(REFEREED RESEARCH)

THERMAL CONDUCTIVITY AND ACOUSTIC PROPERTIES OF NATURAL FIBER MIXED POLYURETHANE COMPOSITES

DOĞAL ELYAF KARIŞIMLI POLİÜRETAN KOMPOZİTLERİN SES VE ISI YALITIM ÖZELLİKLERİNİN İNCELENMESİ

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ABSTRACT

Natural cotton, bamboo and wool fibers were used as reinforcement agents in a polyurethane-based matrix to improve the sound absorption and thermal conductivity properties of the composite. Generally, adding cotton, bamboo or wool fibers to polyurethane foam, improves its sound absorption coefficient. In this study, cotton fibers were observed to provide the best sound absorption coefficient. At higher frequencies, increasing the bamboo or wool fiber content decreases the sound absorption coefficient for the composite. Adding cotton, wool or bamboo fibers to polyurethane foam does not result in a significant change in the thermal conductivity of the material. The best thermal conductivity value was observed with a composite including 4% cotton fiber.

Key Words: Polyurethane, Sound absorption, Thermal properties, Composites, Natural fiber

ÖZET

Bir kompozitin ses yutum ve ısı yalıtım özelliklerini iyileştirmek amacıyla poliüretan esaslı bir matrise doğal pamuk, bambu ve yün elyaf katılmıştır. Bu çalışmanın arkasındaki hedeflerden biri de tekstil atıklarının değerlendirilmesi ve malzeme üretiminde daha az poliüretan kullanılmasıdır. Genel olarak bakıldığında, doğal elyaf ilavesi, poliüretan köpüğün ses yalıtım özelliğini iyileştirmektedir. Yapılan deneylerde, pamuk elyaf destekli poliüretan kompozitin en iyi ses yutum katsayısı değerlerini verdiği gözlenmiştir. Poliüretan köpüğe katılan bambu ve yün elyaf oranlarının arttırılması, kompozitin yüksek frekanslardaki ses yutum katsayı değerlerini azaltmaktadır. Poliüretan köpük içerisine pamuk, bambu veya yün elyaf katılması ısıl özelliklerde kayda değer bir değişim oluşturmamaktadır. En iyi ısıl iletkenlik özelliği %4 pamuk elyaf içeren kompozit için gözlenmiştir.

Anahtar Kelimeler: Poliüretan, Ses yalıtımı, Isıl özellikler, Kompozit, Doğal elyaf.

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1. INTRODUCTION

Polymeric foams have been widely used in industry due to their mechanical, electrical, thermal and acoustic properties (1). As a subfamily of polymers, polyurethane is one of the largest and most versatile products. Changing the chemical composition of polyurethane or adding fiber or other filler reinforcement results in a wide set of materials with different properties. Polyurethane (PU) foam can be rigid or flexible; rigid foams are used for thermal insulation while flexible foams are used as cushioning materials in furnishings, transportation and packing applications.

Several studies have been made composite PU foam materials using synthetic fibers such as glass, carbon, boron, nylon and kevlar as additives. The tensile properties of polyurethane foams were increased using reinforcement agents such as polyester, glass, Kevlar-49 aramid fiber (2-6), carbon nanotubes (1), multi-walled carbon nanotubes (7), methylene-bis-ortho-chloroanilline (MOCA) grafted carbon nanotubes (8), SiO_2 (9), silexil (10), post-consumer PET (polyethylene terephthalate) (11) and mineral fillers such as calcium carbonate and crystallized silica particles (12). Specifically, pure carbon nanotubes (13), multi-walled carbon nanotubes (7) and silexil (10) were observed to improve the Young's modulus of the PU foam composites. Reinforcement of PU foam with glass or Kevlar-49 aramid fibers (7) and impregnating PU between two hollow core piles (14) improved the impact response of the composite. The compressive strength of PU foam composite structures has been improved using free radical polymerized poly (N-isopropylacrylamide) (PNIPA) hydrogels (15); biodegradable interpenetration network structures made from tricalcium phosphate (TCP), hydroxyapatite (HA), and poly(dl-lactide-co-glycolide) (PLGA) (16); expandable graphite pulverized into fine particles within a capsular layer of polymethyl methacrylate (PMMA) (17); a network structure of cement particles formed by hydration (18) and whisker silicon particles (19). The flexural strength of PU foam composites was improved using blocked isocyanate (NCO)-terminated PU prepolymer prepared from ϵ-caprolactam blocked blends of toluene diisocyanate and branched polyester and mixed with various fibers such as glass, carbon and Kevlar-49 aramid fiber (2-4). Composite foams produced by mixing expanded and non-expanded microspheres, packing the dry thermoplastic polyacrylonitrile based microspheres into a fibrous preform in a closed mold, and heating the assembly to expand and weld the microspheres and polyester, aramid and glass fibers together yielded a resistance to crack propagation superior to that of the unreinforced foam structure (5-6). Using unidirectionally oriented glass fiber and carbon fiber as reinforcement materials in polyurethane foam matrix yielded vibrational properties and frequency response characteristics comparable to those of Sitka spruce wood, which is used for conventional soundboard (20,21). Using post-consumer PET (polyethylene terephthalate), PETpc, as a reinforcement filler in flexible polyurethane foams increased the tear resistance (11) of PU composites. Mineral fillers such as calcium carbonate and crystallized silica particles improved the viscoelastic properties of PU foam composite provided that the fillers were larger than the cell size (12). The thermal properties of PU foam composites were improved using polyester, glass, Kevlar-49 aramid fibers (2-4,22) and organically-modified layered silicates, inorganic spherical nanopowder fillers and benzyl-dimethyl-hydrogenated-tallow ammonium salt modified natural montmorillonite fillers (23). Polyurethane filled with expandaple graphite pulverized into fine particles and encapsulated in a layer of poly(methyl methacrylate) (17) and whisker silicon particles (19) improved the flame retardancy of the composite. The sound absorption of the composite has been remarkably improved by incorporating microparticles or micro-porous microspheres into the polyurethane foam matrix (24) using water as a blowing agent and by loading even very low CNT fractions (1). Multilayer construction of polyurethane foam layers with fine powder (white carbon and vermiculite) beds (25) and recycled rubber particles (26) resulted in a high sound absorption performance. A uniform dispersion of carbon nanotubes in the PU foam was achieved to produce a low density conductive foam composite (27). Carbon black and multi-walled carbon nanotubes were added to preimpregnated composite and polyurethane foam as conductive fillers, resulting in a new X-band (8.2-12.4GHz) radar absorbing sandwich structure (28).

Using synthetic fibers as reinforcement agents in PU foam composites poses some disadvantages such as slow deterioration, cost and consumption of nonrenewable resources. The use of renewable raw natural materials as substrates in PU foam composites has attracted the attention of many researchers because of their biodegradability and low cost. Recently, PU–solid wood composites were prepared by impregnating PU prepolymer into low-density, rapidly grown, solid poplar wood and controlling the foaming of the PU prepolymer within the voids in the wood in the presence of the catalysts triethanolamine (TEA), diethylenetriamine (DETA), triethylenediamine (TEDA), or N-methylmorpholine (NMM). Treatment with N-methylmorpholine (NMM) as a catalyst resulted in the best flame resistance because the PU resin formed a foam that extended throughout the wood (29). Nanocomposites of rigid polyurethane foam with unmodified vermiculite clay dispersed in the isocyanate before blending improved the compressive strength and modulus while decreasing the thermal conductivity of the composite (30). Pure and woven sisal fabric resulted in good fracture toughness, while alkali treated coconut fibers improved the toughness of the composite (31). Microcellular crosslinked PU synthesized from a castor oil-based polyol was reinforced with pine wood-fibers or with hemp. Hemp fiber composites generally showed better dynamic flexural properties, and material aging was observed to increase the modulus and toughness of the composite (32). Garnet particles were added to polyurethane foam to increase its abrasiveness, and higher hydroxyl values in the polyol blend increased the surface rougness of the produced composite. (33). Polyurethane-based composites reinforced with woven flax and jute fabrics were prepared to investigate the influence of the type of reinforcing fiber and of the fiber and microvoid contents on the mechanical properties of foams. Increasing the amount of flax and jute fabrics in a polyurethane matrix increased the shear modulus and impact strength, specifically for the composites including woven flax fiber (34). Hard wood cellulose fibers were fibrillated with a high pressure homogenizer form fibers from the micro to nanoscale. The composite materials were prepared using compression molding by stacking the cellulose fiber mats between polyurethane films. The results showed that both cellulose microfibers and nanofibers reinforced the polyurethane and provided better tensile strength and modulus properties (35). The natural and environmentally friendly tea-leaf-fibers (TLF) have been used to improve the sound absorption properties of PU foam composites (36).

Sound absorption constitutes one of the major requirements for modern human comfort. The need for sound insulation in automobiles, manufacturing environments, and equipment requires the development of more efficient and economical ways of producing sound absorption materials. Industrial sound insulation materials generally use materials such as glass wool, foam, mineral fibers and their composites. As an alternative to the natural fiber mixed composites that have recently been widely studied, the current study investigates the effect of cotton, wool and bamboo fiber fillers on the sound absorption and thermal conductivity of polyurethane foams.

2. MATERIALS AND METHODS

2.1. PU Foam Formulation

Two dies of aluminum and stainless steel were manufactured for open molding of PU foam to prepare samples for acoustic measurements.

H2411/1 polyol and isocyanide from BASF were used for PU molding. The polyol component of the foam mixture includes polyol, catalyst and other additives. The isocyanate component includes difenilmetan diisocyanate mixture (ISO PMDI 92140). Table 1 shows the properties of polyol and isocyanate.

Table 1. Properties of Polyol and Isocyanate components

Physical properties	Unit	Polyol	Isocyanate	Standards
density (20 °C)	g/ cm³	1,01	1,24	DIN 51 757
viscosity (20 °C)	mPa.s	400	300	DIN 53 018
NCO content	%	—	31,5	ASTM D 5155-96 A
Storage life	Month	3	6	

The R 525 B separator was used to facilitate the removal of the foam from the die. The dies were carefully cleaned before using the separator.

2.2. Sound Absorption Measurements

PU foam was prepared by mixing the polyol and isocyanate at a 1:1 ratio. Cotton, wool and bamboo were prepared as additives to examine their effects on the sound absorption and thermal conductivity of the foams. Each material was cut to fibers approximately 1 mm long. PU foam mixtures were prepared at three different weight ratios (4%, 8% and 12%) for each fiber material. After pouring the mixture into the die, a driller with a mixer attached was used to bring the composition up to 2000 rev/min to obtain a homogeneous compound. From each mold, 5 different samples with a diameter of 100 mm and thickness of 2 cm were obtained for large tube measurements (Figure 1a). A smaller round piece with a 29 mm diameter and a 2 cm thickness was prepared from each large tube sample for small tube measurements (Figure1b).

The sound absorption measurements were based on a two-microphone transfer-function method according to ISO 10534–2 and ASTM E1050–98 international standards for horizontally mounted orientation-sensitive materials. The testing apparatus was part of a complete acoustic material testing system featuring a Brüel& Kjær PULSE™ interface. Small impedance tube kits consisted of a 29 mm diameter tube (small tube), a sample holder and an extension tube of the same diameter. The large impedance tube kit consisted of a similar tubular apparatus with a diameter of 100 mm. The small and large tube setups were used to measure different acoustical parameters and then large and small tube measurements were combined to determine the sound absorption rate for the frequency range 50 - 6300 Hz.

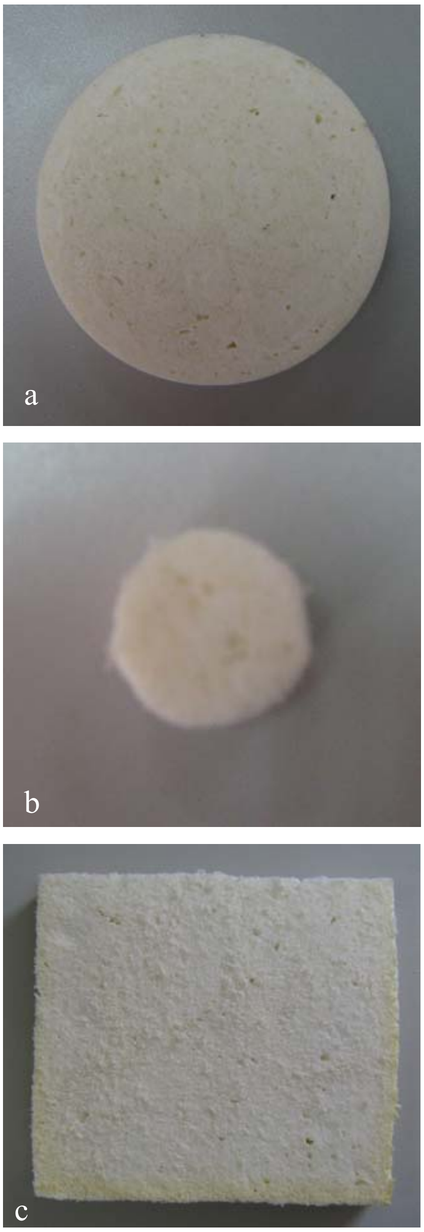

Figure 1. Samples prepared for acoustic and thermal measurements. (a) Sample for large tube sound absorption measurement, (b) sample for small tube sound absorption measurement.

2.3. Scanning Electron Microscope (SEM) Measurements

The morphologies of the various foam samples were examined by a scanning electron microscope (SEM, JEOL JSM-5910 LV, high-resolution) at magnifications of 50x, 250x and 500x. The samples were fractured in liquid nitrogen to avoid structural deformation and then coated with gold by a sputter-coater to impart electrical conductivity and reduce charging artifacts.

2.4. Thermal Conductivity Measurements

A steel die was manufactured for open molding of PU foam to prepare samples for thermal measurements. Pure PU foam alone and three different composites were produced by mixing PU foam with cotton, bamboo or wool. Each fiber was added to PU foam at three different weight ratios of 4, 8 and 12%. Five samples of 20x20x3cm were cut from the free rised PU foam mixtures (Figure 1c). The thermal conductivity of each sample was measured using a LAMBDA-CONTROL A50 thermal conductivity instrument (manufactured by Hesto Elektronik GmbH, Germany) with an upper plate temperature of 36° C and a lower plate temperature of 10° C. The samples were tested according to German Standard DIN 52612.

The measurement of bulk density for short fibers is based on the Archimedes principle in which the volume of water displaced by a solid is equal to the volume of the solid.

3. RESULTS and discusslon

3.1. Impedance Tube Measurements

3.1.1. Impedance tube measurement of PU foam and the cotton composite

The sound absorption coefficients for pure PU foam were plotted against the PU foam composites with cotton fibers at 4% (PU4C), 8% (PU8C) and 12% (PU12C) weight ratios.

The sound absorption of pure PU foam is 0.1 at 50 Hz with a steady increase up to 0.4 at 6.3 kHz.

Including 4% cotton fibers in the polyurethane mixture (PU4C) increases the sound absorption coefficient by as much as 5 times in the frequency range of 50 Hz - 2.5 kHz. The sound absorption of PU4C then decreases to 0.6 at higher frequencies up to 6.3 kHz.

Increasing the cotton content in PU foam to 8 wt% results in an increase in sound absorption by as much as 50% in the frequency range of 50 Hz - 1.5 kHz, when compared to that of PU4C. Between 1.5 kHz and 6.3 kHz, both PU8C and PU4C demonstrate similar sound absorption characteristics except for the frequencies between 3.5 kHz and 5 kHz, where the sound absorption of PU8C becomes 20% higher than that of PU4C.

PU12C results in a 5% increase in sound absorption when compared to PU8C in the frequency range from 50 Hz - 1.5 kHz. For higher frequencies, however, the sound absorption of PU12C is 40% higher than that of PU8C (Figure 2).

3.1.2. Impedance tube measurement of PU foam and bamboo composite

Bamboo fibers mixed with PU foam generally demonstrate higher sound absorption than pure PU foam. Including 4% bamboo fibers in PU foam (PU4B) results in a steadyncrease in the sound absorption coefficient to at most 4 times that of pure PU foam at frequencies from 50 Hz-3.7 kHz. Above 3.7 kHz, the sound absorption of PU4B then decreases from 0.8 to 0.7 and then remains at that level until 6.3 kHz. Increasing the bamboo fiber content in the PU foam composite to 8% (PU8B) does not change the sound absorption characteristics significantly compared to PU4B at frequencies from 50 Hz-3 kHz. After a slight peak around 3 kHz, the sound absorption of PU8B decreases to 0.6 and continues at that level for frequencies from 3.5-6.3 kHz. Including 12% bamboo in PU foam (PU12B), results in similar sound absorption characteristics between at frequencies from 50 Hz-3 kHz. After a slight increase around 3 kHz, the sound absorption of PU12B decreases to 0.6 and then continues at that level for frequencies 3.5-6.3 kHz (Figure 3).

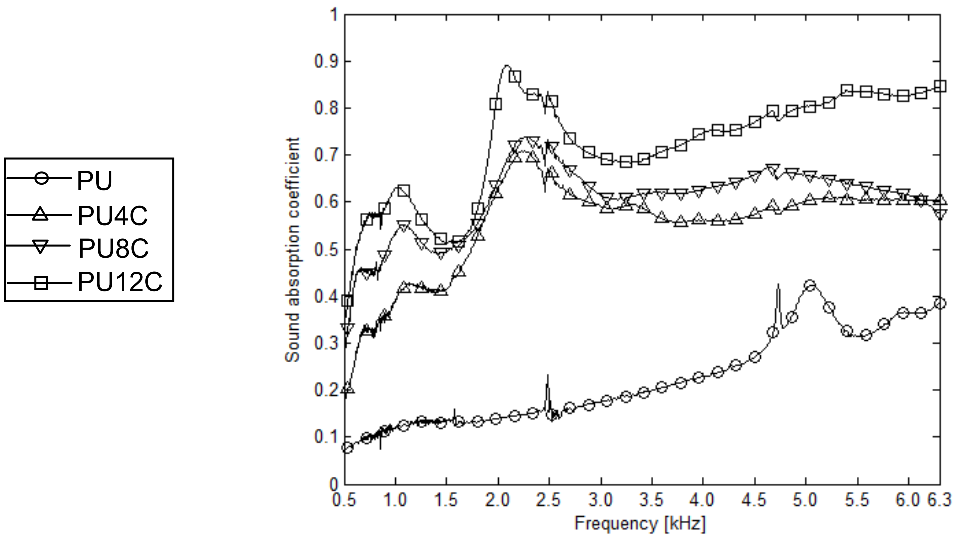

Figure 2. Sound absorption of PU foam and cotton fiber composites

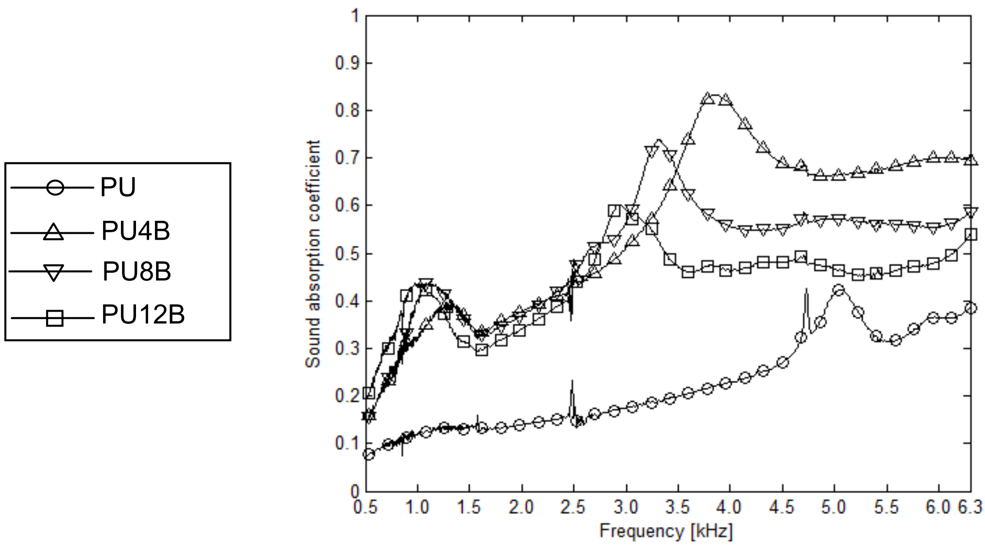

Figure 3. Sound absorption of PU foam and bamboo fiber composites

3.1.3. Impedance tube measurement of PU foam and wool composite

Including wool fibers in PU foam at a 4% weight ratio (PU4W) results in a maximum sound absorption of 0.8 at 2.7 kHz, almost 4 times larger than the absorption of PU foam at the same frequency. The sound absorption of PU4W decreases to 0.6 and then remains steady at that at frequencies from 3-6.3 kHz. Including wool fibers at 8% (PU8W) results in an increase in sound absorption by as much as 67% at frequencies 50 Hz-1.4 kHz and 3-3.5 kHz. However, the sound absorption for PU8W is at most 50% lower than that of PU4W at frequencies between 1.4-3 kHz and 3.5-6.3 kHz. Including 12% wool fiber in the composite (PU12W) results in a 35% increase in sound absorption around 1.1 kHz. However, the sound absorption for PU12W is generally lower than that of PU4W and PU8W for the frequencies between 1.5-6.3 kHz (Figure 4).

3.2. SEM Results

Figures 5-10 present SEM observations of PU foam with different weight percentages (4%, 8% and 12%) of cotton, bamboo and wool fiber. The structure of the PU foam consists of cells with a polyhedron shape. Adding fibers into the structure makes the cells more narrow and the cell sizes become non-uniform (formless). Including 4-8% cotton fiber in the mixture does not damage the PU foam structure, but at 12% cotton, the cell structure is deformed (Figure 5).

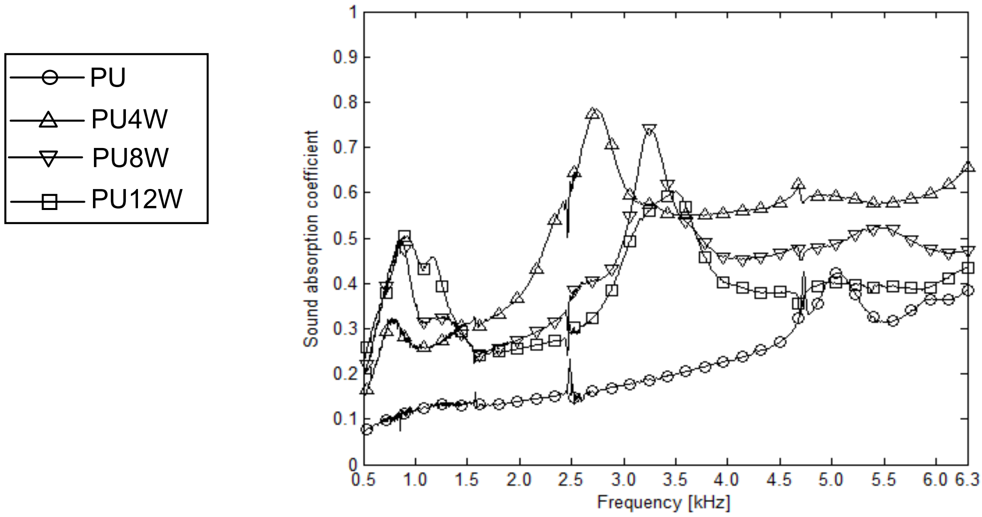

Figure 4. Sound absorption of PU foam and wool fiber composites

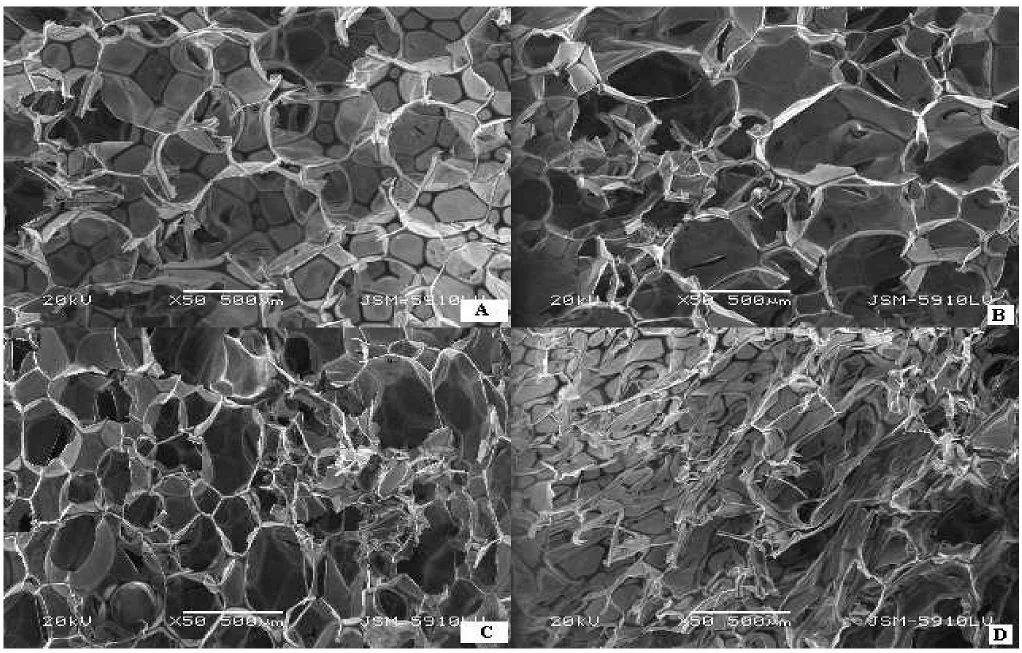

Figure 5. Cross-sectional SEM images of PU foam with cotton fiber additives (PUC). Magnification 50 x. A- standard PU foam, B- PU4C, C-PU8C, D- PU12C.

At greater magnifications (Figure 6) the cotton fibers are observed to cluster on the walls of the cells, thereby protecting the cell structure of the PU foam (especially for PU4C and PU8C).

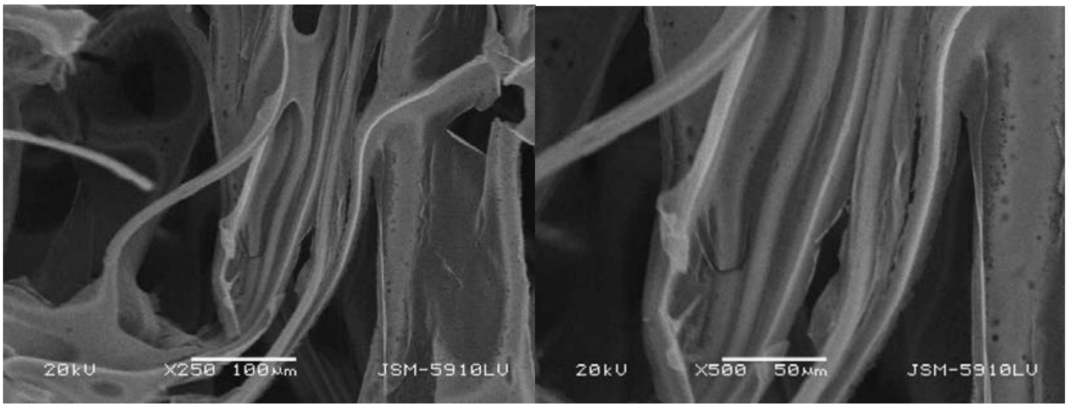

Figure 6. Cross-sectional SEM images of PU foam with cotton fiber additives (PUC). Magnification 250 and 500 x.

Including bamboo fibers at 4% does not damage the cell structure of the foam. Increasing the bamboo content, results in a slight deformation of the cell structure of the foam (Figure 7).

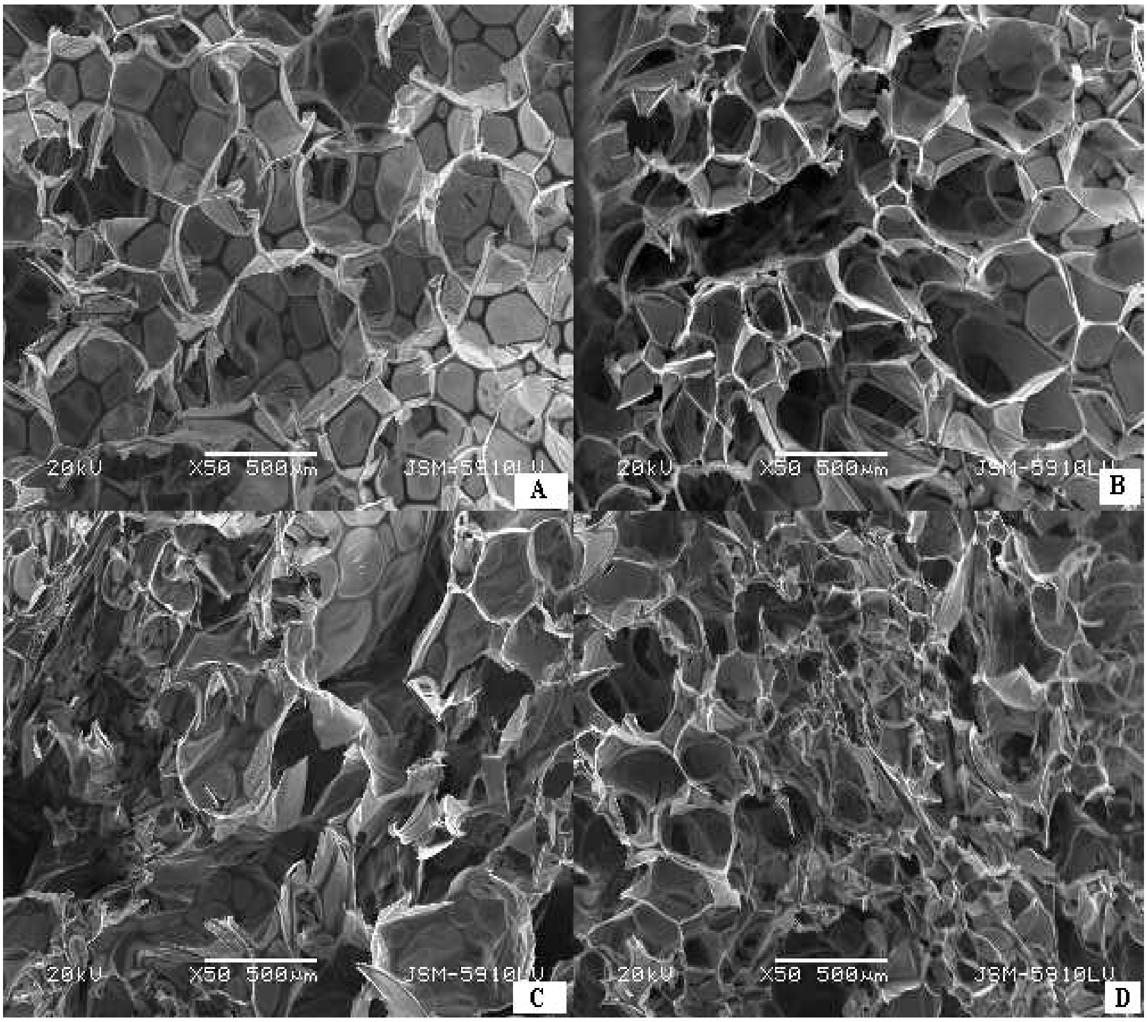

Figure 7. Cross-sectional SEM images of the PU foam with bamboo fiber additives (PUB). Magnification 50 x. A- standard PU foam, B- PU4B, C- PU8B, D- PU12B.

At greater magnifications (Figure 8) the bamboo fibers can be observed to cluster on the walls of the cells. The bamboo fibers were also observed to induce a slight deformation to the cell structure of the foam (especially for PU8B and PU12B).

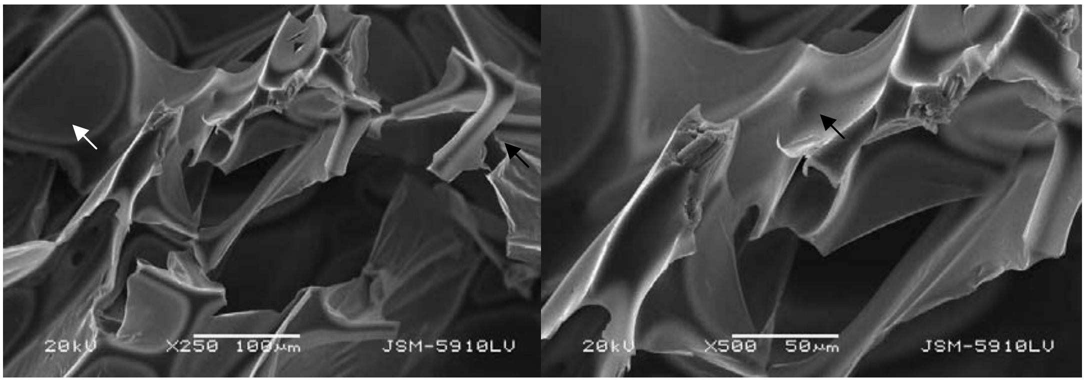

Figure 8. Cross-sectional SEM images of PU foam with bamboo fiber additives (PUB). Magnification 250 and 500 x.

Figure 9 presents SEM images of the composite including wool fibers in three different weight ratios. Wool fiber additives were observed to induce a deformation of the cell structure for all weight ratios.

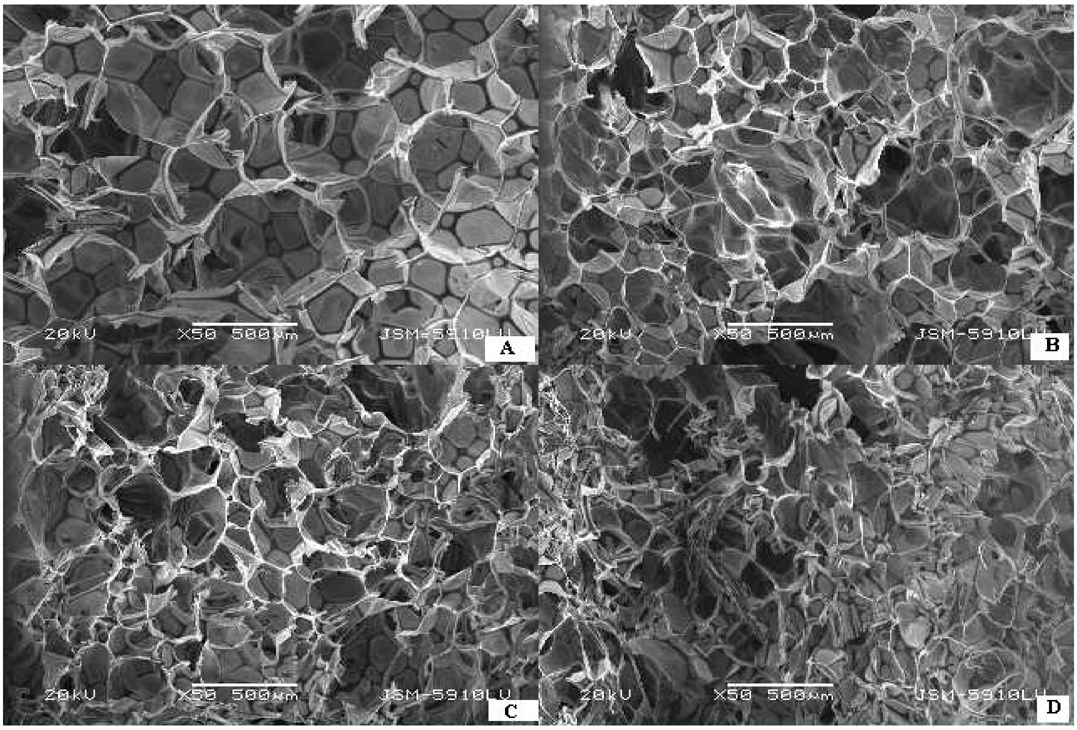

Figure 9. Cross-sectional SEM images of the PU foam with wool fiber additives (PUW). Magnification 50 x. A- standard PU foam, B- PU4W, C- PU8W, D- PU12W.

Wool fibers were observed to penetrate the cells and significantly damage the PU foam structure (Figure 10).

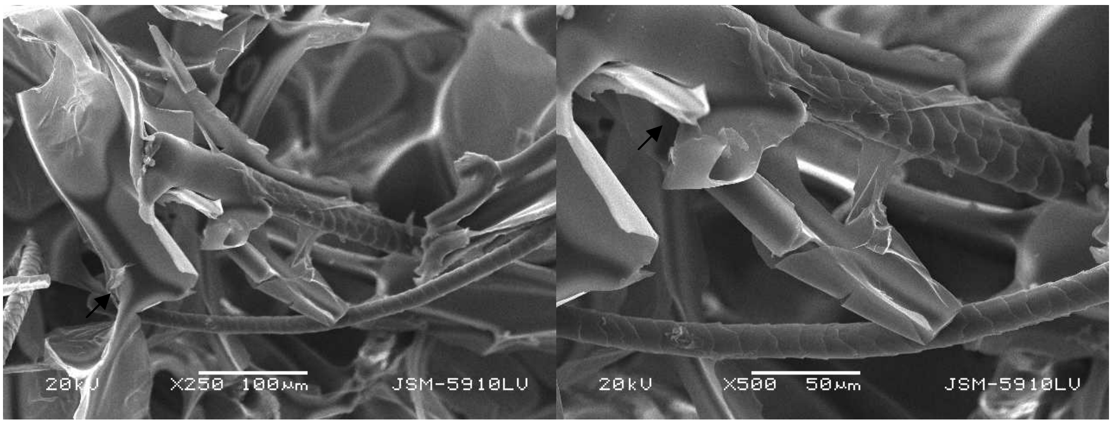

Figure 10. Cross-sectional SEM images of PU foam with wool fiber additives (PUW). Magnification 250 and 500 x.

Table 2. Thermal conductivity of the PU samples

Material	λ*	Density**	Material	λ	Density	Material	λ	Density
PU (pure)	42.5	7.3 10^{-3}						
PU4C	36.4	0.012	PU4W	37.8	0.012	PU4B	37.8	0.010
PU8C	40.5	0.014	PU8W	37.0	0.016	PU8B	37.5	0.015
PU12C	41.7	0.020	PU12W	38.5	0.018	PU12B	38.0	0.017

(*λ = $mW \cdot m^{-1} \cdot K^{-1}$) (** $g\ cm^{-3}$)

3.3. Thermal Conductivity

The thermal conductivity of different natural fiber reinforced composites and corresponding bulk density values are listed in Table 2. Addition of cotton, bamboo and wool fibers to PU foam was found to cause an insignificant change in the λ values for thermal conductivity with the best λ obtained for the composite containing 4% cotton fiber. Increasing the fiber content in the PU foam matrix, resulted in a slight increase in the density of the foam.

4. CONCLUSIONS

This study developed new natural fiber mixed polyurethane based composites and tested their sound absorption and thermal conductivity properties. Adding cotton fibers to PU foam results in a significant increase in sound absorption. PU foam mixed with 12% cotton fibers results in almost 0.8 sound absorption above 2 kHz, which represents a 4 times increase over that of pure PU foam material at certain frequencies. Similarly, including wool fibers in PU foam at a 4% weight ratio results in higher sound absorption characteristics compared to pure PU foam. Except for peaks with a narrow frequency range, PU foam and wool fiber mixed composites result in a maximal sound absorption of 0.6, which is at most a three-fold increase over that of pure PU foam. Below 1,5 kHz, increasing the wool fiber content increases the sound absorption of the composite. When compared to wool fiber mixed composites, cotton fiber mixed PU foam results in greater sound absorption. Except for the peaks in a narrow frequency range, mixing bamboo fibers at 4% by weight in PU foam results in a sound absorption of 0.7, which is three-fold higher than that of pure PU foam at around 4 kHz and continues at that level for a wider range of frequencies between 4-6.3 kHz. Bamboo mixed PU foam composites result in greater sound absorption than wool fiber mixed PU foam composites. For both bamboo and wool mixed composites, increasing the fiber content results in lower sound absorption above 3.5 kHz. This may be due to the disruption of bamboo and wool fibers observed in SEM images of the bamboo and wool fiber mixed PU foam composites. However, increasing the cotton content in the PU foam results in an increase in sound absorption. The SEM images revealed that the cotton fibers oriented around the surface of the foam cells without deforming their structure, while bamboo fibers induce a slight deformation of the cell structure. This disruption may be due to the large surface area of the bamboo fibers resulting in more friction on the walls of the cell structure. Addition of wool fibers to the composite induces significant cell deformation. This may be due to the helical surface structure of the wool fibers causing a drilling effect and hence disruption of the cell structure. Considering all of the natural fiber reinforced composites developed in this study, even 4% fiber reinforcement was sufficient to improve sound absorption when compared to pure PU foam material. Increasing the fiber content in bamboo and wool composites results in a decrease in sound absorption while increasing the amount of cotton fiber improves the sound absorption of cotton reinforced PU composites, and these composites had the best sound absorption of all the fiber reinforced composites. Among the fibers studied, cotton provides the best reinforcement of PU matrix composites. Also, reducing the length of the fibers is expected to improve the acoustic and thermal properties of the PU based composites. Addition of cotton, wool or bamboo fibers to PU foam does not induce a significant change in the thermal conductivity of the composites. Although the differences are not significant, the lowest thermal value was observed for the composite including 4% cotton fibers. As an additional benefit, increasing the fiber content in a composite reduces the amount of PU used, thus lessening the environmental impact and reducing the cost of the developed material.

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