

INTRODUCTION

The advent of natural fibrous insulation materials derived from plant-based resources has proved a potentiality for the new thermal insulation materials used in buildings, in order to meet the demands of energy preservation worldwide. A comprehensive review has been conducted to figure out the factors influencing the thermal conductivity of insulation materials and their possible relationships. Understanding the quantitative relationships between the effective thermal conductivity and influencing factors are essential in determining the thermal performance and energy consumption in buildings. On the other hand, lignocellulose insulation materials can be manufactured without using synthetic resin leading to a reduction in cost, hazardous effects on human health, and the environmental burden imposed by disposal or recycling of the fiberboard. Therefore, binderless thermal insulation materials showed more interest and have been considered as one of the research objectives in the Ph.D. research works. Besides, when natural fiber insulation materials employ in building envelopes, their thermal properties are induced significantly due to the great influence of ambient environmental conditions, such as different temperature distributions in summertime and wintertime or the relative humidity variations, leading to a whole reduction in thermal performance. Accordingly, investigating the thermal property depending on the temperature and relative humidity is always an essential study from a basic research perspective to the advanced investigations.

OBJECTIVES

The doctoral research objectives have been formulated bearing in mind the needs of the current use of natural resources in the context of reducing the energy consumption from traditional sources and enhancing the energy efficiency in construction sector at a building level.

- Development of binderless thermal insulation materials from natural fiber resources.
- Determination of the thermal conductivity coefficient of natural fiber insulation materials and their values regarding the variations of temperature and relative humidity.
- Experimental examination of the water absorption regarding the variations of relative humidity.
- Experimental examination of the influence of temperature and humidity in the thermal conductivity of binderless insulation materials.
- Characterization of natural fiber insulation materials using advanced analytic techniques (SEM, FTIR, TGA).
- Numerical simulation of the heat and moisture transfer in the multi-layered insulation materials used as an exterior wall for building envelope.

MATERIALS

Coir fiber extracted from raw coconut husk and bagasse fiber derived from sugarcane waste were used. Coir materials were pre-treated with NaOH 5% for removing impurities present in the raw plant materials. Later, they were washed to eliminate the excess pollutant particles and then being dried in the oven at 70 °C for 24 h. The sugarcane waste was oven-dried at 70 °C to remove the leftover juice, then they were defibrated using a defibrating machine to extract the bagasse fiber.

SAMPLE PREPARATION

The binderless coir fiber insulation boards (BCIB) were produced by placing the same number of mats in the forming box size of $250 \times 250 \text{ mm}^2$ and hand-formed into homogeneous single layer. After forming, the mats were pressed to compact the materials.

For binderless bagasse insulation fiberboard (BBIB), the wet-forming process was applied for the low-density fiberboard without using binders. After the extraction process, the bagasse fibers were soaked in tap water and defibrated again by adjusting the grain and grinders distance. The mixture was then poured into a round-shaped mold with diameter of 50 cm and the dewatering was done by gravitational force, then the disc shape mats was placed into the oven to dry at $70 \text{ }^\circ\text{C}$ until reaching a constant weight. All the dry specimens were sanded flatly and left in the ambient laboratory condition until further processing. The tested specimens for thermal conductivity measurement were cut from the dry samples into the dimension of $250 \times 250 \times 20 \text{ mm}^3$, $250 \times 250 \times 25 \text{ mm}^3$, and $250 \times 250 \times 30 \text{ mm}^3$ for the thermal conductivity measurement.

METHODS

- Thermal conductivity – Heat flow meter method (EN 12667:2002, ISO 8301:1991/Amd 1:2002).
- Water absorption – Measuring the total mass change of a sample that is exposed to a specified environment using the climatic chamber and the desiccator (ISO 12571:2021(en)).
- Relationship between thermal conductivity value and temperature, humidity – Linear regression technique.
- Surface and morphological analysis – Digital Microscope device and Scanning Electron Microscopy (SEM) equipment Hitachi S-3400N (ISO 21915-1:2020).
- Fourier transform infrared spectroscopy – Transmission mode FT/IR 6300 (ASTM E168, E1252).
- Thermal stability – Thermogravimetric and the derivative thermogravimetric analysis (TGA and DTA) Labsys evo STA 1150 (ASTM D3850).

RESULTS AND THE THESES OF THE DISSERTATION

Theses 1: Factors influencing thermal conductivity of insulation materials

Thermal conductivity of an insulation material is heavily influenced by the operating temperature, moisture content, and density. Other factors also contributed to the heat transfer of insulation materials such as airflow velocity, thickness, pressure, and aging time [1].

The comprehensive review presents general findings on the factors influencing the thermal conductivity of insulation materials commonly used for buildings. The main factors including temperature and moisture content density affecting thermal conductivity values in that higher temperature and higher moisture content always recorded a higher value of thermal conductivity. For the density-dependent, an increase in density may lead to an increase or a reduction in thermal conductivity. In addition, the relationship between the thermal conductivity measured in the steady state and the low range of operating temperature ($-10 \text{ }^\circ\text{C}$; $50 \text{ }^\circ\text{C}$) was always expressed as a linear function. A similar increasing trend was also found for the thermal conductivity-moisture content relationship. Other factors affecting the thermal performance were also reported, namely thickness, airflow velocity, pressure, and aging time. This literature review has contributed to the general research on the thermal conductivity of insulation materials in the construction sector at a building level.

Theses 2: Developing a new thermal insulation material from sugarcane bagasse

The new thermal insulation material was developed from sugarcane bagasse fiber without using binders or additives. Based on the results, it is found that the thermal conductivity values of binderless bagasse fiber insulation boards (BBIB) ranged from 0.0429 W/(m·K) to 0.053 W/(m·K) showing that these boards could be performed as the prominent insulation materials used in buildings.

The binderless bagasse fiber-based insulation materials were produced using the wet process method for low-density fiberboard without using binders. The binderless fiberboard can be manufactured by the activation of the self-bonding feature due to the hydrogen bond formation and adhesive behaviour of lignin and cellulose during the heating and drying process. The thermal conductivity values of the tested specimens were measured at a mean temperature of 20 °C using the heat flow meter method. Accordingly, the λ -values were lower than that of natural fiber reinforced phenol formaldehyde biocomposites made of rice straw, energy reed, and coir fiber which were measured in the Ph.D. works, from 0.06 W/(m·K) to 0.1 W/(m·K). The low value of thermal conductivity of binderless fiber insulation materials came from the low λ -value of raw plant materials and the air presents in the void or open pores of the fiber matrix had low heat conductance. In contrast, the higher values of plant-based biocomposites were due to the high heat conductance of phenolic resin (its thermal conductivity is reported from 0.29 W/(m·K) to 0.32 W/(m·K) and its moisture content was nearly 34%, [2]). As a result, the obtained thermal conductivity values of the tested binderless bagasse fiber insulation materials provided comparatively better results demonstrating that they could perform as prominent building insulation materials.

Theses 3: Water absorption of natural fiber insulation materials regarding the absorbent time

According to the practical examination of water absorption of the binderless bagasse fiber insulation materials at a thickness of 25 mm, the equilibrium state using the desiccator method needs a duration of 28–35 days to be obtained.

This experiment aims to investigate the duration for the water absorption of natural fiber-based samples reaching the equilibrium saturation level. The water absorption percentages of bagasse fiberboard conditioned at the relative humidity of 75% regarding the absorbent time using the desiccator method. Results showed that samples followed typical Fickian diffusion behaviours in that water absorption occurs rapidly at the beginning of exposure with water, however, after time, the absorption rate slows down until reaching the point of equilibrium [3]. As it is seen from Fig. 1, the moisture absorption was relatively high for the first stage of 0–14d, possibly due to a large number of water molecules diffusing through the material starting from the dry state of the absorbency process. From 14 to 28 days, it seems that the pores and capillaries of bagasse fiber which were initially filled with air were steadily replaced by absorbed water leading to a minor change in the moistened weight and almost stable after approximately 28 days. As a result, the saturated level of binderless bagasse fiberboard specimens took approximately 28–35 days to be obtained.

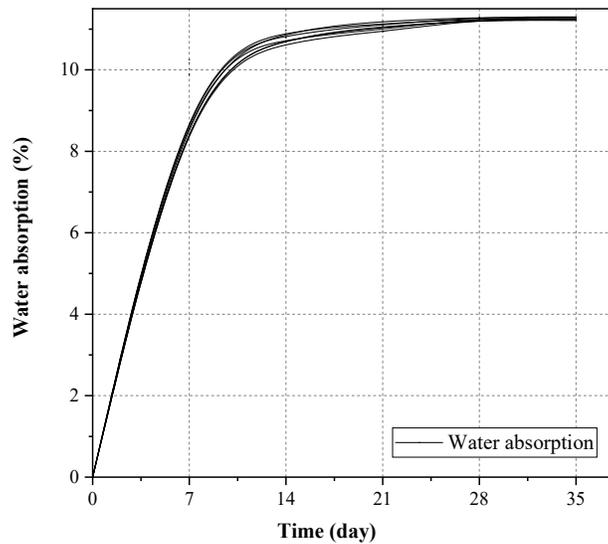


Figure 1. Water absorption percentages of bagasse fiberboard regarding the absorbent time.

Theses 4: Water absorption of binderless natural fiber-based insulation material related to relative humidity levels

The values of water absorbency for the binderless coir fiber insulation boards (BCIB) were from 7.66% at approximately 16.5%RH to the maximum value of 23.54% at around 90.5%RH and the values of binderless bagasse insulation boards (BBIB) were found in the range of 10.5–17.33% as the relative humidity increased from 33 to 96%. As a result, higher relative humidity always revealed higher water absorption.

The water absorption percentages of the BCIB and BBIB samples were conducted at different humidity levels using the climatic chamber and desiccator method, respectively. For the moistened state of BCIB, samples were exposed to five expected levels of humidity (15, 40, 60, 80, and 95%) while the BBIB samples were conditioned in four different humidity levels (33, 57, 75, and 96) in sealed desiccator containing respective saturated solutions. As it is observed from Fig. 2, a similar sorption behaviour was found for three samples of BCIB and three samples of BBIB, and they exhibited a typical behaviour of natural fibers with a high increase of moisture content above 75% RH. Furthermore, the BCIB samples showed a higher percentage of moisture content than the BBIB samples, possibly due to the higher absorption of raw coir fiber than bagasse fiber or because of the higher porosity of BCIB than BBIB during the manufacturing process. As a result, higher water absorption is always associated with higher relative humidity levels, and the moisture sorption isotherm expressed from the experimental data has proved the efficacy of the methods used in this study.

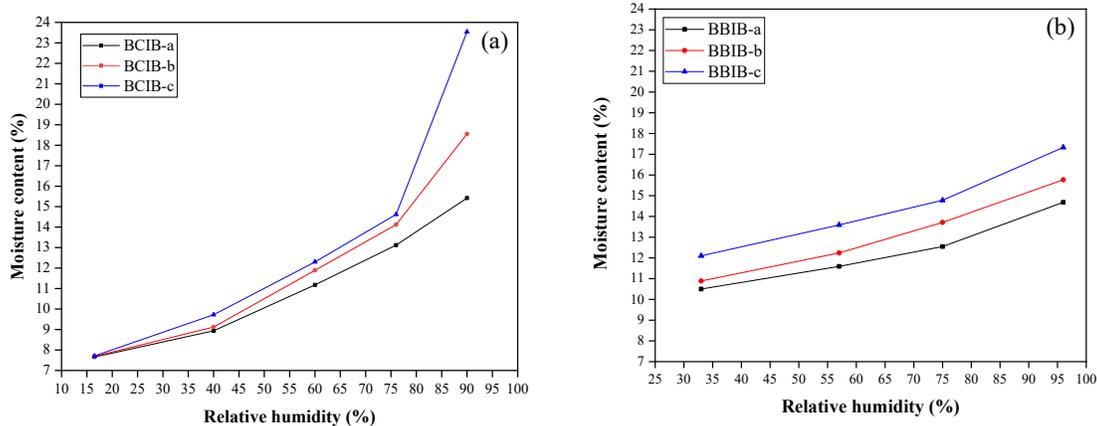


Figure 2. (a) Moisture content of BCIB regarding the increase of relative humidity levels; (b) Moisture content of BBIB regarding the increase of relative humidity levels.

Theses 5: Temperature-dependent thermal conductivity of insulation materials

The thermal conductivity of the binderless bagasse fiber insulation boards (BBIB) increased from 0.041 to 0.057 W/(m·K) while the thermal conductivity of the binderless coir fiber insulation boards increased from 0.037 to 0.066 W/(m·K) as the operating temperature increased from -10 to 50 °C. An increasing tendency in thermal conductivity was always ascribed to increased temperature. The percentage rate of changes in the thermal conductivity values of BCIB was found of 25–45% while the values of BBIB increased approximately by 16–20% showing that different insulation materials record different temperature dependence.

Based on the practical results of temperature-dependent thermal conductivity, it is reported that higher temperatures always recorded higher thermal conductivity values. The temperature dependency of the thermal conductivity values is explained by the basic heat transfer mechanism, as the temperature increase, the heat molecules vibrate faster, allowing for larger heat movements through conductance. According to the European certified reference materials for thermal conductivity measurements, the variations of thermal conductivity typically increase up to 20–30%. Accordingly, the BBIB samples have shown a lower heat consumption than that of BCIB and other bio-based composites. The relationships between the thermal conductivity value and mean temperatures were found and expressed as the linear functions according to the experimental data with high coefficient of determination demonstrating the strong influence of temperature in the thermal conductance (see Fig. 3).

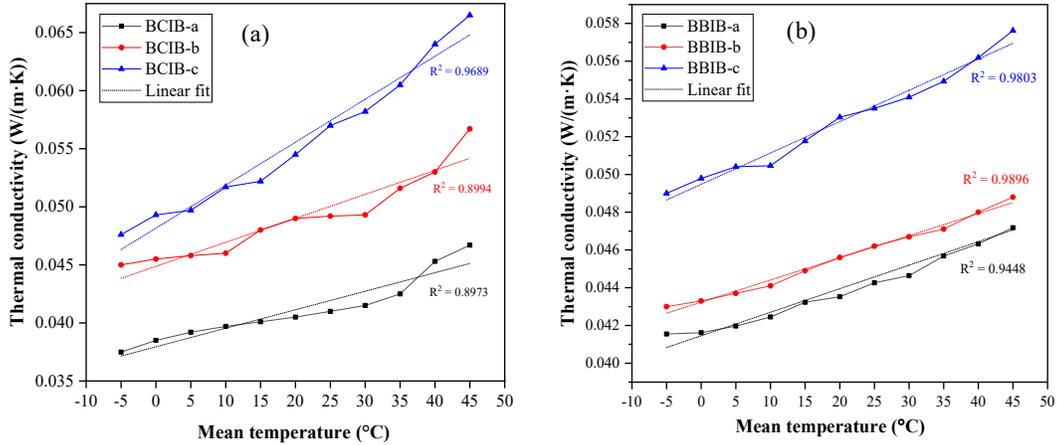


Figure 3. (a) Thermal conductivity of BCIB regarding the increase of mean temperatures; (b) Thermal conductivity of BBIB regarding the increase of mean temperatures.

Theses 6: Relative humidity-dependent thermal conductivity of insulation materials

The thermal conductivity values of three samples of BCIB were recorded in the range of 0.049–0.066 W/(m·K), 0.058–0.094 W/(m·K), and 0.069–0.107 W/(m·K) regarding the humidity range of 16.5–90.5%. Whereas the values of thermal conductivity of three tested specimens of BBIB were found of 0.044–0.049 W/(m·K), 0.046–0.052 W/(m·K), 0.058–0.069 W/(m·K) when the relative humidity increased from 33 to 96%.

Based on the results of thermal conductivity depending on relative humidity, higher relative humidity levels always reported higher values of thermal conductivity. As is seen from the graphs, a similar increasing trend was observed for all tested specimens in that increased relative humidity led to an increase in thermal conductivity values. This is because of the higher water absorption of the tested specimens since they are exposed to humid air conditions at different relative humidity levels. The large heat conductance is also caused by water's high thermal conductivity at approximately 0.6 W/(m·K), especially at above 75% relative humidity. On the other hand, the bulk density of the tested samples may contribute to the water absorption regarding the increase of humidity, therefore, influencing the whole heat conductance. Accordingly, as the density of BCIB increased, the number of voids and open pores in the fibers structure decreased markedly resulting in a reduction of water penetration into the cell walls of the fibers, therefore, decreasing the moisture effect on thermal resistant quality. Whereas, at higher densities, there is more bagasse fiber available to absorb moisture and the moisture uptake of BBIB changes appear to have a greater influence on the results. The relationships between thermal conductivity values and the humidity levels were expressed as linear functions with high coefficient of determination proving the significant effect of relative humidity on the thermal conductance of natural fiber-based insulation materials.

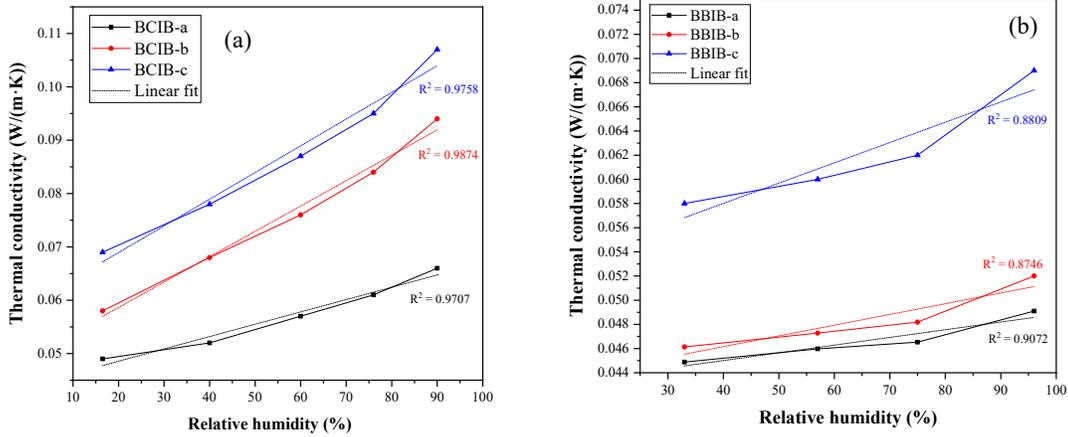


Figure 4. (a) Thermal conductivity of BCIB regarding the increase of mean temperatures; (b) Thermal conductivity of BBIB regarding the increase of mean temperatures.

Theses 7: Numerical simulation of heat and moisture transfer in multi-layered insulation material

The simulation results indicated the general trend of the effective thermal conductivity and moisture content was associated with ambient temperature and relative humidity variations. That is, as outdoor temperature and relative humidity increase or decrease, they will affect the heat and moisture flux across the insulation materials leading to respective changes in the effective thermal conductivity and moisture content levels.

Based on the numerical calculation from the simulation study of heat and moisture transfer in multi-layered insulation materials using the cellulose fiberboard as a core layer, the values of effective thermal conductivity (ETCs) recorded a slight increase in the range of 0.045–0.069 W/(m·K) since the temperature ranged from 5 to 15 °C and the humidity increased from 33 to 75%. The higher temperature and higher humidity always revealed a higher value of thermal conductivity, and a similar increasing trend was found for all simulated cases. Results also showed that changes in the thermal transmittance coefficient are always ascribed to the thickness of the cellulose fiberboard. Accordingly, the U-value decreased from 1.38 W/(m²·K) at the thickness of 50 mm to 0.26 W/(m²·K) at the 200 mm thickness showing that the 50–200 mm thickness could be considered as the critical thickness for designing the multi-layered insulation materials to meet the requirements of a nearly zero-energy building. For the simulation study in the dynamic boundary conditions in that the outside temperature and relative humidity change dynamically for 2 days, the effective thermal conductivity in summer conditions showed a higher value than that in winter conditions due to the great influence of the heat and moisture flux caused by the large difference between the indoor and the ambient temperature as well as the relative humidity of the outdoors. According to the simulation result, the ETCs in the summertime have been remarkably influenced by the heat and moisture flux while there was only the moisture flux contributed to the increase of ETCs in the wintertime. The relative humidity-dependent moisture content over time also showed similar behaviour with the ETCs in that it was significantly influenced by both heat and moisture transfer in the summertime while the variations in the wintertime were mainly from the contribution of moisture flux.

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PUBLICATIONS

Journal articles (published)

1. **D. H. A. Le** and Z. Pásztor (2023). Experimental Study of Thermal Resistance Values of Natural Fiber Insulating Materials under Different Mean Temperatures. *South-east Eur for 11 (1)*: early view. (<https://doi.org/10.15177/seefor.23-03>)
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5. K. M. F. Hasan, P. G. Horváth, M. Bak, **D. H. A. Le**, Z. M. Mucsi, and T. Alpár (2021) Rice straw and energy reed fibers reinforced phenol formaldehyde resin polymeric biocomposites. *Cellulose*, Vol. 28, no. 12, 7859-7875. (<https://doi.org/10.1007/s10570-021-04029-9>)
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Journal articles (submitted)

7. D. H. A. Le, Z. Kóczán, Z. Börcsök, and Z. Pásztor (2023). Development of new insulation material from sugarcane bagasse and examination of the insulation effect depending on temperature and humidity.

Conference paper

1. **D. H. A. Le** and Z. Pásztor (2023). Numerical simulation of the influence of air space thickness in heat transfer of a high-performance glazing system. 2023 9th International Conference on Environment and Renewable Energy, 24-26 February 2023, Hanoi University of Science and Technology, Ha Noi, Viet Nam.

2. **D. H. A. Le** and Z. Pásztor (2022). Experimental Study of Thermal Resistance Values of Natural Fiber Insulating Materials under Different Mean Temperatures. GREEN 2022/4, University of Zagreb, 14-16 September 2022, Zagreb, Croatia.
3. K. M. Faridul Hasan, **Le Duong Hung Anh**, Horváth Péter György, Bak Miklós, Tibor Alpár (2022). Green Insulation Panels Development from Industrial Lignocellulosic Materials Reinforced Cementitious Composites. 5th International Conference on Building Energy and Environment (COBEE 2022). Montreal, Canada, July 26, 2022.
4. Z. M. Mucsi, K. M. F. Hasan, **D. H. A. Le**, P. G. Horváth and T. Alpár (2022). Methylene Diphenyl Diisocyanate and Dement-bonded Insulation Panels Reinforced with Coconut fiber and Energy Reed Straw Mediated by Semi-dry Technology. XXV. SPRING WIND CONFERENCE, Pécs, Hungary.
5. **D. H. A. Le** and Z. Pásztor (2022). Experimental investigation of thermal conductivity values and density dependence of insulation materials from coir fiber. 2022 8th International Conference on Environment and Renewable Energy, 24-26 February 2022, Hanoi University of Science and Technology, Ha Noi, Viet Nam.
6. **D. H. A. Le** and Z. Pásztor (2021) Experimental investigation of the influence of temperature on the thermal conductivity of raw coconut fiber. IV. International scientific and practical conference “ACTUAL PROBLEMS AND DEVELOPMENT PROSPECTS OF FOREST INDUSTRY COMPLEX”, September 8-11, Kostroma State University, Kostroma, Russia, 2021, pp. 34-38.
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Book chapter

1. **D. H. A. Le** and Z. Pásztor (2023). Natural Fiber Reinforced Vinyl Ester Composites Influence of Moisture Absorption on the Physical, Thermal and Mechanical Properties. Vinyl Ester based Biocomposites, CRC Press, Taylor & Francis Group. (Volume 2; ISBN: 978-1-032-22048-2).