadopoulos *et al.* 2019). The key advantages of using nanomaterials include the ability of such materials to penetrate deeper into the wood structure, impregnate more thoroughly, distribute more evenly, and achieve better modification effects compared to the traditional modification methods that involve solids. This may be attributed to its smaller structure and higher surface area to volume ratio. Metal nanoparticles, including copper, gold, titanium, and silver are commonly used due to their outstanding and stable impregnation effect. Apart from synthetic man-made, copper nanoparticles can be extracted naturally from leaves of *Azadirachta indica*, *Pongamia pinnata*, *Lantana camara*, and betung bamboo, as well as fruits such as *Citrus reticulata* (Shiny *et al.* 2019; Rahayu *et al.* 2021). Some studies have been using this sort of impregnating agent, combined with heat treatment (Taghiyari 2011; Dehghanian *et al.* 2020) and densification process to reduce spring-back (Rassam *et al.* 2012). Metal nanoparticles, particularly nano silver, have the potential to form bonds between Ag ions and some functional group of wood components, resulting in dimensional stability enhancement (Rassam *et al.* 2012). Moreover, nano-silica is frequently employed as an impregnating agent due to its affordable price, availability of raw materials, and good modification effect (Bi *et al.* 2021). This type of impregnating agent has excellent water-repellent performance (superhydrophobic), which improves dimensional stability (Bak *et al.* 2018; Rahayu *et al.* 2019; Zhao *et al.* 2021). Other research has combined silica nanoparticles with bonding agents (Bak *et al.* 2018) to enhance bonding properties, as well as monoethylene glycol (MEG) of 0.5 and 1% concentration (Rahayu *et al.* 2019), and nano-CuO with various post-treatment (Zhao *et al.* 2021). Most silica nanoparticles are distributed across cell wall surfaces; however, some deposits and agglomerations can also be seen (Bak *et al.* 2018; Rahayu *et al.* 2019).

## CONCLUSIONS

Impregnation of solid wood can bring about a desired property, especially dimensional stability. This process can use several impregnating agents that act as bulking and crosslinking agents. These types of impregnating agents can be grouped into three classes based upon the bonding efficiency of the impregnating agent to and from the wood cells, namely nonbonded-leachable (NBL), including wax and oil, as well as monomer/polymer; nonbonded-nonleachable (NBNL), which is represented by thermosetting resin; and bonded-nonleachable (BNL), including anhydride and organic acid. The utilization of impregnating agents to enhance dimensional stability is a function of WPG. The impregnating agents in the first and second class gain higher WPG than that of the third class. However, the increasing of WPG in impregnated wood is not always correlated with an increase of wood properties, especially dimensional stability. At extremely high WPG, the ASE can be expected to drop to some extent due to small cracks in the tracheid walls, microcracks in the cell wall layers, major cracks within cells, and even cell wall damage and severe cell wall splitting. Therefore, the utilization of BNL class can be a good choice to produce less weight gain with good dimensional stability, as well as reduce the possibility of cell wall damage after treatment.

Impregnation has limitations. Proper impregnation parameters for each impregnating agent, different methods prior and/or after impregnation process, including the use of catalyst, pressure and thermal, as well as compression should be done in order to accelerate the impregnation process. Types and conditions of wood are undoubtedly essential for predicting impregnation treatability. A combination of factors, including position and morphology, respond to account for their variation.

The emergence of environmental issues has prompted the development of environment friendly impregnating agents as substitution of synthetic man-made. Exploration of renewable impregnating agents, including plant-based and biomass-derived compounds, as well as biological-based chemical materials, has high potential due to their availability in nature and modification effects. In addition, the development of nanotechnology is important in order to solve the current problems of existing traditional impregnation processes.

These approaches serve as a steppingstone for seizing a more environmentally friendly technology that can support long-term economic, ecological, and social growth. To further assess the environmental behavior of treated wood itself and to better understand the mechanism and optimization of the process, more research is required to be done. In addition, issues related to the cost, processing condition, and machinery also hinder efforts to bring the process for scaling up to industrial development.

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