Thermal Properties of Pressurized Sawdust and Natural Rubber Latex Composites

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1. ABSTRACT

This report is based on fabrication and studying the thermal properties of eco-composites based on sawdust and Natural Rubber Latex (NRL). In this work, the sawdust of Jack tree (*Artocarpus heterophyllus)* and Mahogany tree (*Swietenia macrophylla king)* were taken as the basic raw material to fabricate the interested thermal insulating material together with NRL as the binder. Combining two low thermal conductive, eco-friendly materials to produce an affordable building insulating material was the initial motivation of this study. Jack sawdust was focused mainly due to its appreciable results. Sieved and dried fine sawdust with varying weights from 21% - 57% was used to make the composites. A constant volume (5 ml) of NRL having 60% of dry rubber content was used as the binder. When pressurized under 5 tons of force, the respective thermal conductivities of the composites were between the range of 0.2125-0.1773 W m^{-1} K^{-1}. An increment of ~17 % in thermal insulation was vividly seen in the critical composite 44% compared to the lowest and the highest weight percentage composites. The thermal conductivity of this critical composite exhibits an intermediate value between pure NRL 0.1703 W m^{-1} K^{-1} and pure sawdust 0.2550 W m^{-1} K^{-1} pressurized under 5 tons of force. The thermal properties were measured using the Hot Disk Thermal Constants Analyzer (Transient Plane Source 500). Henceforth, this economical and eco-friendly insulation material is a good stand-in thermal insulation of building applications towards efficient energy management.

Key words: Saw dust, Natural Rubber Latex (NRL), Composite, Thermal conductivity.

2. INTRODUCTION

The need to recycle waste from the timber industry is one of the most important environmental issues. Wastes like sawdust, wood shavings, wood chips and fibres have very few profitable uses. Continuous removal of such wastes from the production sites to be transformed into effective and low-price building materials has become necessary. The volume of sawmill waste is directly dependent on the volume of processing of timber for sawn timber. The volume of lumber production on a global scale is more than 400 million m^3 per year [1]. Fine sawdust is considered as one of the agricultural wastes [2], and till now has not been used in the other products. Due to its possession of firing capacity, it is usually used as a fuel source in biomass. To

alleviate environmental problems, sawdust which is a solid waste, has found a wide range of applications, for example, an absorbent to remove heavy metals, dyes, toxic salts and waste oils from water [2]. Apart from this, it is also used as a thermal insulating material. However, much work has not been attempted regarding using these wastes in the production of building materials [3]. Saw dust comprises of cellulose (40-50%), lignin (25-40%), and hemicellulose (10-25%), various functional groups, including hydroxyl, carboxylic, and phenolic groups [4] where the main chemical components are carbon (60.8%), hydrogen (5.2%), oxygen (33.1%), and nitrogen (0.9%) [5]. Many technological models have been developed and practiced for recycling timber raw materials. However, their implementation lags due to the need for sophisticated equipment, significant capital, and skilled workers. For example, chemical processing of timber waste can recycle only 25–30% of their total amount [4,5]. Studies have shown that the proportion of organic waste reaches 50%, but about 40% of them are not included in the recycling program and end up in landfills. Currently, the timber industry mainly uses stem wood. The share in low-quality wood, logging residues, and lumbering is, respectively, 15-40, 30-40 and 19-20% [6]. One of the directions of the utilization of timber waste in the form of sawdust is the production of structural thermal insulation materials in the form of wood. Due to the arising awareness of the impact of materials used on the environment, the attention of researchers is shifting to the development of recyclable and environmentally sustainable composite materials. In general, the demand for the development of natural biodegradable composites from renewable resources for a wide range of applications is increasing due to their advantages, such as eco-friendliness, lightweight, carbon dioxide reduction and biodegradable characteristics [7]. Some examples of interesting work carried out by researchers like fabricating masonry bricks with sawdust [8], sawdust concrete [9], hybrid natural composites [10], wood/natural latex eco-composites cross-linked by Electron Beam Irradiation [11] lead us to do preliminary work to fabricate a thermal insulating material for building application. Natural Rubber Latex (NRL) is harvested from the rubber tree (*Hevea brasiliensis)*. It is a stable colloidal dispersion of polymeric particles in an essentially aqueous medium. NRL concentrates have been commercially available since 1930 and subsequently have been exploited for many products. The remarkable properties of natural rubber (NR) make it preferable for many engineering applications being heavily investigated till to date. It has a long fatigue life and high strength even without reinforcing fillers. For other purposes than for this section, it can be used at approximately 100 ˚C, and sometimes above. In addition, it can maintain its flexibility down to 60 ˚C, has a good creep and stress relaxation resistance, and the cost is low [11]. It is also widely used as a binder due to its desired properties such as its availability, adhesiveness and low insulating properties. The combination of sawdust as the base and NRL just as the binder was carried out to fabricate an eco-friendly composite in order to study its thermal properties.

3. METHODOLOGY

Sawdust of Jack tree obtained from a sawmill was sieved to remove large scraps of wood to obtain fine dust. This sawdust was dried in the oven at 80 ˚C for 6 hours to remove the moisture. The dried sawdust was then sieved again to remove further impurities, if any present. Different weights of this fine sawdust, as shown in Table

1, were mixed with NRL (having a 60% of dry rubber content) to make a series of composites. NRL was stirred at a temperature of 70 ̊C with a 50 - 60 rpm stirring speed to remove ammonia present in it prior to use in the composite fabrication [12]. Each composite was made by blending the measured sawdust with 5 ml of NRL for 30 s until a fine composite was obtained. This blended composite was then kept aside for 5 hours to decrease its moisture content. Blending prevents aggregation of the sawdust, homogeneous dispersion in the rubber matrix and forms a network structure. Each composite was measured and divided into two equal masses to make the pressurized samples. They were made by using the hydraulic pellet press. A force of 5 tons was applied to the blended composite, making a sample of the thickness close to 10 mm

Table 1: Percentage weight of sawdust and NRL

Weight of sawdust (g)	Volume of Latex (ml)
1.00 (21%)	5
1.50 (28%)	5
2.00 (35%)	5
2.50 (40%)	5
3.00 (44%)	5
3.50 (48%)	5
4.00 (51%)	5
5.00 (57%)	5

The prepared samples were placed over a hot plate at 80 ̊C for 6 hours and then left out in the sunlight for two days to confirm the removal of moisture. A trial series of the above composite was made by hand mixing to ensure that unpressurized samples followed the same trend. This mixing was done carefully to avoid coagulation and lump formation of NRL. It was possible to obtain an even composite. The thickness of the samples was nearly 3 mm with different diameters. Unpressurized composites from Mahogany sawdust too were fabricated in the same process for comparison. Unpressurized, acid treated and untreated samples of both sawdust were also fabricated. Acid treatment was done by adding 5 ml of dilute formic acid. The thermal properties of the finally dried composite samples were measured using Hot Disk Thermal Constants Analyser (Transient Plane Source (TPS) 500 S). The TPS method is based on using a transiently heated plane sensor for the measurement of thermal properties. This method can measure the thermal conductivity, thermal diffusivity and volumetric specific heat capacity simultaneously in a short time for isotropic materials over a wide range of temperatures. Figure 1 shows the brief transformation process from sawdust to the measurement of the fabricated composite.

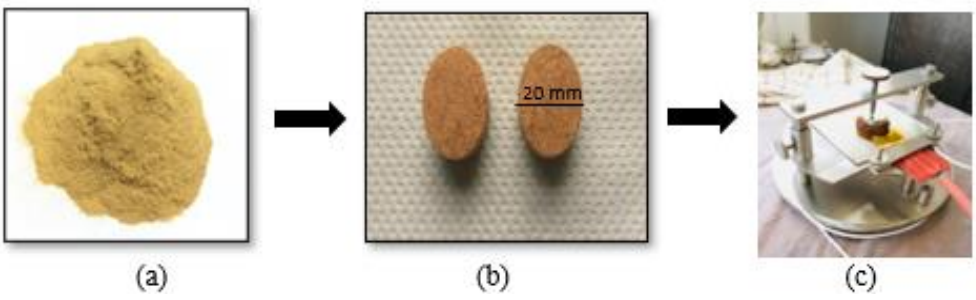

Figure 1: Photographs of (a) Jack saw dust (b) Pressurized sample with saw dust and latex (c) Measuring sample.

4. RESULTS AND DISCUSSIONS

4.1 Thermal Characterization

Firstly, the two set of unpressurised, acid treated and untreated composite samples from the sawdust of Jack and Mahogany were measured. Acid treated composite from Jack and Mahogany sawdust exhibited a thermal conductivity value of 0.2039 W m^{-1} K^{-1} and 0.2118 W m^{-1} K^{-1} respectively. The purpose of acid treatment was to speed up the coagulation of the rubber polymer. It was envisaged that the protein membrane of rubber polymer in rubber particles are negatively charged. The negatively charged rubber particles repel each other, preventing themselves from combining and coagulating. Hydrogen ions from the acid neutralise the negative charges on the surface of the membrane, and a neutral rubber particle is formed. When these neutral particles collide with each other, their outer membrane layers break up. The rubber polymers are set free to coagulate [13]. Acid untreated composite with the same composition as of the acid treated one showed 0.1917 W $m^{-1}K^{-1}$ and 0.1920 W m^{-1} K^{-1} for Jack and Mahogany respectively. This acid untreated composite coagulates naturally by the slow bacterial attack from the air on the protein membrane to produce lactic acid. The ionisation of the lactic acid produces hydrogen ions. The hydrogen ions neutralise the negative charges to form neutral rubber particles, allowing coagulation to occur [13]. From the above results it can be seen that the acid untreated composite sample gave a lesser value for thermal conductivity than the acid treated ones. Since acid untreated Jack sawdust composites exhibited appreciable values, the entire work was carried with Jack sawdust without acid treatment. Thermal properties such as thermal conductivity, thermal diffusivity, and volumetric specific heat capacity of the fabricated composites were measured for characterization. At room temperature under the parameters of heating power 80 mW, measuring time 10s and Kapton 5465 sensor of the Hot Disk Thermal Constant Analyzer 500 S the composites were measured. Figure 2(a) shows the thermal conductivities of both pressurized and unpressurized Jack sawdust composites.

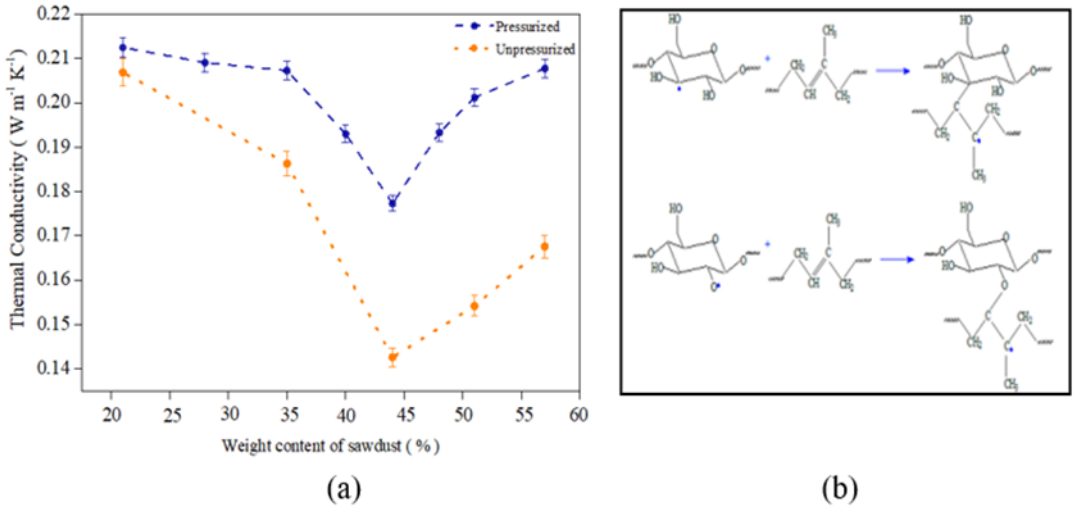

Figure 2: (a) Thermal conductivity variation of both pressurized and unpressurized samples (b) Possible cross-linking mechanism between cellulose from saw dust and NRL molecules.

Both series of the composites follow the same pattern when the weight of the sawdust increases. It could be seen clearly that the thermal conductivity values drop and then rise with the increase in the weight of sawdust. The minimum value of thermal conductivity was observed when the sawdust content weight percentage is closer to 44% in both cases known as the critical composition. This shows an appreciable decrease in thermal conductivity compared to other composites. An increase in thermal insulation property of ~17% compared to the composite with the lowest and the highest weight of sawdust is noticeably seen in the pressurized composite in Figure 2(a). Thermal conductivity of the pressurized critical composition exhibited an intermediate value between the values 0.1703 W m^{-1} K^{-1} and 0.2550 W m^{-1} K^{-1} which were measured for fabricated pure NRL and pure sawdust samples under 5 tons of force. In unpressurized sample, an increase of ~31% of thermal insulation is observed yet, when considering the other thermal and physical properties, it doesn't stand as a good thermal insulating composite. Thermal conductivity value of sawdust in powder form and isotropic thermal conductivity value of a circular wood piece of nearly the same thickness of the fabricated pressurized sample were obtained for comparison. The values obtained were 0.1053 W m^{-1} K^{-1} and 0.4958 W m^{-1} K^{-1} respectively. This shows that the air spaces in between the composite play a vital role in heat conduction. The cross-links and the 3D network structure formed by the NRL molecules also add up to this trapping effect. As the sawdust quantity in blends increases, the cross-link density also increases because the filler action of the sawdust in natural rubber blends leads to their reinforcement until a point [2, 11]. A new lattice structure may be formed due to the force exerted, which must be further clarified in future work. A connection was established between the sawdust loading and the thermal properties of the composites as shown in Figure 2(a), Figure 3(a) and 3(b). Thus, they present a trend in thermal properties as a function of sawdust weight content. Moreover, there is a minimum critical weight percentage above which the fibres do not decrease the thermal conductivity. This may be mainly due to the attribution of the poor interfacial adhesion between the polymeric matrix and hydrophilic lingo-cellulosic fillers which does not allow for efficient heat transfer between the two phases of the material [15]. In addition, these results may also be due to the elastomeric matrix because several studies have shown that latex may be used to improve the interactions at the fibre/matrix interface [7,10,11]. It is compatible with the hydrophobic rubber and the hydrophilic fibres in sawdust. The hydrophilic nature of sawdust may contribute to a significant problem due to the moisture absorbance by its cellulose fibres. The moisture content is dependent on the non-crystalline parts and voids [11]. Natural rubber and cellulose from wood sawdust join together through the C–C and C–O–C bonds, as shown in Figure 2(b). Proper vulcanization has to be done in future work to test ignition property, tensile stress and impact. This study is a preliminary work to find out the insulating property of the composite and the critical composition. Figures 3(a) and 3(b) shows the variation of the respective samples' thermal diffusivity and volumetric specific heat capacity. The thermal diffusivity of pressurized composites was the lowest when the sawdust content was in the region 40 to 50 %. At the same time, the volumetric specific heat is highest for the critical

composition. It is well observed that the volumetric specific heat capacity behaves similar to the thermal conductivity pattern and thermal diffusivity vice versa. It is known that the lower the thermal conductivity and higher the specific heat, the heat will transfer slowly.

This may be the case with typical thermal insulators. By comparing the behaviour of the composites on thermal diffusivity and volumetric specific heat capacity, the pressurized samples appear to act as a good thermal insulator, especially at the critical weight content.

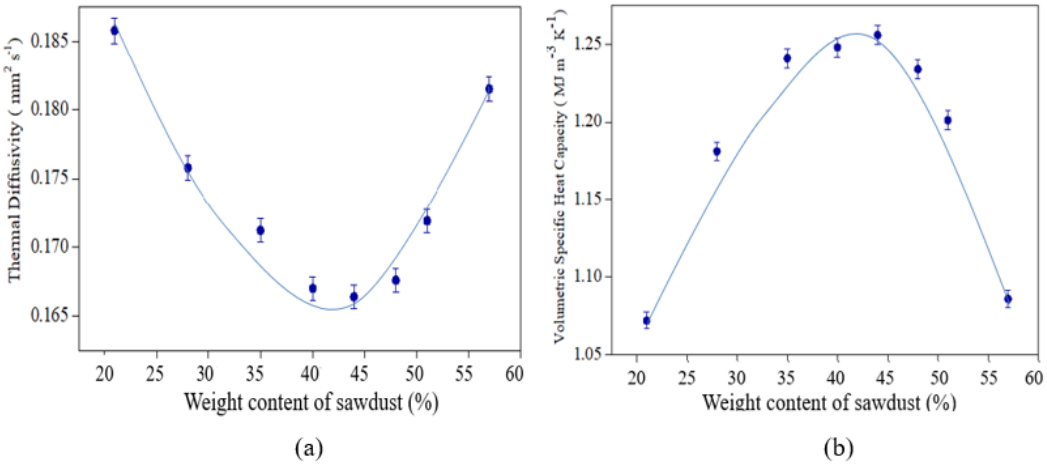

Figure 3: (a) Thermal diffusivity of pressurized samples (b) Volumetric specific heat capacity of the pressurized samples.

The variation of the thermal conductivity with the applied pressure is shown in Figure 4. Initially 5 tons of force was used to fabricate the composites considering it as a reasonable and sufficient force for fabrication. To verify this, thermal conductivity of 3 g of raw sawdust under various force of pressure was observed to track out the pattern. The figure below depicts clearly the increase of thermal conductivity when the force of pressure increases. Until the point of 8 tons, the thermal conductivity increases sharply, whereas increasing thermal conductivity slows down afterwards. It can be seen that after 5 tons of force the increment in thermal conductivity doesn't increase drastically and surprisingly gives nearly the average value of the measured thermal conductivities.

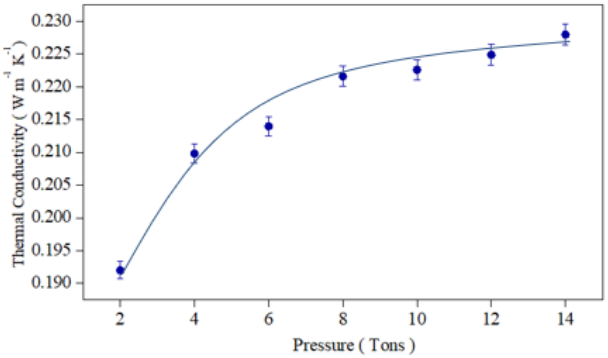

Figure 4: Variation of the thermal conductivity of raw saw dust under various pressure

4.2. Fourier Transform Infrared (FTIR) Characterization

To investigate the molecular chemical structure of the composite, Fourier Transform Infrared (FTIR) characterisation was carried out using Nicolet iS50 (Thermo Scientific) FTIR instrument. For further comparison, pure sawdust and less pressurized samples was also analysed. This is shown below in the Figure 5. Bands from 2940–2840 cm^{-1} are specific to natural rubber and cellulose, lignin or hemicellulose, from the wood sawdust fibres existing in the mixture [16]. Since no chemical treatment was done on sawdust, the peak at 1733 cm^{-1} that belongs to the carbonyl vibration in ester groups appears even after pressurizing [17]. The band at 1231cm^{-1} is observed in the sample (c). The peaks within the range of 1629 cm^{-1} and 1118 cm^{-1} shows a shift towards right in both composite samples and intensified.

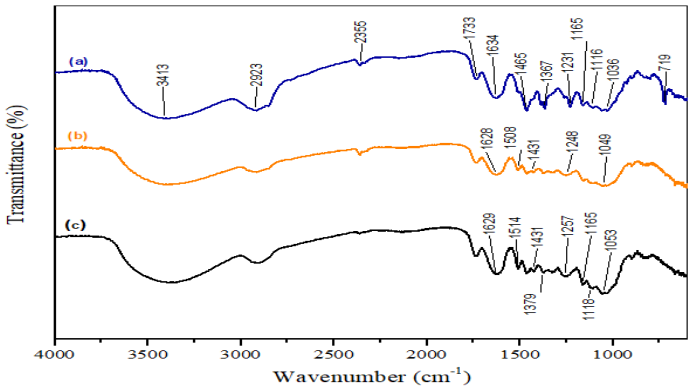

Figure 5: FTIR spectra for (a) Composite under a force of 5 tons (b) Composite under a force of 1 ton (c) Pure sawdust

Table 2: Assignment of the FTIR measured bands for Saw dust

Frequency (cm^{-1})	Assignment	Component	Reference
3413 band 2932	O-H C-H stretching vibration	hydroxyl group in cellulose, carbonyl group of acetyl ester in hemicelluloses, and carbonyl aldehyde in lignin	14, 16
1733	C=O		
2940-2840	C-H methyl and methylene groups	amorphous cellulose in methyl and methylene groups	15
1231		vibrations of the syringyl structure of the lignin	15
1053-1036	C-OH C-O-C	Lignin	16

Table 3: Assignment of the FTIR measured bands for NRL

Frequency (cm^{-1})	Assignment	Reference
3283 band	N-H stretching proteins	17
2961, 2928	$-CH_3$, $-CH_2$ stretching asymmetric	17
2912, 2581	$-CH_3$, $-CH_2$ stretching symmetry	
1733	R1-(C=O)-R2 (lipids)	18
1665-1660	C=C valence vibration	18
1634	amides	18
1465	$-CH_3$ deformation, CH_2 Rocking	19
1367	$-CH_3$ deformation asymmetric	18
1246	C-O-C stretching symmetric	19
1241	$-CH_2$ twisting	19
1036	$-CH_3$ rocking	18

in the composite pressurized at 5 tons as a result of bond degradation and cross linking. Considering NRL, a peak at 2355 cm^{-1} is observed in both the composites and not in the pure saw dust. This may correspond to the new bond formation between the NRL and the saw dust fibres. Peak 1367 cm^{-1} corresponds to deformation asymmetric -CH_3 [17]. The peak at 1231 cm^{-1} in (a) has been shifted to left at 1248 cm^{-1} in (b) and 1257 cm^{-1} in (c). The peaks at 1300–1000 cm^{-1} could be assigned to the C–O stretching vibration of hemicelluloses and lignin of saw dust too [18]. A new peak was observed only in the sample (a) at 719 cm^{-1} which could be formed due to pressure and formation of new saturated C-H bonds and existence of aromatic nuclei of sawdust [13,14,18]. These changes might occur as a result of the elastomer crosslinking and of the consuming of double bonds in lignin. Above results show, due to the matrix formation between NRL and sawdust, voids may be created in various proportion influencing the heat transfer in each composite. Thus, decreasing the thermal conductivity as least of the critical composition as observed.

5. CONCLUSIONS

The main motivation for this study is to produce a low insulating composite combining two low thermally conducting materials and study its thermal properties. With the increase of the weight of sawdust, the thermal conductivity was observed to be decreasing in the composites. The lowest value of 0.1773 W m^{-1} K^{-1} was observed in the pressurized composite sample with sawdust of 44%. This composite further showed an increase in thermal insulation property of ~17% compared to the composite with the lowest and the highest weight content of saw dust. With a further increase in the content of sawdust, the thermal conductivity increases as the sawdust particles tighten to give a new lattice property with less air penetration. The cross-links and the 3D network structure formed by the NRL molecules also add up to this trapping effect. The blended polymeric composite matrix results in efficient heat transfer between the material Henceforth, further work could be planned on this economical and eco-friendly composite for industrial applications.

6. ACKNOWLEDGMENT

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