American University in Cairo AUC Knowledge Fountain

Theses and Dissertations

Student Research

Summer 6-15-2021

## Fabrication and Characterization of Translucent Wood Composite Using Egyptian Hardwoods: Casuarina & Eucalyptus

Sherif Elia The American University in Cairo, sherif88@aucegypt.edu

Follow this and additional works at: https://fount.aucegypt.edu/etds

Part of the Environmental Engineering Commons

#### **Recommended Citation**

#### **APA** Citation

Elia, S. (2021). *Fabrication and Characterization of Translucent Wood Composite Using Egyptian Hardwoods: Casuarina & Eucalyptus* [Master's Thesis, the American University in Cairo]. AUC Knowledge Fountain.

https://fount.aucegypt.edu/etds/1598

#### MLA Citation

Elia, Sherif. Fabrication and Characterization of Translucent Wood Composite Using Egyptian Hardwoods: Casuarina & Eucalyptus. 2021. American University in Cairo, Master's Thesis. AUC Knowledge Fountain. https://fount.aucegypt.edu/etds/1598

This Master's Thesis is brought to you for free and open access by the Student Research at AUC Knowledge Fountain. It has been accepted for inclusion in Theses and Dissertations by an authorized administrator of AUC Knowledge Fountain. For more information, please contact thesisadmin@aucegypt.edu.

# Chool of Sciences and Engineering

Fabrication and Characterization of Translucent Wood Composite using Egyptian Hardwoods: Casuarina & Eucalyptus

A thesis submitted in partial fulfillment of the requirements for the degree of Master of Science in Environmental Engineering

### By

Sherif Hani Zaher Elia

Under Supervision of:

Dr. Salah El-Haggar Professor, Department of Mechanical Engineering, The American University in Cairo Dr. Khaled Nassar Professor, Department of Construction Engineering, The American University in Cairo

January 2021

### Abstract

Due to climate change and global warming, there is an increasing need for sustainable solutions in all industries. As one of the most important and growing industries worldwide, the construction industry has a huge responsibility towards the environment. It is responsible for the consumption of vast amounts of resources and consequently generating massive quantities of waste. Also, as the building sector is responsible for consuming large portions of the world energy and electricity, such buildings' efficiency and sustainability must be considered. While the world is working on balancing between consuming less and recycling more while maintaining the construction materials' effectiveness and efficiency, there is an immense need to develop more sustainable building materials that can increase buildings' efficiency while decreasing the effects on the environment.

Thus, introducing transparent wood composite, which is basically fabricated using delignified wood pieces or fibers with a polymer, can be used in many different applications due to its advantages over other materials [1]. Due to its better thermal insulation compared to regular glass, it can be used in buildings' cladding to reduce its carbon footprint through proper insulation and adequate natural lighting. Accordingly, decreasing the energy and electricity required to ventilate and temperature control as well as the need for natural lighting. Consequently, saving tons of GHG emissions, which significantly contribute to global warming and air pollution and help preserve our environment. Also, it has high potential in solar energy due to its "high optical haze". Finally, on top of the benefits mentioned above, transparent wood is considered a sustainable material, to replace plastics and other materials, due to its better biodegradability [2]. The aim of this study is to fabricate transparent wood using Egyptian hardwoods (Casuarina & Eucalyptus) and optimize the fabrication process. First, a pilot study was conducted to design the experimental setup; then, three chemical treatment recipes were used to determine the most effective method.

Moreover, the lignin modification method was selected for an in-depth study for lignin quantification using Klason lignin for both wood species. Since Casuarina had less lignin content (to begin with (29% compared to 35% for Eucalyptus), it required little to no further treatment after the end of the first treatment process. Consequently, Casuarina wood was selected for the fabrication of transparent wood using the lignin modification method showing good optical properties with 72% transmittance and 95% haziness. Also, TWS was fabricated from Casuarina sawdust using the lignin modification method and showed better overall high optical properties with transmittance around 80% and haziness of 85%. Therefore, a scalable treatment process for sawdust was tried using commercial bleach to introduce a more feasible method. Finally, the optical properties for the TW and TWS were compared to three types of glass (regular, sanded, and rain glass) as an alternative material in which the TWS showed comparable light transmittance compared to regular and rain glass with 85% and 82% respectively. At the same time, TW had similar results to the sanded glass, which had around 72% light transmittance. On the other hand, TW had the highest optical haze with 95%, followed by TWS and sanded glass with 77% showing a better overall behavior for TW and TWS over the glass.

### Acknowledgments

I would like to express my gratitude to all the people who helped me with this research project, and without whom I would not have been able to complete my master's degree. First, I would like to thank my advisors, Dr Salah El Haggar and Dr Khaled Nassar, for their constant support, encouragement, and patience. Also, I would like to thank Dr Walid Fouad, associate professor in the biology department for helping me with the lignin quantification and Dr Nageh Allam, a professor in the physics department for allowing me to use his lab facilities to obtain the optical properties for my samples. I would like to thank Eng. Ahmed Saad, environmental engineering lab engineer and Mohamed Mostafa, the lab aide and Mohamed Saeed, waste management lab instructor for their help and guidance during my lab work. Moreover, I would like to thank Eng. Usama Omar, senior lab supervisor in the mechanical engineering and assembling the polymer infiltration setup (mold, vacuum chamber, and pump).

Furthermore, I would like to thank all my family, my father, my mother, my brother, and my sister for their endless support and without whom I would not have been able to make it through my master's.

I am also grateful for all the help I received during my lab work from my friends especially Sarah Abdel Kader for helping me with my experimental setup, Amal Aboelfotouh for helping me with the lignin quantification, and Manar El Naggar for the helping me with the chemical treatment. I would also like to thank Mohamed Osman for his help in designing the polymer infiltration setup and Yousof Abdel Raouf for his constant support and insights for my research. Also, I would like to thank Eng. Mohamed Ramadan for his help in taking the time to test the optical properties of my material and without whom I would not have finished on time.

Finally, I would like to thank my colleagues Monira Yazeed, Alaa Abbas, Esraa El-Mitainy, Mirette Khorshed, and Lana Mahmoud, for their help and support through my master's degree.

# List of Abbreviations

ATR	Attenuated Total Reflectance
CB	Commercial Bleach
CDW	Construction and Demolition Waste
C&I	Commercial & Industrial
$C_2H_3NaO_2$	Sodium Acetate
CH <sub>3</sub> COOH	Acetic Acid
$C_4H_8O$	Tetrahydrofuran
CLT	Cross Laminated Timber
$CO_2$	Carbon Dioxide
DI	Deionized Water
DL	Delignification Method
DRIFT	Diffuse Reflectance Infrared Fourier Transform
DTPA	Diethylenetriaminepentaacetic Acid
EDTA	Ethylenediaminetetraacetic Acid
FTIR	Fourier Transform Infrared
GHG	Greenhouse Gases
Glulam	Glued-laminated Timber
h	Hour
H2O2	Hydrogen Peroxide
H2SO4	Sulfuric Acid
HB	Hardboard
HVAC	Heating, Ventilation, and Air Conditioning
LM	Lignin Modification Method
LSL	Laminated Strand Lumber
LVL	Laminated Veneer Lumber
М	Moles
MSW	Municipal Solid Waste
MDF	Medium Density Fiberboard
MF	Melamine Formaldehyde
MgSO <sub>4</sub>	Magnesium Sulfate
Na <sub>2</sub> SiO <sub>3</sub>	Sodium Silicate

$Na_2SO_3$	Sodium Sulfite
NaOH	Sodium Hydroxide
NaClO <sub>2</sub>	Sodium Chlorite
OSB	Oriented Strand Board
PB	Particleboard
PEG	Polyethylene glycol
PF	Phenol Formaldehyde
PMMA	Poly (methyl methacrylate)
PSL	Parallel Strand Lumber
SCL	Structural Composite Lumber
THF	Tetrahydrofuran
TW	Transparent Wood/Translucent Wood
TWF	Transparent Wood Fiber
TWS	Transparent Wood Sawdust
UF	Urea-formaldehyde

# **Table of Contents**

A	bstract	: ti
A	cknowl	ledgmentsiv
L	ist of Al	bbreviations
1	. Cha	pter 1: Introduction 1
	1.1.	Construction Industry Development1
	1.2.	Buildings Energy Consumption & Emissions2
	1.3.	Energy Efficient / Green Buildings5
	1.4.	Construction & Wood Waste8
	1.5.	Wood Composites/Engineered Wood11
	1.5.1	. Wood and Natural Fiber Composites11
	1.5.2	. Natural Fiber and Plastic Composites15
	1.6.	Wood Waste Opportunities17
2	. Cha	pter 2: Literature Review18
	2.1.	Wood Types, Structure & Properties18
	2.2.	Lignin Removal & Quantification22
	2.2.1	. Lignin Extraction22
	2.2.2	. Lignin Quantification Methods23
	2.3.	Transparent Wood Composite24
	2.4.	Transparent Wood Fabrication25
	2.4.1	. Wood Species and Sizes26
	2.4.2	Chemical Treatment26
	2.4.3	. Polymer Infiltration
	2.5.	Problem Statement
	2.6.	Objectives
3	. Cha	pter 3: Materials and Methods40
	3.1.	Materials40
	3.2.	Methods40
	3.2.2	. Pilot Study – Chemical Treatment

3.2.	3. Phase I: Performance of Chemical Treatment Methods	45
3.2.	4. Phase II: Lignin Modification Method – Lignin Quantification	46
3.2.	5. Phase III: Transparent Wood from Wood Waste	48
3.2.	6. Phase IV: Casuarina Transparent Wood & Glass	49
3.2.	7. Phase V: Sawdust Treatment using Commercial Bleach	52
3.3.	Characterization & Testing	53
3.3.	1. Lignin Quantification - Klason Lignin	53
3.3.	2. Optical Properties	56
4. Ch	apter 4: Results & Discussion	57
4.1.	Pilot Study Trials & Observations	57
4.1.	1. Chemical Treatment Observations Pre-trials	57
4.1.	2. Plastic Waste Dissolving	59
4.2.	Performance of Chemical Treatment Methods	60
4.3.	Lignin Quantification (Lignin Modification Method)	64
4.4.	Transparent Wood (TW) & Transparent Wood Fibers (TW	/F)66
4.5.	TW – Transparent Wood (TW) & Glass	69
4.6.	Sawdust Treatment using Commercial Bleach	72
5. Ch	apter 5: Conclusion and Recommendations	
5.1.	Conclusion	74
5.2.	Recommendations	76
Referen	ICES	77
Append	ix	

# List of Tables

Table 1: Adhesive classification by structural integrity [19]	15
Table 2: Sodium hydroxide & sodium sulfite recipe journals summary	28
Table 3: Sodium chlorite recipe journals summary	31
Table 4: Lignin retaining recipe journals summary	33
Table 5: Acylated transparent wood journals summary	34
Table 6: Hydrogen peroxide recipe journals summary	36
Table 7: Sodium hypochlorite recipe journals summary	37
Table 8: Casuarina & Eucalyptus lignin quantification results at different intervals	64

# **List of Figures**

Figure 1: World GHG emissions by sector in 2016 showing electricity and heat to be
the biggest contributor in GHG emissions [5]2
Figure 2: EU Annual Energy Consumption by Country in 2009 per floor area [8]4
Figure 3: Breakdown of energy consumption for residential and commercial buildings
in the US [9]5
Figure 4: The percentage of total savings in energy consumption in the EU by country
vs. new households savings showing the effect of retrofitting existing buildings on
energy savings [8]6
Figure 5: The energy consumption in the EU by country for heating per $m^2$ of floor area
in tear 1997 vs 2009 in kilograms of oil equivalent (koe) [8]6
Figure 6: Energy consumption for lighting in EU per country in years 1990, 2002,
&2009 [8]7
Figure 7: Percentage of energy consumption in the EU by country for the commercial
sector [8]7
Figure 8: Construction and demolition waste characterization by weight percentage in
the US showing the wood waste with the highest percentage of 31% [15]9
Figure 9: Wood waste generation percentage per sector in the UK [17]10
Figure 10: Breakdown of wood uses generated from construction and demolition waste
[15]11
Figure 11: The classification of natural fiber composites by raw materials input, length
of major wood element, and density [19]12
Figure 12: General schematic for wood composites manufacturing process [19]13
Figure 13: Laminated veneer lumber (LVL) manufacturing process schematic [19] .14
Figure 14: Classification of petrochemical polymers based on the molecular bonding
property [19]
Figure 15: Wood-plastic composites manufacturing schematics using single or double
screws with different orientations for the setup [19]16
Figure 16: Tree Trunk Cross-section(left) & Growth Ring Cells [23]19
Figure 17: Cross-section of softwood (left) & hardwood (Right) showing vessel size
comparison [23]19

Figure 18: Typical wood cell wall structure showing the layers starting from the middle
lamella, primary wall, and the secondary wall layers & the presence of lignin and
holocellulose in the cell [19]
Figure 19: Eucalyptus tree (left) & Casuarina tree (Right)21
Figure 20: Radial samples vs. longitudinal samples vessels orientation [2]26
Figure 21: TW chemical treatment & fabrication diagram showing the chemical process
setup (left) and the polymer infiltration steps (right) for wood pieces and wood fibers
[2], [51]
Figure 22: Transparent wood samples transparency directly on paper and 5mm above
[49]
Figure 23: Hydrogen peroxide suspended treatment setup [50]
Figure 24: Fabrication process diagram for H <sub>2</sub> O <sub>2</sub> treatment for radially cut wood and
veneer [50]
Figure 25: General fabrication process stages for transparent wood40
Figure 26: Casuarina sample (Left) & Eucalyptus sample (Right)41
Figure 27: Casuarina wood chips & sawdust samples41
Figure 28: Chemical treatment experimental setup using flask and condenser & beakers
on hot plate42
Figure 29: Dehydration setup using vacuum filter container43
Figure 30: Vacuum infiltration setup using vacuum pump and a desiccator43
Figure 31: Polymer infiltration modified setup with a bigger vacuum pump50
Figure 32: Epoxy samples & regular, sanded, and rain glass samples50
Figure 33: Translucent wood fabrication schematics for wood fibers/sawdust (upper
schematic) and radially cut wood (lower schematic) showing the stages from sample
preparation, chemical treatment, and polymer infiltration
Figure 34: Casuarina sawdust treatment using commercial bleach with 3 different
sample to solution ratios
Figure 35: Klason lignin experimental stages starting with sample grinding and
treatment with sulfuric acid to filtration, washing, and drying to constant weight54
Figure 36: Samples (Left) – inside furnace & (Right) - after combustion55
Figure 37: Haze configurations setup using UV spectrophotometer with an integrated
sphere [1]

Figure 38: Treated samples using delignification method, lignin modification method,
and commercial bleach method for Casuarina and Eucalyptus species compared to the
original samples after 6 h of treatment57
Figure 39: Lignin modification samples after 6 h of treatment (Left) & Delignification
method after 6 h (Right) of the second treatment process
Figure 40: Treated samples using delignification method, lignin modification method,
and commercial bleach method for Casuarina and Eucalyptus species compared to the
original samples after 12 h of treatment
Figure 41: Lignin modification samples after 6 h of treatment(left) and 12 h (middle)
& commercial bleach after 30 h of treatment (right) for Casuarina and Eucalyptus
species
Figure 42: Plastic waste mixed in different concentrations of Tetrahydrofuran (THF)
Figure 43: Commercial bleach treatment for Casuarina and Eucalyptus at different
intervals showing the progress of treatment
Figure 44: Delignification treatment for Casuarina and Eucalyptus at different intervals
showing the progress of treatment
Figure 45: Lignin modification treatment for Casuarina and Eucalyptus at different
intervals showing the progress of treatment
Figure 46: FTIR spectrums for untreated Casuarina wood, commercial bleached
sample, lignin modified sample, and delignified sample after 12 h of treatment61
Figure 47: FTIR spectrums for untreated Casuarina wood, commercial bleached
sample, lignin modified sample, and delignified sample at the end of treatment63
Figure 48: FTIR spectrums for untreated Eucalyptus wood, commercial bleached
sample, lignin modified sample, and delignified sample after 12 h of treatment63
Figure 49: FTIR spectrums for untreated Eucalyptus wood, commercial bleached
sample, lignin modified sample, and delignified sample at the end of treatment63
Figure 50: Lignin content for Casuarina & Eucalyptus (using Klason Lignin) during the
treatment process using lignin modification method for the first process (upper) and
second process (lower)65
Figure 51: Transparent wood sawdust samples & wood fibers after polymer infiltration

Figure 52: Epoxy sample (left), TW Casuarina using lignin modification method and epoxy (middle), & TW Sawdust using lignin modification method and epoxy (right) Figure 53: Light transmittance of TW & TWS & Epoxy samples measured using UV Figure 54: Haziness of TWS & Epoxy samples measured using UV spectrophotometer Figure 56: Visibility (transparency) of TW, epoxy, and glass samples compared ......69 Figure 57: Light transmittance of TW, TWS, epoxy, casuarina wood, and glass samples measured using UV spectrophotometer with an integrated sphere......70 Figure 58: Haziness of TW, epoxy, and glass samples measured using UV spectrophotometer with an integrated sphere ......71 Figure 59: Visual comparison for sawdust treatment using Commercial Bleach at different time intervals for three waste to solution ratios 1:20, 1:40, and 1:60 (from left to right in each picture)......72 Figure 60: Treated sawdust after washing at 24, 48, and 72 h for three waste to solution Figure 61: Color comparison for treated sawdust using commercial bleach different waste to solution ratios (1:20, 1:40, and 1:60) at 48 & 72 h of treatment ......73 Figure 62: FTIR spectrums for untreated Casuarina sawdust and commercially bleached sawdust with different waste to solution ratios (1:20, 1:40, and 1:60) at 24 h of treatment. At 48 and 72 h showed the same results with no lignin presence (Figure 63 & Figure 64)......73 Figure 63: FTIR spectrums for untreated Casuarina sawdust and commercially bleached sawdust with different waste to solution ratios (1:20, 1:40, and 1:60) at 48h Figure 64: FTIR spectrums for untreated Casuarina sawdust and commercially bleached sawdust with different waste to solution ratios (1:20, 1:40, and 1:60) at 72h 

### **Chapter 1: Introduction**

#### **1.1.Construction Industry Development**

The construction industry is a growing industry all over the world. The European Union (EU) estimates that the construction industry represents around 28% of its industry sector and about 7% of its economy. As one of the largest sectors, it consumes more resources and energy than any other sector [3]. Consequently, as it is responsible for consuming vast amounts of resources, it generates a lot of waste. Since the world has been facing many challenges due to environmental and climate changes, switching to green alternatives has become necessary. Thus, sustainability is being considered throughout the different stages of building construction, starting from building material selection, buildings operations, and waste disposal.

To begin with, the selection of sustainable building materials is an increasing need all over the world to lessen the effects on the environment from the manufacturing process and waste disposal. Some of the building materials' manufacturing process is energy-intensive, such as cement, steel, and glass. On the other hand, the materials used significantly affect the carbon footprint of buildings. For instance, if the façade cladding for office and commercial buildings is not properly insulated, it would significantly affect the HVAC operation to maintain the appropriate room temperature. Thus, significantly increasing the building's energy consumption and the operational costs and consequently, the environmental impact.

Moreover, tons of construction and demolition waste (CDW) is being produced every year. In 2009, CDW compromised around 25 to 30% of the total generated waste in the EU, over 500 million tons [3]. Although some of these wastes are recycled, massive amounts of waste are discarded, which puts a considerable burden on the environment and the countries' economies. In conclusion, the construction industry contributes greatly to the depletion of resources while overloading landfills with CDW and increasing its contamination. Also, increasing the energy consumption for the manufacturing and transporting of materials besides the buildings' operational needs. Thus, immensely contributing to global warming, which creates health problems due to extreme weathers and air pollution [4].

#### 1.2. Buildings Energy Consumption & Emissions

According to the World Bank, it is estimated that more than half of the global carbon dioxide (CO2) emissions attributed to electricity and heat production. The contribution of each sector's emissions is as follows; 49% electricity and heating, 20% transport, 20% manufacturing industries and construction, 9% residential buildings and commercial & public services, and around 2% from other sectors [5]. Since only around 22% of the global electricity production comes from renewable sources, vast amounts of greenhouse gas (GHG) are emitted every year [6]. To put things in perspective, the GHG emissions in 2016 for electricity & heating, buildings, and waste was estimated to be more than 19 billion tons out of a total of 49 billion tons emitted in the same year. In other words, the construction industry and buildings operation could be directly and indirectly responsible for a large percentage of the 40% of GHG emissions worldwide [5]. Figure 1 shows the world GHG emissions for each sector in 2016.

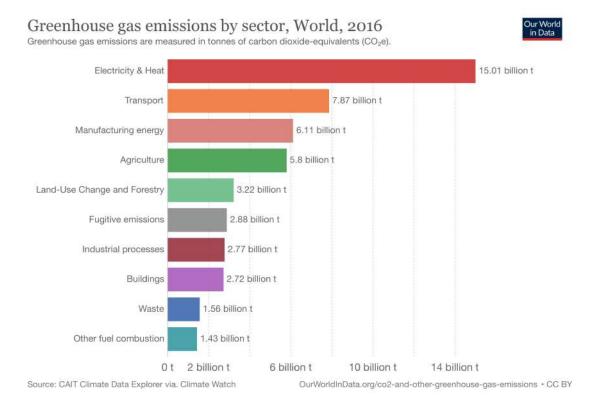


Figure 1: World GHG emissions by sector in 2016 showing electricity and heat to be the biggest contributor in GHG emissions [5]

The three top annual contributors for  $CO_2$  emissions are China, US, and EU28 (28 states in the European Union) [7]. The annual share for the top three is 27%, 15%, and 9.8% respectively. However, if the EU countries were considered separately, the top three contributors would be China, the US, and India, with 27%, 15%, and 6.8%

respectively. When taking the population into account, the US has 16.24 tonnes of CO2 emissions per capita per year, which is the highest followed by Japan and China with 9.45 and 6.98 tons of CO<sub>2</sub> emissions per capita per year. The total CO<sub>2</sub> emissions in 2017 were more than 36 billion tons compared to 2 billion tones in 1990 [5]. Due to this rapid increase in CO<sub>2</sub> emissions in the atmosphere during the twentieth century, there has been a spike in the earth's temperature, causing catastrophic events. Accordingly, the world's efforts have been working on various ways to lessen the emissions and protect the environment. The energy sector is considered one of the most polluting sectors, responsible for approximately half of the CO<sub>2</sub> emissions [5]. In Europe, around 40% of energy consumption is attributed to the buildings sector [8]. The energy consumption depends mainly on the type of building and the activities that take place in it. The EU estimates buildings' total floor area to be around 24 billion m<sup>2</sup> with residential buildings accounting for around 75% and 25% are the non-residential buildings [8]. Non-residential buildings include retail & wholesale trade buildings, office buildings, education, hotels, restaurants, hospitals, sports facilities, and others. Retail and office buildings combined represent more than 50% of the total floor area of the non-residential with 28% and 25% respectively. They are followed by educational buildings accounting for 20% while hotels, restaurants, hospitals, and sports facilities combined accounting for around 22%. Residential buildings annual energy consumption is 200 kWh/m<sup>2</sup> of floor area while the non-residential is around 295 kWh/m<sup>2</sup>, with an average of 220 kWh/m<sup>2</sup>. Obviously, the energy consumption is different in each country in the EU with hotter climate countries (e.g. Spain) consuming less compared to colder climate countries (e.g. Finland); illustrated in Figure 2 [8]. This is because heating is the primary energy use in cold countries, accounting for around 67% of households' total energy consumption. Regarding the non-residential buildings, around 50% of the energy consumption is attributed to the trade (retails & wholesales) and business (offices) sectors. Moreover, on average, lighting energy consumption is about 20% [8].

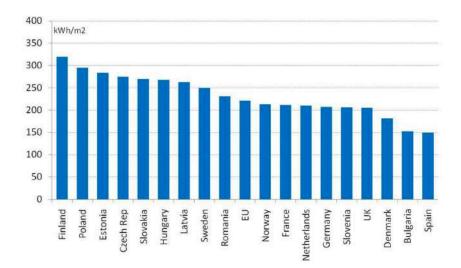


Figure 2: EU Annual Energy Consumption by Country in 2009 per floor area [8]

Similarly, the US commercial and residential buildings consume around 39% of the energy end-use, with residential buildings accounting for 21% and commercial buildings for 19%. More than half of the energy consumption for residential and commercial buildings goes to the HVAC & illumination, in which thermal comfort (heating & cooling) accounts for around 36% while lighting accounts for 18% [9]. Figure 3 shows the energy use in the US for industry, transportation, and buildings sectors. Moreover, it illustrates the percentage of building types and energy consumption for each activity. Comparing different developed countries for residential energy use, the energy consumption of heating is the largest. For instance, some developed countries' energy consumption, such as US, Canada, Germany, UK, France, and Japan, for the residential building's energy required for heating is relatively high compared to other demands [9]. The most energy-consuming commercial buildings in the US are retail, offices, and hotels, with 32%, 18%, & 14% respectively, compared to 22%, 33%, 30% in Spain and 22%, 17%, & 16% in the UK [10]. On average, the US's energy consumption for such commercial buildings is 280 kWh/m2 annually, which is relatively higher compared to residential buildings with around 150 kWh/m2 annually. Taking an in-depth analysis for office buildings energy consumption, the HVAC consumes 48% while the lighting consumes 22% compared to 55% & 17% for the UK and 52% & 33% in Spain [10]. This means that in office buildings, HVAC & lighting accounts for more than 70% of the energy consumption. Thus, concluding that HVAC and lighting are the most influencing factors in energy consumption in the commercial and residential buildings.

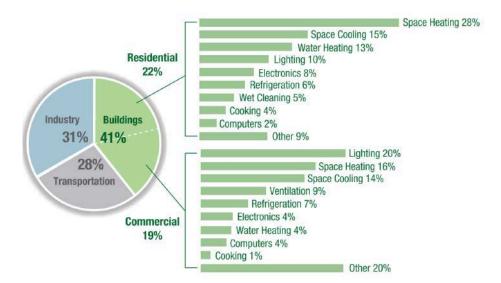


Figure 3: Breakdown of energy consumption for residential and commercial buildings in the US [9]

According to the European Commission report in 2019, the total global emissions for fossil CO<sub>2</sub> exceeded 35 gigatons since 2012. The power industry and buildings CO<sub>2</sub> emissions are more than 50% of the total global CO<sub>2</sub> emissions, with around 20 gigatons in 2018. Based on the report, the power industry and buildings are also responsible for around half of the fossil CO<sub>2</sub> emissions in Egypt, with total emissions of more than 320 megatons of GHG out of which 238 megatons were CO<sub>2</sub> emissions [7].

#### 1.3. Energy Efficient / Green Buildings

In light of the increasing demand for energy consumption as well as GHG emissions, various efficiency trends have been adopted around the world. Various building codes have been introduced to certify buildings as green or energy efficient in order to meet countries' sustainability goals. Building certifications and codes, such as Leadership in Energy and Environmental Design (LEED), Building Research Establishment Environmental Assessment Method (BREEAM), Energy Efficiency Building Code (EEBC), and Tarsheed & Green Pyramid in Egypt have become crucial tools in the fight for sustainability [11]. These codes aim to give inclusive guidelines for more sustainable buildings throughout its lifecycle, starting from construction to operations efficiency. Some of the categories included in the codes are regarding water efficiency, energy efficiency, CO<sub>2</sub> emissions, resources impact on the environment, and indoor air quality. Each code has different certification levels depending on the final scoring of the building from each of the required categories. These guidelines are

applicable in newly constructed buildings and existing buildings as well. Over the course of 12 years in the EU, due to the efficient methods adopted, the energy consumption per  $m^2$  for households decreased by around 15%. In the period of 20 years, the EU has upgraded the standards for new buildings, which resulted in decreasing the consumption per  $m^2$  by approximately 40% [8]. However, unfortunately, strict standards for new buildings cannot solve the problem alone without considering existing ones. Since it depends on the percentage of new developments in each country to affect the decrease in overall consumption, targets cannot be met without modifying existing buildings. That is why countries have been issuing regulations for existing building retrofitting in order to be able to reach their set targets. Figure 4 shows the contribution of new households in decreasing energy consumption vs the total saving for the period of 19 years (from 1990 to 2009) [8].

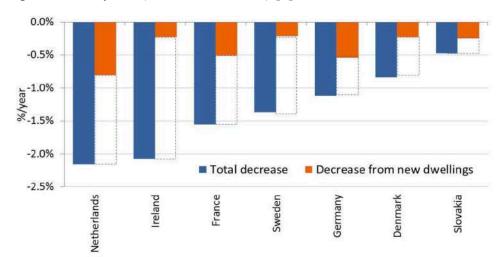
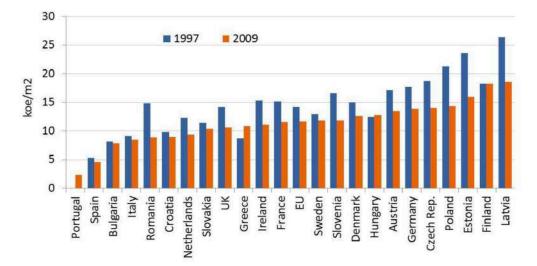


Figure 4: The percentage of total savings in energy consumption in the EU by country vs. new households savings showing the effect of retrofitting existing buildings on energy savings [8]



*Figure 5: The energy consumption in the EU by country for heating per m<sup>2</sup> of floor area in tear 1997 vs 2009 in kilograms of oil equivalent (koe)* [8]

On the other hand, due to global warming, air conditioning in hotter countries in the EU has increased in the past years. Southern countries in the EU, such as Croatia, Bulgaria, Malta, and Cyprus, are among the high energy-consuming countries for air conditioning. It was estimated that the average household in such countries consumes energy for air conditioning ranging from 450 to 650 kWh in 2009 [8]. Furthermore, lighting is a significant source of energy consumption in households that should be considered. The average EU consumption per household for lighting is 450 kWh per year ranging from 200 kWh for Slovakia to 900 kWh for Sweden [8].

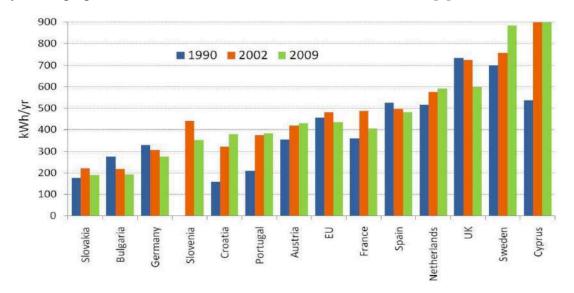


Figure 6: Energy consumption for lighting in EU per country in years 1990, 2002, & 2009 [8]

Regarding commercial building, in some countries, such as Germany and France, space heating consumption represents an average of 65% of the total energy consumption, with trading and office buildings as the most significant [8].

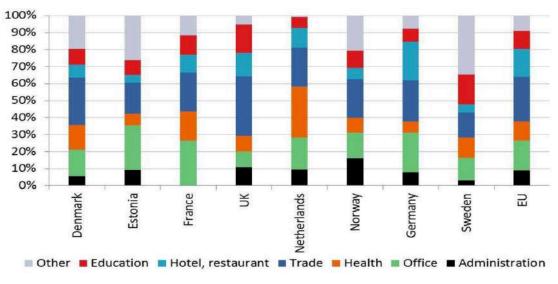


Figure 7: Percentage of energy consumption in the EU by country for the commercial sector [8]

#### 1.4. Construction & Wood Waste

Due to its uses in many applications and sustainability, wood is heavily consumed in the construction industry as a primary source for various applications, such as homes, furniture, paper, cardboard, and many other supplies; thus it impacts the society's lifestyle [12]. Due to the high consumption of natural wood, vast amounts of trimming, cut-offs, sawdust, and other wood wastes are produced due to wood processing. For instance, about 40 to 50 % of the wood mass is considered waste after the tree harvesting operation. This waste includes the branches, knots, and roots of the trees are most likely left behind in the forest. Some of the waste is left in the soil to preserve its nutrients while the rest can be used as fuel in various forms, such as wood chips, pellets, and firewood. Out of the 50 to 60 % of the wood mass harvested, around 40 to 60 % is considered to be waste during the timber sawing process. Afterwards, what is left out of the tree that can be used in its intended purpose is about 25% [13]. Thus, leaving massive amounts of wood waste that can cause many problems ranging from their disposal cost to their danger to the environment if not disposed of properly. Although there are many uses for wood waste, their disposal is still very difficult because it is a source of great hazard because of its flammability. Therefore, the best approach is to reduce wood waste as much as possible while using the right means to recycle the remaining waste to decrease our consumption of the raw material and properly dispose of the residuals.

There are various wood waste sources, such as industrial and commercial activities, construction and demolition activities, and part of the municipal solid waste streams [14]. Industrial activities include all the various processing phases of wood to convert from a tree to different products. These phases are divided into primary processing and secondary processing. Primary processing includes in-forest processing, such as cutting down trees, trimming, and saw-milling. In order to be processed, only 2/3 of the tree is taken while the rest is considered to be waste and only 25% of the tree is used after processing [13]. While secondary processing includes all processes that convert timber logs to lumber and various finished products, construction and demolition activities also produce various types of wastes, including wood waste. According to a study in the US, on average, 31% of the construction and demolition waste (CDW) is wood waste [15]. The study results expressed the C&D waste characterization by weight percent and are illustrated in Figure 8.

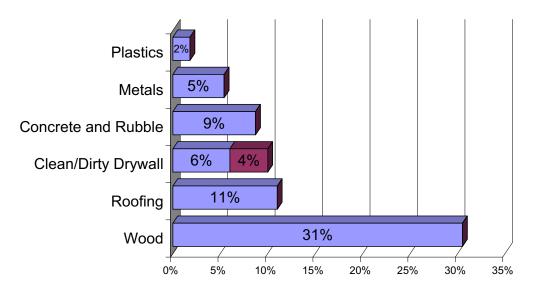


Figure 8: Construction and demolition waste characterization by weight percentage in the US showing the wood waste with the highest percentage of 31% [15]

It is estimated that the CDW generated in the US per year is more than 100 million tons of CDW while contributing to 29% of the total solid wastes generated in the country and around 23% in Hong Kong (total of 20 million tons of CDW) [16]. Similarly, in the UK and Australia, the CDW compromises 50% and 30% of the total landfilled solid wastes. While contributing 38%, 19%, and 14% in Hong Kong, Germany, and Finland, respectively [16]. In the UK, 32% of wood waste is considered construction and demolition waste while around 42% from commercial and industrial sectors (C&I). The wood waste produced composition is categorized as follows; solid wood, particleboard, MDF, plywood...etc. More than 55% of the wood waste is considered solid wood, followed by particleboard and MDF with a combined percentage of around 20%. Also, there are sums of wood waste generated from residential wastes that are either collected separately or within the mixed Municipal Solid Waste (MSW). Although people might believe that the amounts of wood waste in the MSW are minimal, some statistics show otherwise. For instance, in the UK, statistics show that MSW contributes up to 19% of the total wood waste collected in the country [17]. Figure 9 shows the wood waste generated by each activity annually in the UK.

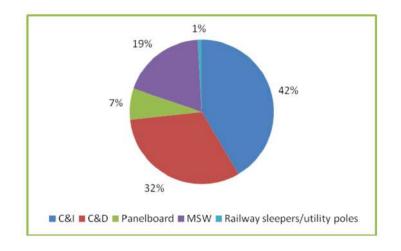


Figure 9: Wood waste generation percentage per sector in the UK [17]

According to a study in Shanghai, CDW generated in 2012 was approximately 13.5 million tons with the construction waste responsible for more than two-thirds of that amount [18]. The waste's main components were concrete and bricks, forming around 80% of the total amount followed by wood waste ranging from 8 to 11%, followed by steel and gypsum. Moreover, the generated waste is divided almost evenly between the residential and non-residential buildings although in recent years compared to a decade ago, the residential buildings generated more than 70% of the CDW[18]. Recycling of wood waste is very popular among developed and developing countries. For instance, sawdust is used in many applications like diet for rabbits and mixed with animal wastes to be used as fertilizers for some plants while cut-offs (scrap) can be used as wood for fireplaces. These simple applications cannot consume the vast amount generated every year. That is why industries have taken part in recycling waste to produce products that can decrease our natural resource consumption. For example, wood composites or sometimes called engineered wood, such as particleboard, plywood, MDF.... etc. Some of these products are manufactured using wood waste, such as the wood chips [15], [19]. Engineered wood has become very popular worldwide in manufacturing furniture, construction forms, and packaging. There are several types of wood composites, such as counter boards made of small pieces of wood waste (cut-offs), chipboards made of sawdust, and MDF boards, which are made of paper waste. Although different kinds of wood waste from secondary processing can be recycled, the massive forest residuals form a huge problem in countries with a high density of forests as well as the wood waste generated from CDW and MSW [15]. Figure 10 shows the amount of utilized wood waste in CDW vs the amount disposed of.

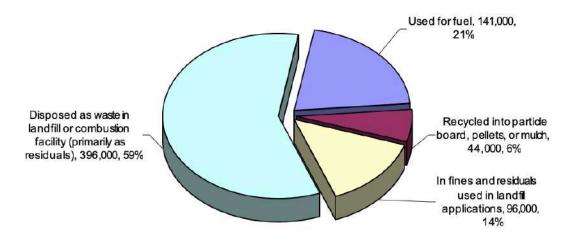


Figure 10: Breakdown of wood uses generated from construction and demolition waste [15]

The common use of wood waste is divided between being used as fuel, produce engineered wood, or in landfill covers. Unfortunately, most of it is being disposed of in landfills or incinerated [15]. However, many other opportunities can be explored to recycle or upcycle wood waste to produce high-value products, which can lead to the preservation of forests. Thus decreasing the deforestation effect on the environment, which include reducing pollution, fighting global warming, reducing desertification, protecting biodiversity, and improving air, water, and soil quality [20].

#### **1.5.Wood Composites/Engineered Wood**

For the above-mentioned needs, industries have taken part in recycling the waste to produce engineered wood/wood composites that can decrease our natural resource consumption. For example, wood composites have become very popular worldwide in manufacturing furniture, construction forms, and packaging. There are two main categories for the wood composites, wood and natural fiber-based composites & natural fiber and plastic composites.

#### 1.5.1. Wood and Natural Fiber Composites

To begin with, wood and natural fiber composites are made out of wood and an organic polymer. There are several examples of it, such as counter boards made of small pieces of wood waste (cut-offs), particle boards made of wood chips, MDF boards made of paper waste, and plywood & laminated. According to APA–The Engineered Wood Association which "is a nonprofit trade association representing manufacturers of engineered wood products in the United States and Canada glued-laminated timber

(Glulam), plywood, oriented strand board (OSB), structural composite lumber (SCL), I-joists, crack-resistant panel siding, speciality products, including radiant barrier sheathing, furniture frames, truck and recreational vehicle bodies, and signs" are all examples of engineered wood products. Figure 11 shows the approximate density of the different engineered wood products with the length of the wood used in the manufacturing process for each [19].

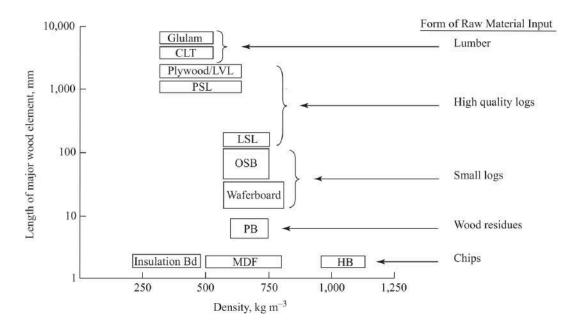


Figure 11: The classification of natural fiber composites by raw materials input, length of major wood element, and density [19]

"Engineered wood products as a material class are dependent upon effective adhesive bonding for their manufacture and superior performance as compared to solid wood" [19]. Generally, wood composites are designed to have better mechanical properties and utilize the high quality of wood resources. The manufacturing process for engineered wood may differ depending on the raw material input, adhesive type, and intended end product. However, there are general manufacturing processes that are common between the different engineered wood products/wood composites, such as the preparation of input material (sawing & sanding), drying (moisture content control), adhesive application, forming, and sawing and sanding of the finished product. Figure 12 shows the general processes for the above-mentioned engineered wood products [19].

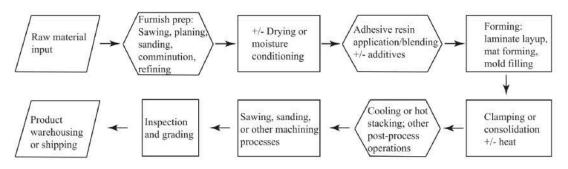


Figure 12: General schematic for wood composites manufacturing process [19]

Wood and natural fiber composites can be classified into five main categories: laminated timber, plywood/laminated veneer lumber, strand composites, particleboards, and medium density fibers boards. First, laminated timber, the word laminated basically means building up in layers. There are two types of laminated timber, Glued laminated timber (Glulam) and Cross-laminated timber (CLT). Glulam is layering of plywood and veneers glued together to create structural elements, such as columns, beams, ...etc. Glulam elements' dimensions can be as long as 30m and 1.8m in depth, which are made out of 3.5 - 3.8 cm thick. Typically, the moisture content of the Glulam is between 12 - 16 % in order to minimize the dimensional changes. The CLT is similar to the Glulam, but its layers are laid perpendicular to each other like plywood. Second, plywood/laminated veneer lumber is basically like the laminated timber, but instead of using layers of lumber, layers of veneers are used. Plywood is made out of layers of veneers glued perpendicular to each other while the laminated veneer is glued parallel to each other. Figure 13 shows the typical production line for the laminated veneer lumber (LVL). Third, strand composites are mainly flakes of wood, glued together to form boards called oriented strand boards that can be glued together in layers to create laminated strand lumber and oriented strand lumber [19].

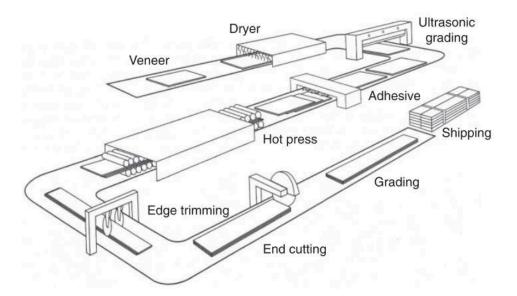


Figure 13: Laminated veneer lumber (LVL) manufacturing process schematic [19]

Fourth, is the particleboard, which is called "nonstructural adhesive-bonded composites". Particleboards are made out of wood waste residue broken down to small sizes and dried to 2-8% moisture content and then mixed with resin and compressed under high temperatures (150 C - 220 C). Finally, medium density fiberboards are made out of pulp chips with moisture content 8-12% and compressed with resin (8-10%) at high temperatures (150C - 190C). Similarly, the hardboard has a higher density, thinner thicknesses, and less resin content (2%). Three main types of resin are used in the manufacturing of the engineered wood, urea-formaldehyde (UF), melamineformaldehyde (MF), and phenol-formaldehyde (PF) resin. Generally, there are two types of adhesives used to bond wood/wood fiber, natural (naturally occurring) and synthetic (man-made) adhesives. "Adhesives or adhesive raw materials of natural origin include blood protein, milk protein, starch, tannins, and lignin. Examples of synthetic adhesives are urea and phenol-formaldehyde resins, isocyanates, and polyvinyl acetate" [19]. Natural adhesives are usually used to produce wood and natural fiber composites while synthetic adhesives (such as plastics) produce natural fiber plastic composites. The classification of adhesives by origin and structural integrity can be shown in Table 1.

Structural integrity	Service environment	Adhesive type
Structural	Fully exterior (withstands long-term water soaking and drying)	Phenol formaldehyde Resorcinol formaldehyde Phenol-resorcinol formaldehyde Emulsion polymer/isocyanate Melamine formaldehyde
	Limited exterior (withstands short-term water soaking)	Melamine-urea formaldehyde Isocyanate/pMDI Epoxy
	Interior (withstands short-term high humidity)	Urea formaldehyde Casein
Semistructural	Limited exterior	Cross-linked polyvinyl acetate Polyurethane
Nonstructural	Interior	Polyvinyl acetate (PVAc) Animal Soybean Elastomeric construction Elastomeric contact Hot-melt Starch

Table 1: Adhesive classification by structural integrity [19]

#### **1.5.2.** Natural Fiber and Plastic Composites

Natural fiber plastic composites are made out of natural fibers glued together using plastic-based adhesives. Synthetic petrochemical polymers can mainly be classified as plastics, thermoplastics, and rubbers [19]. Figure 14 shows the classification of petrochemical polymers based on the molecular bonding property.

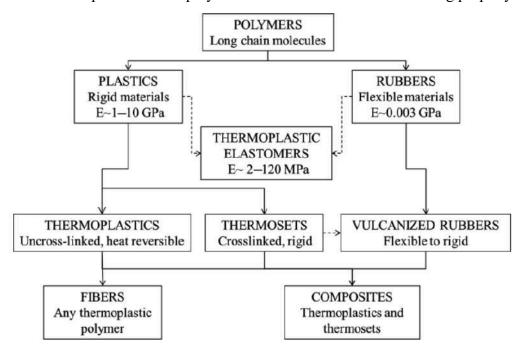


Figure 14: Classification of petrochemical polymers based on the molecular bonding property [19]

There are four main technologies to manufacture plastic composites. To begin with, extrusion is mainly the use of an extruding screw to push the material through a heated unit to melt the polymer and pumping it from the other side. The extrusion rate, feeding rate, and temperature are the manufacturing process parameters adjusted as desired. This method can be applied using single or double screws to produce fiber plastic composite. Also, different polymers can be used and mixed using this method, and it is called coextrusion, which like extrusion can use single or double screws or a combination of both. Second, is the injection method in which the material is pressed in a heating unit with a screw then injected in the desired mold and cooled.

Similarly, to the extrusion, the injection method is basically heating the polymer through heating units which creates the polymer paste that is then injected into the mold with the desired shape. The process is usually used for thermoplastic pellets. Third, is compression molding, which is mainly compressing the heated polymer into a preheated mold with the desired additives. This method can be used for different forms of plastics, such as powder or pellets and accordingly, the time, temperature, and force are selected. If thermoset is used, the sample is cured in the hot mold and then removed from the mold and finally, the thermal forming of thermoplastic sheets into desired shapes. The process involves heating the plastic sheets and pressed into the desired molds until it is cooled down and then removed. The process can be executed through mechanical thermoforming, air thermoforming, or vacuum thermoforming [19]. Woodplastic composite is widely used as a building material and mainly manufactured using the extrusion method. The manufacturing process is selected depending on the final product desired and the type of recycled wood and plastic waste mix used. The three main methods used in the manufacturing process can be illustrated in Figure 15, showing the main technology used is extrusion using single or double screws with different orientations for the setup [19].

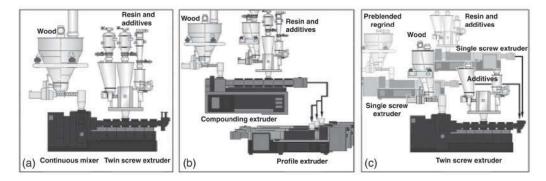


Figure 15: Wood-plastic composites manufacturing schematics using single or double screws with different orientations for the setup [19]

#### **1.6.Wood Waste Opportunities**

In this regard, the rapid rate of deforestation can cause the depletion of timber and paper products that form about 51% of global GDP. Moreover, it is discussed that losing biodiversity causes the deterioration of people's well-being and drives them into poverty. The preservation of forests protect our natural resources and preserves "80% of terrestrial species" [21]. Another benefit is improving the quality of life in general; as mentioned before it improves air, water, and soil qualities that directly affect people's health. That brings us to the social and economic benefits as it can significantly affect our society in various ways.

To begin with, governments, especially in developing countries, have a huge burden regarding health care costs. Protecting our forests could contribute to decreasing this burden while maintaining a better quality of life. Also, lower carbon emission is one of the most critical environmental benefits because it enhances air quality by reducing both pollutions and temperature increase. This could be easily achieved through adopting renewable energy systems. Global energy emission is expected to increase by 45% by 2030, while the global temperature is expected to rise 6 degrees [21]. Renewable energy from wood waste will not only protect the environment by decreasing the above-mentioned concerns but also to create 20 million jobs in the renewable energy sector (biofuels, solar PV, and wind) alone by 2030. It is expected that if the world invested 630 billion USD, it would create 20 million green jobs by 2030 [21]. Thus, if the world invested in renewable energy systems and green services and products production and exporting will help create job opportunities while contributing to public health[20]. As a result of the above measures, sustainable development, economic development, social equity, and poverty reduction could be achieved [22]. Therefore, through recycling and renewable energy, green jobs are created that can minimize unemployment rates as it has been a massive problem in many developing and developed countries in recent years, causing a burden on governments and societies.

### **Chapter 2: Literature Review**

Wood is one of the most consumed renewable organic building materials. It is widely used in the construction sector due to its properties and sustainability, which is currently crucial in any industry. The appealing properties of wood, such as relatively low density (compared to other construction materials), high strength, and low thermal conductivity, makes it versatile and widely used in countless applications [1].

#### 2.1. Wood Types, Structure & Properties

Natural wood is divided into two main categories, softwoods and hardwoods. Softwoods come from cone-bearing trees with needle-like leaves that do not produce seeds (conifer trees), while hardwoods come from broad-leaved trees that produce seeds (flowering plants). Although hardwood trees are slow growing, making it denser than softwoods, some hardwoods can be softer and lighter than softwoods and vice-versa. Generally, softwood and hardwood trees trunks consist of two main sections, the inner layers, which are called heartwood and the outer layers, which are called sapwood. Young trees can have sapwood throughout its cross-section, but as the tree grows, the old sapwood turns to heartwood, which is usually darker in color. This is due to the increase of the extractive content in the wood cells. Sapwood cells then turn to inactive cells, and the heartwood is formed, which is less permeable and less prone to decay. Since the channels present in the wood is clogged, it is more difficult to treat heartwood compared to sapwood. Wood usually form identifiable cell clusters in a growing season, forming what is called growth rings or annular rings, from which the age of the tree can be estimated [23]–[25]. While the tree is growing, new cells are produced which form the growth ring. The cells produced and the start of the growth cycle form a specific cluster that differs from those produced at the end of the cycle. The first cell cluster produced is called earlywood, and the following ones are called latewood. This phenomenon is because each cluster of cells is formed in a different season in different temperature and climate conditions [23].

Figure 16 shows a typical cross-section of a tree trunk illustrating the sapwood and heartwood sections as well as the difference in formation for the earlywood and latewood for both softwood and hardwood.

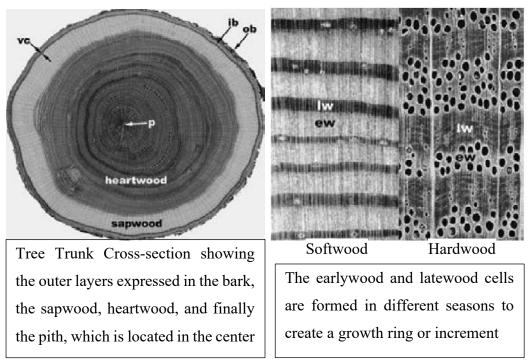


Figure 16: Tree Trunk Cross-section(left) & Growth Ring Cells [23]

One of the most distinguishable features of hardwoods is the presence of vessels, which contributes to a more porous surface compared to softwoods. These vessels are aligned together and form channel-like pores that can be observed in a tree trunk cross-section. These vessels could vary in size from 30 to 300  $\mu$ m in diameter and 100 to 1200  $\mu$ m in length [23]. The vessels can be distinguishably identified in a cross-section sample under the microscope for hardwood compared to the softwood, as shown in Figure 17.

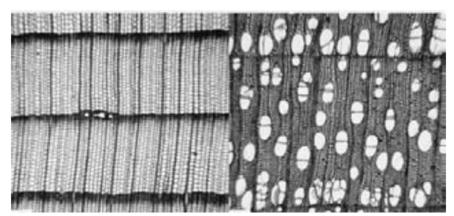
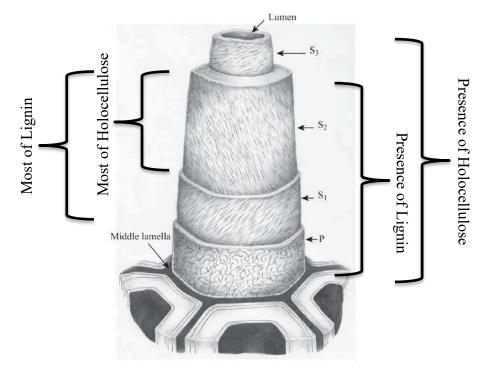


Figure 17: Cross-section of softwood (left) & hardwood (Right) showing vessel size comparison [23]

Mature wood cells are majorly composed of the cell wall and cell lumen. First, the cell lumen is basically a void, which is mainly responsible for the transportation of water throughout the tree. While the cell wall is the main structure of wood cells and is divided into three sections: the middle lamella and the primary & secondary walls. The

three main components of the wood structure of the cell wall are cellulose, hemicellulose, and lignin. The cellulose and hemicellulose combined are the major carbohydrate components of the wood cell and are referred to as holocellulose, which are the wood cells' main structure. While lignin is the organic polymer, which is the is the binding agent that glues the holocellulose together. Collectively, the holocellulose and lignin are referred to as lignocellulose. In addition to these components, there are small amounts of organic substances called extractives and inorganics, which are called ash (relatively small percentages). The sections of the cell wall are illustrated in Figure 18. As mentioned above, the lumen, which is the cell's void, is surrounded by the secondary wall, then the primary wall, and finally the middle lamella. The secondary wall is divided into three layers, called S1, S2, and S3. S2 is the thickest of the three layers and is considered the most important as it contributes greatly to the properties of wood [19], [23]–[26]. Although the lignin can be found throughout the cell wall from the middle lamella to the layers of secondary wall (except for the third layer S3 which has minimal to non-lignin), most of the lignin is mostly located in the first and second layers of the secondary wall. At the same time, most of the holocellulose is located in the second layer of the secondary wall. Thus, making the middle lamella and the primary wall mostly constitute of lignin while S1 is divided between the lignin and holocellulose and finally, the S2 & S3 layers majorly composed of holocellulose [26].



*Figure 18: Typical wood cell wall structure showing the layers starting from the middle lamella, primary wall, and the secondary wall layers & the presence of lignin and holocellulose in the cell [19]* 

In general, to illustrate the components of the whole wood cell structure for American softwoods and hardwoods, the holocellulose constitutes around 65-70% (by dry weight) of the wood while the lignin ranges from around 20-30%. While the inorganic content called ash is less than 1%, which is a relatively small percentage [26]. The characteristics of the components of the wood cells have a significant influence on wood properties. For instance, the amount of cellulose, the angle of the fibers, and the thickness of the cell walls greatly affect the mechanical properties (strength) as well as the physical properties (density) of the wood. While the organic extractives content affects the wood color, decay resistance, flammability, and density as well [23].

There are a few wood species that are locally grown in Egypt. Two of Egypt's most popular species are Casuarina and Eucalyptus, which are usually planted as windbreaks around farms. Both species are classified as fast-growing hardwood trees [27], [28]. According to the global forest resource assessment for Egypt in 2015, Casuarina is the most planted species in Egypt with more than 43 thousand hectares in 2009 followed by Eucalyptus with more than nine thousand hectares [29]. Both species have a dark red-brownish color and a relatively high density of around 500 – 700 kg/m3 depending on the subspecies [29], [30]. First, Casuarina has a chemical composition of around 65% holocellulose, 29% lignin, and 6% extractives. While Eucalyptus has around 56% holocellulose, 34% lignin, and 10% extractives [28].



Figure 19: Eucalyptus tree (left) & Casuarina tree (Right)

#### 2.2. Lignin Removal & Quantification

#### 2.2.1. Lignin Extraction

Lignin extraction from cellulose-based materials is a standard process in the paper and pulp manufacturing industry. The raw materials used can be wood fibers (in the form of wood chips and sawdust) or non-wood fibers, such as rice straw, bagasse, and recycled paper [31]. The main processes for pulp manufacturing are raw materials preparation, pulping, chemical recovery, bleaching, and papermaking. The main processes responsible for the lignin dissolving and whitening of the fibers are the chemical/semi-chemical/mechanical pulping and bleaching. First, the chemical pulping process is mainly dissolving lignin components through the use of chemicals [32]. The three main methods used in pulping are the soda, sulfite, and Kraft (sulfate) methods. In case of wood fibers, the soda and sulfite treatments are not commonly used and are replaced by the Kraft pulping [33]. The soda and Kraft methods are classified under alkaline pulping, in which both use a high alkaline solution containing sodium hydroxide, but in Kraft pulping, sodium sulfide is added to the solution. The process is basically cooking the wood fibers in the alkaline solution at high temperature and pressure, which breaks the lignin bonds. In Kraft pulping, the lignin removal percentage can reach 95% while leaving the cellulose fibers intact.

Regarding the sulfite pulping, sulfite is used for the same purpose in which sulfurous acid or sulfur dioxide are used for delignification but compared to the Kraft pulping, it is not as efficient [32], [34]. Other chemicals used in the pulping process are sodium sulfite and sodium sulfate [35]. Secondly, the bleaching process is mainly responsible for removing the lignin residuals in the pulp (to prevent yellowing of paper) as well as increasing its brightness, which is an important quality parameter. There are different bleaching methods, depending on the fibers used and the pulping method. The bleaching process is divided into stages, which can vary from three to seven stages, with a washing step after each stage [31]–[33]. There are various chemicals that can be used in the bleaching process, such as chlorine, hypochlorite, chlorine dioxide, oxygen, and hydrogen peroxide [35]. Generally, bleaching can be categorized into oxygen with magnesium oxide and caustic soda at high temperature and pressure to bleach the fibers then treated with chlorine and chlorine dioxide to complete the bleaching [33].

On the other hand, conventional bleaching used after Kraft pulping has several stages that include sequential use of chlorination, alkali, and chlorine dioxide solutions, while for sulphite pulping bleaching requires fewer stages. For mechanical pulping, bleaching can be conducted on one stage using hydrogen peroxide, sodium hydro sulphate, or sodium peroxide [33]. Moreover, sometimes sodium hypochlorite can be used for bleaching preceded by EDTA treatment to eliminate metal ions [36].

#### 2.2.2. Lignin Quantification Methods

Lignin quantification is an important test as a characterization test for pulp and paper manufacturing industry [37]. There are various quantitative and qualitative methods to measure lignin content in wood and pulp samples. Quantitative methods include acetyl bromide and acid-insoluble lignin (also referred to as Klason lignin), which is the most commonly used method, especially for wood samples. Unlike the acetyl bromide, the Klason lignin is considered a direct method, in which the carbohydrate components are dissolved, leaving the lignin components for measuring. While indirect methods include acetyl bromide and spectrophotometric methods, which depend on the calculation of lignin from the difference between the original sample and the polysaccharide content (carbohydrate components) or by measuring some structural characteristics and forming a relationship to the lignin concentration [37], [38]. First, Klason lignin, as mentioned above, is dissolving the non-lignin components using sulfuric acid ( $H_2SO_4$ ), which leaves the lignin residues to be measured. This process is conducted on two steps; first, the sample is treated in cold 72% H<sub>2</sub>SO<sub>4</sub> followed by boiling it in diluted 3% H<sub>2</sub>SO<sub>4</sub>, leaving the insoluble lignin residues. The oven-dry weight of lignin is then obtained to calculate the lignin content in the sample [37]. The procedure for Klason lignin is illustrated in TAPPI 222 "acid-insoluble lignin in wood and pulp", which states that an extractive free sample should be used for this method [39]. The extractive free sample's preparation process could lead to the loss of some lignin, which affects the lignin content's accuracy.

On the other hand, Klason lignin does not determine the soluble lignin content, which could range from 0.3% to 5% depending on the sample type [37]. That is why sometimes Klason lignin is combined with the spectrophotometric method (for the decanted solution from the second stage of Klason lignin) to determine the soluble lignin components [38]. Spectrophotometric methods are based on measuring the absorbance at a certain wavelength (ranging from 210 to 280 nm) and relating it to the

lignin concentration. Similarly, UV micro-spectrophotometric method was developed to measure the absorbance by logging the light intensity micro-photometrically. Although both methods are quick, non-destructive and require small samples, it is not recommended for hardwoods due to the hardwood lignin composition, which has different absorbances. Other spectrophotometric methods involve dissolving the sample in a solvent, such as sulfuric acid, nitric acid, sodium chlorite solution, and acetyl bromide in acetic acid, and measuring the absorbance at 280nm wavelength [37]. In this regarded, the most popular method used is the acetyl bromide due to its simple and fast procedure as well as small sample size and does not require any correction for the acid-soluble lignin [37], [38]. However, the acetyl bromide method is as accurate as the lignin standard used for calibration [38]. Unlike UV spectroscopy, Fourier transform infrared (FTIR) spectroscopy measures lignin components through light reflectance. FTIR can be conducted using two methods, attenuated total reflectance (ATR), or diffuse reflectance infrared Fourier transform (DRIFT). ATR-FTIR method is conducted through starting the infrared beam that passes through a crystal and recording the emitted photons wavelength. While DRIFT is mostly used on pressed powder samples with purified salt, which the infrared beam passes through [40]. However, these are qualitative methods not quantitative like Klason lignin. Moreover, it measures the functional groups only on the sample's surface and is not considered efficient in detecting small lignin content differences [40], [41].

## 2.3. Transparent Wood Composite

Transparent wood composite is basically fabricated using delignified wood pieces or fibers with a polymer, which can then be used in many different applications due to its advantages over other materials [1]. To begin with, it has better thermal insulation compared to regular glass. Moreover, with some processing, transparent wood can be stronger than steel, which allows it to replace the two primary materials used in modern construction (glass & steel). Also, it has high potential in solar energy due to its "high optical haze". Finally, on top of the benefits mentioned above, it is considered a sustainable material, to replace plastics and other materials, due to its better biodegradability [2]. Currently, environmentally friendly materials are becoming very popular due to the rising problem of global warming and climate change. As a renewable, biodegradable, and environmentally friendly material, wood has been used

in many applications in construction, manufacturing, and many others. In that respect, "recently, there is a great interest in abundant cellulose nanofibers (CNF) and cellulose nanocrystals extracted from wood in electronics, energy, and a range of other types of applications due to their nanostructures, excellent mechanical, optical properties, ...etc." [12]. According to a recent study, flexible transparent conductive paper can be used in "flexible consumer electronics, smart clothing, energy harvesting, and sensors in the future" [42]. An example of the use of transparent wood in electronics is optical technologies. Transparent wood samples were manufactured and tested as an organic light-transmitting diode (OLED). These samples were prepared using different types of resins to study the correlation between the strength and ductility of the sample and its transparency properties. The samples were prepared using nanofibers of wood, which is basically sawdust, of a specific size. First, the lignin was removed using sodium chlorite (NaClO<sub>2</sub>) and then the hemicelluloses were removed using potassium hydroxide. Second, the samples were impregnated with different types of resin, as mentioned above. Finally, the transmittances of the samples were observed, as well as their strength [43]. Another approach has focused on manufacturing layers of artificial transparent wood layers with self-healing properties. The transparent artificial wood nanostructures were prepared using hydrogen peroxide with various other chemicals and adhesive materials induced between its layers. The prepared samples' transparency, strength, and self-healing properties were tested and evaluated [44].

# 2.4. Transparent Wood Fabrication

The fabrication process for transparent wood is divided into two main stages: chemical treatment and polymer infiltration. According to the literature, there are different reported methods to fabricate transparent wood starting with the selected wood species, woodcut and size, chemicals used in treatment, and finally, the selected polymer. First, the samples used are either wood pieces (with various dimensions and thicknesses), veneer, or wood particles. Afterwards, the samples are chemically treated, in which the samples are bleached, and its lignin components are removed (which gives the wood its yellowish color). Thus, the samples turn white are washed and dehydrated to prepare it for the next step. Finally, the samples are infiltrated with the selected polymer (mostly PMMA or Epoxy), which replaces the lignin and the hollow channels in the samples to obtain transparent wood substrates.

#### 2.4.1. Wood Species and Sizes

The wood species used in the fabrication of transparent wood were either wood pieces, wood veneers, or wood particles (fibers). The most commonly used wood species were basswood, balsa, pine, beech, birch, and poplar. The wood samples used were either radially cut or longitudinally cut with thicknesses ranging from 1mm to 10mm. The sample sizes used were squared pieces ranged from 2cm to 10cm, and in one case, up to 20cm. The radially cut samples had an advantage over the longitudinal cut samples due to the shorter channels, which facilitated faster treatment. On the other hand, the longitudinal had a better overall cohesiveness due to the strength of the collective fibers [1], [2], [45]–[50]. The wood samples' treatment time depended on the wood species, thickness, and size [1].

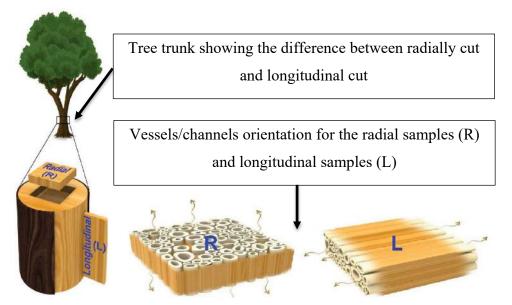


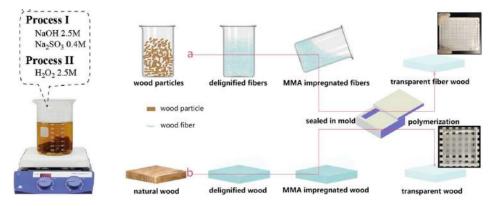
Figure 20: Radial samples vs. longitudinal samples vessels orientation [2]

#### 2.4.2. Chemical Treatment

Most techniques depend on a two-stage process, while others use a single-stage process. In most cases, the treatment requires heating the solutions at various temperatures depending on the method used. However, there is one method that did not require heating and instead depended on soaking the samples in the solution at room temperature for more extended periods. There are four main treatment methods reported in the literature, which use one or combination of the following chemicals: sodium hydroxide, sodium sulfite, sodium chlorite, sodium hypochlorite, and hydrogen peroxide. The details for each treatment method and the concentration of the solution are mentioned in the appropriate sections.

#### 2.4.2.1. Sodium Hydroxide & Sodium Sulfite

The first method is a two-stage process, in which a mixture of sodium hydroxide (NaOH) and sodium sulfite (Na<sub>2</sub>SO<sub>3</sub>) is used to delignify wood samples, which are commonly used in the paper and pulp industry. The mixture is prepared using a concentration of 2.5 mol/L of NaOH and 0.4 mol/L of Na<sub>2</sub>SO<sub>3</sub> in deionized water. The wood samples were immersed in the solution and kept boiling for 12 hours to get rid of lignin. The samples were then rinsed with hot distilled water and transferred to the bleaching solution. The bleaching solution is prepared using hydrogen peroxide  $(H_2O_2)$ with concertation of 2.5 mol/L in deionized water. The solution kept boiling until the wood sample turned white. Afterwards, the samples were rinsed to wash off the chemicals and then dehydrated using ethanol to get rid of all moisture in the wood samples [2], [45], [51]. Thus, the samples were ready for the polymer infiltration, which was conducted using either epoxy resin or poly (methyl methacrylate) (PMMA); and in one case Polyvinylpyrrolidone (PVP) was used as the polymer[46]. It was noticeably clear that after the delignification treatment (solution 1), the wood samples lost a lot of its strength, easily broken up into little pieces during the H<sub>2</sub>O<sub>2</sub> treatment, especially at boiling temperature. This treatment method was mainly used for wood pieces while another approach treated poplar wood particles (fibers) with sizes ranging from 2 to 5 mm using the same concentration for the NaOH and Na<sub>2</sub>SO<sub>3</sub> solution, but 30 wt% H<sub>2</sub>O<sub>2</sub> was used for bleaching. The wood particles were stirred into fibers and infiltrated using PMMA to produce large-sized transparent wood with dimensions of  $10 \text{ cm} \times 10 \text{ cm}$ [51]. Unfortunately, this chemical process produced concentrated black liquor due to removing up to 90% of the lignin [49]. Thus, creating a challenge on the environmental treatment process for such wastes that are very difficult to recycle or dispose of.



*Figure 21: TW chemical treatment & fabrication diagram showing the chemical process setup (left) and the polymer infiltration steps (right) for wood pieces and wood fibers* [2], [51]

	Samples Used		Chemical Treatment						Polymer Infiltration				
Defense			Proces	Process 1		Process 2		Debudention	Polymer				
Reference	Type of Wood	Size	Chemicals used & Concentrations	Temperature & Duration	Chemicals used & Concentrations	Temperature & Duration	Washing	Dehydration	Polymer used	Preparation method	Vacuum	After Vacuum	
[2]	Basswood (R- wood & L- wood)	50 mm × 50 mm × 3 mm	Sodium hydroxide 100 g/L & Sodium sulphite 50 g/L	Boiling for 12 h	Hydrogen peroxide 2.5 mol/L	Boiling without stirring until samples were white	Rinsed with hot distilled water (3 times)	Ethanol	Epoxy resin	Sample immersed in the liquid resin	5min (3 Times) @ 200 Pa	30 °C for 12 h	
[45]	Basswood	5mm Thick	Sodium hydroxide 100 g/L & Sodium sulphite 50 g/L	Boiling for 3 h	Hydrogen peroxide 2.5 mol/L	Boiling for 2–3 hrs until samples were white	-	-	Epoxy resin	Sample immersed in the liquid resin	-	-	
[51]	Poplar	Particles were 2–5 mm	NaOH (10 wt%) and Na2 SO3 (5 wt%) in deionized water	boiled for 2–4 h	H2O2 (30 wt%)	Boiling until samples were white	Stirred into fibers and filtered three times with deionized water.	-	PMMA	Pre-polymerized 80°C for 15 min	10m	Glass mold at 60 °C for 8 h	
[46]	Basswood	30 mm × 22 mm × 1 mm	NaOH and Na2SO3 in DI water resulting in a concentration of 2.5 mol/L and 0.4 mol/L respectively	Kept boiling for 12 h	H2O2, 2.5 mol/L in DI water	Kept boiling without stirring until yellow color disappeared	Hot distilled water three times	Ethanol	PVP	Was dissolved in ethanol at a concentration of 15% by mass of polymer (The solution depth was approximately ten times greater than the wood thickness)	10 min @ 200 Pa	Solution placed on a hot plate at 60 °C. The sample was peeled off of the bottom of the dish after the solvent evaporated	

## *Table 2: Sodium hydroxide & sodium sulfite recipe journals summary*

#### 2.4.2.2. <u>Sodium Chlorite</u>

The second method is a one stage process using sodium chlorite (NaClO<sub>2</sub>) to remove lignin from wood. The solution was prepared using 1 wt.% of sodium chlorite in an acetate buffer solution with pH 4.6. at 80°C from 3 to 12 h or until it turned white, which depends on the thickness of the wood samples [1], [47], [48], [52], [53]. Like the previous method, the wood samples were easily broken after treatment due to lignin's removal, which is the wood fibers' binding agent. Consequently, a combination of both methods was used to help in keeping the samples intact by decreasing the treatment time by the sodium chlorite and using a bleaching solution to turn the sample into the desired white color. The bleaching solution used was similar to the previous method, which is H<sub>2</sub>O<sub>2</sub> solution but with a concentration of 5mol/L in deionized water. The solution kept boiling from 1 to 4 hours, depending on the samples used [52], [54]–[56]. Afterwards, the samples were washed using deionized or distilled water to wash off the chemicals and was ready for dehydration. The dehydration process was done using ethanol and acetone and/or a mixture of both. Some journals dehydrated the wood samples using pure ethanol then 1:1 ratio of ethanol and acetone, and finally with pure acetone; each step was repeated three times [1], [53], [57], [58]. In contrast, others used pure ethanol and pure acetone sequentially and were repeated three times without using a mixture of both [47], [59], [60]. Then the samples were ready for the polymer infiltration, which was mostly done using PMMA [1], [47], [53], [57]–[60]. Although after adding the bleaching stage, the treatment process produced black liquor as the previously stated method, which is undesired for environmental purposes.

Another approach was adopted to further enhance the structural strength during the chemical treatment and decrease the black liquor produced due to the lignin removal solution. The main concept is producing lignin-retaining transparent wood by modifying and enhancing the treatment process. First, unlike the previous two methods, the samples were bleached and then treated with the delignification solution. Second, the bleaching solution is a highly alkaline solution prepared by mixing the chemicals in the following order: 3 wt.% of sodium silicate (Na<sub>2</sub>SiO<sub>3</sub>), 3 wt.% of sodium hydroxide (NaOH), 0.1 wt.% of magnesium sulfate (MgSO<sub>4</sub>), 0.1 wt.% of DTPA, and finally 4 wt.% of hydrogen peroxide (H<sub>2</sub>O<sub>2</sub>). Sodium silicate was used to enhance the stability of the H<sub>2</sub>O<sub>2</sub> while the sodium hydroxide was used to increase the alkalinity of the solution. Also, the DTPA was used to decrease the metal ions in the solution and

enhance the efficiency of the H<sub>2</sub>O<sub>2</sub>. The wood samples were immersed in the alkaline solution and placed on the hot plate at 70°C until the samples turned white [49]. This process aims to bleach the wood sample turning them white without removing much of the lignin. Afterwards, the sample was rinsed and transferred to the second solution, which is the sodium chlorite delignification. Similar to the previously mentioned method, the solution was prepared using 1 wt.% of sodium chlorite in an acetate buffer solution with pH 4.6 and placed on the hot plate at 80°C. This treatment method's main advantage is that the transparent wood retains around 80% of the lignin and reduces the black liquor output from the process. Consequently, the transparent wood's mechanical strength should increase due to keeping the most of lignin, which, as mentioned before, is the binging agent for the wood. Thus, minimizing the environmental impact of the fabrication process by reducing the toxic effluents, which deems it a green process compared to the previously stated methods [49].

Although the fabricated samples using the recipes mentioned above were transparent, it was not a see-through material. As shown in Figure 22, although when the transparent wood samples were placed on any text, it was readable, when it was placed a few millimeters above it, the text was fuzzy and unreadable in most cases. Thus, a further treatment step was introduced to enhance visibility from higher distances. The transparent wood templated where acetylated using acetic anhydride in N-methyl-2pyrrolidone solvent and pyridine as a catalyst with a weight ratio of 7:100:6 [61]. The reaction time was about 6 hours at 80°C with wood template to a mixture ratio of 2:1. Before the acetylation, the wood sample was first treated using 1 wt.% of sodium chlorite in an acetate buffer solution with pH 4.6 and placed on the hot plate at 80°C for 6 to 12 hours. After acetylation, the sample was further treated with sodium chlorite until it turned white. The samples were then washed with deionized water and dehydrated using ethanol and acetone. Transparent wood was then fabricated using PMMA and compared to non-acylated transparent wood. The visibility of the acetylated TW has increased as well as the light transmittance compared to the non-acetylated TW [61].

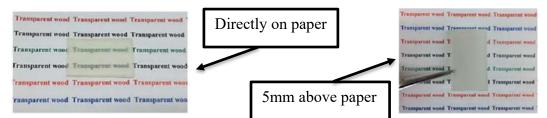


Figure 22: Transparent wood samples transparency directly on paper and 5mm above [49]

	Samples Used		Chemical Treatment						Polymer Infiltration				
Defense	Sample	es Used	Proces	Process 1		ess 2	Washing	Debudanting		Polymer			
Reference	Type of Wood	Size	Chemicals used & Concentrations	Temperature & Duration	Chemicals used & Concentrations	Temperature & Duration	wasning	Dehydration	Polymer used	Preparation method	Vacuum	After Vacuum	
[1]	Balsa wood	20 mm × 20 mm × range (0.6 - 8mm)	1 wt.% of sodium chlorite with acetate buffer solution (pH 4.6)	80°C for 6 - 12 h	12		Deionized water	1. Pure ethanol - 2. 1:1 mixture of ethanol and acetone - 3. pure acetone	PMMA	Prepolymerized at 75°C for 15 min in two-necked round- bottom flask with 0.3 wt. % 2,2'-Azobis	30 m (3 Times)	Sandwiched between two glass slides and packaged in aluminum foil	
			solution (pri 4.0)					Each step was repeated 3 times		Heated in an oven at 70 °C for 4 h		70°C for 4 h	
[57]	Beech wood	0.1 mm - 0.7 mm Thick	5 wt.% sodium chlorite in acetate buffer solution (pH 4.6)	95°C for 12 h	-		Deionized water	1. Pure ethanol - 2. 1:1 mixture of ethanol and acetone - 3. pure acetone	PMMA	Pre-polymerized 90°C for 5 min	1 h (3 Times) @ 0.1 bar	Sandwiched between two glass slides and packaged in aluminum foil - 85°C for 12 h	
[52]	Balsa wood & basswood	20 mm * 20 mm range (1 – 5 mm)	1 wt. % of sodium chlorite with acetate buffer solution (pH 4.6)	80°C for 3 - 12 h	hydrogen peroxide (H2O2, 5 mol/L)	90°C for 1 hr	Deionized water	Left in Ethanol for 8 hrs	Epoxy resin	Sample immersed in the liquid resin	2 h @ 1000 Pa	Rolled up with silicone paper and placed in a petri dish and then dried naturally at atmospheric pressure for 12 h	
[48]	Tangential veneers of balsa wood	2 cm×2 cm and 6 cm×6 cm (0.6–0.8 mm Thickness)	1 wt % NaClO2 in acetate buffer solution (pH=4.6)	80°C for 6 h	Samples were freeze-dried	After totally dried, the delignified wood samples were compressed by 75 kN for 25 min under 25°C	Washed with deionized water three times	-	PMMA	-	-	Heated in an oven at 70°C for 4 h	
[62]	Basswood (Tilia) veneers	20 mm × 20 mm × 0.4 mm	2 wt.% NaClO2, 0.1 wt.% Acetic acid glacial, and 97.9 wt.% Ultrapure water	an oscillation fre	ator at 80 °C with quency of 40 rpm 20, and 150 min	-	Ultra-pure water	Ethanol	PMMA	-	-	-	
[53]	Poplar wood (P. adenopoda Maxim)	25 mm × 25 mm × 1 mm and 50 mm × 50 mm × 1 mm	1.5 wt.% NaClO2 with acetate buffer solution (pH 4.6)	80°C for 8 h	-		Deionized water	1. Pure ethanol - 2. 1:1 mixture of ethanol and acetone - 3. pure acetone	PMMA	Pre-polymerized 75°C for 20 min with 0.3 wt.% 2,2'-Azobis	30 min @ 5 kPa	The infiltrated wood was sandwiched between two glass slides and polymerized in an	

## Table 3: Sodium chlorite recipe journals summary

											oven at 70°C for 4 h
[59]	Balsa wood	15 mm × 25 mm × 1.0 mm	l wt.% of sodium chlorite in acetate buffer solution (pH 4.6)	80°C until the wood became totally white	-	Deionized water	Ethanol and acetone sequentially (repeated 3 times)	PMMA	MMA monomer was first pre-polymerized at 75°C for 15 min with 0.3 wt.% 2,2'- azobis (2- methylpropionitrile) (AIBN) as initiator and cooled down to room temperature	-	The infiltrated wood was sandwiched between two glass slides and polymerized in an oven at 70°C for 4 h
[58]	Basswood (Veneers)	100 mm × 100 mm × 1.6 mm	Sodium chlorite at a concentration of 0.5 g/g of wood in a 1 N acetate buffer solution (sodium acetate and acetic acid to maintain a pH ~ 4.6)	80°C (~12 h)	-	Hot deionized water three times	From ethanol to ethanol/acetone (1:1 by volume) to acetone - Each step of the solvent exchange was carried out twice for approximately 30 min	PMMA	-	at 70°C for at least 12 hours	For one layer - Placed between two glass plates and tightly secured with four medium (31.75 mm) binder clips
[60]	Wood veneer (balsa)	2 cm × 2 cm × 0.2 cm	l wt.% of sodium chlorite in acetate buffer solution (pH 4.6)	80°C until almost uniformly white	-	Deionized water	Ethanol and acetone sequentially (repeated 3 times)	PMMA	The prepolymerization was completed by heating the MMA at 75°C for 15 min with 0.3 wt.% 2,2'-azobis (2-methyl- propionitrile) followed by cooling to room temperature. Then MMA/QDs were vacuumed	-	Finally, the infiltrated wood was sandwiched between two glass slides, wrapped with aluminum foil, and heated in an oven at 70°C for 4 h in ambient atmosphere
[63]	Silver birch wood	2 cm × 2 cm	NaClO2 (1 wt. %) in an acetate buffer solution (pH 4.6)	80°C until samples became white	-	Deionized water (several times)	Ethanol and acetone sequentially (repeated 3 times)	PMMA & PEG 1000	PEG was heated at 45 °C until complete dissolution. MMA (30 wt %) was added to the molten PEG (70 wt %) solution and kept under stirring at 45 °C for 10 min. The initiator (0.3 wt % based on MMA), was then added to the PEG/MMA solution, and the blend was continuously stirred at 45 °C for 10 min	15 min (three times)	The blend solution containing the DW samples was heated at 40°C for a few minutes between each infiltration to delay PEG crystallization. The infiltrated DW template was finally sandwiched between two glass slides, packaged with aluminum foil, and polymerized in an oven at 70°C for 4 h

	Samples Used			Chemical Treatment					Polymer Infiltration				
D . 6			Proces	Process 1		Process 2				Polymer			
Reference	Type of Wood	Size	Chemicals used & Concentrations	Temperature & Duration	Chemicals used & Concentrations	Temperature & Duration	Washing	Dehydration	Polymer used	Preparation method	Vacuum	After Vacuum	
[49]	Pine, birch, and ash wood veneer	100 mm × 100 mm × 1.5 mm	Deionized water, sodium silicate (3.0 wt.%), sodium hydroxide solution (3.0 wt.%), magnesium sulfate (0.1 wt.%), DTPA (0.1 wt.%), and then H2O2 (4.0 wt.%)	70°C until samples were white	lwt.% sodium chlorite in acetate buffer solution (pH 4.6)	80°C until samples were white	Deionized water	Ethanol and acetone sequentially. Each solvent- exchange step was repeated 3 times	РММА	Pre-polymerized 75°C for 15 min	Vacuum	Sandwiched between two glass slides and packaged in aluminum foil - 75°C for 4 h	
[64]	Balsa wood chip	200 mm × 200 mm × 1 mm	Bleaching solution was prepared by mixing chemicals of deionized water, sodium silicate (3 wt %), sodium hydroxide solution (3 wt %), magnesium sulfate (0.1 wt %), and H2O2 (4 wt %)	Kept boiling until the yellow color of the wood slice disappeared	-		Rinsed with cold water	Ethanol	PAA solution	Acrylic acid was diluted with deionized water to a concentration of 40 wt.%. Subsequently, ammonium persulfate (1 wt.%) was added to the mixture	10 min @ 200 Pa	Finally, the monomer- infiltrated wood sample was sandwiched between two glass slides, wrapped with aluminum foil, and heated in an oven at 75°C for 4 h in air atmosphere for polymerization	

## Table 4: Lignin retaining recipe journals summary

	Samples Used			Chemical	Treatment				l	Polymer Infi	ltration	
Reference	Samples	Samples Osed		Process 1		ess 2	Washing	Dehydration		Polymer		
Reference	Type of Wood	Size	Chemicals used & Concentrations	Temperature & Duration	Chemicals used & Concentrations	Temperature & Duration	wasning	Denydration	Polymer used	Preparation method	Vacuum	After Vacuum
[47]	Balsa wood & birch	0.7, 1.5, 3, 7, 10 mm Thick	l wt.% of sodium chlorite with acetate buffer solution (pH 4.6)	80°C until samples were white	Acetate anhydride with pyridine as the catalyst and N- methyl-2- pyrrolidone (NMP is 7 : 6 : 100)	(60°C, 80°C, 100°C, 120°C), and time (1 h, 2 h, 3 h, 4 h, 5 h, 6 h, 8 h, 10 h, 12 h)	Deionized water / Dehydration:- Ethanol and acetone sequentially. Each solvent- exchange step was repeated 3 times	Ethanol and acetone sequentially. Each solvent- exchange step was repeated 3 times	PMMA	Pre-polymerized 75°C for 15 min	-	Sandwiched between two glass slides and packaged in aluminum foil - 70°C for 4 h
[43]	Wood powder sieved through a 60 mesh sieve	-	Acidified sodium chlorite solution	70°C for 1 h (the process was repeated 5 times)	5 wt.% potassium hydroxide solution at room temperature for 12 h	& at the same concentration at 80 °C for an- other 2 h in order to leach hemicelluloses	Distilled water	Acetone	Acrylic resins	Under a reduced pressure	12 h @ 0.09 MPa	
[61]	Balsa wood longitudinal section	20 × 20 mm × (0.24, 0.32, 0.42, 0.51, and 0.65 cm)	l wt % of sodium chlorite in an acetate buffer solution (pH 4.6)	at 80°C for 6 h (12 h for 0.51 and 0.65 cm, to make sure all the templates have a similar composition)	acetic anhydride (Sigma- Aldrich) in the solvent of N- methyl-2- pyrrolidone (NMP, Sigma- Aldrich) with pyridine (Sigma- Aldrich) as the catalyst. The ratio of wood template (g):acetate anhydride (mL):pyridine (mL):NMP (mL) is 2:7:6:100. The reaction was performed under 80°C for 6 h	Then the acetylated wood was treated with NaClO2 again until white	washed with deionized water, ethanol, and finally acetone	-	PMMA	Finally, the delignified wood templates were then infiltrated with PMMA solution for 4 h (12 h for 0.51 and 0.65 cm) under vacuum and were heating in an oven at 45 °C for 24 h, and then 70 °C for 6 h to complete the polymerization process. MMA prepolymerization reaction was performed in a round- bottom flask at 75 °C for 15 min with initiator (2- methylpropionitrile, 0.3 wt.%, AIBN, Sigma-Aldrich) and terminated with ice-water bath.	4 – 12 h	The infiltrated wood templates were covered with two glass slides on both sides and packaged in the aluminum foil before heating in an oven

## Table 5: Acylated transparent wood journals summary

## 2.4.2.3. <u>Hydrogen peroxide</u>

The third method used H<sub>2</sub>O<sub>2</sub> solution for the chemical treatment in different setups to fabricate thicker transparent wood samples. The thickness ranged from 5mm to 50mm using grids to place the samples on top of the solution in the beaker and boil the solution to treat the wood samples without damaging it, as shown in Figure 23. The first approach was by boiling 30 wt.% H<sub>2</sub>O<sub>2</sub> solution from 2 to 24 hours (depending on the thickness) and let the fumes infiltrate the suspended wood samples to turn it white. The samples were then washed and dehydrated using ethanol and impregnated using epoxy resin. A similar process was followed by a pretreatment step before the H<sub>2</sub>O<sub>2</sub> solution treatment where the samples were immersed in 2.5 mol/L NaOH solution at 100°C for about 12 hours. The wood samples were then transferred to the boiling H<sub>2</sub>O<sub>2</sub> solution on a suspended grid to complete the treatment. Followed by washing in DI water and then dehydration using pure ethanol then 1:1 ratio of ethanol and acetone, and finally with pure acetone. Then the samples were finally impregnated using epoxy resin [50], [65]. The fabrication process diagram can be shown in Figure 24.

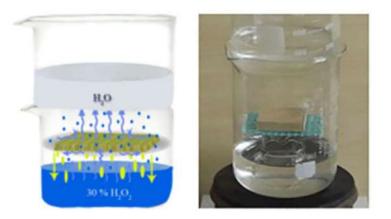


Figure 23: Hydrogen peroxide suspended treatment setup [50]

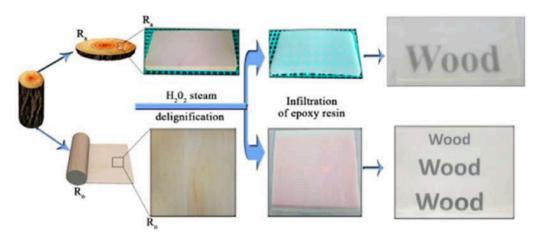


Figure 24: Fabrication process diagram for H<sub>2</sub>O<sub>2</sub> treatment for radially cut wood and veneer [50]

	Samples Used		Chemical Treatment					Polymer Ir	nfiltration		
Reference	Samples C	Samples Osed		reatment	Washing	Dehydration	Р	olymer			
	Type of Wood	Size	Chemicals used & Temperature & Concentrations Duration				Polymer used	Preparation method	Vacuum	After Vacuum	
	Basswood/pine (R-	50/100 × 50 mm	Boiling H2O2	2–12 h until	Cold water			Sample immersed	20 m (3		
[50]	wood & L-wood)	210 mm × 190 mm × (0.8 - 2 mm)	aqueous solution (30 wt.%)	yellow color disappeared	and ethanol	Ethanol	Epoxy resin	in the liquid resin	Times) @ 0.08 MPa	30°C for 24 h	
[66]	Basswood	20 mm × 20 mm × 0.4 mm	6 wt.% H2O2, 1 wt.% trisodium citrate dihydrate, 1 wt.% NaOH, and 92 wt.% ultrapure water	60 °C for (30min to 150min)	Ultrapure water	Ethanol	РММА	pure MMA monomer was uniformly mixed with AIBN initiator (0.5 wt.% solution) and pre- polymerized at 75°C for 15 min and then cooled to room temperature	30 min	Sandwiched between two pieces of glass and transparent wood (TW) samples were obtained by heated at 70°C for 5 h	
[65]	Basswood pieces	11 × 7 × 0.5 or 2 or 5 cm3	placed them on grids with a unit size of 5 × 5 cm 3. 30% H2O2 aqueous solution	Boiling for approximately 4 and 12 h (5 cm - approximately 24 h)	Rinsed with DI water	dehydration with ethanol, then a 1:1 (volume ratio) mixture of ethanol and acetone, and finally, pure acetone (step by step) Each step was repeated many times until the water was completely removed	Epoxy resin	the delignified wood was immersed in the liquid resin	20 m repeated three times	Finally, the wood was put between two glass plates and kept static at 30°C for 24 h	

## Table 6: Hydrogen peroxide recipe journals summary

## 2.4.2.4. <u>Sodium Hypochlorite</u>

Lastly, sodium hypochlorite treatment is the least energy-consuming method. This treatment method is a one stage process in which the wood samples are placed in 5 wt.% sodium hypochlorite with a mass ratio of wood sample to solution ranging from 1:40 to 1:60 and left at room temperature until the samples turned white. The sample was then washed and dehydrated using water and ethanol solution and was ready for the polymer infiltration [67], [68].

	Samples U	Inod	Chamical T	Chemical Treatment			Polymer Infiltration				
Reference	Samples C	Jseu	Chemical I Cathlent		Washing	Dehydration	Р	olymer			
	Type of Wood	Size	Chemicals used & Concentrations	Temperature & Duration	Ŭ	, i	Polymer used	Preparation method	Vacuum	After Vacuum	
[67]	Basswood and pine wood	$50 \text{ mm} \times 50 \\ \text{mm} \times 3 \\ \text{mm}$	sodium hypochlorite (wood: NaClO = 1:40 wt)	3–5 h (Room Temperature)	-	solution of water and ethanol (1:1 wt) for 3 times	Microporous filtering film and filter paper	Pressed at different pressure for about 1–3 h	-	-	
[68]	Basswood	44.5 mm × 44.5 mm × 0.8 mm	Sodium hypochlorite solution 5wt% (The mass ratio of Basswood and NaClO solution was 1:60)	Ambient temperature until the wood slices became white completely (3 h - 9 h)	Ethanol water solution (50 wt%) and washed for three times to remove the remaining chemicals	-	PET film, and covered with a microporous membrane filter	The wood fibers were forced down to one direction by rolling a glass rod which was placed upon the wood slice with a constant pressure	After that, the aligned wood slice was covered by some pieces of filter papers and pressed at room temperature for 5 h	-	

Table 7: Sodium	hvpochlorite	recipe	iournals	summarv
10010 /	nypoentorne	recipe,	journans	Stunnen y

#### 2.4.3. Polymer Infiltration

The transparent wood fabrication was mainly conducted using either epoxy resin or PMMA. To begin with, the epoxy resin used was ultra-transparent with the recommended ratio of resin to hardener recommended by the manufacturer. The treated wood was immersed in the epoxy and then vacuumed on several cycles to ensure full infiltration of the epoxy in the wood templates. According to the literature, the working vacuuming pressure ranged from 200 Pa to 0.08 MPa and the vacuum time ranged 5 to 20 min. The sample was then placed between either two glass plates of silicon paper and let static to harden [2], [50], [52][65].

While the PMMA was prepared using MMA monomer and 2,2' -azobis (2methylpropionitrile) as an initiator. The pre-polymerized MMA was prepared using 0.3 wt.% of the initiator and left for 15 min at 75°C and then cooled to room temperature using a water bath. The treated wood was then infiltrated under vacuum on several cycles to ensure full infiltration with time ranging from 15 min to 1 h. Finally, the samples were sandwiched between two glass plates and covered with aluminum foil and placed for 4 hours at 60 to 85°C [1], [47], [49], [51], [69].

In order to produce thermal energy storing transparent wood with a reversible optical transmittance, a mixture of PMMA and PEG was used as the polymer. The mixture of 30 wt.% of MMA to 70 wt.% of PEG with a molecular weight of 1000 g mol<sup>-1</sup> was used, and 0.3 wt.% (from MMA) of 2,2′ -azobis (2-methylpropionitrile) was used as an initiator. The treated wood was then infiltrated under vacuum using the PEG/PMMA mixture and then sandwiched between two glass plates and covered with aluminum foil and placed for 4 hours at 70°C [63].

Lastly, PVP with a molecular weight of 13000000 and dissolved in ethanol (15% concentration) was used and infiltrated under vacuum. Then it was placed on the hot plate at 60°C until the solution was evaporated and the sample peeled off [46].

## **2.5.Problem Statement**

Due to climate change and global warming, there is an increasing need for sustainable solutions in all industries. As one of the most important and growing industries worldwide, the construction industry has a huge responsibility towards the environment. It is responsible for consuming vast amounts of resources and consequently generating massive quantities of waste. Also, as the building sector is responsible for consuming large portions of the world energy and electricity, such buildings' efficiency and sustainability must be considered. While the world is working on balancing between consuming less and recycling while maintaining the construction materials' effectiveness and efficiency, there is an immense need to develop more sustainable building materials that can increase the efficiency of buildings while decreasing the effects on the environment. Thus, introducing a translucent wood-plastic composite made out of wood (possibly wood waste) waste and plastic polymer (and possibly a percentage of recycled plastic waste) to be used in buildings cladding to reduce its carbon footprint through proper insulation and adequate natural lighting. Accordingly, decreasing the energy and electricity required to ventilate and temperature control and the need for natural lighting. Finally, saving tons of GHG emissions contributes to reducing global warming and air pollution and preserving our environment.

# 2.6.Objectives

- 1. To develop translucent wood composite from locally grown wood in Egypt.
- 2. To select the most efficient chemical treatment for locally grown wood species Casuarina & Eucalypts.
- 3. To optimize the fabrication process for translucent wood to be low energy consuming as possible and least hazardous effluents.
- 4. To fabricate translucent wood composite using wood waste.
- 5. To introduce a scalable chemical treatment method for translucent wood.
- 6. To compare the optical properties for translucent wood, pure epoxy, and glass.

# **Chapter 3: Materials and Methods**

## **3.1.** Materials

Sodium hydroxide (NaOH, Laboratory reagent grade, Fisher Scientific UK), sodium sulfite (Na<sub>2</sub>SO<sub>3</sub>, Analytical reagent grade, Fisher Scientific UK), hydrogen peroxide (H<sub>2</sub>O<sub>2</sub>, 50%, El Salam for Chemicals Industries), sodium silicate (Na<sub>2</sub>SiO<sub>3</sub>, Extra pure, Oxford Lab Fine Chem LLP), magnesium sulfate (MgSO<sub>4</sub>, Laboratory reagent grade, Fisher Scientific UK), diethylenetriaminepentaacetic acid (DTPA, Extra pure, Sisco Research Laboratories-India), Ethylenediaminetetraacetic acid (EDTA, Pure Lab. Chemicals, Free Trade Egypt), sodium chlorite (NaClO<sub>2</sub>, 80% extra pure, Loba Chemie-India), sodium acetate (C<sub>2</sub>H<sub>3</sub>NaO<sub>2</sub>, Anhydrous, Free Trade Egypt), acetic acid (CH<sub>3</sub>COOH, 96% pure reagent for analysis, Free Trade Egypt), bleach (Clorox-Egypt), ethanol (C<sub>2</sub>H<sub>5</sub>OH, 99.9%, ALAMIA-Egypt), acetone (C<sub>3</sub>H<sub>6</sub>O, 99.9%, Diachem Chemicals), sulfuric acid (H<sub>2</sub>SO<sub>4</sub>, for analysis, ADWIC), Tetrahydrofuran (C<sub>4</sub>H<sub>8</sub>O, stabilized with 0.1% quinol, Loba Chemie-India), Glass Microfiber filters (Grade G F/A (1.6 µm), 1820-047, Whatman), epoxy (3D, CMB-Egypt), and Eucalyptus & Casuarina samples were obtained from locally harvested tree logs.

## **3.2.Methods**

The general fabrication process can be shown in Figure 25. It starts with the wood samples preparation, chemical treatment (whether composed of one or two-stage process), then washing, dehydration, and finally the polymer infiltration. The samples, recipes, and methods used in each phase are mentioned in their appropriate sections.

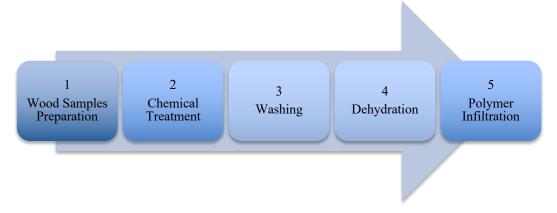


Figure 25: General fabrication process stages for transparent wood

## 3.2.1.1. <u>Wood Samples Preparation</u>

To begin with the wood samples preparation, the wood sample was prepared as follows in all the experimental phases. Two different logs were obtained from the different trees' species, one Casuarina & one Eucalyptus. These logs were plainsawn to obtain squared rods with dimensions of 4 cm by 4 cm by 50 cm. The squared rods were then cut radially (against the grain) to obtain wood samples with a 3 mm thickness; the obtained wood samples were 40 mm by 40 mm with a thickness of 3 mm from each log. The shape and size of the obtained samples can be shown in Figure 26. Similar samples were obtained with larger sizes of 60 mm by 60 mm and 80 mm by 80 mm with a thickness of 3 mm to test the effectivity of the process on larger sizes to investigate the scalability of the process.



Figure 26: Casuarina sample (Left) & Eucalyptus sample (Right)

Another set of wood waste samples from Casuarina were obtained to be used in this study. The samples obtained were from wood chips and sawdust. The wood chips were crushed and sieved, and two sets of sizes were retrieved; first is particles size between 2.25 & 4.75 mm and second with a particles size less than 2mm. The two different samples sizes can be shown in Figure 27. The last set of samples were Casuarina sawdust samples that were a product of the sawmilling, and it was sieved on sieve #4 (425  $\mu$ m), and the sample was collected from the passed-through sawdust.



Figure 27: Casuarina wood chips & sawdust samples

#### 3.2.1.2. Chemical Treatment & Polymer Infiltration

The second step is the chemical treatment, depending on the recipe followed, the treatment can be conducted on one stage or two stages. The chemical treatment aims to get rid of the lignin, which gives the wood samples its brownish color and prepares the samples for the polymer infiltration. The samples were bleached and delignified whether the treatment was a one or two-stage process. The details for each recipe used throughout the experimental work can be found below in the following sections. The third step is the samples washing, after the chemical treatment the samples were washed off the chemicals using distilled or deionized water to obtain clean white samples (at this stage, the cellulose and hemicellulose are the main components). The equipment used in the chemical treatment and washing were a hot plate, a flask and a condenser for the boiling solutions and beakers and parafilm for the nonboiling solutions. The setup can be illustrated in Figure 28.



Figure 28: Chemical treatment experimental setup using flask and condenser & beakers on hot plate

Afterwards the samples were completely dehydrated in the fourth step using ethanol and/or acetone. The samples were dehydrated on vacuum filter containers either using pure ethanol and pure acetone sequentially for three times or using four different concentrations of ethanol, starting with 25%, 50%, 75% and then pure ethanol. Afterwards, the samples were stored in pure ethanol and left in the fridge until use.



Figure 29: Dehydration setup using vacuum filter container

Finally, is the fifth step, which is the polymer infiltration. After dehydration, the samples were immersed in the polymer and vacuumed at 0.7-0.8 bar for five minutes and then depressurized using the setup shown in Figure 30. The vacuuming process was repeated at least three times to ensure the total infiltration of the samples. After the samples were completely infiltrated, the samples were kept in an oven at 30°C and left to dry for 8 h. The fabricated samples were left static for at least 24 h to harden.

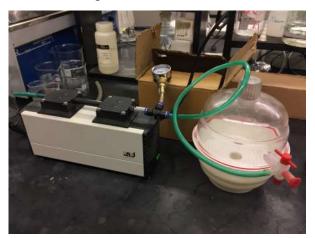


Figure 30: Vacuum infiltration setup using vacuum pump and a desiccator

The experimental plan was divided into four phases, the first phase is the pilot study for the chemical treatment in which different recipes were used on both wood species to optimize the experimental design and setup. The second phase was designed to test the three recipes' performance on the two wood species (Casuarina & Eucalyptus) and select the most promising one. The third phase is an in-depth study on the selected treatment method in which the amounts of lignin were measured, and the most promising experimental parameters were observed. Also, the samples were fabricated using epoxy resin and tested for their properties. In the third phase, an upscaling method was introduced by treating different wood wastes (different particle sizes), and samples were fabricated also using epoxy resin. Finally, in the fourth and last phase, transparent wood samples were fabricated and compared to glass samples.

#### **3.2.2.** Pilot Study – Chemical Treatment

For the first phase of the experimental work, three different methods were used to assess each recipe's effectiveness on the locally grown Egyptian wood species (Casuarina & Eucalyptus). Three recipes were selected from the literature and were used in this study. Two of the most commonly used methods in the literature and the substitution of the third one with commercially available bleach were used on wood samples. The first recipe used was the delignification method, which consists of a twostage process using sodium hydroxide & sodium sulfite for delignification and hydrogen peroxide solution for bleaching [2]. The second recipe used was the lignin modification method, which consists of a two stages process using alkaline hydrogen peroxide solution (H<sub>2</sub>O<sub>2</sub>), as the main component for bleaching, and sodium chlorite (NaClO<sub>2</sub>) for delignification [49]. The third recipe was the commercial bleach method using Clorox, based on the literature using a sodium hypochlorite solution (NaClO) as a one stage process [68]. The details for each recipe are as follows.

#### **Delignification** Method:

First is the delignification method, the delignification solution (first process) was prepared by dissolving 2.5 mol per liter of sodium hydroxide (NaOH) & 0.4 mol per liter of sodium sulfite (Na<sub>2</sub>SO<sub>3</sub>) in deionized water (DI); and the solution's pH was recorded. Two samples (40 mm by 40 mm by 3 mm) from each species were immersed in the solution then placed on the hot plate and kept boiling for at least 12 h. One sample from each species was taken out of the solution at 6 h and the other set of samples at the end of the treatment. The color of the samples was observed for each species at different time intervals.

After the samples were extracted from the solution, the samples were washed using boiling DI water to wash off the chemicals for 1 h. Afterwards, the bleaching solution (second process) was prepared by mixing 2.5 mol per liter of hydrogen peroxide ( $H_2O_2$ ) in deionized water. The samples were transferred and immersed in the bleaching solution and kept boiling for about 3 h.

#### Lignin Modification Method:

Second is the lignin modification method, the alkaline hydrogen peroxide solution (first process) was prepared by dissolving the following chemicals in deionized water (DI) at 70°C for at least 12 h. The chemicals were mixed in the following order: 3 wt.% of sodium silicate (Na<sub>2</sub>SiO<sub>3</sub>), 3 wt.% of sodium hydroxide (NaOH), 0.1 wt.% of

magnesium sulfate (MgSO<sub>4</sub>), 0.1 wt.% of EDTA (instead of the DTPA because it wasn't available in the market), and finally 4 wt.% of hydrogen peroxide (H<sub>2</sub>O<sub>2</sub>); and the solution's pH was recorded. Two samples (40 mm by 40 mm by 3 mm) from each species were then immersed in the solution. Similar to the previous methods, one sample from each species was taken out of the solution at 6 h and the other set of samples at the end of the treatment.

As for the delignification process (second process), 1 wt.% of sodium chlorite (NaClO<sub>2</sub>) was dissolved in an acetate buffer solution with a pH of 4.6. Since the available sodium chlorite has an 80% purity, the weight was corrected in the solution preparation calculations. According to the literature, the acetate buffer solution was prepared by preparing 0.2 M (moles) of sodium acetate (1.64g of sodium acetate in every 100ml DI) and 0.2 M of acetic acid (1.143 ml of acetic acid in every 100ml DI). Both solutions were then mixed, starting with the sodium acetate and adding the acetic acid until the desired pH of 4.6 was reached. Afterwards, the sodium chlorite was added and mixed in the buffer solution until total dissolving. The samples were then immersed in the delignification solution and placed on the hot plate at 80°C for about 3 h.

## Commercial Bleach (Clorox):

Finally, the sodium hypochlorite solution, instead of preparing the 5 wt.% solution, commercial bleach (Clorox) with the same concentration, was used. One sample (40 mm by 40 mm by 3 mm) from each species were immersed in the solution and left in room temperature for 30 h.

#### 3.2.3. Phase I: Performance of Chemical Treatment Methods

Based on the outcomes of the previous stage, some modifications have been made to the experimental design. The same concentrations for the three recipes as stated in the previous stages have been used except for the modified method recipe in which the DTPA was used instead of the EDTA as mentioned in the literature [49]. Also, the solution to the sample ratio for the commercial bleach was adjusted according to the literature.

In this phase, samples from both species (Casuarina & Eucalyptus) were treated by the three recipes to compare the effectiveness of the treatment. For the delignification method, ten samples from each species (Casuarina with a total weight of 29.65 g & Eucalyptus with a total weight of 31 g) were immersed in the delignification solution (first solution) in two separate beakers and placed on the hot plate and kept boiling for 12 hours. A sample from each species was taken out every three hours; a total of four samples from process one. To ensure the efficiency of the treatment, the solution was replaced with a fresh one once the reaction stopped in which the solution was saturated. The solution was changed for the Eucalyptus 6 times in total at time points 30 min, 1 h, 2 h, 3 h, 6 h and 9 h of treatment. While for the Casuarina samples, the solution was changed 5 times in total at time points 1 h, 2 h, 3 h, 6 h and 9 h of treatment although it was not totally consumed in the last time point. The rest of the samples were rinsed with DI water then transferred to the bleaching solution (second solution) for six hours, and a sample was taken out every two hours; a total of three samples. The solution was changed 3 times in total at time points 30 min, 90 min, and 4 h of treatment.

Similar to the delignification method, the modified method bleaching solution (first solution) was prepared (using DTPA). Ten samples from each species (Casuarina with a total weight of 29.5 g & Eucalyptus with a total weight of 30.5 g) were immersed in it and left on the hot plate at 70°C for 12 hours. A sample from each species was taken out every three hours; a total of four samples. The solution for the Eucalyptus was changed 2 times in total at time points 2 h and 6 h of treatment. While for the Casuarina samples, the solution was changed 2 times in total at time points 5 h and 9 h of treatment. Afterwards, the rest of the samples were rinsed with DI water transferred to the delignification solution (second solution) and placed on the hot plate at 80°C for six hours. A sample was taken out every two hours, with a total of three samples, and the solution was changed 1 time for each species.

Finally, the commercial bleach was used to treat eight samples from each species with the sample to solution ratio of 1:60 according to the literature [68]. The eight Casuarina samples' weight was 23.67 g, so 1420.2 g of Clorox were added while the Eucalyptus samples weighted 24.36 g, so 1461.6 g of Clorox were added. Both beakers were covered and left at room temperature for 24 hours, and a sample was taken out every three hours; a total of eight samples.

#### 3.2.4. Phase II: Lignin Modification Method – Lignin Quantification

## 3.2.4.1. <u>Chemical Treatment</u>

According to the first phase observations, the lignin modification method was selected as it was the most promising one. In this phase, the selected method was used to treat both wood species and study the change of lignin content during the treatment process. As mentioned in the pilot phase, the solutions have been prepared (using EDTA) and mixed in the same order. For each species, 15 samples were used in the treatment in which a sample was extracted every 3 h from the  $H_2O_2$  alkaline solution for a total of seven samples to be tested for lignin. The total treatment time for the first process was terminated after 21 h. To ensure the chemical treatment's efficiency, the solution was replaced with a fresh one once the reaction stopped. The solution was changed four times in total at time points 1, 4, 9, and 15 h of treatment. The extracted samples were then washed in boiling DI water for 1 h and dehydrated for 3 h using pure ethanol. Afterwards, the rest of the samples were transferred to the delignification solution, and a sample was extracted every hour for a total of 5 samples for the lignin quantification. The second process's total time was 5 h in which the solution was changed 3 times in total at time points 1, 2, and 4 h of treatment. The extracted samples were then washed in boiling DI water for 1 h and dehydrated for 1 h using pure ethanol. A total of 12 samples were extracted from the two-stage treatment process for each species' lignin quantification.

#### 3.2.4.2. Lignin Quantification

The lignin quantification was conducted according to the Klason lignin method in TAPPI T 222 om-02 "Acid-insoluble lignin in wood and pulp" and corrected for the ash content according to TAPPI T 413 om-93 "Ash in wood, pulp, paper and paperboard: combustion at 900°C"[39], [70]

## 3.2.4.3. Washing & Dehydration

After the chemical treatment, the samples must be washed to get rid of the chemicals. Thus, the chemicals were washed off by boiling the samples in deionized water for 2 hours. The samples were then dehydrated using different concentrations of ethanol, starting with 25%, 50%, 75% and then pure ethanol. The samples were left in each solution for 15min and transferred to the next one then was transferred in the pure ethanol solution and stored in the refrigerator until use.

## 3.2.4.4. Polymer Infiltration

One sample from each species was infiltrated using epoxy resin. The epoxy was prepared according to the manufacturer's recommendation of mixing the polymer (A) and the initiator (B) with a ratio of 3A:1B. The samples were immersed in the epoxy (using silicone molds) and vacuumed using a vacuum desiccator and a vacuuming pump for 5 minutes and then depressurized. The cycle was repeated at least three times

to ensure full infiltration. Finally, the infiltrated samples were placed in an oven at 30°C for 8 h and left to the next day to dry (samples were left static for 24 h to harden) and 5mm thick samples were obtained.

#### 3.2.5. Phase III: Transparent Wood from Wood Waste

After the trials on wood pieces, wood waste was used to study the process's scalability instead of relying on small wood pieces. Two types of wood waste were used, wood fibers and sawdust with sizes ranging from 4.75 mm to fine sawdust particles. In this phase, the samples were fabricated using epoxy resin to be comparable to the wood pieces fabricated in the previous phase. The fabrication process steps can be illustrated in Figure 33.

## 3.2.5.1. Wood Fibers

As mentioned above, two sizes of wood chips were used, a sample with a particle size between 2.25 mm & 4.75 mm and another with particle size less than 425 µm. Both samples were treated using the modified recipe for the chemical treatment and epoxy resin as the polymer for the infiltration. The chemical treatment was conducted using the previously stated two-stage modified recipe method. The H<sub>2</sub>O<sub>2</sub> alkaline solution was prepared as previously stated and placed on the hot plate at 70°C for 15 h. The solution was discarded and replaced with a new solution when the reaction stopped. The solution was changed 5 times in total at time points 1, 4, 7, 9, and 11 h of treatment. The samples were then washed for 4 h in boiling DI water to wash off the chemicals. For the delignification process, the solution was prepared according to the previously stated methods and placed on the hot plate at 80°C for 5 h. The solution was changed two times in total at time points 1.5 and 3.5 h of treatment. The samples were then washed for 2 h in boiling DI water to wash off the chemicals. After dehydration, the samples were stored in pure ethanol in the refrigerator until use. The samples were infiltrated using two methods. First, a sample was vacuumed on a filter container to form a thin disk and then infiltrated with the epoxy as the previously stated method. Second, after dehydration, the samples were mixed with the epoxy and vacuumed (with 20 wt.% of the wood sample). In both cases, after the vacuuming at - 0.8 bar, the samples were placed in an oven at 30°C for 8 h and left static for a total 24 h to harden and 5mm thick samples were obtained. The samples were fabricated using silicon molds, as mentioned before.

#### 3.2.5.2. <u>Sawdust</u>

Similar to the wood chips, the sawdust was treated using the modified recipe and fabricated using epoxy resin. The sawdust used was a product from sawmilling sieved on sieve #40 (425  $\mu$ m). Similar to the above-stated methods, the sawdust samples were fabricated by mixing it with the epoxy and compared to the wood chips samples.

#### 3.2.6. Phase IV: Casuarina Transparent Wood & Glass

### 3.2.6.1. Chemical Treatment

According to the previous phases' observations, the lignin modification method was used to treat Casuarina wood pieces using DTPA instead of the EDTA, as mentioned in the literature[49]. The solutions have been prepared as mentioned in the previous phase and mixed in the same order. In this phase, 12 Casuarina samples with a total weight of 28.7 g were used in the treatment for a total of 14 hours in the H<sub>2</sub>O<sub>2</sub> alkaline solution at 70-80°C. To ensure the efficiency of the treatment, the solution was replaced with a fresh one once the reaction stopped. The solution was changed three times in total at time points 3, 6, 12 h of treatment. Afterwards, the samples were transferred to the delignification solution at 80-85°C. The second process's total time was five hours in which the solution was changed one time in at time point 2 h of treatment. The extracted samples were then washed in boiling DI water for 1 h and dehydrated using pure ethanol and left in the fridge till use.

## 3.2.6.2. Polymer Infiltration

Six treated wood samples were infiltrated using epoxy resin. The epoxy was prepared according to the manufacturer's recommendation of mixing the polymer (A) and the initiator (B) with a ratio of 3A:1B. The samples were immersed in the epoxy (using rectangular silicon molds) and vacuumed using a vacuum desiccator and a vacuuming pump for 5 minutes at -0.9 bar and then depressurized. The cycle was repeated 3 - 5 times to ensure full infiltration. Finally, the infiltrated samples were placed in an oven at 30°C for 8 h and left to the next day to dry (the samples were left static for at least 24 h to harden). The resulting samples were around 60 mm × 40 mm × 5mm (the TW was about 40 mm × 30 mm). Similarly, six pure epoxy samples were poured as a reference with the same dimensions. In this phase a bigger vacuuming pump was used to ensure better polymer infiltration as shown in Figure 31.

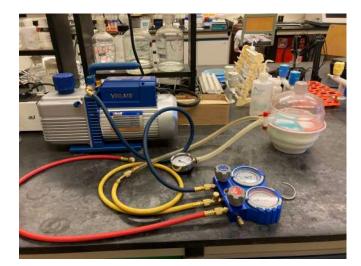


Figure 31: Polymer infiltration modified setup with a bigger vacuum pump

# 3.2.6.3. Epoxy & Glass Samples

Six samples from three different types of glass were obtained: regular transparent glass, sanded glass, and rain glass as shown in Figure 32. Samples were 40 mm  $\times$  60 mm  $\times$  5mm. Also, as a reference for transparent wood samples, six pure epoxy samples were prepared. The epoxy was prepared according to the manufacturer's recommendation of mixing the polymer (A) and the initiator (B) with a ratio of 3A:1B. The epoxy was poured in rectangular silicon molds and vacuumed using a vacuum desiccator and a vacuuming pump for 5 minutes at -0.9 bar and then depressurized.



Figure 32: Epoxy samples & regular, sanded, and rain glass samples

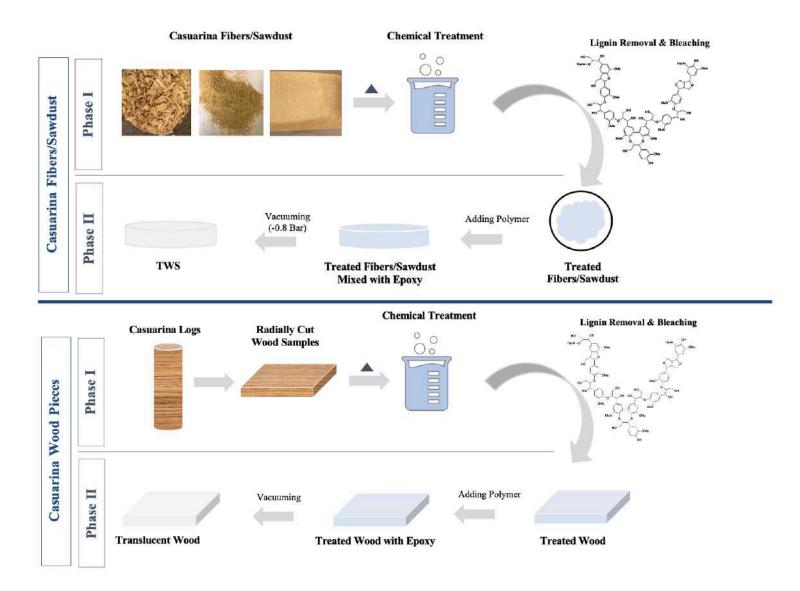


Figure 33: Translucent wood fabrication schematics for wood fibers/sawdust (upper schematic) and radially cut wood (lower schematic) showing the stages from sample preparation, chemical treatment, and polymer infiltration.

#### 3.2.7. Phase V: Sawdust Treatment using Commercial Bleach

In order to introduce a scalable process for the fabrication of transparent wood, two factors have to be considered. First, transparent wood fabricated using wood pieces or veneers is limited to the available sizes of such materials and an increasingly challenging chemical treatment setups to treat the desired sizes. Second, energy-intensive and expensive chemical treatment methods are obstacles in the way of producing feasible transparent wood. Thus, in this phase, an efficient chemical treatment process was used to treat sawdust samples to produce a feasible and easy to upscale the fabrication process. Since sodium hypochlorite is the main component in commercial bleach (Clorox), an experiment was designed to test the efficiency of the treatment on sawdust samples. The Clorox used was in the first three months of production date to ensure the solution's efficiency. Three different solution to sawdust ratios were used to minimize the amount of solution used, starting with 1:20, 1:40, and finally, 1:60, as mentioned in the literature [68]. Three flasks containing 5g of Casuarina sawdust were used, and then each one was filled with a different ratio of sawdust to bleach, as shown in Figure 34.

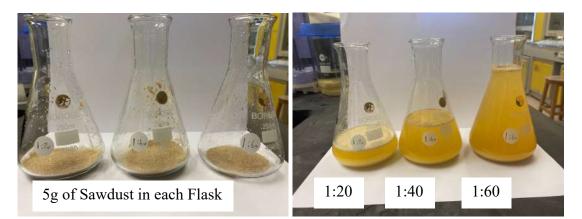


Figure 34: Casuarina sawdust treatment using commercial bleach with 3 different sample to solution ratios

The setup has been repeated three times for three different time points of treatment starting with 24 h, 48 h, and finally 72 h. The sample colors were visually observed at different time intervals, and then an FTIR spectroscopy was conducted at the end of each treatment for the nine runs and compared to the initial sample. Since FTIR is a qualitative method, the main aim was to detect the amount of lignin present at the end of each treatment for the three ratios at the different treatment times and compare it to the treated samples' color. The samples were was for 1 h in boiling DI water at the end of the treatment and air dried for the FTIR spectroscopy.

### 3.3. Characterization & Testing

#### 3.3.1. Lignin Quantification - Klason Lignin

#### Lignin Content

The lignin quantification was conducted according to the Klason lignin method in TAPPI T 222 om-02 "Acid-insoluble lignin in wood and pulp". In summary, the samples were grinded and sieved to the desired particle size, then sulfuric acid (H<sub>2</sub>SO<sub>4</sub>) was used to dissolve the cellulose and hemicellulose. After the dissolving process is finished, the samples were filtered and washed using glass microfiber and glass crucible to collect the lignin left in the sample and then dried to constant weight and lignin content was calculated. The percentage of lignin was calculated according to the following formula [39]:

$$Lignin\ content = \frac{weight\ of\ sample\ (g)}{oven-dry\ weight\ of\ sample\ (g)}$$
(Equation 1)

The detailed process of determining the lignin is as follows. First, the samples were grinded and sieved on sieve #40 (425  $\mu$ m). From each test specimen, two samples were weight (0.2 g each, which is a deviation from the protocol that suggests 1 g sample) to be used as a technical replica and another sample was taken to determine the moisture content. After weighing, the samples were placed in autoclave bottles (250 ml), and 72% sulfuric acid was added (3 ml, corrected for the 0.2 g sample) in each bottle and mixed with the sample. Afterwards, the autoclave bottles were placed in a water bath at  $20 \pm 1^{\circ}$ C for two hours with occasional mixing. Then distilled water was added (112 ml corrected for the 0.2 g sample) to each bottle to decrease the concentration of the sulfuric acid to 3%. After decreasing the sulfuric acid concentration, the bottles were placed inside an autoclave for a 30min cycle (another deviation from the protocol, which states four hours boiling on a hotplate). The autoclave temperature reached 122°C and kept for 30 min; the pressure was 16 psi. The autoclave bottles were then taken out of the autoclave and left overnight to settle. According to the followed protocol, the flask (in our case bottles) should be kept inclined for the lignin particles to settle. The purpose of this step was to ease the vacuum filtering process by decanting the supernatant into the filter before the lignin to avoid the blockage of the filter. Since the samples were five times smaller, this process was unnecessary, so the bottles were kept upright to settle overnight. The following day after the samples settled, the supernatant for each sample was decanted into a glass crucible with glass microfiber filter (1.6 μm pores)

then the lignin, which was washed three times with hot distilled water. Finally, the glass microfiber filters containing the lignin were placed in an oven at  $105 \pm 3^{\circ}$ C until the weights recorded were constant. For the process to be as accurate as possible, a correction has been made for the moisture content of the samples and the ash content. The detailed process can be illustrated in Figure 35.



Figure 35: Klason lignin experimental stages starting with sample grinding and treatment with sulfuric acid to filtration, washing, and drying to constant weight

## Moisture Content

The remaining of the grinded samples were collected and weighted for each time interval and dried. The samples were dried at  $105 \pm 3^{\circ}$ C using an oven until the weights recorded were constant. The moisture content was calculated using the following equation:

Moisture content (%) =  $\frac{\text{initial weight of sample (g)} - \text{sample weight after drying (g)}}{\text{sample weight after drying (g)}} \times 100$  (Equation 2)

## Ash Content

The determination of ash was conducted according to TAPPI T 413 om-93 "Ash in wood, pulp, paper and paperboard: combustion at 900°C" [70]. Four samples from each species were obtained for this test with a particle size between 4.75 & 2.25 mm. Three samples were used for the ash content determination while the fourth sample was used for moisture content. The samples were inserted into ceramics crucibles with covers and then placed inside the furnace and ignited at 900°C for a total of 8 h. The percentage of ash was calculated based on the following equation [70].

Ash (%) = 
$$\frac{\text{weight of ash (g)}}{\text{oven-dry weight of sample (g)}} \times 100$$
 (Equation 3)



Figure 36: Samples (Left) – inside furnace & (Right) - after combustion

### 3.3.2. Optical Properties

#### 3.3.2.1. <u>Transmittance</u>

The transmittance was measured using a two-beam light UV spectrophotometer with an integrated sphere using barium sulfate as the white standard. The equipment used was SHIMADZU UV-2600i (UV-VIS spectrophotometer) with a range from 220 to 1500 nm. First, a calibration was made with the white standard to make sure 100 percent was obtained, and then the samples were placed in front of the integrated sphere, and the transmittance was measured.

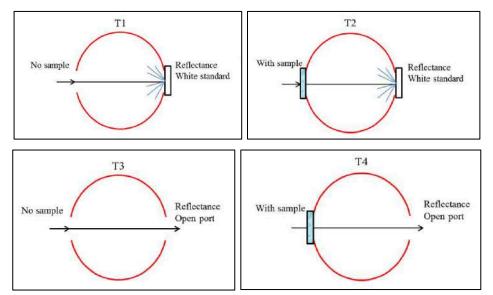
### 3.3.2.2. <u>Haziness</u>

Similar to the transmittance, the same equipment was used as well as the white barium sulfate standard. The measurements were done according to ASTM D1003 "Standard Method for Haze and Luminous Transmittance of Transparent Plastics". The result can be expressed in **Error! Reference source not found.**:

Haze (%) = 
$$\left[\frac{T4}{T2} - \frac{T3}{T1}\right] \times 100$$
 (Equation 4)

In summary, four readings were obtained at different setups, and then the haze was calculated accordingly. The four different setups are as follows:

- T1 was recorded with the white standard without the sample.
- T2 was recorded with the sample and the white standard.
- T3 was recorded after removing both the sample and the white standard
- T4 was recorded with the sample and without the white standard.



*Figure 37: Haze configurations setup using UV spectrophotometer with an integrated sphere* [1]

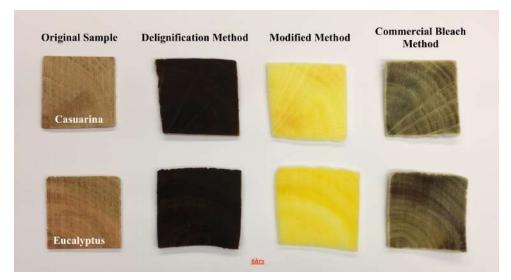
# **Chapter 4: Results & Discussion**

# 4.1. Pilot Study Trials & Observations

The pilot study aimed to test the effectiveness of different treatment methods for both Egyptian wood species (Casuarina & Eucalyptus) and optimize the parameters for the chemical treatment setup. Moreover, the possibility of dissolving plastic waste to be incorporated with the polymer used for infiltration was investigated. This phase was divided into two stages. The first stage was the pre-trial, in which the treatment for the samples was monitored, and the color of the samples was observed. The second stage was dissolving plastic waste in THF with different waste to solvent ratios, and the results were visually observed.

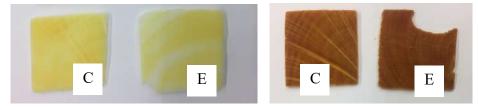
## 4.1.1. Chemical Treatment Observations Pre-trials

The three chemical treatment recipes were used to treat both wood species (Casuarina & Eucalyptus) and evaluated accordingly. First, two pieces of each wood species were immersed in the three different recipes, and the results were compared visually after 6 h and 12 h of treatment for the first solution. After 6 h, one piece of each wood species was taken out of the first solution and immersed in the second solution (for each recipe except for the bleach) while the other pieces were left in the solution for another 6 h. Figure 38 shows the change in color of the first set of samples after 6 h treatment before the second solution treatment. The initial observation showed that the modified method had the best treatment compared to the other methods.



*Figure 38: Treated samples using delignification method, lignin modification method, and commercial bleach method for Casuarina and Eucalyptus species compared to the original samples after 6 h of treatment* 

When comparing the color change, the delignification samples turned to darker brown color compared to the original samples, and the modified samples turned to a yellowish color, while the bleached samples turned to faded but darker brown color. Afterwards, the first set of samples (6 h treatment) of the modified and delignification methods were transferred to the second solution for each recipe and left for 3 h, while the bleached samples were transferred to the bleach again. The color of the samples of the first two methods was observed compared to each other. The 6 h modified samples were lighter yellowish in color compared to the delignification samples were light brownish. Moreover, the delignified samples were fragile and broke easily. The results can be shown in Figure 39.



*Figure 39: Lignin modification samples after 6 h of treatment (Left) & Delignification method after 6 h (Right)* of the second treatment process

Figure 40 shows the change in color of the first set of samples after 12 h treatment before the second solution treatment. The samples had the same pattern as the 6 h treated samples regarding its color compared to the original samples showing the modified method as the lightest in color. However, the 12 h samples of the delignification and modified samples were darker in color compared to the 6 h samples, which contradicts the purpose of more extended treatment. In order to confirm the observations, the samples were washed in DI water to see if there was a change of color. In Figure 40, the delignified samples showed no change in color after washing, which meant that the treatment process was responsible for the darker color. On the other hand, the commercial bleach samples showed improvement (lighter color), but all samples were far from turning white.

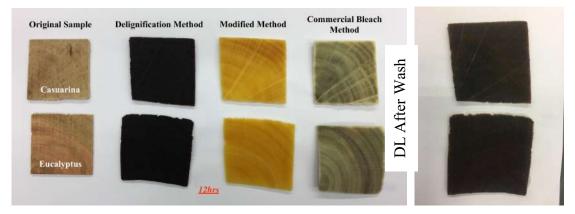


Figure 40: Treated samples using delignification method, lignin modification method, and commercial bleach method for Casuarina and Eucalyptus species compared to the original samples after 12 h of treatment

Furthermore, the modified 12 h samples were transferred to the delignification solution (second solution) to complete the treatment and compared to the fully treated 6 h samples. Comparing the two sets of samples, the 12 h samples showed better treatment with a light yellowish color, but the samples looked stained and were not turning white even when left for a longer time. Moving to the commercial bleach samples, the samples were left for a total of 30 h, and parts of it turned to a light yellowish color while brown strokes remained visible in both species. The results can be shown in Figure 41.

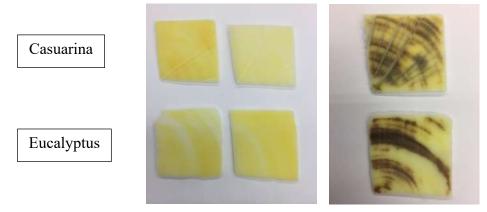


Figure 41: Lignin modification samples after 6 h of treatment(left) and 12 h (middle) & commercial bleach after 30 h of treatment (right) for Casuarina and Eucalyptus species

#### 4.1.2. Plastic Waste Dissolving

In this phase, polystyrene plastic waste was dissolved in THF using four different waste to solvent ratios. The plastic waste to solvent ratios were 1:2, 1:5, 1:8, 1:12 and was mixed until it totally dissolved at room temperature. The solution was poured in a silicone mold and vacuumed and left to dry overnight. The observations can be illustrated in Figure 42, showing a tradeoff between wrinkling and transparency. However, the solution was too viscous and very cloudy to be used as the polymer since the polymer desired to be very transparent and less viscous to infiltrate the wood samples properly. Thus, discounting the plastic waste dissolving and epoxy resin were used in future trials.

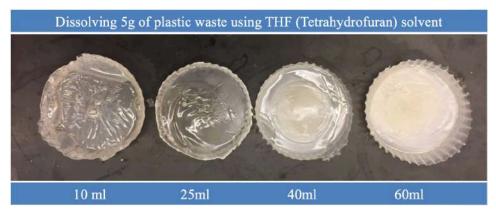


Figure 42: Plastic waste mixed in different concentrations of Tetrahydrofuran (THF)

# 4.2. Performance of Chemical Treatment Methods

The delignification method, lignin modification method, and commercial bleach were used to treat the Egyptian hardwoods (Casuarina & Eucalyptus) samples to evaluate their effectivity. Based on the previous stage observations, the solutions were replaced once the reactions stopped to avoid the samples staining and ensure effective treatment. Regarding the delignification and lignin modification treatments, the first solution process is indicated as I and the second process as II in the figures below. Also, regarding the commercial bleach, wood to solution ratio of 1:60 was used instead of just covering up the samples with the solution and was left for 24 h. The samples comparisons can be shown in the figures below, showing each treatment method's progress at comparable time points.

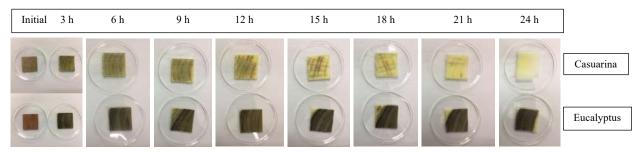


Figure 43: Commercial bleach treatment for Casuarina and Eucalyptus at different intervals showing the progress of treatment

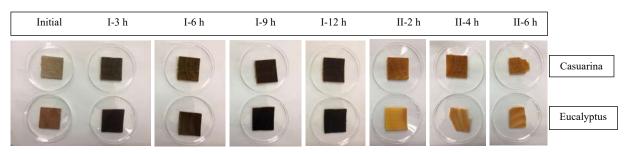


Figure 44: Delignification treatment for Casuarina and Eucalyptus at different intervals showing the progress of treatment

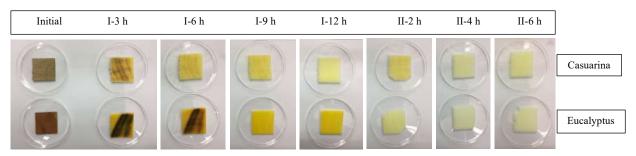


Figure 45: Lignin modification treatment for Casuarina and Eucalyptus at different intervals showing the progress of treatment

The commercial bleach treatment showed promising results but was only effective for Casuarina and had minimal effect on the Eucalyptus, especially on the heartwood areas. Also, at the end of the treatment (after 24 h), the samples were very fragile and easily broken and still had a yellowish fade color. Moving to the delignification method, at the end of the treatment, both species did not turn white and were broken up into little pieces due to the removal of lignin. Although it was more effective on the Eucalyptus samples, the results were not encouraging to proceed with. On the other hand, the lignin modification method showed very promising outcome since it worked on both species and the samples turned white much faster compared to the two other recipes even for the heartwood. Moreover, the process didn't produce as much as black liquor as the delignification method for both species compared to the delignification method, which had far less treatment effect on both species and the lignin modification method, which was more efficient to treat Casuarina samples only. To sum up, the lignin modification method was the most efficient on both species with the least treatment time and least black liquor produced compared to delignification method.

According to the literature, the lignin's presence is a characteristic of aromatic compounds peaks at 1505 cm<sup>-1</sup> in the FTIR spectrum [49], [71]. Thus, the absorbance peaks at wavenumber 1505 cm<sup>-1</sup> were observed to compare the lignin presence in each sample. Based on the results after 12 hours of treatment for the three recipes compared to the untreated Casuarina wood, there was no presence of lignin in the commercial bleach method while the lignin modification method had less amount of lignin than the delignification method. Theoretically, this should not have been the case if the wood samples were fully treated by the lignin modification method at a shorter time than the delignification method. Figure 46 illustrates the FTIR spectrums for the three recipes after 12 h of treatment compared to the untreated Casuarina wood.

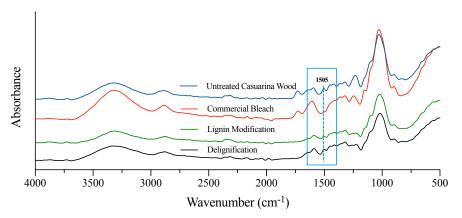


Figure 46: FTIR spectrums for untreated Casuarina wood, commercial bleached sample, lignin modified sample, and delignified sample after 12 h of treatment.

After 18 h of treatment by the lignin modification method and the delignification method, the FTIR showed no presence of lignin in the lignin modification sample and very little lignin in the delignification sample. Moreover, after 24 h, the commercial bleach treatment showed no presence of lignin like the lignin modification method. Figure 47 shows the FTIR spectrums for the three recipes compared to the untreated Casuarina wood. Although the lignin was removed by the commercial bleach and lignin modification methods, no black liquor was produced due to the treatment process as the delignification process produced. Thus, confirming that the lignin modification method and the commercial bleach method are the most promising treatments for Casuarina.

Similar to Casuarina, after 12 hours of treatment for Eucalyptus, the commercial bleach and lignin modification methods showed lower lignin presence than the delignification method. Figure 48 shows the difference in peaks at 1505 cm-1 for the three recipes and the untreated Eucalyptus wood. On the other hand, at the end of the treatment (18 h for LM & DL, and 24 h for CB), the lignin modification treatment method showed no presence of lignin. In contrast, both the commercial bleach (unlike for Casuarina) and delignification methods still had presence of lignin. Thus, confirming the visual observations that showed the lignin modification method to be the most effective method for both wood species. Figure 49 illustrated the FTIR results for untreated Eucalypts wood and the three treatment methods mentioned above.

Linking the visual observations with the FTIR results, it can be concluded that the delignification method is not efficient for treating Casuarina and Eucalyptus wood species since the treated samples did not turn white nor the lignin was totally removed. Also, black liquor was produced as a result of the treatment process, which is considered not environmentally friendly due to its difficult disposal. Furthermore, the commercial bleach treatment was much more efficient for Casuarina (samples turned white) than for Eucalyptus, which had very little change in color through the treatment process. In addition, the samples lost a lot of its strength at the end of the process due to the long treatment time, which made it very difficult to wash and dehydrate the samples to prepare it for polymer infiltration. Finally, the lignin modification method proved to be the most effective treatment method for both species by turning the sample white and removing the lignin while retaining a little bit of the sample's strength. Moreover, no black liquor was produced from the process, which required constant boiling of the solutions through the treatment process.

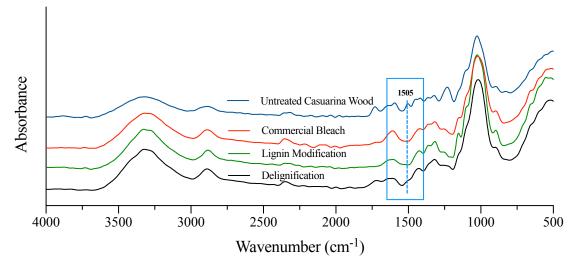


Figure 47: FTIR spectrums for untreated Casuarina wood, commercial bleached sample, lignin modified sample, and delignified sample at the end of treatment.

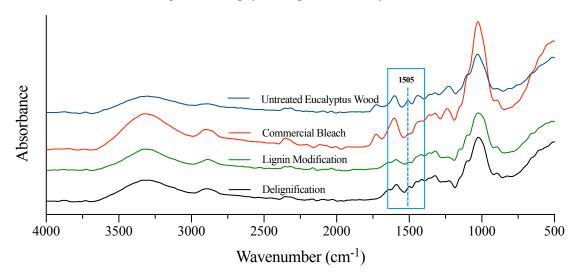


Figure 48: FTIR spectrums for untreated Eucalyptus wood, commercial bleached sample, lignin modified sample, and delignified sample after 12 h of treatment.

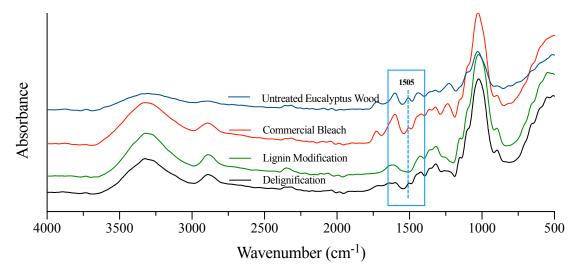


Figure 49: FTIR spectrums for untreated Eucalyptus wood, commercial bleached sample, lignin modified sample, and delignified sample at the end of treatment.

### 4.3. Lignin Quantification (Lignin Modification Method)

To better understand the chemical treatment effect, both species' lignin content was quantified using Klason lignin throughout the lignin modification chemical treatment. First, the initial lignin content of Casuarina and Eucalyptus were measured and corrected for the moisture content and the ash content. The ash content for Casuarina was about 1.18% (ash content for Eucalyptus was assumed to be the same as Casuarina). The total treatment time was 26 h for both solutions; the bleaching solution treatment time was 21 h while the delignification solution was 5 h. Seven intervals were selected for the bleaching process (every 3 h) and analyzed and five intervals for delignification process (every hour). The first solution process (H<sub>2</sub>O<sub>2</sub> alkaline bleaching) is indicated as I and the second process (sodium chlorite delignification) as II. The lignin content at the 12 different time points during the chemical treatment was obtained and recorded in Table 8. The initial lignin content for Eucalyptus was around 6% more than the Casuarina, which indicates a longer treatment time. However, the drop in lignin content in both species was comparable throughout the treatment time until towards the end of the first process and beginning of the second process. Starting the second process, the Casuarina samples almost lost all its lignin content while the Eucalyptus still required further treatment. After the first hour, the lignin content for the Casuarina samples could not be detected. On the other hand, the Eucalyptus still had some lignin content until the end of the second process. Figure 50 illustrates the change in lignin content for both species.

Treatment Time (h)	Lignin (%)	
	Eucalyptus	Casuarina
Initial	35%	29%
I-3	23%	20%
I-6	20%	20%
I-9	16%	18%
I-12	15%	15%
I-15	16%	13%
I-18	13%	10%
I-21	12%	8%
II-1	10%	7%
II-2	8%	1%
II-3	4%	0%
II-4	3%	0%
II-5	2%	0%

Table 8: Casuarina & Eucalyptus lignin quantification results at different intervals

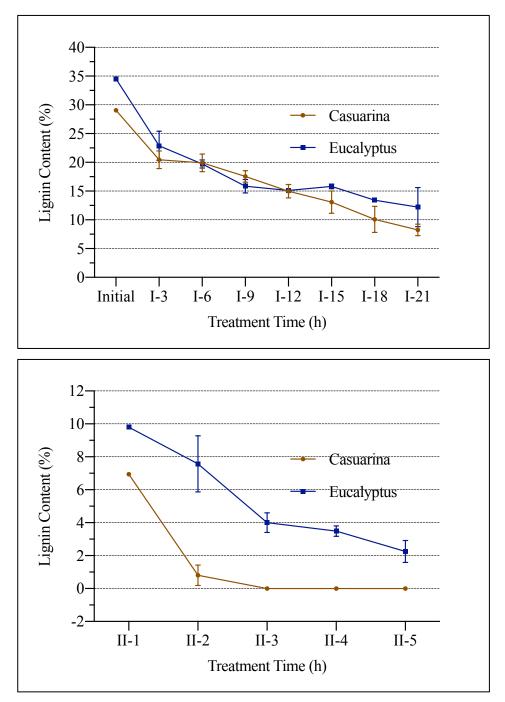


Figure 50: Lignin content for Casuarina & Eucalyptus (using Klason Lignin) during the treatment process using lignin modification method for the first process (upper) and second process (lower)

According to the obtained results, Casuarina samples were easier to treat than the Eucalyptus samples, especially that it has lower initial lignin content. After the bleaching stage, the casuarina samples were almost treated to the point which required little to no further treatment with the delignification solution. The Casuarina samples almost had no lignin content after the first hour of the delignification process after the 21 h of bleaching. Showing a better overall behavior over the Eucalyptus; thus, choosing Casuarina for further investigation.

### 4.4. Transparent Wood (TW) & Transparent Wood Fibers (TWF)

Transparent wood was fabricated using Casuarina wood treated using the lignin modification method and infiltrated using epoxy resin. While transparent wood fiber was fabricated using the same recipe and adding 20 wt.% of sawdust to the epoxy resin. Finally, pure epoxy samples were poured as a reference with the same dimensions. The results of this phase can be shown in Figure 51 labelled as mentioned below.

Regarding the wood waste samples, two infiltration methods were used to fabricate transparent wood samples. First, as mentioned in the literature, a thin disc made from the wood fibers (between 2.25 & 4.75 mm) and then immersed in the epoxy and vacuumed (S1). Unfortunately, the binding was unsuccessful, and it made a totally white sample with no visibility. Accordingly, for the second sample, the wood fibers (between 2.25 & 4.75 mm) were mixed with the epoxy and vacuumed (S2), but it had the same issues as the previous one. Thus, excluding the wood fibers between 2.25 & 4.75 mm from future trials.

Regarding the wood fibers with particle size less than 2 mm, the samples were fabricated using both infiltration methods. The thin disc infiltration method produced an epoxy sample with the wood fibers clearly visible inside (in white – S3) while the mixing technique produced an opaque sample that showed clusters of fibers that obstructed its transparency (S4). Finally, the sawdust samples were fabricated using both methods and compared. The thin disc method made the sample cloudier (S5) than the mixing method (S6), which was the most promising one due to its visibility. Thus, choosing the sawdust and the mixing method for further trials.



Figure 51: Transparent wood sawdust samples & wood fibers after polymer infiltration

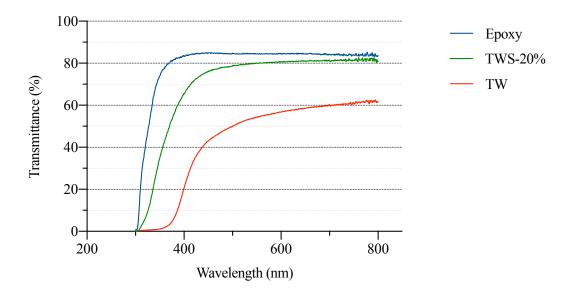
Two square silicone molds were used for the epoxy and Casuarina TW samples while a circular mold was used for the sawdust. The Casuarina TW color was yellowish compared to the other two samples but still was translucent, as shown in Figure 52.



Figure 52: Epoxy sample (left), TW Casuarina using lignin modification method and epoxy (middle), & TW Sawdust using lignin modification method and epoxy (right)

The three fabricated samples' optical properties were measured in the visible light range (400 - 800nm) using UV spectrophotometer with an integrated sphere. The transmittance for pure epoxy was the highest with around 84% while TWS had around 80% transmittance and lastly was the Casuarina transparent wood sample with around 62%. The results for TW, TWS, and epoxy can be illustrated in Figure 53. Although 84% is relatively high, pure epoxy was expected to have a higher transmittance; especially it is an ultra-transparent epoxy used for coating wall art and paintings. The cause of such phenomena might be attributed to the impurities in the mix during the fabrication process or due to the sanded finish of the square silicone molds used in this experiment. Unlike the squared silicone mold, the circular mold had a very smooth finish, which might explain the little difference between TWS and the pure epoxy. Thus, the silicone molds were used. Aside from the epoxy sample finish problem, the transparent wood fabricated using sawdust showed very promising results.

To sum up, regarding the polymer infiltration step, the mixing technique was selected over the thin disk due to the incomplete polymer infiltration as well as the clouding effect displayed in the sawdust sample (S5) in Figure 51. Also, the sawdust showed much better results compared to the two sizes of the wood fibers. Thus, choosing to proceed with sawdust over the wood fibers using the mixing technique to ensure better polymer infiltration. Moreover, the silicone molds used were replaced with new similar silicone molds with smooth surfaces to ensure better sample finish and comparable results for all samples.



*Figure 53: Light transmittance of TW & TWS & Epoxy samples measured using UV spectrophotometer with an integrated sphere* 

According to **Error! Reference source not found.**, haziness was calculated using UV spectrophotometer with an integrated sphere in the visible light range (400 - 800nm). The results for epoxy and TWS can be illustrated in Figure 54. Similar to the transmittance, the haziness results were affected by the sanded finish of the epoxy sample. Although the pure epoxy should not be scattering light, the haziness was about 68%, which is relatively high for such clear materials. On the other hand, TWS had haziness of 85%, which is considerably good despite using a smooth finished silicone mold in the fabrication process. Thus, showing very promising optical properties (transmittance and haziness) for TWS.

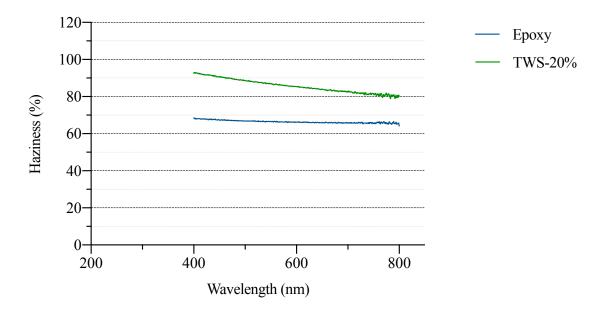


Figure 54: Haziness of TWS & Epoxy samples measured using UV spectrophotometer with an integrated sphere

## 4.5.TW - Transparent Wood (TW) & Glass

When placed on a document, the fabricated translucent wood showed good translucency in which the text was readable through it. However, once lifted from the document, the text was not readable, which is similar to the literature[68]. The fabricated translucent wood samples (using Casuarina) can be shown in Figure 55.



Figure 55: Fabricated translucent wood (Casuarina)

To put things into perspective, the fabricated translucent wood samples are compared to the pure epoxy and the three types of glass (regular, sanded, and rain glass). In Figure 56, all samples were placed directly on the document, and the transparency was observed. The epoxy and regular glass had the most visibility, followed by the sanded glass with very clear transparency. Finally, the translucent wood and rain glass had the least visibility compared to the other materials.

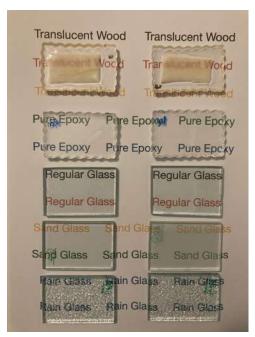


Figure 56: Visibility (transparency) of TW, epoxy, and glass samples compared

In this phase, the fabricated transparent wood optical properties were compared to untreated pieces of wood, pure epoxy and three types of glass: regular transparent glass, sanded glass, and rain glass. In addition, the results obtained for TWS from the previous phase was included in the comparison.

The transmittance was measured in the visible light range (400 – 800 nm). To begin with, the pure epoxy had the highest transmittance with around 92%, followed by regular glass and rain glass with around 85% and 82% respectively. Finally, the sanded glass and transparent wood had a comparable result with 72% and 70% respectively. Untreated Casuarina wood transmittance was measured to be included as a reference to the fabricated Casuarina transparent wood. The transmittance results for epoxy, TW, TWS, Casuarina wood, and glass can be illustrated in Figure 57. Although the visibility of sanded glass was better than the rain glass, the light transmittance did not follow the same trend. Finally, the translucent wood sawdust was around 80% placing it between epoxy, regular, and rain glass and translucent wood, and sanded glass. Although high light transmittance is a desirable feature, there is a tradeoff between the light transmittance and light scatter. In order to determine the appropriate application for each material, the haziness has to be measured. Unfortunately, it is usually a tradeoff between the two properties; the higher the light is transmitted, the less light is scattered.

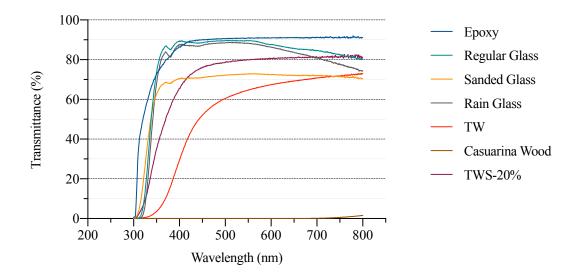


Figure 57: Light transmittance of TW, TWS, epoxy, casuarina wood, and glass samples measured using UV spectrophotometer with an integrated sphere

Unlike for the light transmittance, epoxy, regular, and rain glass had the lowest haziness compared to TW, TWS, and sanded glass. Transparent wood had the highest light scatter with around 95% haze compared to only 77% & 27% for the sanded glass and rain glass respectively. In comparison, epoxy and regular glass had almost zero values for haziness due to their high visibility. The haze results for TW, epoxy, and glass are illustrated in Figure 58. Comparing the obtained results from this phase to the results of TWS obtained in the previous phase, it shows that TWS has a high potential in replacing sanded glass. TWS had a transmittance of 80% and haziness of 85% while the sanded glass had 72% and 77% respectively. While in other applications, which does not require high visibility, transparent wood has an edge in offering better thermal insulation and high haziness without compromising much the amount of light transmitted.

Since the transparent wood fabricated using sawdust showed very promising results compared to other glass materials as well as the transparent wood fabricated from wood pieces, a scalable chemical treatment is introduced in the next phase. The next phase aims to test the efficiency of using commercial bleach in the sawdust's chemical treatment since the sample losing its strength is not an issue for the sawdust; like it was in the previous phases for the wood pieces. Also, the commercial bleach is not energy-intensive treatment method and could be the cheapest compared to relatively more expensive chemicals used in the other two processes.

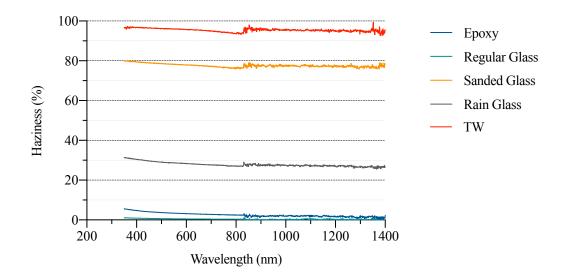


Figure 58: Haziness of TW, epoxy, and glass samples measured using UV spectrophotometer with an integrated sphere

### 4.6. Sawdust Treatment using Commercial Bleach

In this phase, three waste to solution ratios were used to treat Casuarina sawdust for three different time intervals 24, 48, and 72 h. The ratios used were 1:20, 1:40, and 1:60 and the change of color was observed over the first five hours of treatment and then at the selected time points. The samples were then washed in boiling DI water, and the colors were observed as well. Finally, the samples were air-dried, and FTIR spectrum was obtained to determine lignin's presence versus the color change. As shown in Figure 59, the samples were bleached in the first few hours of the treatment, especially for the 1:60 ratio. However, when the samples were washed, as shown in Figure 60, the sawdust turned to a brownish color for the 1:20 ratio and yellowish color for the 1:40. On the other hand, the 1:60 ratio showed the best color changes compared to the other two ratios, and the color did not change a lot after increasing the treatment time. However, comparing the 1:40 ratio after 48 and 72 hours, it showed comparable results to the 1:60, which could save on the solution used to treat the samples. Thus, further investigation was conducted through FTIR to determine the presence of lignin for each ratio at the three different time intervals.

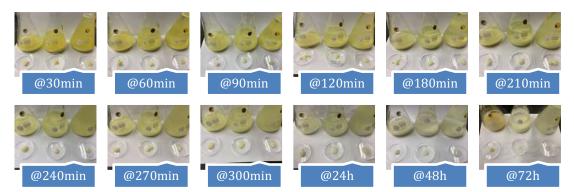


Figure 59: Visual comparison for sawdust treatment using Commercial Bleach at different time intervals for three waste to solution ratios 1:20, 1:40, and 1:60 (from left to right in each picture)



*Figure 60: Treated sawdust after washing at 24, 48, and 72 h for three waste to solution ratios 1:20, 1:40, and 1:60 (from left to right in each picture* 

The bleached air-dried sawdust samples can be shown in Figure 61, with the 1:60 ratio at 48 and 72 h having the whitest color compared to the 1:20 and 1:40. However, according to Figure 62, the FTIR showed no presence of lignin for all three ratios after only 24 hours of treatment, which does not explain the color of the sample treated with ratio 1:20. Although FTIR showed no presence of lignin, some sample needed further treatment, especially after washing in boiling DI. The sample could have stained from the washing step, but this was not common behavior for all ratios, thus requiring more investigation with lignin quantification (Klason lignin) to verify the results. Also, to create a correlation between the lignin content and the color of the sample, a brightness test can be conducted to explain the treatment effect better.

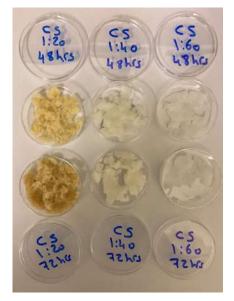


Figure 61: Color comparison for treated sawdust using commercial bleach different waste to solution ratios (1:20, 1:40, and 1:60) at 48 & 72 h of treatment

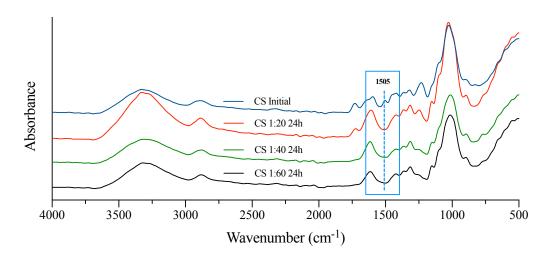


Figure 62: FTIR spectrums for untreated Casuarina sawdust and commercially bleached sawdust with different waste to solution ratios (1:20, 1:40, and 1:60) at 24 h of treatment. At 48 and 72 h showed the same results with no lignin presence (Figure 63 & Figure 64)

# **Chapter 5: Conclusion and Recommendations**

#### **5.1.**Conclusion

Transparent wood can be used as an alternative material in many applications to either replace less environmentally friendly materials or increase the desired system's efficiency. Replacing glass building claddings with transparent wood can increase the overall efficiency of the HVAC and lighting systems of the building. Better efficiency can be achieved due to its better thermal insulation compared to glass and its high optical haze that reduces the need for artificial lighting during the day.

There are different fabrication methods for transparent wood, which can be used depending on the wood species and size. During this study, three chemical treatment methods were used to test the efficiency of such treatment on the Egyptian hardwoods (casuarina & eucalyptus); these methods are the delignification method, the lignin modification method, and the commercial bleach treatment. In theory, the chemical treatment main aim is to remove the lignin content from the wood (which gives it its color) and replace it with a transparent polymer. First, the delignification treatment method consists of two processes. The first process removes most of the lignin, and the second process bleaches the wood while removing some of the lignin content. In the first process, the resultant is concentrated black liquor and weak samples, and in the second process, the sample could easily be broken. Although it was an effective method for some wood species, such as basswood, it did not show promising results with the Egyptian hardwoods. Also, the commercial bleach had minimal effect on the eucalyptus samples, and by the end of the casuarina treatment, the samples still had a yellow color and easily broken.

On the other hand, the lignin modification method showed better treatment behavior for both wood species and stronger wood templates ready for the polymer infiltration. The treatment was faster and easier compared to the other two methods and did not produce black liquor in the treatment process, which is more environmentally friendly. Thus, the lignin modification method was selected to proceed with for future phases.

In the next phase of the study, an in-depth analysis has been conducted on the lignin modification treatment method for both wood species to select the optimum treatment parameters and the wood species with the better treatment behavior. Klason

lignin was used to determine the lignin content change throughout the treatment process and analyze both wood species' treatment effectiveness. Although the eucalyptus had higher lignin to start with, both wood species had comparable results in the first half of the treatment process. However, towards the end of the first treatment process (the alkaline H<sub>2</sub>O<sub>2</sub> solution), the casuarina samples were almost fully treated and required little to no delignification treatment (second treatment process). Accordingly, reducing the treatment time as well as the chemical solutions needed for the treatment of casuarina. Thus, selecting casuarina wood for the next phase.

Casuarina wood fibers and sawdust were treated using the lignin modification and infiltrated using epoxy resin to fabricate the transparent wood samples in this phase. After the epoxy infiltration, the wood fibers were not properly infiltrated using two different methods (disk and mixed), but the sample had clear white cloud and no transparency. On the other hand, the sawdust samples had clear visibility and could easily read through it using either method. However, the disk method has cloudier visibility compared to the mixed sample. Casuarina wood pieces and sawdust were treated using lignin modification method and infiltrated using epoxy resin to fabricate transparent wood samples in this phase. To put things into perspective, transparent wood fabricated using casuarina wood pieces, and sawdust was compared to pure epoxy to evaluate the optical properties. Comparing the three sample, the pure epoxy transmittance was the highest with around 84%, followed by the sawdust (TWS) with around 80% and finally the transparent wood (TW) with 62%. The silicone molds used for the epoxy and TW were squared molds with the sanded finish while the TWS had a smooth finish. This could have been a factor decreasing the actual values for both the epoxy and TW; thus, excluded from future work and replaced by new silicone molds with a smooth finish.

After fabricating TW and TWS (20%) samples, their optical properties were compared with pure epoxy and three types of glass (regular glass, sanded glass, and rain glass). The pure epoxy had the highest transmittance, with around 92%, followed by regular glass and rain glass with around 85% and 82% respectively. Finally, the sanded glass and transparent wood had a comparable result with 72% and 70% respectively. Moving to haziness, transparent wood had the highest light scatter with around 95% haze compared to only 77% & 27% for the sanded glass and rain glass respectively and almost zero percent for both the pure epoxy and regular glass. Thus, transparent wood showed fairly good light transmittance compared to pure epoxy and

glass, but on the other hand, it had the highest haziness with around 95%. Also, since the TWS had a high light transmittance (~80%) as well as high haziness (~85%), it paves the way for a scalable process to produce transparent wood with superior optical properties. Moreover, the commercial bleach showed promising results in sawdust treatment, even though for a longer period of time but without heating, which helps produce feasible transparent wood that can be used in many applications.

Finally, commercial bleach treatment method showed very promising results, especially for higher waste to solution ratios which paves the way for a scalable and cheap treatment method for fabricating TWS. However, further research is required to determine the optimum parameters for the treatment and test its optical properties and study its effect compared to the fabricated samples using lignin modification method.

### **5.2. Recommendations**

According to the results obtained from this research, the following points can be covered in future work:

- Investigate the effect of using DTPA in the lignin modification method by conducting lignin quantification (Klason lignin) for the treatment of casuarina and eucalyptus samples and compared to the EDTA results.
- Study the effect on the optical properties for TWS using different ratios of sawdust to polymer, as mentioned in the literature. A range from 10 to 30% of sawdust to epoxy ratio can be used and compared.
- Study the effect of using different polymers on the optical properties of transparent wood or TWS.
- Investigate the effect of using eucalyptus sawdust to fabricate TWS vs using casuarina sawdust with the commercial bleach method (using different solution to sawdust ratios and different treatment durations).
- Study the mechanical properties for TW and TWS fabricated using different polymers and compare it to natural wood, glass, and other plastic polymers.

# References

- Y. Li, Q. Fu, S. Yu, M. Yan, and L. Berglund, "Optically Transparent Wood from a Nanoporous Cellulosic Template: Combining Functional and Structural Performance," *Biomacromolecules*, vol. 17, no. 4, pp. 1358–1364, 2016.
- M. Zhu *et al.*, "Highly Anisotropic, Highly Transparent Wood Composites," *Adv. Mater.*, vol. 28, no. 26, pp. 5181–5187, 2016.
- [3] M. Mália, J. De Brito, M. D. Pinheiro, and M. Bravo, "Construction and demolition waste indicators," *Waste Manag. Res.*, vol. 31, no. 3, pp. 241–255, 2013.
- [4] M. Marzouk and S. Azab, "Environmental and economic impact assessment of construction and demolition waste disposal using system dynamics," *Resour. Conserv. Recycl.*, vol. 82, no. JANUARY 2014, pp. 41–49, 2014.
- [5] H. Ritchie and M. Roser, "CO<sub>2</sub> and Greenhouse Gas Emissions," *Our World in Data*, 2020. [Online]. Available: https://ourworldindata.org/co2-and-other-greenhouse-gas-emissions.
- [6] H. Ritchie and M. Roser, "Energy," *Our World in Data*, 2020. [Online]. Available: https://ourworldindata.org/energy.
- [7] M. Crippa *et al.*, "Fossil CO2 and GHG emissions of all world countries," 2019.
- [8] "Energy Efficiency Trends in the EU."
- [9] GEA, "Global Energy Assessment: toward a sustainable future Chapter 5 -Energy Security," *Glob. Energy Assess.*, p. 1884, 2016.
- [10] L. Pérez-Lombard, J. Ortiz, and C. Pout, "A review on buildings energy consumption information," *Energy Build.*, vol. 40, no. 3, pp. 394–398, 2008.
- [11] "Egypt GBC | guidelines." [Online]. Available: https://www.egyptgbc.org/pages/guidelines. [Accessed: 22-Jan-2021].
- [12] H. Zhu *et al.*, "Wood-Derived Materials for Green Electronics, Biological Devices, and Energy Applications," *Chem. Rev.*, vol. 116, no. 16, pp. 9305–9374, 2016.
- [13] L. F. BOTANNINI, WOOD: TYPES, PROPERTIES, AND USES. .
- [14] Zero Waste South Australia, "Stage 1 Final Report Study on the South Australian Plastics Packaging Resource Recovery Sector," no. April, 2012.
- [15] DSM Environmental Services, "MASSACHUSETTS CONSTRUCTION AND

DEMOLITION DEBRIS INDUSTRY STUDY," 2007.

- [16] W. Lu and H. Yuan, "A framework for understanding waste management studies in construction," *Waste Manag.*, vol. 31, no. 6, pp. 1252–1260, 2011.
- [17] "Wood waste : A short review of recent," no. July, 2012.
- [18] T. Ding and J. Xiao, "Estimation of building-related construction and demolition waste in Shanghai," *Waste Manag.*, vol. 34, no. 11, pp. 2327–2334, 2014.
- [19] D. D. STOKKE, Q. WU, and G. HAN, Introduction to Wood and Natural Fiber Composites. John Wiley & Sons, 2014.
- [20] J. A. Ocampo, A. Cosbey, and M. Khor, "The Transition to a Green Economy: Benefits, Challenges and Risks from a Sustainable Development Perspective," 2013.
- [21] F. Bushehri, "Green Economy In The Arab Region," 2012.
- [22] S. M. El-Haggar *et al.*, Sustainable Industrial Design and Waste Management, no. 2007. 2007.
- [23] A. C. Wiedenhoeft and R. B. Miller, "Structure and function of wood," in Handbook of Wood Chemistry and Wood Composites, Second Edition, no. 2005, R. M. Rowell, Ed. CRC Press, 2012, pp. 9–33.
- [24] R. A. Parham, R. L. Gray, and I. T. T. Rayonier, "Formation and Structure of Wood," pp. 3–56, 1984.
- [25] R. B. Miller, "Structure of wood," in Wood handbook—Wood as an engineering material, 1999.
- [26] R. Rowell, R. Rowell, R. Pettersen, and M. Tshabalala, *Cell Wall Chemistry*. 2012.
- [27] W. A. Whistler and C. R. Elevitch, "Species Profiles for Pacific Island Agroforestry C . cunninghamiana (river she-oak)," *Species Profiles Pacific Isl. Agrofor.*, vol. 2.1, no. April, 2006.
- [28] A. Mohareb and M. Badawy, "Chemical Composition and Resistance to Decay of Thermally Modified Wood from Casuarina glauca and Eucalyptus camaldulensis Grown in Egypt," *Alexandria Sci. Exch. J. An Int. Q. J. Sci. Agric. Environ.*, vol. 38, no. April-June, pp. 159–167, 2017.
- [29] Food and Agriculture Organsiation of the United Nations, Global Forest Resources Assessment 2015. 2015.
- [30] J. Rencoret, A. Gutiérrez, and J. Carlos del Rio, "CHEMICAL COMPOSITION OF DIFFERENT EUCALYPTUS WOOD SPECIES USED FOR PAPER PULP

MANUFACTURING Jorge Rencoret, Ana Guti{é}rrez and Jos{é} Carlos del R{í}o Instituto de Recursos Naturales y Agrobiolog{í}a de Sevilla, CSIC, PO Box 1052, E-41080," pp. 1–4, 2005.

- [31] P. Objectives, "Inspection Manual: Pulp and Paper Industry," 2002.
- [32] P. Bajpai, "Paper and Paperboard Industry," Pulp Pap. Ind., pp. 13–24, 2015.
- [33] A. Latha, M.C. Arivukarasi, C. M. Keerthana, R. Subashri, and V.Vishnu Priya,
  "Paper and Pulp Industry Manufacturing and Treatment Processes A Review," *Int. J. Eng. Res.*, vol. V6, no. 02, 2018.
- [34] Z. Liu, H. Wang, and L. Hui, "Pulping and Papermaking of Non-Wood Fibers," *Pulp Pap. Process.*, pp. 3–32, 2018.
- [35] R. Alén, *Pulp and paper*, vol. 2. 2019.
- [36] A. Shawky Sol, M. Sakran She, F. Ahmad, and M. Abdel-Atty, "Evaluation of Paper Pulp and Paper Making Characteristics Produced from Different African Woody Trees Grown in Egypt," *Res. J. For.*, vol. 11, no. 2, pp. 19–27, 2017.
- [37] B. S. Mózo, Methods in Lignin Chemistry, vol. 53, no. 9. 2017.
- [38] K. V. Fagerstedt, P. Saranpää, T. Tapanila, J. Immanen, J. A. A. Serra, and K. Nieminen, "Determining the composition of lignins in different tissues of silver birch," *Plants*, vol. 4, no. 2, pp. 183–195, 2015.
- [39] Technical Association of the Pulp and Paper Industry, "Acid-insoluble Lignin in Wood and Pulp. Test method T222 Om-02," 2002.
- [40] N. M. Stark, D. J. Yelle, and U. P. Agarwal, "Techniques for Characterizing Lignin," *Lignin Polym. Compos.*, pp. 49–66, 2016.
- [41] J. Rodrigues, O. Faix, and H. Pereira, "Determination of lignin content of Eucalyptus globulus wood using FTIR spectroscopy," *Holzforschung*, vol. 52, no. 1, pp. 46–50, 1998.
- [42] K. Gao *et al.*, "Cellulose nanofibers/reduced graphene oxide flexible transparent conductive paper," *Carbohydr. Polym.*, vol. 97, no. 1, pp. 243–251, 2013.
- [43] Y. Okahisa, A. Yoshida, S. Miyaguchi, and H. Yano, "Optically transparent wood-cellulose nanocomposite as a base substrate for flexible organic lightemitting diode displays," *Compos. Sci. Technol.*, vol. 69, no. 11–12, pp. 1958– 1961, 2009.
- [44] S. Diabang, O. Felix, T. Roland, and C. Gauthier, "Bio-Inspired Multiproperty Materials :," no. 2, pp. 1127–1136, 2015.
- [45] T. Li et al., "Wood Composite as an Energy Efficient Building Material: Guided

Sunlight Transmittance and Effective Thermal Insulation," *Adv. Energy Mater.*, vol. 6, no. 22, 2016.

- [46] M. Zhu *et al.*, "Transparent and haze wood composites for highly efficient broadband light management in solar cells," *Nano Energy*, vol. 26, pp. 332–339, 2016.
- [47] Y. Li, X. Yang, Q. Fu, R. Rojas, M. Yan, and L. Berglund, "Towards centimeter thick transparent wood through interface manipulation," *J. Mater. Chem. A*, vol. 6, no. 3, pp. 1094–1101, 2018.
- [48] Q. Fu, M. Yan, E. Jungstedt, X. Yang, Y. Li, and L. A. Berglund, "Transparent plywood as a load-bearing and luminescent biocomposite," *Compos. Sci. Technol.*, vol. 164, no. April, pp. 296–303, 2018.
- [49] Y. Li, Q. Fu, R. Rojas, M. Yan, M. Lawoko, and L. Berglund, "Lignin-Retaining Transparent Wood," *ChemSusChem*, vol. 10, no. 17, pp. 3445–3451, 2017.
- [50] H. Li, X. Guo, Y. He, and R. Zheng, "A green steam-modified delignification method to prepare low-lignin delignified wood for thick, large highly transparent wood composites," *J. Mater. Res.*, pp. 1–9, 2019.
- [51] X. Wang *et al.*, "Large-Size Transparent Wood for Energy-Saving Building Applications," *ChemSusChem*, pp. 1–9, 2018.
- [52] J. Qin *et al.*, "Optimization of delignification process for efficient preparation of transparent wood with high strength and high transmittance," *Vacuum*, vol. 158, no. 26, pp. 158–165, 2018.
- [53] Z. Qiu *et al.*, "Transparent wood bearing a shielding effect to infrared heat and ultraviolet via incorporation of modified antimony-doped tin oxide nanoparticles," *Compos. Sci. Technol.*, vol. 172, no. January, pp. 43–48, 2019.
- [54] T. Feng, J. Qin, Y. Shao, L. Jia, Q. Li, and Y. Hu, "Size-Controlled Transparent Jute Fiber for Replacing Transparent Wood in Industry Production Area," *Coatings*, vol. 9, no. 7, p. 433, 2019.
- [55] W. Gan, L. Gao, S. Xiao, W. Zhang, X. Zhan, and J. Li, "Transparent magnetic wood composites based on immobilizing Fe3O4nanoparticles into a delignified wood template," *J. Mater. Sci.*, vol. 52, no. 6, pp. 3321–3329, 2017.
- [56] W. Gan, S. Xiao, L. Gao, R. Gao, J. Li, and X. Zhan, "Luminescent and Transparent Wood Composites Fabricated by Poly(methyl methacrylate) and γ-Fe2O3@YVO4:Eu3+ Nanoparticle Impregnation," ACS Sustain. Chem. Eng., vol. 5, no. 5, pp. 3855–3862, 2017.

- [57] H. S. Yaddanapudi, N. Hickerson, S. Saini, and A. Tiwari, "Fabrication and characterization of transparent wood for next generation smart building applications," *Vacuum*, vol. 146, pp. 649–654, 2017.
- [58] K. E. O. Foster, K. M. Hess, G. M. Miyake, and W. V. Srubar, "Optical Properties and Mechanical Modeling of Acetylated Transparent Wood Composite Laminates," *Materials (Basel).*, vol. 12, no. 14, p. 2256, 2019.
- [59] Y. Li, M. Cheng, E. Jungstedt, B. Xu, L. Sun, and L. Berglund, "Optically Transparent Wood Substrate for Perovskite Solar Cells," ACS Sustain. Chem. Eng., vol. 7, no. 6, pp. 6061–6067, 2019.
- [60] Y. Li, S. Yu, J. G. C. Veinot, J. Linnros, L. Berglund, and I. Sychugov, "Luminescent Transparent Wood," *Adv. Opt. Mater.*, vol. 5, no. 1, pp. 3–7, 2017.
- [61] H. Chen *et al.*, "Thickness Dependence of Optical Transmittance of Transparent Wood: Chemical Modification Effects," *ACS Appl. Mater. Interfaces*, vol. 11, no. 38, pp. 35451–35457, 2019.
- [62] J. Wu, Y. Wu, F. Yang, C. Tang, Q. Huang, and J. Zhang, "Impact of delignification on morphological, optical and mechanical properties of transparent wood," *Compos. Part A Appl. Sci. Manuf.*, vol. 117, no. November 2018, pp. 324–331, 2019.
- [63] C. Montanari, Y. Li, H. Chen, M. Yan, and L. A. Berglund, "Transparent Wood for Thermal Energy Storage and Reversible Optical Transmittance," ACS Appl. Mater. Interfaces, vol. 11, no. 22, pp. 20465–20472, 2019.
- [64] Z. Bi, T. Li, H. Su, Y. Ni, and L. Yan, "Transparent Wood Film Incorporating Carbon Dots as Encapsulating Material for White Light-Emitting Diodes," ACS Sustain. Chem. Eng., vol. 6, no. 7, pp. 9314–9323, 2018.
- [65] H. Li, X. Guo, Y. He, and R. Zheng, "House model with 2–5 cm thick translucent wood walls and its indoor light performance," *Eur. J. Wood Wood Prod.*, vol. 77, no. 5, pp. 843–851, 2019.
- [66] Y. Wu, J. Wu, F. Yang, C. Tang, and Q. Huang, "Effect of H2O2 bleaching treatment on the properties of finished transparent wood," *Polymers (Basel).*, vol. 11, no. 5, pp. 1–14, 2019.
- [67] M. Zhu et al., "Anisotropic, Transparent Films with Aligned Cellulose Nanofibers," Adv. Mater., vol. 29, no. 21, 2017.
- [68] C. Jia *et al.*, "Scalable, anisotropic transparent paper directly from wood for light management in solar cells," *Nano Energy*, vol. 36, no. March, pp. 366–373,

2017.

- [69] Y. Wu, J. Zhou, Q. Huang, F. Yang, Y. Wang, and J. Wang, "Study on the properties of partially transparentwood under different delignification processes," *Polymers (Basel).*, vol. 12, no. 3, pp. 1–14, 2020.
- [70] S. This, "Ash in wood , pulp , paper and paperboard : combustion at 900 ° C," *Tappi*, 2006.
- [71] K. K. Pandey and A. J. Pitman, "FTIR studies of the changes in wood chemistry following decay by brown-rot and white-rot fungi," *Int. Biodeterior. Biodegrad.*, vol. 52, no. 3, pp. 151–160, 2003.

# Appendix

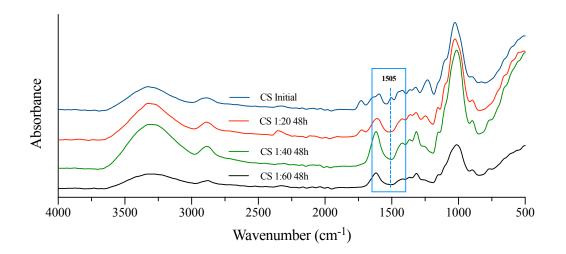


Figure 63: FTIR spectrums for untreated Casuarina sawdust and commercially bleached sawdust with different waste to solution ratios (1:20, 1:40, and 1:60) at 48h of treatment.

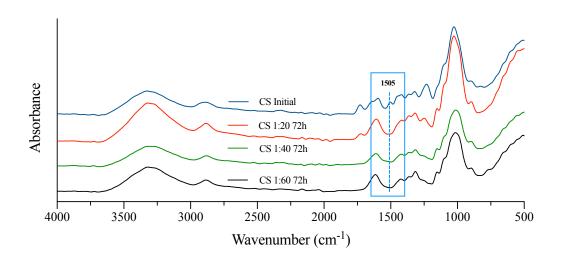


Figure 64: FTIR spectrums for untreated Casuarina sawdust and commercially bleached sawdust with different waste to solution ratios (1:20, 1:40, and 1:60) at 72h of treatment.