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RESEARCH ARTICLE

Composite Materials Based on Rice Straw and Natural Rubber for Thermal Insulation Applications

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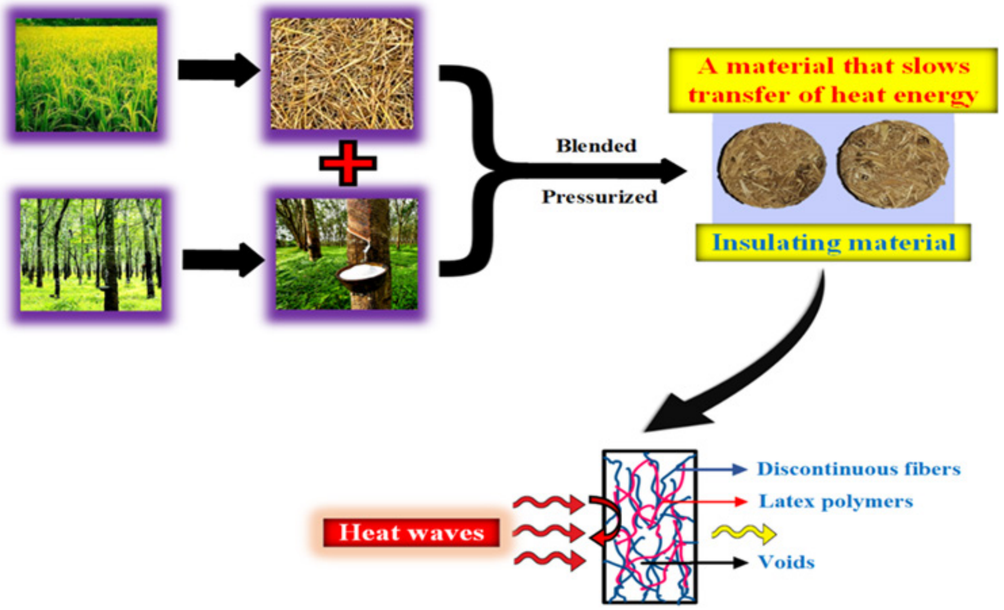

Highlights

- Thermally insulating composites with rice straw blended with natural rubber latex is a promising value-added waste material.
- The thermal conductivities ranged from 0.1902- 0.1526 W m^{-1} K^{-1}.
- An acceptable thermal insulating composite was found at 25% of the weight content of rice straw.

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Composite Materials Based on Rice Straw and Natural Rubber for Thermal Insulation Applications

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Abstract: Thermal properties of composites fabricated with coalescence of two low thermal conducting materials, rice straw and natural latex were investigated. Composites constituting various weight content of oven-dried blended rice straw mixed with a constant volume of natural latex were fabricated with a surface area of 31 cm^2 and their thermal properties were compared by pressurizing under a force of 5 tons. Hot Disk Thermal Constants Analyzer Transient Plane Source (TPS) 500S was used to measure the thermal properties such as thermal conductivity, volumetric specific heat capacity, and thermal diffusivity of the aforementioned composites. The lowest thermal conductivity was achieved for both the unpressurized and pressurized composites with 25% of the rice straw's weight content, which was recorded as 0.0636 and 0.1526 Wm^{-1}K^{-1} respectively. High Specific heat capacity and less thermal diffusivity were also seen in the pressurized samples with 25% of rice straw, whereas this behaviour was the opposite in the unpressurized sample. The effect of the applied pressure on the thermal properties of the composite is also studied and it was observed that the thermal conductivity increases up to 8 tons on 31 cm^2 in the composite with increasing pressure and then decreases while it continues to increase in the sample made of rice straw alone. Since high specific heat and low thermal diffusivity are the desired features of a thermally insulating material other than the low thermal conductivity, this economical and eco-friendly composite pressurized up to a certain limit could be used by further processing with preservatives toward efficient energy management as a good material for thermal insulation of building applications.

***Keywords**:* Thermal insulator; Rice straw; Natural latex; Composite; Thermal properties.

INTRODUCTION

The awareness of experts about global warming, the greenhouse effect, and climatic changes has led to more efforts to improve energy conservation and environmental friendliness. This impact has drawn the attention of researchers specifically on agricultural and industrial natural waste materials, particularly those comprised of natural fibres because of their eco-friendliness. To assess the outstanding ways to gain from these wastes and utilize them by avoiding hazards resulting from burning or leaving them in the field to decompose, researchers require information about their physical, mechanical, and thermal properties. The use of natural fibre composites or natural fibre toughen composites under various environmental factors has become famous in recent years. Compared to synthetic fibres, natural fibres have special advantages, such as biodegradability, low cost, non-abrasiveness, low energy consumption for process and low density. Nevertheless, some drawbacks, such as brittleness and moisture absorption are possessed by natural fibres (Mishra *et al.*, 2001). The effective use of this agricultural waste has attracted the attention of countries in which crop cultivation is their main revenue. A various number of "byproducts" are produced in varying quantities of chaff, straw, and weed seeds when cereal crops are processed after harvest (Hillman, 1981, 1984; Jones, 1984, 1987). Straw, a renewable resource grown annually is the stalks remaining after grain harvest. It is just a tenable economic material suitable for building almost any structure like free acoustic and safety walls whilst also being a residue to be burnt. It is also alike to the chemical composition of wood. In many applications straws from grass, wheat, oats sugarcane and more could be utilized. A by-product after paddy harvest is rice straw and each kilogram of milled rice roughly produces 0.7 to 1.4 kg of rice straw depending on the rice variety (IRRI, 2017). It comprises cellulose, hemicellulose, silica, and lignin. Cellulose and hemicelluloses are fibre organics, while lignin is in the cell wall of the biomass (Chen, 2014). Therefore, on average natural rice straw contains 33-40% cellulose, 24-28% of hemicellulose and 2-25% of lignin (Mukherjee *et al.*, 2018).

Although this rice straw is considered as a waste material, on the other hand, from ancient times it has also been used as a building material for the roof of houses to cool and comfort the interior (Asdrubali *et al.,* 2015). Architecture in ancient times usually presents an intuitive procedure to build designs for local environmental conditions. However, this practice has been neglected in new technology since its desired interior environment requires conventional fuels to be provided. Recently, these ancient concepts fusing with new materials have led to a new stream known as "Passive Buildings". However, its industries employ millions of people, especially in developing countries. Insulating buildings with good quality indoor air with proper circulation is a significant challenge for energy management in construction. Apart from this, the key role

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is played by the cost and type of insulation material when it comes to the application (Al-Homoud, 2005). The novelty and principal motive of this study is to engineer an eco-friendly composite material with low thermal conductivity. In this work, rice straw and Natural Rubber Latex (NRL) both of that have low thermal conductivity, are cost-effective and abundantly found in Sri Lanka, were used to fabricate an eco-friendly composite, where straw is the base, and the NRL is the binder.

The rubber tree (*Hevea brasiliensis)* is the source of Natural Rubber Latex (NRL). It is a stable aqueous medium of polymeric particles distributed in a colloidal manner. NRL has been exploited for many products commercially since 1930. For many engineering applications the remarkable properties which have been extensively investigated to date of natural rubber (NR) make it preferable. For purposes other than for this application, it can be used at approximately 100 ℃, and sometimes at higher temperatures up to 180 ˚C. Even without reinforcing fillers, it has high strength and a long fatigue life. NRL melts and becomes sticky when heated to temperatures above 50 ˚ C. At approximately 120 ˚ C it begins to decompose. Due to its desired properties such as its availability, adhesiveness, low insulating properties and its ability to bond to a wide variety of materials, including fabric and metals, it is also widely used as a binder.

The physical properties of any composite fabricated out of rice straw and the NRL depend on the matrix, size and shape of the fibres, and their interfacial bonds between the fibre and the NRL. Adhesion between a matrix in composite materials and strengthen fibres plays a vital role in the final mechanical and thermal properties of the composite (Mallick, 2007). Currently, the thermophysical properties of natural fibre–reinforced polymer composites have attracted the attention of various researchers and material scientists from all over the world. (Indicula *et* al., 2006; Sgriccia and Hawley, 2007; Li *et al.,* 2008; Kim *et al.,* 2006).

By considering the above facts, in this work we explore the possibilities of fabricating an eco-friendly low-cost insulating composite by studying the thermal properties of different compositions of rice straw and NRL, as well as the influence of varying conditions of pressure. .

MATERIALS AND METHODS

Raw rice straws obtained from the paddy fields were dried, cleaned, and cut into small lengths of nearly 5 – 6 cm and subjected to oven drying at 80° C for 4 hours. These dried cut pieces of rice straw were then blended using a kitchen blender to get fine strands to make the composite samples. Different amounts of this blended straw were mixed with a constant 5 ml volume of NRL having 60% of dry rubber content to make a composite series, as shown in Table 1. Prior to the addition, with a stirring speed of 50 - 60 rpm, the NRL was stirred at 70 ˚ C in order to remove ammonia present in it (Chanatip and Somchai, 2014).

The blended mixture was then subjected to pressure using the hydraulic pellet press (Specac, GS25011 Model) with a force of 5 tons on a surface area of 31 cm^2, with a sample thickness of 5 mm. The prepared pellets were oven-dried at 60° C for 6 hours and for further removal of water the samples were kept in a desiccator under sunlight for two days. This was ensured by weighing the samples at regular time intervals until a constant weight was observed for each sample. Figure 1 shows the photograph of a fabricated eco-composite. Apart from that, unpressurized composites were fabricated by hand mixing the straw and latex and moulded to a sample with a thickness close to 5 mm and a diameter of 60 mm, ensuring that the NRL was well bound to the straw. The Hot Disk Thermal Constants Analyser (Keithley, Transient Plane Source (TPS) 500S) was used to measure the thermal properties of the finally dried composite samples.

Table 1: The ratio of straw and Natural Rubber Latex (NRL) in the fabricated composites.

Weight of straw (g)	Volume of NRL (ml)
0.50 (12%)	5
0.75 (17%)	5
1.00 (21%)	5
1.25 (25%)	5
1.50 (30%)	5
1.75 (32%)	5

Figure 1: Photograph of fabricated composite with rice straw and latex.

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 within a short time for isotropic materials over a wide range of temperatures. One of the advantages of this analyzer is that all those three parameters could be obtained at one sample measurement. The Hot Disk sensor has an etched out double spiral shape pattern of a thin (Nickel) foil for electrical conductivity sandwiched between two thin sheets of an insulating material (Kapton). A sensor is placed between two pieces of the measuring sample where the surface facing the sensor has to be smooth when performing a measurement. To increase the temperature of the sensor between ranges of a fraction to several degrees, a strong electrical current is passed through it. Simultaneously, the increase of resistance with temperature is recorded together as a function of time. Thus, making the Hot Disk sensor act both as a dynamic temperature sensor and a heat source too. The transient recording must be obstructed as soon as any effect from the outside boundaries of the two sample pieces is recorded by the sensor. This is due to the assumption based on the solution of the thermal conductivity equation as the Hot Disk sensor is located in an infinite medium. Here, the samples were measured under the parameters of heating power of 50 mW and measuring time of 10 s using the Kapton 5465 sensor at room temperature.

RESULTS AND DISCUSSION

The variation of the thermal conductivities of the unpressurized and pressurized samples of the fabricated composites with their rice straw content is shown in Figure 2. Both series reveal the same trend in their values as the weight of the rice straw increases. Initially, 5 tons/31 cm^2 of force was used to fabricate the pressurized composites considering it as a reasonable and sufficient force for fabrication. However, the thermal conductivity values of the pressurized samples are higher than those of the unpressurized samples. It is vividly seen that with the increase in the weight of the straw, the values of thermal conductivity drop and then rise. A significant decrease in thermal conductivity is seen in both series when increasing the amount of rice straw in the composites until it reaches around 25%, which is the critical composition. Compared to the composite with the lowest weight of rice straw in the pressurized composite, an increase of ~20% in thermal insulation is noticeable. The value exhibited by this composite is between 0.1588 W/m K and 0.0954 W/m K which represents the thermal conductivity of the pure latex and raw rice straw alone fabricated and pressurized under a force of 5 tons, respectively. An increase of ~71% in thermal insulation was exhibited by the unpressurized sample at the critical composition, compared to the lowest straw-weight composite in the series. The unpressurized composite possesses a value lower than the value obtained for pure latex and pure straw, which is an interesting observation in this study. As a trial and error, another composite was also fabricated by hand mixing but pressed under a workshop roller for comparison. Its value was found to be 0.1506 W m^{-1} K^{-1}. This value fell within the range of values of pressurized and unpressurized composites. The observed results lead us to conclude that a vital role in heat conduction is played by the air spaces in-between the composite.

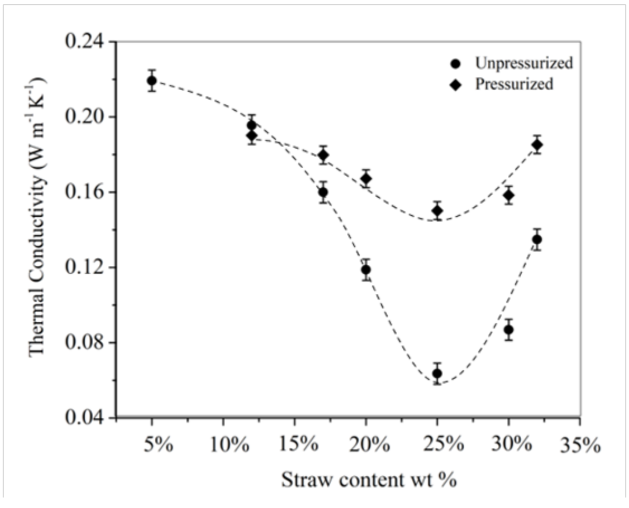


Figure 2: Variation of the thermal conductivity with different weights of rice straw of the pressurized and unpressurized samples.

The reason for the observation of the critical composite could be due to the crosslinks of latex macromolecules and the three-dimensional network giving a unique property to the composites in comparison to the other unpressurized samples. Microstructural factors such as fibre length, orientation, distribution, packing arrangement, diameter and volume fraction influence the properties of fibrous composite materials. Moreover, the bond strength between the latex macromolecule matrix and the fibres accumulates to enhance material performances (Paul *et al.,* 2008). Due to the electrostatic forces or chemical bonds in the presence of internal stresses, immobilization of the matrix chains, voids or micro-cracks in the interlayer, different properties are possessed by the polymer network in proximity with the fibre surface (Karger-Kocsis, 1995). While blending, the latex particles could be mixed uniformly with the layers and interpenetrated with each other. The three-dimensional meshwork and the cross-links formed by the NRL molecules sum up the trapping effect of air. Further, the disorientation of the straw trapping more air spaces could have prevented continuous heat transfer throughout the sample. After the critical composition, the thermal conductivity shows an increase with the weight of the straw. This could be due to the silica and other materials which support the thermal conducting property present in the strands giving a cumulative contribution to this behaviour, inhibiting the insulation effect. Apart from this, the penetration of air through the composite may be inhibited with the increase in the weight of the straw due to the drastic decrease of air spaces while compressing and tightening the straw and latex. Further investigations into properties such as density, the volume of voids, FTIR and XRD are needed to understand this behaviour completely. Until a certain point, the filler action of the fibre in natural rubber blends leads to their reinforcement as the straw quantity in blends increases which in turn increases the cross-link density (Kaewpruk *et al.*, 2021; Khedari *et al.*, 2004). Further clarification must be carried out since there are high chances of a new lattice structure being formed due to the force exerted. However, it was also observed that a minimum critical weight percentage of straws is required to increase the thermal insulation.

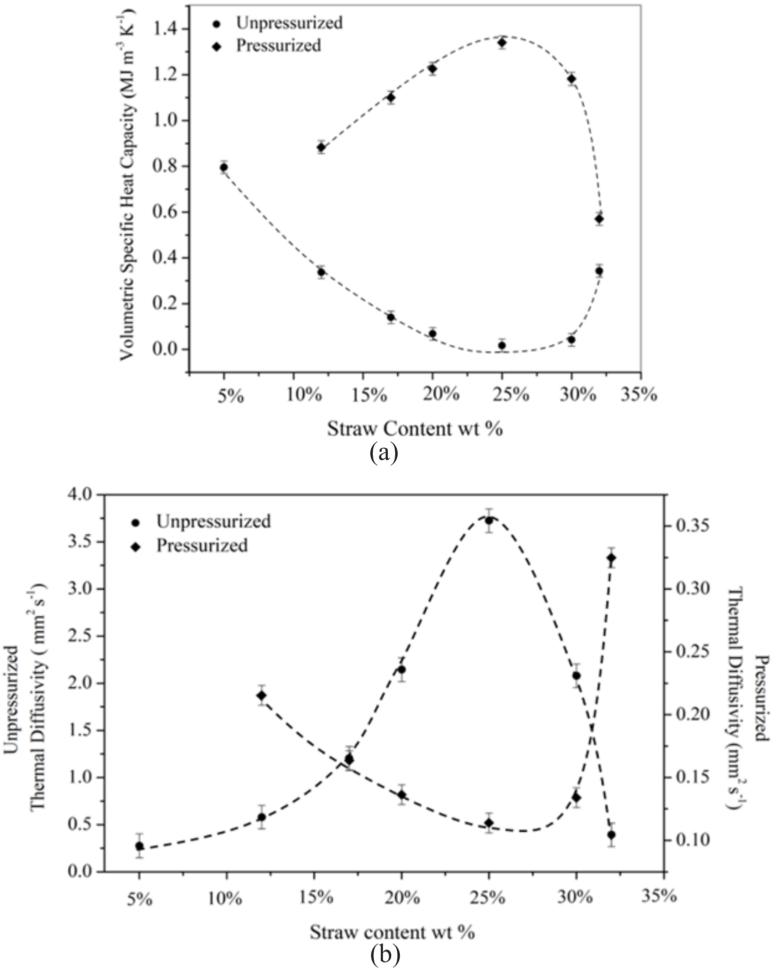
This might be a result predominantly due to the hydrophilic lingo-cellulosic fibres and polymeric matrix attribution of the mediocre interfacial adhesion which does not allow for efficient heat transfer between the two phases of the material (Rahman, 2010). In addition, several studies have shown that latex may be used to improve the interactions at the fibre/matrix interface. These results may also be due to the restriction of the mobility of the elastomeric matrix/ polymer chains because (Njoku *et al.,* 2012; Manaila *et al.,* 2016; Khedari *et al.,* 2004). The hydrophilic fibres in straw are consistent with the hydrophobic rubber. Bonds like C–C and C–O–C are formed between natural rubber and cellulose straw while the voids and non-crystalline parts govern the moisture content (Ismail *et al.,* 2002). The cellulose fibre's affinity to moisture gives the hydrophilic nature of straw and causes a significant problem too. Mechanical properties such as ignition, tensile stress and impact test must be done in future work after vulcanization. Yet further study is being progressed as this is an extended work of our preliminary work finding out the insulating property of the composite and the critical composition. Another notable feature observed overall is that at the regions of lesser and higher weight of straw content, the values of the thermal conductivities seem to converge. Table 2 shows the comparison of thermal conductivity values of the composites fabricated in this work with other reported thermal insulation materials.

Interestingly, the unpressurized rice straw with NRL had a lower thermal conductivity range than that of the particle board from the mixture of durian peel and coconut coir and is in the order with the other two thermal insulation materials mentioned in Table 2. The pressurized straw and NRL sample show a value greater than all. However, this shows that the rice straw with NRL could be considered as one of the suitable insulating materials for industrial purposes. Apart from the thermal conductivity, volumetric specific heat capacity and thermal diffusivity were obtained simultaneously for unpressurized samples and pressurized samples. Figures 3(a) and 3(b) shows the variation of the aforementioned samples' volumetric specific heat capacity and thermal diffusivity.

This study on volumetric-specific heat capacity and the thermal diffusivity behaviours with the straw content reveals that the thermal diffusivity behaviour of an unpressurized sample follows the same trend as thermal conductivity

Table 2: Tabulated values for thermal conductivity of fabricated thermal insulation materials.

Sample boards	Thermal conductivity ($W m^{-1} K^{-1}$)	Source
Mixture of durian peel and coconut coir	0.0728	Khedari *et al.* (2004)
Straw and NRL composite (Unpressurized) (Pressurized)	0.0636 0.1526	 Present work
Water hyacinth fiber with NRL	0.0246	Chanatip and Somchai (2014)
Pineapple leaf fiber with NRL	0.0223	Suankaeo (2011)



(a)

(b)

Figure 3: Pressurized and unpressurized sample's (a) Volumetric Specific Heat Capacity and (b) Thermal Diffusivity.

and vice versa for volumetric-specific heat capacity. During changes in temperature with time the associated propagation of heat into the medium gives the physical significance of thermal diffusivity. The smaller the thermal diffusivity, the more time is required for heat to penetrate the solid. With less thermal diffusivity and a high Specific heat capacity is a good quality thermal insulator. This behaviour is seen in the pressurized samples. Therefore, the pressurized samples could be taken as a better stand for an insulating material than the unpressurized composite in the fabrication of green insulating composites. To ascertain the affinity between pressure and thermal conductivity, two more series of raw straw and composite samples were fabricated with the critical composition. Figure 4 shows the variation of the thermal conductivity with the pressure used to fabricate the pelletized samples.

The thermal conductivity of the raw rice straw series increases with the pressure gradually whereas the composite shows a pattern with a maximum as the pressure increase. The raw straw sample shows a continuous increment in values as the pressure increases. This may be due to the tightening of fibres and fewer air spaces available as explained earlier. But an interesting observation was seen in the critical composite. The thermal conductivity value increases until the force of 8 tons and then gradually decreases.

One of the notable feature in the figure 4 is that the thermal conductivity values for both the series at a force of 8 tons/31 cm^2 are nearly the same. The thermal conductivity values go apart after this point. The decrement may be due to the breakage of bonds in elastomer in the composites when a huge force is applied. Figure 5 shows the volumetric specific heat capacity and thermal diffusivity; the other two thermal properties of the composite.

As mentioned earlier high specific heat and low thermal diffusivity are the desired features of any thermally insulating material. From the above results, it could be deduced that at higher pressure the composite losses its insulating property gradually comparably to the lower pressure region. Although the two ends of the graph of Figure 4 show less thermal conductivity, the change in values is not drastic. But the change in volumetric specific heat capacity gives a wide range of changes in its value. Comparing the two ends of Figure 5(a) it is vividly seen that the lower side has greater values than the higher side. This depicts that the composite in the lower pressure region has more insulating properties than the composites in the higher region. Figure 5(b) too supports this relationship. Thus, a lesser pressurized region could be suggested for further fabrication. Therefore, taking all these parameters into account an effective insulating composite material could be made for the construction industry in the market with appropriate finishing.

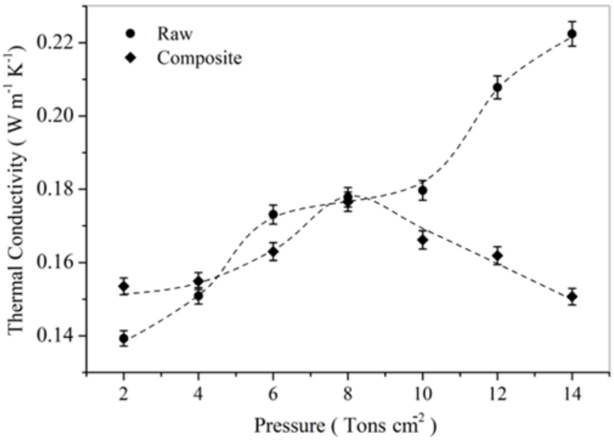
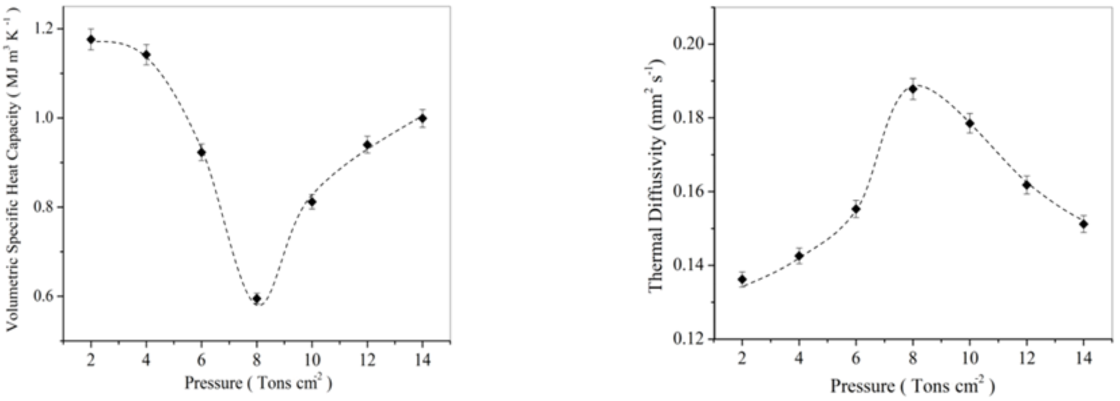


Figure 4: Thermal conductivity of the raw straw and composite pellets under various pressure.



(a) (b)

Figure 5: Observation of the effect of pressure on thermal properties of critical composite (a) Volumetric Specific Heat Capacity and (b) Thermal Diffusivity.

CONCLUSION

The principal motive of this study is to engineer a low-insulating composite combining two low thermally conducting materials, straw and natural rubber, and study its thermal properties. With increasing straw weight, the thermal conductivity was observed to be decreasing until the critical composition. The lowest observed value for the pressurized composite was 0.1526 W m^{-1} K^{-1} with a straw content of 25%. Furthermore, this composite showed an increase of ~20% in thermal insulation property compared to the composite with the lowest straw weight. Increasing further the content of straw, the thermal conductivity increases as the cumulative effect of silica appears with less air penetration. Henceforth, taking all these parameters into account, further work could be planned on this economical and eco-friendly composite for industrial applications**.** Concerning the relationship between the applied pressure and the thermal properties, the thermal conductivity of raw rice straw gradually increased with the pressure, but the critical composite initially increased and then started to decrease with increasing pressure. The increment may be due to the tightening of the fibres and the less air space available, whereas the decrement may be caused by the breakage of bonds in the elastomer in the composites when a huge force is applied. A study is also in progress on the thermal conductivity of alkaline-treated fibres where the amorphous hemicellulose and lignin content in straw is removed expecting better results.

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DECLARATION OF CONFLICT OF INTEREST

The authors have no competing interests to declare that are relevant to the content of this article.

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