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Characterization of the electrical, acoustic, and thermal insulating properties of biodegradable chito-cellulosic structural composites

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ABSTRACT

Insulation materials are essential for engineering efficiency, as they directly reduce energy losses or eliminate the need for extra components to compensate for them. However, most insulation materials are non-biodegradable polymers that significantly contribute to environmental degradation during production and disposal. This study explores the use of bioinspired chito-cellulosic materials —a family of biological composites known in structural applications for their low cost, versatile manufacturing, and ecological integration— as sustainable insulation. The study compares three chito-cellulosic variants with different cellulose compositions and evaluates their electrical, thermal, and acoustic insulation capabilities, as well as flammability, biodegradability, environmental impact, and mechanical properties. The insulating results are compared to conventional polyurethane foams, demonstrating lower thermal insulation capabilities, similar electrical insulation, and better acoustic insulation. Moreover, they offer the advantages of being 3D-printable, fully biodegradable in environmental conditions, and fireproof, highlighting their potential as a viable green alternative to synthetic insulators.

1. Introduction

Insulation materials play a crucial role in engineering by containing energy within a specified area, whether isolating electricity in a conductor, preventing unwanted noise from escaping or maintaining heat within designated boundaries. These materials are integral to the efficiency of engineering systems, as they reduce the need for additional energy to compensate for losses in various applications [1]. As we move from an unchecked technological and industrial growth era to one focused on sustainability, insulating materials have become key to enhancing performance and reducing environmental impact across sectors such as buildings, transportation, and electronics [2,3].

The demand for advanced insulating materials is expected to grow by more than 6 % annually over the next decade. However, the widespread use of these materials, particularly polyurethane foams, which dominate the market [4], comes with significant drawbacks. These foams primarily use non-biodegradable polymers, contributing to global health and environmental issues [5,6]. Efforts to combine them with biological materials such as chitin, cellulose, or wood flour have resulted in composites that pose even more significant challenges at the end of their lifecycle. These composites remain persistent pollutants, and the inclusion of organic matter hinders their recycling, even in the rare instances when they are recovered [7]. Their high buoyancy exacerbates this problem, as uncollected waste becomes marine litter, circulating without boundaries in the ocean, accumulating in remote areas, and acting as vectors for invasive species and pathogens [8,9]. Given that recovery strategies are insufficient to slow down the accumulation of synthetic waste in Earth's ecosystems [10], there is an urgent need to develop materials with insulating capabilities that can enhance technological efficiency while being fully integrated into Earth's ecological cycles without human intervention [11].

In recent years, cellulose and cellulosic composite panels have emerged as potential biodegradable insulators due to cellulose's inherent physical properties, abundance, and renewability [12–16].

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However, the need for additional additives and crosslinkers to impart functional properties, such as reduced flammability, compromises their environmental integration [17]. Moreover, their limited shape-forming versatility restricts their use to low-precision applications like wall panels. These limitations can be overcome through bioinspired manufacturing technologies, which replicate the native assembly processes of materials, enabling the production of complex structures with unmodified biological components [11].

Bioinspired manufacturing deviates from traditional approaches by developing tools grounded in biological principles and conditions, preserving the native form of the components. This approach emulates the complex synergies and functionalities evolved by various organisms, allowing for the unassisted integration of the resulting composites into Earth's ecology. A prime example of this approach is the bioinspired chito-cellulosic composites known as Fungal-like Adhesive Materials (FLAMs) [18]. These composites replicate the oomycete wall using its native components (i.e., cellulose and chitin), the first and second most abundant organic components on Earth, respectively [19]. FLAMs have been adapted for additive manufacturing and casting, forming complex geometries using a water-based process, ion transport, and simple chemistry [20,21], resulting in a significantly lower environmental impact than synthetic foams with equivalent mechanical characteristics [18,22]. In addition to their seamless integration into Earth's ecological cycles, these composites can be produced as part of a circular economy through the bioconversion of organic components from various sources, particularly urban waste [23]. Despite their demonstrated potential in structural applications, the insulative capabilities of FLAMs and chitinous cellulosic composites remain unexplored.

This study evaluates the insulation capabilities of three FLAM variants against electricity, heat, and sound and their flammability, biodegradability, environmental impact, and mechanical properties. These are essential functional properties needed to replace hydrocarbon-based insulative materials in most engineering applications. FLAMs have been produced with various sources of cellulose, from raw wood to pure cellulose from recycled or raw sources, to adapt the material to available resources and balance mechanical properties and cost [18]. Here, we specifically produced and tested high-quality composites with purified cellulose, cost-effective composites with wood flour (a byproduct of the lumber and woodworking industries), and composites with equal parts cellulose and wood flour. Our results demonstrate that these bioinspired composites are cost-effective and scalable sustainable insulators with better suitability than the synthetic counterparts for modern manufacturing techniques such as additive manufacturing.

2. Experimental section

2.1. Materials

High-purity cellulose fibers (JELUCEL® HM400X) and wood flour fibers (JELUXYL® WEHO) were purchased from JELU-WERK Josef Ehrler GmbH & Co. KG (Germany). Chitosan of medium molecular weight, around 75 % to 85 % degree of deacetylation, was purchased from I-CHESS Chemicals Pvt. Ltd. (India). Glacial Acetic Acid was purchased from Sigma-Aldrich (Singapore). Soft (23 kg/m^3, 2425 $/m^3)and high-performance rigid (105 kg/m^3, 13,735 $/m^3) polyurethane foams were purchased from local suppliers. Commercial polyethylene acoustic insulation foam with an average density of 85 kg/m^3 and an approximate cost of 4800 $/m^3 was purchased from RS Components Singapore Pte Ltd. (Singapore).

2.2. Production of chitinous cellulosic composites

As a biopolymeric binder, a 3 % (*w/v*) Chitosan solution was prepared by dissolving, as received, powdered chitosan in 1 % (*v/v*) Glacial Acetic Acid. Different chitinous cellulosic composites were produced by mixing as received fibrous fillers (cellulose fibers, wood fibers, or an equal part of both fiber types) with the chitinous binder. Wet samples were cast into molds and dried in ambient conditions. Required samples were machined to size accordingly for each test. The density of the samples ranged from 283.46 ± 14.17 kg/m^3 for the wood-based sample to 432.25 ± 21.61 kg/m^3 for the cellulose-based sample. The 1:1 mix had a density of 311.31 ± 15.56 kg/m^3. The manufacturing cost varied from approximately 490 $/m^3 for the composite using commercial wood flour to 1200 $/m^3 for the cellulose-based composite.

2.3. Porosity of the samples

Porosity was measured using Scanning Electron Microscope images of the foams, which were automatically analyzed using an FFT bandpass filter implemented in Image J [24]. The porosity of the samples was determined to be 18.9 %, 20.65 %, and 12.65 % for the cellulose, wood, and hybrid FLAMs, respectively, and 19.66 % and 53.74 % for the rigid and soft PU.

2.4. Additive manufacturing

The additive manufacturing system uses a six-axis robot capable of reaching 1.65 m horizontally and carrying a 20 kg payload. It was mounted on a hydraulic scissor-lift platform for an extended work envelope. The extrusion system assembled at the robotic arm's end includes a 50 L pump and a precision dispensing unit, both designed for highly viscous materials. Custom software and hardware manage communication between the robot's controller and extrusion system. A parametric design software was also developed to translate design geometry to machine instructions. Material compaction and expansion were encoded to create a preset model [25]. The nozzle used has a 7 mm internal diameter and a 3 mm layer height for interlayer adhesion, resulting in a 13 mm wide bead profile. The flow rate is approximately 2.8 ml/s with a 50 mm/s feed rate. Refer to [18,26] for a more detailed description and characterization of the printing setup.

Scanning electron microscopy

A Field Emission Scanning Electron Microscope, JEOL JSM-7600-F, was used to study the surface morphologies of samples. Samples were mounted onto an aluminum stub using carbon tape and sputter coated with gold before imaging.

2.5. Flexural test

For the flexural test, rectangular samples of 100 mm by 16 mm by 6 mm were cast in a mold and dried in ambient conditions. Triplicates for each chitinous cellulosic were prepared and further dried in a 50 °C oven to ensure it was completely dried before being subjected to a three-point bending test where a span length of 60 mm and a crosshead speed of 4.0 mm/min was applied using a 1kN Universal Testing Machine, Instron 5943.

2.6. Sound transmission and absorption test

Circular discs in triplicates, 100 mm and 30 mm in diameter, were prepared for each composite type and subjected to acoustic testing in an Impedance Tube Kit (Brüel & Kjær Type 4206). For sound absorption measurement, the two-microphone method was performed following the ISO10534-2 and ASTM E1050–12 standards [27,28]. The method involves decomposing a broadband signal using stationary random noise into its incident and reflected components; consequently, the absorption coefficient can be obtained.

The experiments used in the sound Transmission Loss (TL) measurement were identical to the absorption coefficient measurement except that TL measurement requires two additional microphones with an extension of the tube. The measurement follows the procedures stated in ASTM E1050–12 and ASTM E2611 standards [28,29]. Likewise, the loudspeaker emits a stationary random noise from one end of the tube.

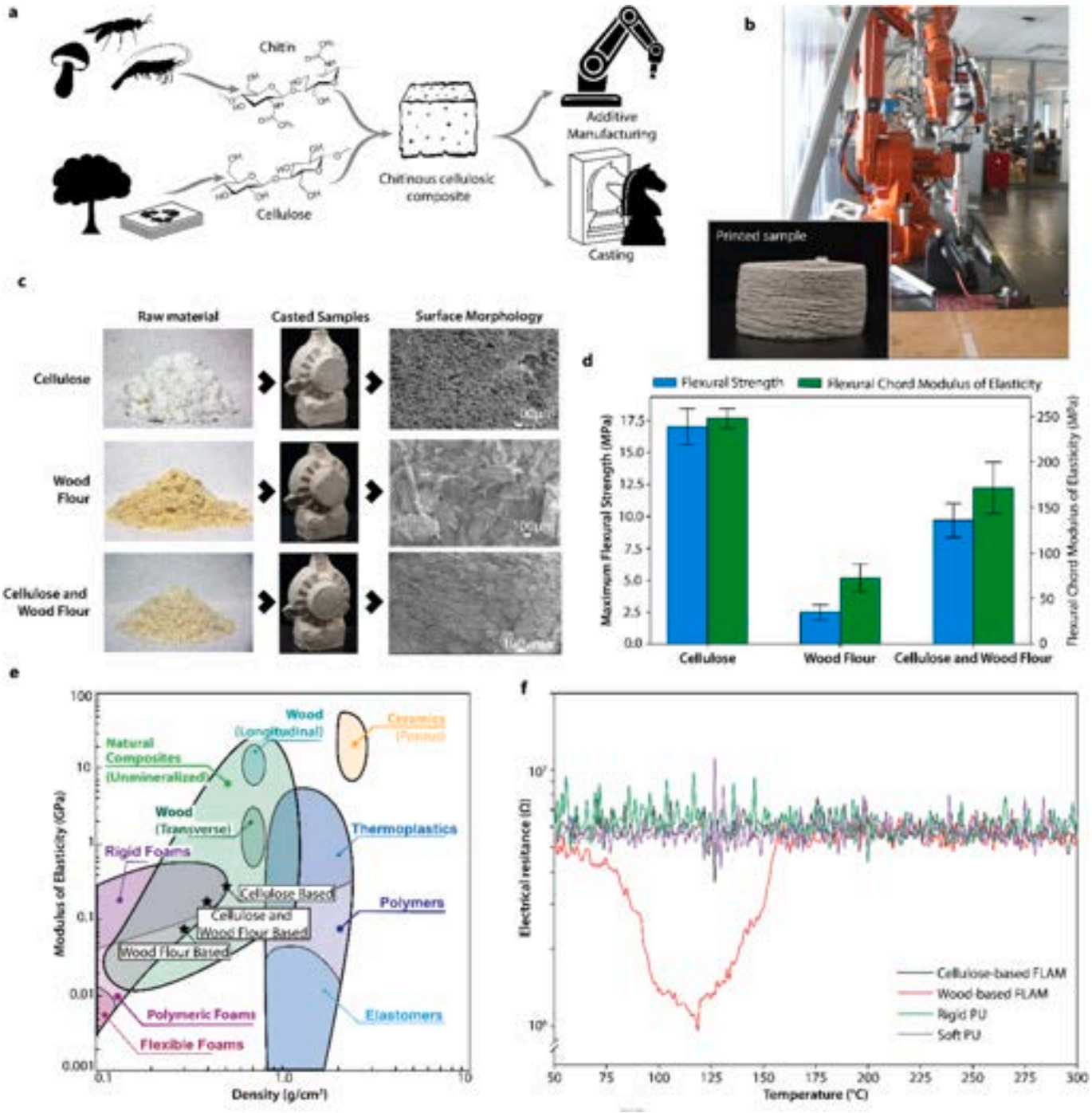


Fig. 1. Overview of the chitinous cellulosic composite, including potential applications and bulk mechanical properties. (a) The bioinspired chitinous cellulosic composites (i.e., FLAMs), made with chitosan and cellulose, were tested in additive and conventional manufacturing. (b) The pneumatic system is assembled on a robotic arm for extruding FLAMs into a diverse range of 3D printed products, from simple to complex geometries. The inset showcases the cylinder produced to test the manufacturability of the material. (c) The composites can be made with different qualities of cellulose, from high-purity cellulose to industrial waste such as wood dust. (d) The bulk mechanical properties of the bioinspired composites, comparable to those of petroleum-based rigid polyurethane foam, are highly dependent on the quality of the cellulosic component used. (e) Ashby plot showcasing the modulus of elasticity of low-density materials of synthetic and natural origin. The relative positions of the three versions of chito-cellulosic composites tested for their insulating capabilities are highlighted. (f) Measurements of the electrical resistance of the chito-cellulosic composites and their polyurethane counterparts at different temperatures show that all samples behave as insulators across all temperature ranges.

The test sample was placed in the middle, separating the tube into upstream and downstream sections based on their position relative to the sound source. The transfer matrix element obtained from the two-load implementation (i.e., open and anechoic conditions) from the four microphones allows the sound transmission loss to be obtained.

It is worth noting that two different tube sizes were used for both sound absorption and TL measurements to measure different frequency ranges. The large tube with an internal diameter of 100 mm allows a test measurement from 200 Hz to 1600 Hz, and a smaller tube with an internal diameter of 30 mm will be able to test from 500 Hz to 6400 Hz. The results from both tubes were then combined at the overlapped frequencies. In order to reduce the possibility of statistical error, an

average of three repetitive tests were carried out for each sample to obtain the sound absorption coefficient and TL. Each test run took around 60 s with 16 averaging of the signal.

2.7. Thermal conductivity test

The thermal conductivity of the materials was ascertained using a Heat Flow Meter (TA Instruments, FOX 50) in accordance with ASTM C518–98 standard. Samples of 50 mm diameter and 25 mm thickness were prepared and sandwiched between two heated plates with controlled temperatures ranging from 260 K to 340 K (−13.15 °C to 66.85 °C). For the high-density PU foams, the thermal conductivity at low temperatures, while in agreement with previous reports, falls below the nominal range of the apparatus specifications.

2.8. Flammability test

The flammability of the chito-cellulosic composites was assessed using the UL-94 standard flammability test for plastic materials. Composite strips were fabricated and exposed to an open flame for 30 s or until the burning material exceeded a length of 25 mm. Observations included the self-extinguishing properties of the flames, the extent of fire damage, and the combustion duration.

2.9. Thermogravimetric analysis

Using a thermogravimetric analyzer (TA Instruments, TA Q50), samples placed in aluminum pans were subject to thermal degradation from temperatures of 25 °C to 600 °C, increasing at a rate of 10 °C/min, with an inert gas (Nitrogen) flow of 50 ml/min.

2.10. Electrical conductivity test

The electrical resistance of the chito-cellulosic and polyurethane samples was measured by heating them in air with a Torrey Pines hotplate, a Keithley 2410 SMU, and a 4PP with spring-loaded tungsten tips. The tips were 5 mm apart on a line, and a glass slide was used to isolate the samples from the aluminum hotplate. Contact resistance issues were prevented by applying current to the samples through the outer probes of the 4PP and measuring the potential difference across the inner probes. The Keithley 2410 kept a constant voltage of 2.1 V across the inner probes by adjusting the current through the outer probes. To examine the temperature effect on the electrical resistance of the samples, we computed the sheet resistance as $\pi\Delta V/\ln(2)I$ and increased the hotplate temperature by five °C/min until 350 °C.

2.11. Biodegradation test

The biodegradability of the samples was evaluated using a soil burial biodegradability test. The samples were cut into blocks measuring 20 mm × 20 mm × 5 mm, dried under room conditions until they reached a constant weight (w0), and then buried in compost soil. The compost soil was a mixture of equal parts organic compost, organic material, and topsoil. Three samples for each material and time point (60 samples in total) were buried at a depth of 8 cm in the compost soil mixture. Every ten days, three samples of each of the five materials were retrieved, cleaned, washed with DI water, and dried at 60 °C until a stable weight (wt) was achieved. The weight loss percentage was calculated as 100 (w0-wt)/w0.

2.12. Life cycle assessment (LCA)

The standardized life cycle assessment methodology was employed to evaluate the environmental attributes of the different composite variants [30]. A cradle-to-gate life cycle attributional study was conducted to quantify the environmental attributes of material production. This study aims to inform potential industry users of the impacts compared to polyurethane rigid foam, which is widely used as an insulating material and has mechanical characteristics closely resembling those of chito-cellulosic composites. As raw materials such as wood flour and chitosan can be sourced from waste resources of low or no economic value from the timber and fishing industries, the recyclable materials were assumed to be "cut off" without environmental credit or burden from the originating process. A functional unit of 1 kg of end material was used to compare systems. Primary data on composite productions were based on actual measurements for the base scenario. Additional scenarios highlighting the use of wood dust were incorporated here compared to previously studied scenarios with international and regional supplies of cellulose and chitosan [31]. The inventory data is included in Supporting Information Table 1. OpenLCA software was used to conduct the calculations, where background data from the Ecoinvent 3.5 database was imported, and the emissions were translated to midpoint indications using the characterization factors within the ReCiPe 2016 methodology [32].

3. Results and discussion

3.1. Physical characterization

FLAMs have been used in additive manufacturing, subtractive manufacturing, and mold casting, either alone or in combination. To corroborate that our production of FLAMs was successful and had the demonstrated versatility to form structures previously reported, we checked their suitability for additive manufacturing using an industrial robot (Fig. 1a). Our results printing simple geometries (Fig. 1b) confirmed that our synthesis resulted in a material with similar manufacturability to earlier reports. Separately, the molding properties of the composites were tested by producing scaled replicas of our robotic arm's base with the three different composites (Fig. 1c). Qualitatively, we found the reproduction of the geometries using cellulose-based FLAM more consistent, straightforward to produce, and of higher accuracy than those made using composites containing any amount of wood flour. Scanning electron microscopy (SEM) was used to investigate the morphology of the composites in depth (Fig. 1c). All three varieties (cellulose, wood, and hybrid) display a highly porous structure consistent with previous observations. However, the cellulose-based composites show the characteristic microfiber structure of the cellulose, which is absent in those composites incorporating wood. The cellulose fibers appear entangled, which could contribute to the sturdiness of this composite when compared to those including wood, which at the microscopic level resemble an aggregate of particles rather than an intertwined network.

The quantitative measurement of mechanical characteristics corroborated the initial qualitative observations. As shown in Fig. 1d, composite samples of pure cellulose exhibited the highest flexural strength of 17.03 ± 1.407 MPa, with an elastic modulus of 247.73 ± 10.83 MPa, consistent with previous results [18]. The introduction of wood flour significantly reduced the composite's flexural strength due to the microparticles' varied morphological geometry and contaminants, such as lignin and hemicellulose, which do not contribute to chitosan-cellulose binding. This reduction in mechanical properties for a lower material cost was evident when wood flour was introduced in equal parts and when only wood flour was used, suggesting a steady transition from the costlier but stronger cellulose-based material to the less expensive but weaker wood-based one. Interestingly, compared to the mechanical properties of various material classes (Fig. 1e), the chito-cellulosic composites spanned the entire range of mechanical properties exhibited by polyurethane foams; wood-based composites demonstrated properties akin to soft polyurethane foam, while pure cellulose-based composites were structurally similar to rigid polyurethane foams. This result highlights the remarkable mechanical versatility of FLAMs, comparable to that of the most adaptable synthetic foams. It also

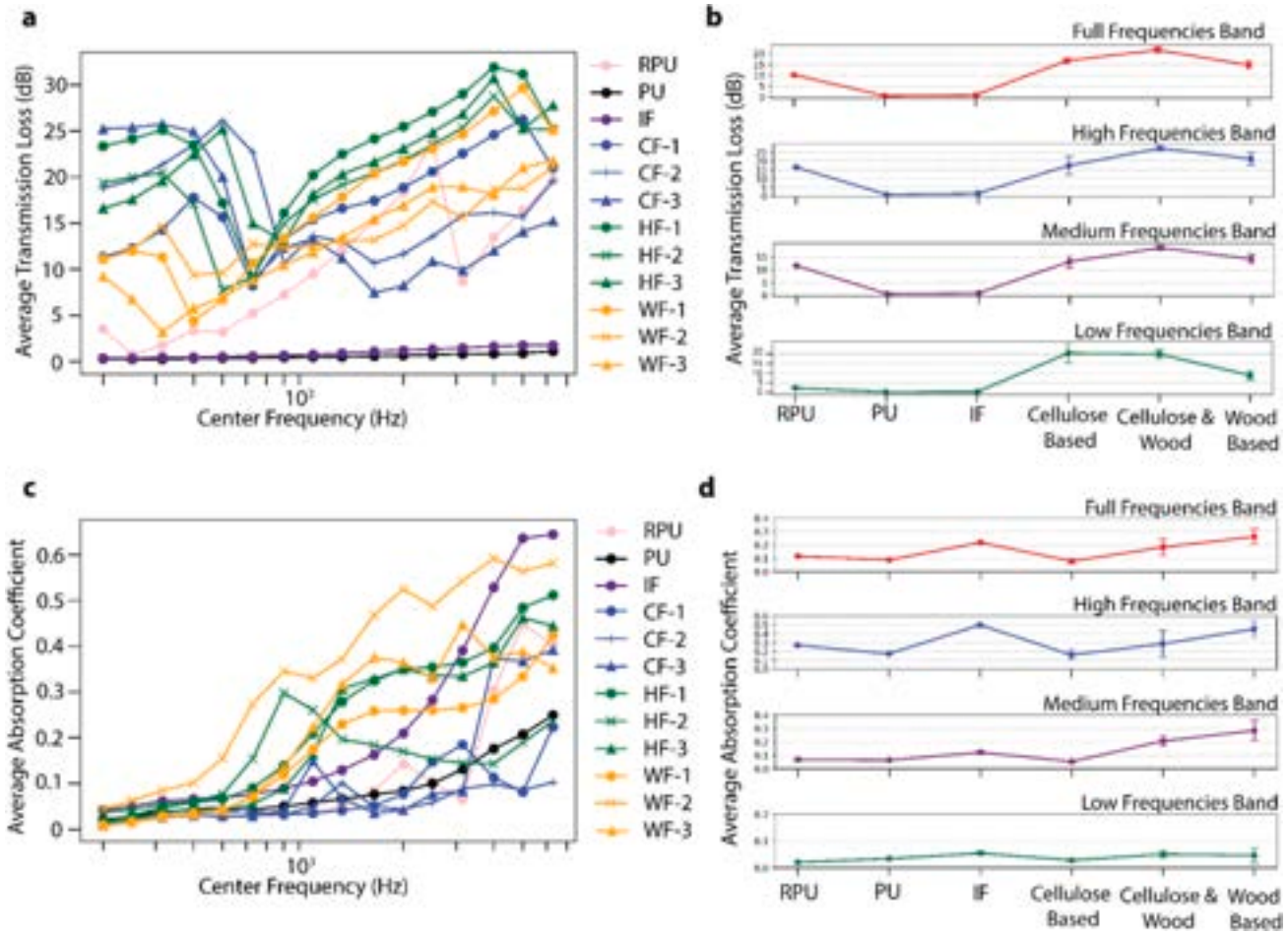


Fig. 2. Insulating properties of chitinous cellulosic composite. (a-b) Across all frequency bands, cellulose and wood-based composites performed better as sound barriers when compared to petroleum-based counterparts. Possibly due to the difference in material stiffness. (c-d) As a sound absorption material in the low and medium frequency bands, adding wood dust fillers enabled cellulose-wood composites to be comparable to commercial noise insulation foam made with polyurethane foam. In the high frequencies band, the higher porosity, wood dust-based composite can be possible alternatives to commercial insulation foams.

suggests their potential to function as an environmentally sustainable alternative in a wide range of engineering applications.

3.2. Electrical insulation properties

We measured the electrical resistance of the samples across their surfaces (Fig. 1f). All the elements used, whether organic (i.e., chitosan, cellulose, wood flour) or synthetic (polyurethane), are known insulators, with resistivities ranging from 10^{12} Ω/m (polyurethane) to 10^{14} Ω/m (wood). These values are well above the 10^{8} Ω/m threshold commonly accepted as the boundary between semiconductors and insulators. Therefore, we did not anticipate a significant difference in the electrical insulating capabilities of both materials under normal conditions. However, we examined how these insulating capabilities responded to heat, simulating electrical components' regular operation and malfunction. To do this, we placed the samples on a programmable hot plate and gradually increased the temperature from room conditions to 300 °C while monitoring the conductivity across the surface of the samples.

All the samples maintained their original electrical insulating capabilities across all temperature ranges, with no significant differences observed between samples or temperatures. It is worth noting that while measurements for polyurethane and cellulose composites were very consistent, composites containing wood showed more significant variability between samples, which might be representative of their less consistent composition. Notably, the wood-based composites exhibit a limited increase in conductivity at around 100 °C, which later stabilizes. Given the localized nature of this change —ruling out the possibility that it results from a permanent change in the material — we hypothesized that the samples containing wood, due to their porosity or composition, retain significantly higher amounts of water in their structure than those composed solely of cellulose and polyurethane foams. As the sample is heated, this water migrates from the inner areas of the sample to the surface, increasing the conductivity until all the water has been expelled and the sample stabilizes.

3.3. Sound insulation properties

Sound transmission loss, the measure of a material's ability to prevent airborne sound transmission from one region to another, is quantified as the difference between incident and transmitted acoustic energy. We assessed the transmission loss in the three chito-cellulosic composites, the rigid and soft polyurethane foams used as benchmarks in all tests, and a commercial polyethylene foam sample specifically designed and commercialized for sound insulation (Fig. 2a). For all the biological composites, transmission loss across the material decreased as frequency increased to the first panel resonant frequency. This behavior in the stiffness-dominated region aligns with the mechanical characterization of the materials, where cellulose-based composites had the highest modulus of elasticity and resonant frequency, followed by cellulose-wood-based composites and, finally, wood-based composites. Beyond this frequency, the transmission loss enters a region dominated by panel mass effects. In this region, the transmission loss increased at 6

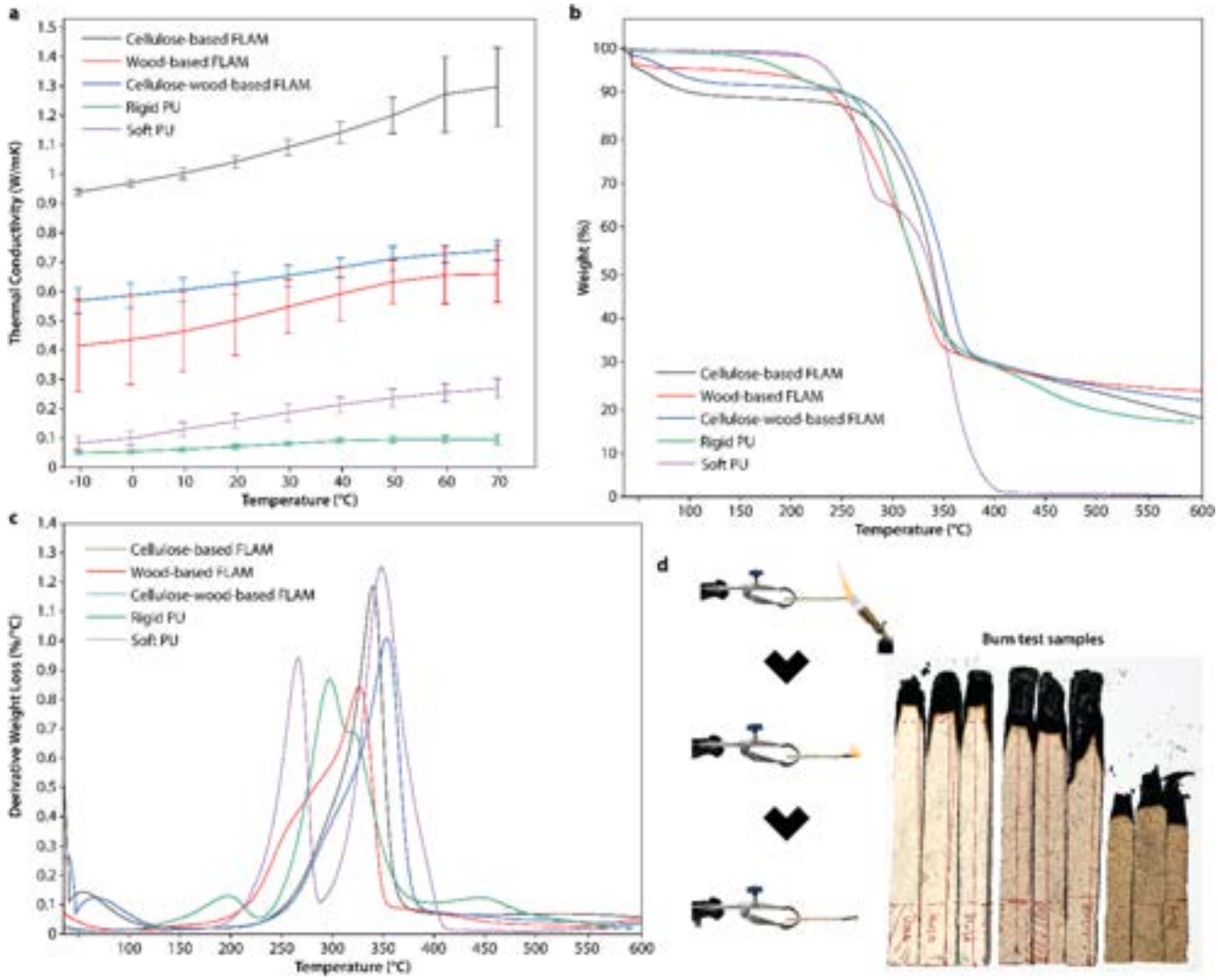


Fig. 3. Thermal properties of the chitinous cellulosic composite. (a) While chito-cellulosic composites and polyurethane foams have comparable thermal conductivities, the latter are marginally better insulators. In porous materials, these measurements are highly dependent on the material structure and the characteristics of their porosity, which influence airflow through the material, and not only their composition. Note that for PU foam, the thermal conductivity measurements at low temperatures, while in agreement with previous reports, fall below the nominal measuring range of the heat flow meter. (b-c) Weight loss and its rate as temperature increases. FLAM, made with naturally hydrophilic polymers, holds higher moisture content, which explains the initial weight drop as the water evaporates. Subsequent significant losses in all polymers correspond to the degradation of the functional groups first and the main polymer backbone later. (d) In an adapted horizontal burn test, the materials demonstrated high resistance to open flames, particularly the cellulose-based composites, and comparable to the UL94 HB rating classification.

dB per octave, suggesting that the tested samples are predominantly isotropic [33].

The overall transmission loss across low, medium, and high-frequency bands is encapsulated in Fig. 2b. In the low-frequency band ($\leq$ 500 Hz), all variants of chito-cellulosic composites outperformed the rigid and soft polyurethane foams and the commercial polyethylene insulation foam. In the medium and high-frequency ranges, the composites also surpassed soft polyurethane and commercial insulation foams' performance while matching the performance of rigid polyurethane foam.

Unlike transmission loss, which measures the energy transmitted between two distinct spaces, the sound absorption coefficient quantifies a material's capacity to diminish sound reflections within an enclosed space. As depicted in Fig. 2c and d, all variants of polyurethane foams previously tested, the commercial sound insulating foam (polyethylene-based), and the biological composites exhibited minimal or no absorption in the low-frequency range of less than 500 Hz. As the frequency range increases to the medium frequencies band, the polyurethane and the pure cellulose-based composite remain poor sound absorbers. In contrast, the absorbance of the commercial insulating foam and the composites containing wood significantly increases. This capacity to minimize sound reflections is directly proportional to the wood content. This trait is maintained at higher frequencies, where wood-based composites remain a viable alternative to commercial insulation foam.

In conclusion, all tested chito-cellulosic samples demonstrated superior performance in preventing sound transmission and reflection compared to regular polyurethane and commercial polyethylene sound insulation foams. Despite the inferior mechanical properties of wood-based composites relative to those based on cellulose, the former significantly excelled in minimizing sound reflection, performing on par with commercial sound insulation foams.

3.4. *Thermal insulation properties*

The thermal conductivity of a material, indicative of its heat transmission ability, was measured by placing samples of the different

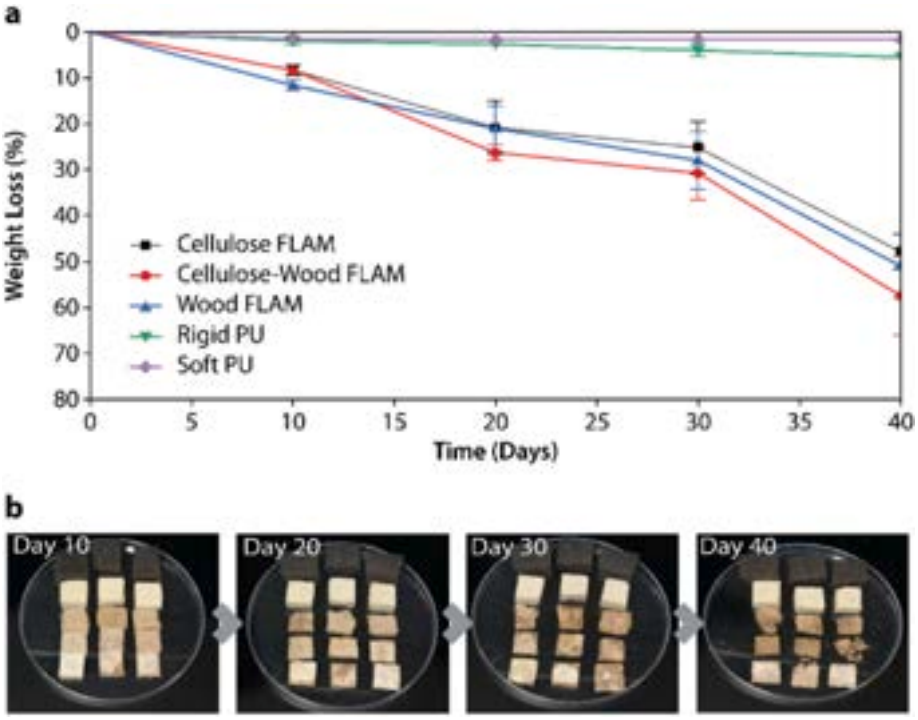


Fig. 4. Biodegradability test of chito-cellulosic and polyurethane-based composites. (a) The 40-day-long soil burial test showed that the FLAM composites retain the biodegradability of their unmodified raw components (i.e., cellulose and chitosan). This degradation accelerates in the later stages when the samples start losing their integrity and breaks apart. (b) Photographs of the samples at different time points, demonstrating the loss of material integrity in the biological samples. From top to bottom rows, the samples are: Soft Polyurethane, Rigid Polyurethane, wood dust-based FLAM, Wood dust-cellulose-based FLAM, and cellulose based FLAM.

materials between two plates at different temperatures and measuring the heat flow through the specimen [34]. Fig. 3a shows the temperature versus thermal conductivity curves for Polyurethane foams and chito-cellulosic samples. The thermal conductivity values for all samples increased with temperature, stabilizing at approximately 343 K (70 °C). Among the biological samples, wood-based composites exhibited the lowest thermal conductivity (0.06–0.08 W/K m), while cellulose and the 1:1 mix FLAMs also demonstrated relatively low thermal conductivity values of 0.10–0.13 W/K m and 0.07–0.08 W/K m, respectively. These values, approximately three times higher than those of the best insulating polyurethane foams [35], indicate that chito-cellulosic composites require comparatively higher thickness for effective thermal insulation. While this is a drawback in many applications, it is an acceptable trade-off in thermal insulation applications considering the other advantages of the biological composites, such as mechanical strength, manufacturability by additive manufacturing, lower cost, and ecological integration. It is worth noting that thermal conductivity is not inherent to the composition of these porous materials, as it is strongly influenced by their porosity and its effect on the airflow within the material [36]. Therefore, it is unsurprising that high-quality and expensive rigid polyurethane foam performs better than soft foam due to its significantly smaller porosity and average pore size. Consequently, while all the chito-cellulosic composites tested are suitable for thermal insulation, wood-based composites would be marginally better in situations where mechanical strength is not a priority due to their lower cost. The minor differences in thermal conductivity between samples are more likely dependent on the microparticle size and distribution rather than the composition of the material.

In addition to assessing the ability of various porous materials to prevent heat flow, we analyzed their high-temperature stability by measuring thermal degradation (Figs. 3b and c). The most notable difference between natural composites and synthetic insulators is the initial mass loss in the former due to water release at around 100 °C. This reversible effect, absent in polyurethane foams, is not a product of degradation [37]. Despite this difference, all synthetic and biological polymers begin to degrade at about 300 °C due to depolymerization and, in the case of biological composites, the destruction of the crystallite structure. In an open atmosphere, oxidative degradation will also contribute to this degradation [38,39]. The solid residue percentage was around 20 % for all samples, except for the low-density polyurethane foam, which ultimately burned out. Therefore, all chito-cellulosic composites exhibit similar thermal stability to high-end synthetic insulators.

3.5. *Flammability*

While flammability does not directly affect the functionality of insulating materials for heat or noise-generating systems, it is crucial for their safety and heavily regulated, determining their potential use in engineering applications. We assessed the flammability of the various composites using a horizontal burn test adapted from the UL-94 standard flammability test for plastic materials [40]. This test, released by Underwriters Laboratories, measures the ability of plastic materials to extinguish or spread a flame after ignition and their dripping behavior in response to a small open flame under controlled laboratory conditions. We fabricated composite strips and exposed them to an open flame for 30 s or until the material burning exceeded a length of 25 mm. We recorded observations of whether the flames were self-extinguishing, the length of fire damage, and combustion time as part of the characterization (Supporting Information Table 2).

Cellulose and wood flour are combustible on their own. However, when combined with chitosan, all variants of the chitinous-cellulosic composites become self-extinguishing. As depicted in Fig. 3d, all samples ceased burning before reaching the reference mark and exhibited a burn rate slower than the requirement for HB classification. The cellulose-based composites' high burning resistance, which withstood a propane torch for 30 s without any damage beyond the direct flame area, is particularly noteworthy. In accordance with the UL-94 HB rating, all

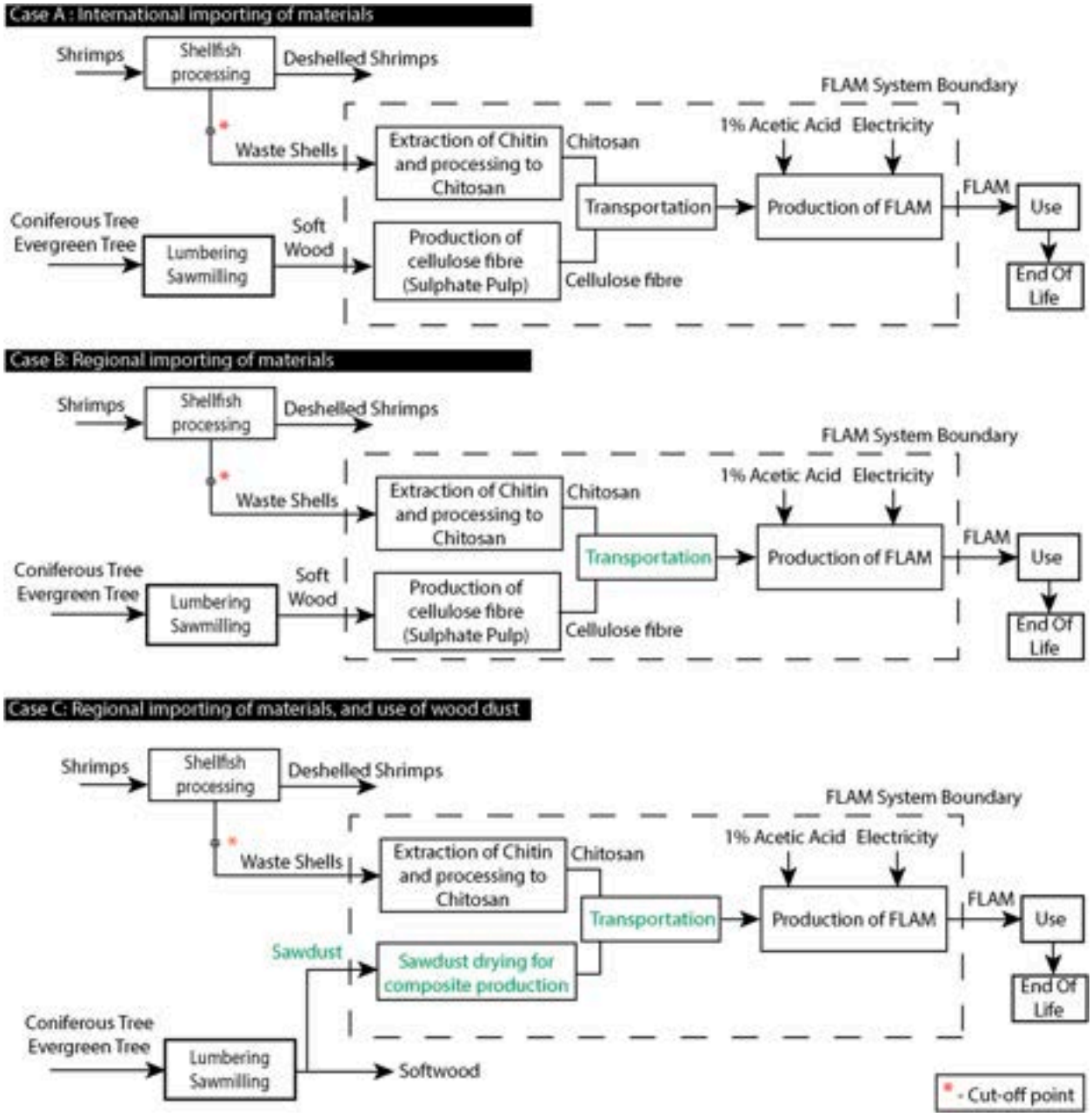


Fig. 5. System boundary of different production scenarios considered during life cycle assessment. The three scenarios contemplate various material sources and sourcing methods, exploring opportunities to reduce environmental impact. The main differences between the scenarios lie in the reduced transportation distance associated with regional raw material sourcing and the use of sawdust/wood flour, a byproduct of lumbering and woodworking, for producing the chito-cellulosic composites, as opposed to using virgin cellulose pulp.

chito-cellulosic composites are suitable for portable, attended, intermittent-duty household appliance enclosures (e.g., hair dryer) and decorative parts.

3.6. Biodegradability

The biodegradation of the chito-cellulosic composites and polyurethane foam samples was investigated using a soil burial biodegradability test under ambient conditions, simulating the scenario when the foams are not part of a waste management system and end up in natural ecosystems [41]. We buried three replicates of each sample and tracked their weight loss in 10-day intervals over 40 days. Fig. 4a illustrates the state of the samples collected at each time point. All composites exhibited similar characteristics, losing about 60 % of their mass during the 40 days they were buried (Fig. 4b). As expected, the polyurethane foams showed negligible mass loss during the same period. It is important to note that while the biodegradation of both chitosan and cellulose is primarily driven by the action of enzymes secreted by soil microorganisms, the mass loss in the case of polyurethane foams is more of a biodeterioration rather than a biodegradation, as it results from partial disintegration of the material due to abiotic factors (e.g., moisture) [42]. Therefore, as expected, the chito-cellulosic composites retain the same biodegradability as their components, resulting in materials fully integrated into Earth's ecological cycles without needing recovery or processing in specialized facilities for biodegradation.

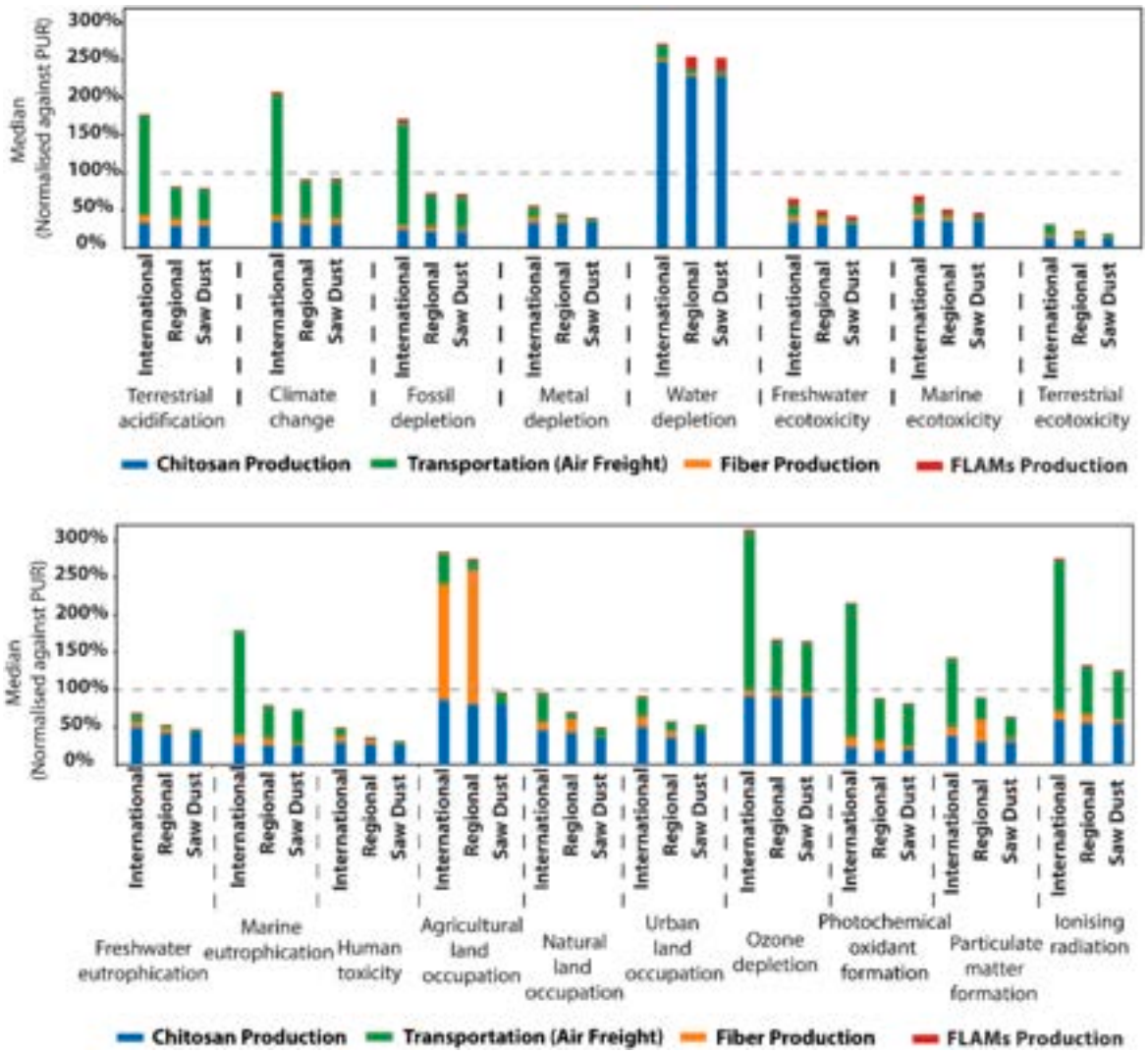


Fig. 6. Emissions attributed to various environmental indicators normalized to rigid polyurethane foam production emissions. Compared to rigid polyurethane foam, the base scenario of producing cellulose-based composites while sourcing materials from international suppliers improves several environmental indicators. Furthermore, given the wide availability of cellulose and chitosan, producing the same composite using regionally available suppliers allows the environmental impact of chitinous cellulosic composites to be noticeably better than rigid polyurethane foam across almost all indicators. As expected, the use of sawdust significantly reduces the environmental impact associated with the large land requirements of forestry.

3.7. Environmental attributes

To compare the environmental impact of producing chitinous cellulosic composites with different material sourcing options (Fig. 5), we normalized the emissions impacting the various environmental indicators against rigid polyurethane foam as it is the broadly used insulator with the closest mechanical characteristics to the chito-cellulosic composites (Fig. 6). Thus, less than 100 % normalized emissions would indicate that the studied scenarios are more environmentally friendly than rigid polyurethane foam. The system boundaries for the life cycle assessments commence with the sourcing of raw materials—shrimp shells and wood. Consequently, the environmental analysis encompasses the chemical processes that convert these materials into chitosan and cellulose. In the base scenario, where chito-cellulosic composites are produced at the lab scale without considering the optimization of the material source and efficiency of the logistics during its procurement, less than half of all environmental indicators (eight out of eighteen) improved respect rigid polyurethane foam. Those indicators worsening are primarily contributed by the emissions from transporting raw materials via air freight internationally, resulting in the significant use of aviation fuel. Additionally, significant emissions associated with water depletion contributed to the environmental impact of chitosan production in laboratory conditions, where deionized water was used to dilute solvents or wash demineralized, deproteinized, or deacetylated products.

While emissions can potentially be reduced by opting for sea freight for material transportation, the associated transportation distances can be significantly minimized to a level unachievable with synthetic polymers, as both cellulose and chitosan are ubiquitous biopolymers that can be sourced regionally or even locally from urban waste sources [23]. This improvement is evident in the second scenario, where emissions from regional sourcing are lower than those from rigid polyurethane foam for most environmental indicators (fourteen out of eighteen).

Two environmental indicators that remain high in the production of chito-cellulosic composites are water depletion and agricultural land occupation. Interestingly, when sawdust or wood flour, considered byproducts of the lumber industry, are utilized, the emissions associated with agricultural land occupation are drastically reduced, as they are attributed to primary lumbering products. Along with the opportunities in alternative chitosan extraction methods, such as enzymatic processes where water or solvents can be minimized, the environmental attributes of chitinous cellulosic composites can be further optimized. Meanwhile,

with the current industrial production methods, chitinous cellulosic composites are a superior choice regarding relevant environmental indicators of currently heightened concerns, such as climate change and fossil depletion.

4. Conclusions

In conclusion, this study demonstrates that bioinspired chito-cellulosic composites, specifically Fungal-like Adhesive Materials (FLAMs), made with the two most ubiquitous organic molecules on Earth and exhibiting exceptional manufacturability, present a viable and sustainable option for insulation applications. We have shown the advantages and trade-offs in insulating capabilities, mechanical properties, flammability, and environmental impact of composites made with different qualities of cellulose, from highly purified cellulose to powdered wood. Although the thermal conductivity of these composites is approximately three times higher than that of polyurethane foams, necessitating increased material thickness for similar thermal insulation, this drawback is offset by their superior mechanical strength, manufacturability through additive manufacturing, lower cost, and ecological integration. Additionally, FLAMs exhibit electrical and acoustic insulating properties on par with the best current options, biodegradability, and reduced environmental impact, making them suitable for various engineering applications where sustainability is a priority. Their seamless integration into Earth's native ecological cycles, coupled with a production rate several orders of magnitude higher than the global demand for all plastics, positions these sustainable materials as a realistic option for insulating foams. The results highlight the potential of these biological composites to contribute to a new era of technological efficiency and environmental responsibility, aligning with the growing demand for sustainable materials in the industry.

CRediT authorship contribution statement

Ajay Kumar P.V.: Writing – review & editing, Investigation, Formal analysis, Data curation. **Shiwei Ng:** Writing – review & editing, Formal analysis, Data curation, Conceptualization. **Yung Boon Chong:** Writing – review & editing, Methodology, Investigation, Formal analysis. **Heow Pueh Lee:** Writing – review & editing, Supervision, Methodology. **Stylianos Dritsas:** Writing – review & editing, Software, Methodology, Formal analysis. **Javier G. Fernandez:** Conceptualization, Data curation, Formal analysis, Resources, Writing – review & editing, Supervision, Methodology, Project administration, Funding acquisition.

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

Javier Gomez Fernandez reports was provided by Singapore University of Technology and Design. Javier Gomez Fernandez has patent #Apparatus for large scale additive manufacturing, and a material for manufacturing (US Patent 11,820,077) licensed to Singapore University of Technology and Design. If there are other authors, they declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.susmat.2025.e01394.

Data availability

All data is included in the main document and supplementary files

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