

## Sick building syndrome: The effects of animal and plant-based adhesive in wood furniture

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ARTICLE INFO	ABSTRACT
<p><i>Article history:</i> Received July 13, 2021 Received in revised form Dec. 08, 2022 Accepted January 05, 2023 Available online April 01, 2023</p> <p><i>Keywords:</i> Adhesives Animal Plant Sick building syndrome</p> <p>*Corresponding author: A. Ghafar Ahmad School of House Building and Planning, Universiti Sains Malaysia Email: <a href="mailto:aghafar@usm.my">aghafar@usm.my</a> ORCID: <a href="https://orcid.org/0000-0001-7969-7928">https://orcid.org/0000-0001-7969-7928</a></p>	<p><i>Sick building syndrome (SBS) is a condition that afflicts occupants of a building, typically a workplace, and is linked to time spent in the building. The precise cause for SBS is still unknown. However, there are a few possibilities that is believed to be the cause of SBS such as poor ventilation, bad lighting design, presence of mold, fungus and formaldehydes which is usually found in wood furniture. This research paper focuses on the furniture factor of SBS. Certain material and finishes used in furniture production are harmful to the users as it has the potential to emit chemical substances from the wood composite products which could affect the indoor air quality. Furniture production may not sound harmful, but if the processes were observed in detail, we can see animal exploitations along the progress. Adhesives used to join the furniture may have substances that are derived from animals. The objective of this research is to study the chemical component in an animal-based adhesive and plant-based adhesive in wood furniture that is commonly used in lobby or lounge area (sofa, coffee table, etc.). With the chemical components identified, it is used to see how it contributes to the Sick Building Syndrome and which adhesives will be a better option for furniture production. This research also aims to compare animal-based and plant-based adhesives in terms of quality and cost. This research uses the qualitative method in which we look at past research first-hand observations, interviews, data, etc. The outcome by the end of this research will achieve all research objectives and suggest a better adhesive in wood furniture production whether it is animal-based or plant-based adhesives that has the least effect to SBS. This research can be beneficial to all furniture companies that are finding an alternative for a safer and greener choice of adhesive in wood furniture production. This research can also be a guideline to help improve the quality in furniture production and reduce the risk of SBS.</i></p>

### Introduction

Due to the global pandemic Covid-19, people are advised to stay indoors most of the time to reduce the risk of infection. Since people stay indoors more, they are more likely to be exposed to indoor air pollutants which effects the indoor air quality (IAQ) of the building. Based on a study in a

laboratory in Nigeria, it is reported that the 21.7% of users experienced mucosal related symptom, 18.6% experienced general related symptom and 37.7% experienced respiratory related symptom and all of these are associated with IAQ (Reuben et al. 2019). A poor IAQ could affect the health of the indoor occupants although it is not very severe (Hosseini, Fouladi-Fard, and Aali 2020), but if the

issue was not taken seriously, it could affect not only health but also the work quality and productivity of the indoor occupant (Wu et al. 2021).

It is important to know what causes VOC emission. Since most of the furniture used in any buildings be it in homes, offices, retail shops and etc. are made of wood, thus more people are exposed to VOC and formaldehyde emission without notice since it is a topic not widely educated about. Indoor Air Quality (IAQ) is said to be one of the contributors to Sick Building Syndrome (SBS) where a person may experience acute health or comfort problems due to being in a building or a closed space for a certain amount of time and it is usually associated with inadequate heating, air conditioning system, bad indoor air quality, etc. (Arikan, Tekin, and Erbas 2018). It is now well known that the main causes of SBS are poor indoor air quality due to inadequate ventilation or fresh air intake, biological contamination of the indoor environment, and an accumulation of noxious compounds such as formaldehyde and volatile organic compounds (W. J. Kim et al. 2002) which are usually found in adhesives used in furniture, carpentry, wood flooring, etc. Formaldehyde emission is dangerous as it acts as an indoor air pollutant and one of the causes for mucosal irritative symptoms (W. J. Kim et al. 2002).

Past researchers has found alternatives to replace adhesives that are harmful to the building occupants and two alternatives proposed are adhesives that are derived from plant proteins such as soybean protein (Bacigalupe et al. 2017), tannin, cashew nut liquid (Lee, Jeon, and Kim 2011) and adhesives derived from animal protein such as catfish protein (Cheng et al. 2021), spent hen protein (Wang and Wu 2012) and mussel protein (Kord Forooshani and Lee 2017). Past researchers agreed that these two alternatives are considered as bio-based adhesives as they are not chemically derived from petroleum like formaldehyde adhesive. Although petroleum-based adhesives are already proven to emit chemical pollutants to the air as it emits volatile organic component (VOC) and formaldehyde particle, the chemical components in a plant-based and animal-based adhesive and their effect to sick building syndrome (SBS) remain in question.

Since there are two alternative adhesives proposed by researchers, what are the differences between these two adhesives in terms of cost,

quality, and production and which will be a better choice to replace the adhesives used currently? The objective of this research is to study the chemical components in animal-based and plant-based wood adhesives and do comparison between these two adhesives in terms of cost, quality, and production. With this research paper, a better alternative for adhesive will be proposed for the use of carpentry industry that can reduce the risk of SBS and is environment friendly. The theoretical framework is that a plant-based adhesive is a better option to be used in wood furniture production as it is better in terms of cost, production, and quality.

#### Literature review

Sick Building Syndrome (SBS) is often associated with various health problem when an individual is in a closed space in a building for a certain amount of time (Thach et al. 2019). There is no clear cause to what causes SBS but it is believed that one of the factors are Indoor Air Quality (IAQ). Volatile organic compounds (VOCs) are one of the factors that effects IAQ (D.Maskell, C. F. da Silva, K. Mower, C. Rana, A. Dengel, R. J. Ball, M. P. Ansell, P. J. Walker 2015) and they are important indoor air pollutants produced by evaporation at room temperature from diverse sources, such as building materials, paints, cleaning agents, furnishings, adhesives, combustion materials, floor, and wall coverings (Kwon et al. 2018).

VOCs are a large group of various compounds including natural compounds such as formaldehydes, phenol, benzene, toluene, vinyl acetate, methyl acetate ethanol, methanol and many more (Łebkowska, Załęska-Radziwiłł, and Tabernacka 2017). These natural and carbonyl compounds are the main pollutants present in indoor air and effects the indoor air quality (Adamová, Hradecký, and Pánek 2020). VOC emission are harmful because they have carcinogenic and mutagenic properties that can cause headache, irritation to the eyes, nasal, etc. (Kwon et al. 2018) and can cause respiratory problems to an individual (Kwon et al. 2018). In wood adhesives, the most common type of VOCs that are emitted to the indoor surrounding is formaldehyde.

Formaldehyde is a type of VOC gas that comes from urea formaldehyde (UF) which is a type of adhesive resins that is derived from petroleum and are the main binders used in the industry because it is lower in cost and easier to

produce. Because of formaldehyde emissions, it has been proven that formaldehyde-based adhesives are not environmental-friendly products (Vineeth, Gadhav, and Gadekar 2019). If a person is exposed to formaldehyde gas too long, they might develop allergies, migraines, irritation to the eyes, nasal passages, mouth, and lungs, and could cause respiratory problems (Łebkowska, Załęska-Radziwiłł, and Tabernacka 2017). Other types of VOCs are acetaldehyde, acetone, hexanaldehyde, toluene, ethylbenzene and etc. (Navarrete et al. 2013). All these harmful gases could cause health problems such as headaches, respiratory problems etc. but it is not as serious as formaldehyde. To avoid effects on sensitive people in non-industrial buildings, the World Health Organization (WHO) has recommended that the concentration in air should be below  $0.1 \text{ mgm}^{-3}$  as a 30-min average (Yu and Kim 2012).

Researchers have successfully found alternatives or replacements for petroleum-based adhesives which are nature and environment friendly bio-adhesive and those are animal-based and plant-based adhesives. The most common bio-adhesive that are being used and developed are adhesives made from plants and animals. Animals are also considered as a bio-adhesives as they are also part of the nature and are from nature origin just like plants are (Virginia, Suhasman, and Agussalim 2020). These bio-adhesives are said to be free from formaldehyde thus does not bring harm to the indoor air quality. For plant-based adhesives, few plants that can be used as a substitute to UF are soy-bean protein, camelina protein, lignin, tannin and many more.

One of the plant-based adhesives that is widely being studied is Soy-bean protein and it has a high potential to replace UF. Soybeans are a species of legumes and the seeds of a low-growing field vine. These vines are ancient in culture; the written record of their domestication in China dates back almost 5000 years (Pen Ts'ao Kong Mu, 2838 B.C.) (Jeon, Lee, and Kim 2011). The modified soybean-based adhesive exhibited sufficient mechanical properties to use as an adhesive for composite wood products (Jeon, Lee, and Kim 2011) such as having good solubility and adhesive strength for binding pigments in paper coatings and water-based paints (Jeon, Lee, and Kim 2011). Like other alternatives plant-based adhesives, few tests are needed to be done in order to assure that these bio-adhesives can be used

such as having good shear adhesion strength from 0.28 to 1.42 Mpa (Zhu et al. 2017) and have good water resistance.

Another option for bio-adhesives are adhesives derived from animals, specifically animal protein. Proteins have been used for centuries to prepare adhesives especially during ancient times, while petroleum-based adhesives have started to dominate the market about a few decades ago due to their affordable cost and satisfactory performance (Wang and Wu 2012). Compared to petroleum-based adhesives in terms of performance, traditional animal-based adhesives are still at a disadvantage due to its weakness against moisture (J.-H. Kim et al. 2017) and have low adhesive strength and poor water resistance. Studies have been done to improve the chemical properties of animal-based adhesives so that it can meet the wood and furniture industry requirements and expectations. Experiments like automated bonding evaluation system (ABES) were used to determine the shear strength of animal adhesives (Wang and Wu 2012) and water resistance of the adhesive was measured according to the ASTM Standard D1151-00 using the wet strength and soaked strength (Wang and Wu 2012).

Example of animal-based adhesives are adhesives that are derived from catfish protein (Cheng et al. 2021) and spent hen (Wang and Wu 2012). The proteins of these animals are extracted from their skin, blood, gluten, hides, bones, etc. and during testing, these proteins are mixed with appropriate volumes of deionized water for 2 hours to produce the solid content of about 6% weight. This solid content was chosen in order to produce a smooth coating onto the wood surface (Wang and Wu 2012).

The similarity between animal-based and plant-based adhesives are both have proteins which is an essential component for bio-adhesives. Although these alternatives are present, it is hard for them to enter the mainstream market as they are higher in cost due to its more complicated production compared to petroleum-based adhesives (Hemmilä et al. 2017). Researchers and scientists are still developing more bio-based adhesives so that one day they could be used widely especially in construction and carpentry industry. But, soy-bean based adhesive are slowly being used and launched commercially for plywood manufacturing (Hemmilä et al. 2017) thus this shows that bio-

based adhesives is still possible and have a chance to be widely used.

Although past studies were done on VOC, formaldehyde emission and alternatives to switch from petroleum-based adhesive to an environmental-friendly bio-adhesives, there are still no study on how using bio-adhesives can help reduce the risk of SBS. Thus, the research gap for this study is to study how using bio-adhesives can help reduce the risk of SBS. The conceptual framework is that since both animal and plant-based adhesives are considered as bio-based adhesive, both have the potential to be used in the industry and can replace petroleum-based adhesive but plant-based adhesive would be a better option compared to animal-based adhesive in terms of quality, cost and production.

## Method

This research uses qualitative methodology using the Case Study method. Data and information about sick building syndrome, plant-based adhesives and animal-based adhesives were collected and analyzed. The data and information are taken from past papers and research that are connected to this issue. Case Study is also used in this research to collect data information on existing research, study and real experiments done regarding the topic. Using the case study method, the chemical component of animal and plant-based adhesives were identified along with the characteristics on what makes it a better adhesive.

### Case study

#### 1. Preparation and evaluation of catfish protein as a wood adhesive

Information that is extracted from this case study are the chemical component in catfish protein adhesive and its quality/productivity in terms of sheer strength and PH resistance, etc.

Fish is very well known for containing a variety of proteins, such as structural proteins, sarcoplasmic proteins, connective tissue proteins in the fish muscle, and have a high amount of collagen from its skin (Cheng et al. 2021). In this research paper, catfish protein is the study object where it was extracted and undergoes through chemical processes for it to be produced as adhesives. Also, a few tests and experiments were done to see whether these animal-based adhesives

can be used in the industry with the variable of sheer strength, water resistance and pressure. But in order to see the quality and performance of the adhesive, the tests were also done on cottonseed protein isolate (CPI) and their blends for comparison.

Before the tests and experiment were taken place, the catfish protein were extracted from it's skin and placed on ice. The catfish skin were then treated with few chemical modification to extract the skin and gelatin(protein) mixture were filtered. Gelatin are proteins and proteins are made up of amino acids. Thus, in table 1 it shows the amino acid composition in skin sample G, sample GE and sample SKPD. In the table 2, it can be seen that glycine has the highest amount in each sample with approximately 23% followed by proline (approx. 12%) and hydroxyproline (approx. 9%)

**Table 1.** Amino acid composition of catfish skin gelatin (G), hydrolyzed catfish skin gelatin (GE) and dried skin sample (SKPD)

Amino acid	G	GE	SKPD
HYP (Z)	9.36	9.26	8.55
ASP (D)	5.65	5.80	6.28
THR (T)	2.47	2.49	2.74
SER (S)	3.80	3.83	4.00
GLU (E)	9.52	8.90	9.75
PRO (P)	12.69	12.65	11.93
GLY (G)	23.30	23.27	21.94
ALA (A)	10.05	10.05	9.57
VAL (V)	2.55	2.60	2.88
MET (M)	1.28	1.27	1.20
ILE (I)	1.36	1.38	1.69
LEU (L)	2.32	2.34	2.99
TYR (Y)	0.52	0.59	0.82
PHE (F)	1.77	1.94	2.00
HIS (H)	0.86	0.87	0.95
Hlys	0.63	0.82	0.67
LYS (K)	3.60	3.62	3.90
ARG (R)	8.29	8.32	8.15
Total (w/w%)	100.02	100.00	100.01

In order the test the performance of the adhesives made from catfish protein, the adhesive sample were tested with a few variables. The first test is testing the adhesive strength of the adhesive by applying it on wood and apply hot pressure to the adhesives. The variables are the number of layers applied to the wood sample, the time taken and the temperature applied and the result were recorded in table 2. As we can see in table 2, sample 2 has the highest tensile strength with 2 layers of adhesive coating with temperature of 120-degree celcius applied and in 10 minutes. The catfish protein adhesive performance were also

tested with pH test. But from the result that we can see in table 3, there are not much drastic changes to the adhesive strength of the adhesives.

**Table 2.** Adhesive performance of catfish protein as a function of hot-pressing temperature and time and layers of protein solution applied to the wood strips

Sample	Layers	Temp (°C)	Time (min)	Tensile strength (MPa)*
S1	2	120	20	1.73 ± 0.35 <sup>a</sup>
S2	2	120	10	1.85 ± 0.37 <sup>a</sup>
S3	2	120	5	1.76 ± 0.33 <sup>a</sup>
S4	1	120	5	0.81 ± 0.38 <sup>b</sup>
S3	2	120	5	1.76 ± 0.33 <sup>a</sup>
S5	3	120	5	1.81 ± 0.80 <sup>a</sup>
S6	2	100	5	1.38 ± 0.52 <sup>a, b</sup>
S3	2	120	5	1.76 ± 0.33 <sup>a</sup>
S7	2	130	5	1.68 ± 0.68 <sup>a</sup>

\*Data are given as average ± standard deviations. The data in this column have been subjected to statistical analysis; values with different superscript letters are significantly different at  $p < 0.05$  in the analysis of variance

**Table 3.** Effects of pH on the performance of catfish protein adhesive

Sample	Layers	Temp (°C)	Time (min)	Tensile strength (MPa)*
S3	2	120	5	1.76 ± 0.33 <sup>a</sup>
C1	2	120	5	2.30 ± 0.51 <sup>a</sup>
T1	2	120	5	2.11 ± 0.39 <sup>a</sup>
C2	2	120	5	2.21 ± 0.59 <sup>b</sup>
C3	2	120	5	1.83 ± 0.35 <sup>a</sup>

\*Data are given as average ± standard deviations. The data in this column have been subjected to statistical analysis; values with different superscript letters are significantly different at  $p < 0.05$  in the analysis of variance

Catfish protein adhesive were tested again with the same variables alongside with CPI and their variables for comparison. This comparison is important to see whether catfish protein adhesive is as good as CPI as CPI have been proved to perform well as a wood adhesive. The result are as seen in table 4 and there is not much different in the tensile strength although with different variables.

**Table 4.** Comparison of adhesive performance of catfish protein, cottonseed protein isolate (CPI), and their blends; all data observed at pH 4.8

No	Sample	Layers	Temp (°C)	Time (min)	Tensile strength (MPa)*
T1	Fish gelatin only	2	120	5	2.11 ± 0.39 <sup>a</sup>
T2	Gelatin/CPI 1:1	2	120	5	2.17 ± 0.81 <sup>a</sup>
T3	Gelatin/CPI 0.75:0.25	2	120	5	2.33 ± 0.92 <sup>a</sup>
T4	Gelatin/CPI 0.25:0.27	2	120	5	2.28 ± 0.77 <sup>b</sup>
T5	CPI only	2	120	5	1.58 ± 0.94 <sup>a</sup>

\*Data are given as average ± standard deviations. The data in this column have been subjected to statistical analysis; values with different superscript letters are significantly different at  $p < 0.05$  in the analysis of variance

**Table 5.** Hot water test for catfish protein, cottonseed protein isolate (CPI), and their blends; all data observed at pH 4.8

No	Sample	Layers	Temp (°C)	Time (min)	Tensile strength (MPa)*
H1	Cpi only	2	120	5	0.69 ± 0.43 <sup>a</sup>
H2	Fish gelatin only	2	120	5	0.82 ± 0.31 <sup>a</sup>
H3	Gelatin/CPI 1:1	2	120	5	1.82 ± 0.66 <sup>a</sup>

\*Data are given as average ± standard deviations. The data in this column have been subjected to statistical analysis; values with different superscript letters are significantly different at  $p < 0.05$  in the analysis of variance

Based on the table 4 and 5 above, it can be seen that catfish gelatin adhesive performance and CPI performance are nearly the same and does not show any insignificant different. But catfish gelatin clearly shows a slightly better result than that of CPI which proves that catfish gelatin have a better performance and have a chance to be used as wood adhesive in the market. But when CPI and catfish gelatin mixture is combined, it shows a better adhesive strength performance than catfish gelatin alone. This might be because the protein in CPI (plant-based protein) and protein in

catfish gelatin (animal-based protein) interact with each other and could enhance the adhesive property of the adhesives. The amount of certain amino acid in CPI might be higher than catfish gelatin and a certain amount of amino acid in catfish gelatin might be higher than that of CPI. Thus, the interaction between the amino acids in these two proteins have helped build a stronger bonding between the amino acids and produces an adhesive with stronger adhesive strength.

The chemical component in catfish protein adhesives are mostly amino acids which are

glycine, proline and hydroxyproline is the highest amount of amino acids in a catfish protein. Based on this experiment and study, it is proven that catfish protein can be used as a bio-adhesives with tensile/sheer strength of 1.81MPa, have pH resistance with strength of 1.83MPa when highest pH were tested (10). In this test it is also proven that catfish protein adhesive have a better performance than CPI which is already proven as a functioning adhesive.

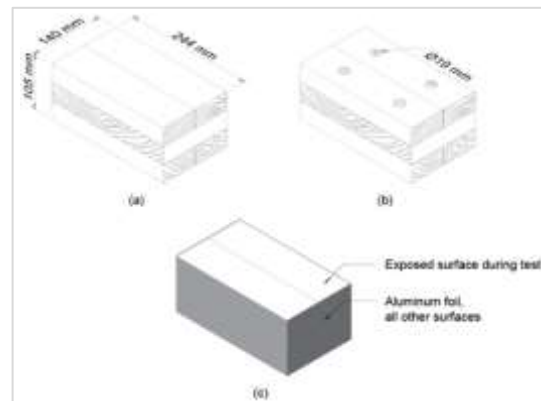
## 2. Evaluating volatile organic compound emissions from cross-laminated timber bonded with a soy-based adhesive

The information extracted from this paper is the difference between bio-adhesive and petroleum-based adhesive and its contribution to VOC emission. The chemical components is observed in order to see whether it has the potential to effect the IAQ of a building.

This study aims to evaluate VOC emission from four types of adhesives that is applied to cross laminated timber (CLT) which is a soy-bean based adhesive, melamine formaldehyde (MF) adhesive, polyurethane adhesive and another one is not applied any adhesive(dowel). To evaluate these adhesives, the method of small-chamber testing using the protocols outlined by the California Department of Public Health (CDPH) is used at an accredited facility following a standardized method. By using this method, a full characterization of the VOCs emitted can be obtained.

All of the four samples were applied on cross-laminated timber (CLT) where three of the samples have similar thickness of 105mm, 3 lamellae panels, produced in laboratory, mechanically cold-pressed panels and glued on the face. Sample 1 was bonded with a proprietary soy flour with Magnesium Oxide (MgO), sample 2 was bonded with polyurethane adhesive (PUR) and sample 3 was not bonded with any adhesive but assembled with 19mm diameter beech dowels pressed in. The fourth sample was cut from a commercially-produced CLT panel that was fabricated about eight months before the preparation of the other samples and was bonded with a two-part melamine formaldehyde (MF) adhesive. All of the samples were then cut into 244mm x 140 mm to have a larger surface area of CLT exposed and have a single joint centred at each lamella. Sample 3 assembly is shown as in figure 1 with four dowels and during the test, a

single lamella face area was left exposed as a finished ceiling.



**Figure 1.** Cross-laminated timber (CLT) sample dimensions (b) doweled CLT sample assembled with four beech dowels (c) exposed surface area of samples during the test

When the samples arrived at the laboratory, they were kept at a conditioning period first for then days and then placed in the small-chamber test for four days. This is to evaluate the VOC emission of a new building after 14 days as it is the number of days that a new building might already be occupied after construction/renovation finished. Table 6 shows the testing conditions of the samples in conditioning period and test period.

**Table 6.** Small-chamber environmental testing conditions

Criteria	Conditioning period	The period
Duration	10 days $\pm$ 5 h	4 days $\pm$ 2 h
Temperature	23 $^{\circ}$ C $\pm$ 2 $^{\circ}$ C	23 $^{\circ}$ C $\pm$ 1 $^{\circ}$ C
Relative humidity	50% $\pm$ 10%	50% $\pm$ 5%
Air exchange rate	2 ACH minimum	1 ACH $\pm$ 0.05 ACH

After the experiment finished, there are a number of chemical emissions recorded as seen in table 7. Dowel sample has most of the chemical emission recorded and this is because woods also releases chemical emission naturally while soy-bean and PUR adhesive does not emit some of the air chemical emission. The acceptance criterion for formaldehyde emission in a building is  $<9.0 \mu\text{g}/\text{m}^3$  (Ersion 2010). Based on the table above, dowel, soy-bean adhesive and polyurethane adhesives has achieved formaldehyde emission that is at or below  $2.5 \mu\text{g}/\text{m}^3$ . But MF adhesive is different because it has a significantly high amount of formaldehyde emission of  $54.4 \mu\text{g}/\text{m}^3$ .

**Table 7.** Estimated indoor air concentrations of individual chemical compounds for a private office

Chemical	CAS No.	Estimated office indoor air concentration $\mu\text{g}/\text{m}^3$				CDPH method allowable conc. ( $\mu\text{g}/\text{m}^3$ )
		Dowel	Soy	PUR	MF*	
Formaldehyde	50-00-0	2.3	2.5	1.4	54.4	9
Acetaldehyde	75-07-0	7.1	4.5	-	5.0	70
Toluene	108-88-3	2.9	-	-	-	150
Acetic acid	64-19-7	30.5	44.7	12.0	9.5	-
2-Propanone (acetone)	67-64-1	12.6	1.8	-	2.3	-
Hexanal	66-25-1	24.5	11.7	4.2	-	-
Pentanal	110-62-3	4.1	2.1	-	-	-
2-Furancarboxaldehyde (furfural)	98-01-1	-	4.6	3.7	3.9	-
(+/-)-alpha-Pinene	80-56-8	862.0	146.4	101.3	71.6	-
beta-Pinene	127-91-3	9.5	4.4	-	-	-
d-Limonene	5989-27-5	75.3	5.9	-	6.3	-
beta Myrcene	123-35-3	9.0	-	-	-	-
3-Carene	13466-78-9	10.4	-	-	-	-
p-Cymene	99-87-6	15.9	-	-	-	-
alpha-Terpineol	10482-56-1	-	2.3	-	2.4	-
Tricyclene	508-32-7	6.8	-	-	-	-
Camphene	79-92-3	7.4	-	-	-	-
(+)-Camphene	5794-03-6	52.4	-	-	-	-
a-Phellandrene	99-83-2	2.2	-	-	-	-
beta-Phellandrene	555-10-2	2.9	-	-	-	-
Benzene, 1-methyl-2-(1-methylethyl-; (o-cymene)	527-84-4	2.2	-	-	-	-
Dehydrosabinene	36262-09-6	4.0	-	-	-	-
Terpinolene	586-62-9	15.5	-	-	3.5	-
1-Pentanol	71-41-0	2.4	-	-	-	-
p-(1 propenyl)-toluene	104-46-1	17.7	-	-	-	-
o-Isopropenyltoluene	7399-49-7	29.1	-	-	-	-
Triethylphosphate	78-40-0	2.1	2.1	-	-	-
Methyl acetate	79-20-9	-	-	-	23.8	-
2,2,4-Trimethyl-1,3-pentanediol monoisobutyrate (Texanol)	25265-77-4	-	-	2.2	-	-

\*Melamine formaldehyde (MF) sample commercially-produced and aged eight months prior to testing

Based on table 7, although dowel does not use any adhesives, it still releases a number of VOCs which other adhesives does not emit those VOCs but the VCOs emitted does not have the Chronic Reference Exposure Level (CREL) thus it is not harmful to the environment. Formaldehyde, acetaldehyde and toluene are VOCs that are that are identified with CREL thus they are harmful to the indoor environment. Soy-bean adhesive emitted formaldehyde, acetaldehyde and toluene with the air concentration of  $2.5 \mu\text{g}/\text{m}^3$ ,  $4.5 \mu\text{g}/\text{m}^3$  and  $0.0 \mu\text{g}/\text{m}^3$  respectively. While for polyurethane adhesive only formaldehyde is emitted with air concentration of  $1.4 \mu\text{g}/\text{m}^3$  meanwhile for dowel, it has emitted all of the VOCs that is identified with CREL with  $2.3 \mu\text{g}/\text{m}^3$  amount of formaldehyde,  $7.1 \mu\text{g}/\text{m}^3$  amount of acetaldehyde and  $2.9 \mu\text{g}/\text{m}^3$  amount of toluene.

Although MF does not emit toluene, the amount of formaldehyde in the air concentration

exceeds the formaldehyde emission limit and the amount is very high. When compared MF to dowel, soybean adhesive and PUR, the difference in their formaldehyde is very big thus proves that MF usage is very harmful for the indoor environment. Although dowel, soybean adhesive and PUR do emit VOCs that are quite harmful, but the amount it emitted is not high and VOC emission cannot be controlled fully but at least there are less risk of developing any kind of building related sickness among the occupants. Thus, with this result it is proven that dowel, soy-bean adhesive, and polyurethane adhesives are better options to replace the current petroleum-based MF adhesive that is widely used in the wood and furniture industry.

- Bio-based wood adhesive from camelina protein (a biodiesel residue) and depolymerized lignin with improved water resistance

From this research paper, the information that is extracted is the chemical component in camelina protein adhesive and its quality in terms of sheer strength, water resistance, etc.

Camelinas are easy growing oilseed crop that grows on dry land. In the past few decades, Camelina has drawn increasing attention due to its abundance in poly - unsaturated fatty acids, which were known for aviation fuels and health benefits (Zhu et al. 2017) and can be an alternative to soy-bean adhesives as soy-bean have high competition to be used as foods and non-foods uses (Zhu et al. 2017). But the concern for a camelina protein (CP) adhesives is that it has low mechanical strength because of weak protein-protein interaction and poor water resistance. Thus, this study paper aims to develop CP, an environmental-friendly adhesive using lignin and improve its shear adhesive strength and water resistance.

In order to improve CP as an adhesive, classic protein modification techniques is used to increase and improve the protein's hydrophobicity, reactivity and cross-linking degree. The use of lignin is studied to improve the quality of CP adhesive where it is proved that lignin has the potential to improve the mechanical, thermal and hydrophobic properties of CP adhesive. Lignin is a biopolymer that has the properties of abundant aromatic structures, and phenolic hydroxyls that are appropriate to acts as copolymer to modify a protein's hydrophobicity, mechanical properties and reactivity. The hydrophobic materials in the protein modification would help to stabilize the protein structure of CP thus making the adhesive more resistance to water.

But lignin also needs to be modified due to its inert nature that limits its solubility and reactivity which could affect its further application. The lignin needs to be modified and depolymerized to ensure its reactivity and solubility. As seen in figure 2, unmodified lignin shows less intermolecular reaction between CP and lignin because of its hydrophobic nature, it resulted in low reactivity and miscibility. However, with modified depolymerized protein, although the particle size has been reduced, it has allowed for more depolymerized lignin group to react and conjugate with CP thus increases the cohesion and hydrophobicity which resulted in enhanced adhesion strength especially under wet condition.

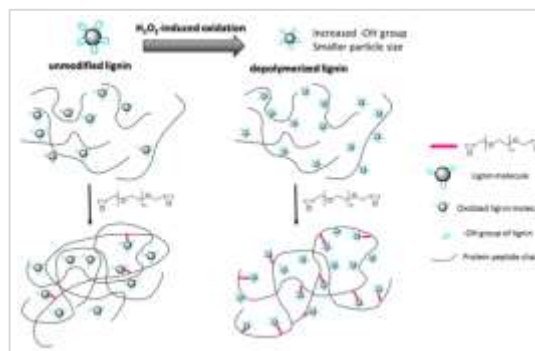


Figure 2. Reaction between the protein and oxidized lignin

Because there are newly appeared -OH groups at the surface of depolymerized lignin at oxidize lignin (OL) molecules, there will be a reaction of -OH groups with the functional group (mostly -OH and -COOH) at the wood surface. This will contribute to the interfacial adhesion strength between the wood and adhesives. Shortly, the camelina protein and oxidized lignin copolymer are expected to increase hydrophobicity and help to enhance the cohesion and the interfacial adhesion strength. Consequently, the water resistance of the adhesive would also be improved. Figure 3 shows the protein-lignin adhesives bonding between wood panels.

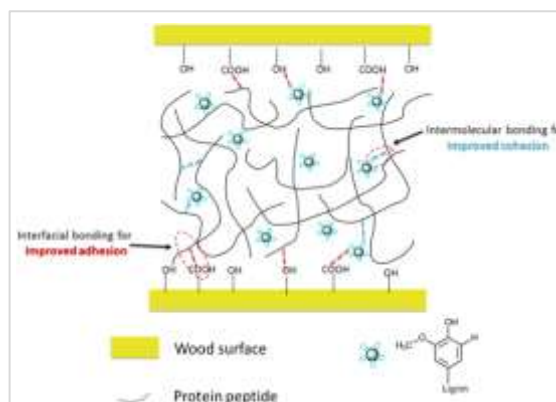
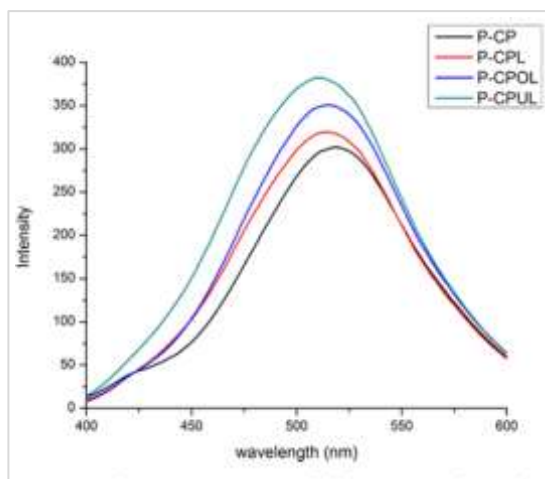


Figure 3. Protein-lignin adhesives bonding between wood panels

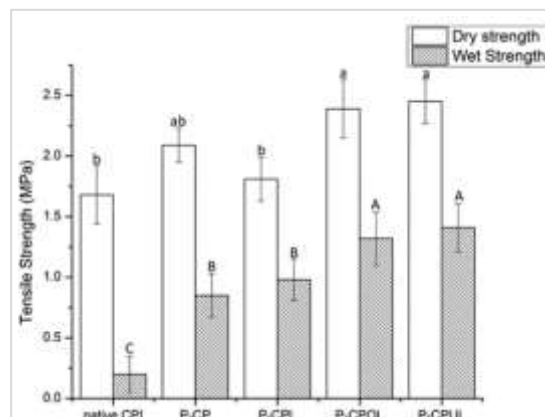
The test and experiment was continued with different type of depolymerized lignin using fluorescence intensity (FI) to see the protein-lignin interaction. In order to verify the conjugating effect of CP on different types of lignin, the hydrophobicity between CP-lignin was evaluated. FI in the presence of 1-anilino-8-naphthalenesulfonate (ANS) can determine the

hydrophobicity of CP-lignin, since Lignin that has a large amount of aromatic groups that contributed to CP's hydrophobicity. Increased FI shows that there are increased protein-lignin interaction. The four types of depolymerized lignin used in this test are P-CP, P-CPL, P-CPUL, P-CPOL and CPI which is the unmodified CP. As seen in figure 4, P-CP showed to have the lowest FI which means that it has the least hydrophobicity, followed by P-CPL, P-CPOL and lastly P-CPUL which has the highest FI thus it has the highest hydrophobicity showing strongest CP-lignin interaction.



**Figure 4.** Fluorescence intensity of different protein-lignin dispersions of P-CP, P-CPL, P-CPOL and P-CPUL

The four adhesives with different types of lignin were then tested to see its sheer tensile strength in dry condition and wet condition. The results are as shown in figure 5 and as we can see from there, CPI has the lowest sheer strength in dry and wet condition where the tensile strength for dry strength is 1.68 MPa and for wet strength is 0.2 MPa thus this shows that CPI are not water resistant. And the highest result for tensile strength is P-CPUL with tensile strength of 2.35 MPa and 1.43 MPa for dry strength and wet strength respectively. This shows that P-CPUL has the highest water resistance compared to other samples due to its hydroxyl group that were able to bond stronger hydrogen bond and covalent bond. From figure 3, we can also conclude that the water resistance increased with the sheer strength consequently.



**Figure 5.** Dry shear strength and wet shear strength of different adhesive samples

With this result, it can be concluded that CP adhesives can be improved with depolymerized lignin especially lignin that are modified to and depolymerized to P-CPUL that has the highest reaction and water resistance thus CP adhesives has the potential to be used in the industry.

At the end of this experiment, camelina protein has similar amino acid content to any other plant protein but could differ in terms of its sequence. The adhesive P-CPUL which have the chemical component of glutelin polypeptides and lignin have a strong tensile strength of 2.5 MPa and have the water resistance with strength 1.43 MPa.

## Result and discussion

Based on the case study 2, it can be seen that petroleum-based adhesives contributes a lot to the indoor air pollution compared to soybean adhesive which is plant-based adhesive. Although based on production, MF is much cheaper and less complicated as it does not require to undergo a harder chemical modification compared to soybean adhesive. Soybean adhesives are proven able to replace MF in the carpentry industry but it requires a lot of chemical modification due to its nature of weak sheer strength and low water resistance (Yue et al. 2020), just like camelina protein which in the case study 3 it needs to be chemically modified with depolymerized lignin.

But it is safe to say that based on the last results in case study 2, bio-adhesives proves to be a safer option to be used in the mainstream industry compared to MF as the amount of formaldehyde it releases are way less than that of MF with the result of of 2.5  $\mu\text{g}/\text{m}^3$  compared to

MF having the result of 54.4  $\mu\text{g}/\text{m}^3$ . Since bio-adhesive emit VOCs less thus they are safer to be used and have less risk to sick building syndrome.

Both animal-based adhesive and plant-based adhesive have one similarity which is to produce adhesives, the protein must be extracted. This is because proteins are made up of hundreds or thousands amino acids and it has the properties that is needed for adhesion and sheer strength. The sequence, amount and type of amino acids that exist in animal and plant protein are different. Based on this case study, the highest amount of amino acid in catfish protein are glycine, proline and hydroxyproline and since camelina protein have similar to soybean adhesive (Liu, Bean, and Sun 2018) thus they consist of albumin, glutelin and globulins (Boyle et al. 2018). Since there are

difference in protein composition in both subjects thus it affects the sheer strength, water resistance and adhesion of the adhesives and the amount of protein compound in animals and plants are also different.

Meanwhile for plant-based adhesives, there are no specific numbers of protein content for soy-bean protein, but the industrial grades are generally blended to yield a uniform protein content of 44~52%, depending on the source. The other principal constituents of soybean meal are carbo-hydrates, totaling about 30% and ash, at 5 or 6% (Jeon, Lee, and Kim 2011). The table below will further compare animal-based adhesives and plant-based adhesives in terms of sheer strength, water resistance, production.

**Table 8.** Small-chamber environmental testing conditions

Criteria	Animal-based (catfish protein) adhesive	Plant based (camelina protein) adhesive
Shear strength	1.73 $\pm$ 0.35a MPa as a function of hot-pressing temperature, time and layers of protein solution applied to the wood strips.	2.13 MPa for camelina protein copolymerized with oxidized kraft lignin(P-CPUL)
Water resistance	1.82 MPa	1.43 MPa camelina protein copolymerized with oxidized kraft lignin(P-CPUL)
Production	Catfish skin was mechanically removed from the fillet. Few processes were followed including washing, rinsing, drying with certain chemicals to yield the gelatin and produce the adhesive.	Need to be mixed with certain amount of soy-bean adhesive and depolymerized lignin for it to have function and good performance. A bit complicated since need to extract soy-bean protein and lignin.

## Conclusion

Indoor Air Quality (IAQ) is one of the factors that contributes to Sick Building Syndrome (SBS). VOC and formaldehyde emission has been one of the main contributors to pollutant in the indoor environment. Plant-based adhesives and animal-based adhesives could be an alternative to replace petroleum-based adhesives that is harmful to the indoor occupants as they emit little to no formaldehyde into the indoor air. Bio-adhesive emit less formaldehyde and the amount of formaldehyde release are less than the acceptance criterion for formaldehyde emission in a building which must be equal or less than 9.0  $\mu\text{g}/\text{m}^3$  thus it can be concluded that bio-adhesive have less contribution to sick building syndrome.

Both animal-based and plant based are suitable to be used as wood adhesives, but animal-based adhesive might be better in terms of its quality which is they have a quite better sheer strength, higher water resistance and easier production as it does not need to go through a

harsher chemical modification compared to plant adhesive although soy-bean adhesive are widely being studied now. With this research, we can see that an option for bio-adhesives are possible as it has less contribution to sick building syndrome and are more environmental friendly. This research can help more people see the contribution bio-adhesive have for sick building syndrome in hopes that in future researches, bio-adhesive can be modified until it is more cost efficient thus can be used in the mainstream carpentry industry.

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#### Author(s) contribution

**Zahidah binti Hamdi** contribute to the research concepts preparation and literature reviews, data analysis, of article drafts preparation and validation.

**A. Ghafar Ahmad** contributed to the research concepts preparation, methodologies, investigations, data analysis, visualization, articles drafting and revisions.