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Morphological and Thermo-Mechanical Characterization of Open-Cell Spray Polyurethane Foamed Wall Insulation Modified With Cellulose Fiber

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Abstract

Open-cell spray polyurethane foam insulation was prepared using the soy-based polyol and water as blowing agent. Cellulose fiber was embedded in polymer matrix as reinforcement and the effects of fiber on morphological changes, as well as thermal and mechanical properties of the foam insulation were investigated. The foam was characterized at cellular level by FTIR and SEM and it was demonstrated that incorporation of fiber in open-cell foam insulation altered the foaming structure. Cell density increased and became more homogeneous. Bulk density and compressive strength of the composite foam system were improved. Thermal effectiveness of the composite foam was improved at lower fiber concentration. Moisture permeability was reduced. However, at higher fiber content the reinforcement effect weakened due to agglomeration of fiber.

Key words: Polyurethane; Polyol; Isocyanate; Cellulose fiber; Cross-linking; Urethane; Urea; Hard and soft segments; Open-cell; Composite foam

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INTRODUCTION

Polyurethanes were developed by Bayer Company around the year 1950. It was a success story and formed the basis for a present multi-billion dollar business in the world today^[1]. Since then manufacturers and pioneers of polyurethane dedicated a great deal of research for improvements, but in spite of many discoveries, there has been little change to the original work. Polyurethane foams are currently used in many applications depending on their categories such as flexible, semi-rigid and rigid. They are mainly used in transportation, furniture, and construction industries. The control of parameters such as chemical composition, functionality of the materials and molecular weight would produce a wide variety of foam types with significant differences in properties.

Spray polyurethane foam (SPF) is a type of rigid foam widely used as wall insulation in residential and commercial buildings. It is applied in-situ with a special blending machine and a spray gun. It has two components, component "A" as isocyanate and component "B" as polyol. They come together at the nozzle of a gun, which is then sprayed on the wall. It quickly expands to many times its original volume and solidifies in seconds, effectively sealing the wall. Although it sets in about an hour, full curing is reached after 24 hours. It adheres well and has the ability to reduce heat loss through walls, thus saving energy for home owners. It acts asan air and moisture barrier, and provides effective thermal insulation. Its performance is superior to commonly used fiber-glass batt insulation. It also contributes to the structural stability of the building, due to its superior adhesion properties. Many studies in USA indicated that the racking strength of building structures can be increased by applying SPF insulation.

Main raw materials used in the preparation of PU spray foam are derived from petro-chemical products. Efforts to replace them with alternative natural and

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sustainable resources have been accelerated during recent years. Polyols derived from vegetable oils are considered a potential replacement due to their attractive properties. As spray foam insulation is gaining popularity, enhancement of mechanical properties becomes an important consideration as they are usually weak. Many researchers have demonstrated that addition of fiber has contributed to the foam quality in flexible and rigid foaming, but little is known about introducing fiber in spray foam insulation. In this study, anattempt has been made to produce SPF from soy-based polyol and water as blowing agent with incorporated cellulose fiber in polymer matrix as reinforcement. The potential improvement of SPF properties and the effects and interaction of fiber in foaming process were investigated. The benefits would be to encourage the use of sustainable and green materials to increase safety and comfort in buildings.

1. OPEN-CELL AND CLOSED-CELL FOAM

SPF is either closed-cell or open-cell, referring to the internal structure of the foam. In closed-cell foamalmost all the bubbles remain closed and the gas that forms during the foaming process gets permanently trapped in the cells. In open-cell foam the bubbles are mostly open or rupturedduring the foaming process and the diffused gas could escape from the cells. The advantages of closed-cell foam with respect to open-cell foam include its strength, higher R-value, and greater resistance to the leakage of air or water vapor. The disadvantages are that it is denser, requires more material, and is, therefore, more expensive. The choice of foam is based on the requirements of

 $\begin{array}{lll} \text{Scheme I} & & \rightarrow \\ \text{R-NCO} + \text{R'-OH} & \rightarrow \\ \text{R-NCO} + \text{H}_2\text{O} & \rightarrow \\ \text{R-NH-CO-OH} & \rightarrow \\ \text{R-NCO} + \text{R-NH}_2 & \rightarrow \end{array}$

PU foam reaction is a unique process in which polymerization and foam blowing occurs simultaneously. Polymer structure forms rapidly in order to support the fragile foam, but not fast enough to burst the bubbles.

2.2 Secondary Reactions

Isocyanate reactive groups further react with urethane and urea groups to form allophanates and substituted biurets

Scheme II R-NH-CO-O-R'+R-NCO (Urethane) R-NH-CO-NH-R+R-NCO (Urea) performance or application specific characteristics such as strength, vapor control, available space and so forth. Open-cell SPF has an R-value around 3.3 per inch and typically weighs about 0.4-0.6 lb/cu. ft. and is usually used indoors. Closed-cell SPF has an R-value of around 6.0 per inch andtypically weighs about 1.5-2 lb/cu.ft. and is mainly used outdoors.

2. POLYURETHANE FOAM REACTIONS

The isocyanate group (-N=C=O) is an unsaturated and highly reactive group containing two cumulative double bonds. It can react with both electron acceptor and electron donor functional groups. The main groups reacting with isocyanate are hydroxyl, carboxyl and amino groups. Isocyanate undergoes two types of reactions in the foaming process - primary and secondary.

2.1 Primary Reactions

Primary reactions are fast and performed at lower temperatures compared to secondary reactions. At first, the isocyanate (-NCO) group reacts with the hydroxyl (-OH) functional group of polyol to form urethane (-NH-CO-O-). This reaction is an exothermic polymerization reaction. During this exothermic process, the isocyanate reacts with a blowing agent such as water to generate carbon dioxide, as per the reaction scheme I below, to cause the foam to rise during polymerization reaction. First isocyanate group(-NCO) reacts with water to generate Carbamic acid which is unstable and immediately decomposes, forming an amine and carbon dioxide. The amine (R-NH₂) reacts again with another isocyanate group to produce Urea.

R-NH-CO-O-R' (Urethane) R-NH-CO-OH (Carbamic Acid) R-NH₂+ CO₂ R-NH-CO-NH-R (Urea)

respectively^[2]. The secondary reactions of isocyanate leads to cross-linking, and the resulting polyurethane becomes more rigid. Thus temperature control during polyurethane synthesis is critical to control secondary reactions and thereby cross-linking. Allophanate linkages may or may not be reversible, depending on many factors such as the nature of catalyst or NCO/OH ratio as schematically shown below (Scheme II).

→ R-NH-CO-NR-CO-O-R'
(Allophanate)
→ R-NH-CO-NR-CO-NH-R
(Biuret)

3. HARD AND SOFT SEGMENTS

The structure of PU contains hard and soft segments which contribute to its rigidity and elastomeric properties, depending on the ratio of these segments in copolymers. In the early stages of polymerization, the reaction of polyol with isocyanate is slower than that of isocyanate with water due to secondary hydroxyls present in soy-based polyol. The generated urea from the later reaction is able to start phase separation (hard segments) from the liquid matrix due to its ability to form strong hydrogen bonds between urea linkages. As soon as the polyol-isocyanate reaction proceeds, the soft phase formation starts and urethane links bond the soft and hard segments together.

The hard segments are mainly crystalline, having urethane groups that are cross-linked through hydrogen bonding with urea and other urethane linkages^[3]. The soft segments are the fatty acid linear sections of polyol structure stretched in coil shape within the polymer profile, and provide elastomeric quality within the matrix network. While hard segments are highly crystalline, the soft segments are comparatively mobile. The presence of these two phases in the same molecule can alter the properties of flexible and rigid foaming. Upon mechanical deformation, the soft segments are stressed by uncoiling, and the hard phases become aligned in the stress direction. This reorientation of hard segments and powerful hydrogen bonding contributes to high strength, elongation, and tear resistance of the foam structure^[4, 5].

4. FIBER AS REINFORCEMENT

Polyurethanes are reinforced with synthetic or natural fibers to enhance their properties. Synthetic fibers such as nylon and glass fibers are made of petro-chemicals with a good elasticity and strength. As societybecomes more environmentally conscious, natural fibers such as wood and cellulose fibers replace synthetic fibers. Their benefits as reinforcement in composite materials are sustainability, low cost, biodegradability and low energy consumption. The effect of reinforcement depends on a variety of parameters, such as nature of fiber, aspect ratio and interaction of fiber with the polymer.

In recent years, a great deal of research has been dedicated to the use of cellulose fiber as reinforcement in polymers. The objectives of this study were toincorporate cellulose fiber in spray foaminsulation at low and high concentrations and investigate its impacts in foaming. It was the first attempt to introduce fiber in such foam insulation. The average length of cellulose fiber was 350 micron with aspect ratio of about 26 which provided the best compromise between viscosity increase and reinforcement.

5. EXPERIMENTAL

5.1 Raw Materials

Multi-functional soy-based polyol (bio-polyol-X500) was purchased from Cargill Industrial Oils & Lubricants, Chicago, USA. Its viscosity @ 25°C was 3200 cps with hydroxyl value 56 mg KOH/g. The polymeric diphenylmethane di-isocyanate (Robinate M) was donated by Huntsman PU Geismar, LA, USA having NCO content 31.5%, viscosity @ 25°C 190 cps, and functionality of 2.7. The catalysts I & II, as well as the surfactant, were donated by Air Products and Chemicals, Allentown, USA. Blowing agent used was distilled water. The cellulose fiber was purchased from Sigma-Aldrich having the average length of 350 μm .

5.2 Foam Preparation

Spray foam specimens were prepared in the laboratory by mixing soy-based polyol and other ingredients for 5 minutes. Then added the isocyanate and mix it foradditional 30 seconds. The mixture was poured into a mould for free rise. After an hour, the sample was removed from the mould and left for 48 hours for full curing under ambient temperature. In case of fiber foam (composite foam) cellulose fiber was embedded in foaming in 13 php, 26 php, and 40 php concentration. At first fiber was pre-mixed thoroughly with polyol for 20 minutes before other ingredients and isocyanate were added. Attempt was made to increase the concentration of cellulose fiber as much as possible to make the biofoam more environmentally friendly and to reduce the amount of raw materials. It may be mentioned that the quantity of all ingredients used in the formulation was expressed as per hundred grams of polyol (php).

5.3 Characterization

The bulk density of the foam was measured in accordance with ASTM-D1622 with sample size of 50 mm \times 50 mm \times 25 mm, and an average of six measurements was recorded. The density was calculated by dividing the weight of the sample by its volume. Water vapor transmission of foam was measured using a desiccator containing saturated calcium nitrate for maintaining a relative humidity of 50% at all times. A foam specimen of 25 mm thickness was sealed to the open mouth of an impermeable dish containing water and placed inside the desiccator under ambient temperature. The test dish mouth had an area of 126 mm × 126 mm, defined as the area of the specimen exposed to water vapor. Periodic weighing of dish assembly and the test method specified in ASTM-E96 determined the rate of water flow through the specimen. The cellular morphologies of the foam system samples were investigated by Hitachi S-2500 as well as Hitachi TM-1000 Table-top scanning electron microscopy (SEM). Samples were freeze-fractured in liquid nitrogen and coated with gold before observation. Image J software was used to determine cell sizes. Viscosityof polyol components was measured under normal temperature of 25 °C using Brookfield Viscosity Machine (BVT). The ingredients were polyol, catalysts, blowing agent, surfactant and the fiber(where applicable). The mixture was poured into a glass jar, placed under the viscometer, and the test performed for 30 seconds with spindle number 64 and running speed of 50 and 100 RPM, and then averaged. Fourier transform infrared (FTIR) spectra of the foam samples were recorded using Brucker infrared spectrometer to measure the concentration of functional groups in polymer structure. A total of 32 scans were taken with a resolution of 4 cm⁻¹ in frequency range of 4,000-400 cm⁻¹. The thermal resistance and R-value of the specimens was measured using Laser Comp FOX314 Heat Flow Meter instrument according to ASTM-C518. The compressive strength and modulus of the foam system was measured by INSTRON 3,367 equipment in accordance with ASTM-D1621. The crosshead speed was 2.5 mm/min, and cell load of 2 kN with sample dimensions of 50 mm × 50 mm × 25 mm. The force required for 10% deformation of the original thickness has been considered as the compressive strength of the foam.

6. RESULTS AND DISCUSSION

6.1 Effect of Viscosity During Foaming Process

Viscosity of a liquid is its resistance to flow. The presence of fiber in the polymer reduced the ease of flow and increased the viscosity of the matrix to a tarlike consistency. High viscosity affected the rising height and gel time of the foaming process. Gel time is the starting point of gel formation and production of urethane cross-links till the full expansion of the foam. Gel time increased with fiber concentration in foaming, suggesting that diffusion was reduced across the interfaces^[6]. The rising height is the height of a free-rise cellular plastic to achieve its ultimate expansion under certain conditions^[7]. The rising height of the foam samples were examined and recorded after each curing time. The results indicated that as the fiber content was increased, the rising height of the fiber foam system was reduced proportionately compared to the neat foam. This obviously led to the foam volume reduction as a whole. The viscosity increase offiber embedded polyol were measured by Brookfield viscometer and shown in Table 1.

Table 1
Impact of Cellulose Fiber Concentration as Reinforcement in Foam Insulation

Item	Neat foam	13 php cellulose foam	26 php cellulose foam	40 php cellulose foam	
Rising height, (cm)	15.5	15	13	11	
Reduction in volume (%)	0	3	13	28	
Gel time (second)	40	50	60	75	
Viscosity increase (%) (polyol component)	0	10	16	> 100	

The orientation, concentration and aspect ratio of fiber would affect the performance of the reinforced composite foam. Although longer fiber length would provide better reinforcement, the optimum fiber length was found to be around 400 micron in which to obtain reasonably good fiber dispersion without increasing the matrix viscosity significantly. The results showed that if the fiber concentration remained below 26 php, the viscosity increase would be about 16%, and it would still possible to spray it using conventional spray guns. Incorporation of fiber more than this amount requires technological advancement to the present spray foam system.

6.2 Bulk Density and Compressive Strength

Both neat and composite foam samples were prepared with the same formulation and analyzed to investigate the changes in density and compressive strength properties. They are illustrated in Figures 1-4 and presented in Table 2. Incorporation of cellulose fiber increased the bulk density of the composite foam significantly (up to 60%) compared to the neat foam. The effect of fibre on density is more pronounced for lower dosage of fibre, and this effect becomes practically insignificant with further increase in the fibre content beyond 26%. The initial increase in foam

density might be attributed to cell nucleation and heavier mass of the fiber. It is hypothesized that the presence of fiber led to the process of heterogeneous nucleation, producing smaller homogeneous cell sizes within cellular foam and thus increasing the cell density[8, 9, 10]. Surfaces promote nucleation because of wetting. Using Image J software, the average diameter of the cells in composite foam was observed to be smaller than the neat onesby up to 8%. Secondly, when fiber was embedded in foaming, although there was interaction on the surface of fiber with the polymer, fiber remained as a heavier solid material within the foam and couldcontribute to the density increase too[11]. As density was determined by mass over volume, the increase in foam mass was proportional to the fiber content. The fiber massalone was estimated to be ten times heavier than the neat foam mass.

It was further evidenced that addition of fiber led to the shrinkage of the composite foam compared to foam without fiber. The rise-height was reduced by 3% for 13 php, 13% for 26 php, and 28% for 40 php in the fiber foam system. It appeared that fiber influenced the molecular chain packing and mobility in the composite foam system.

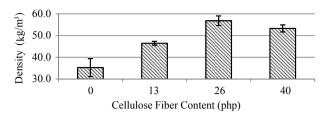


Figure 1
Effect of Fiber Content on Density of the Foam

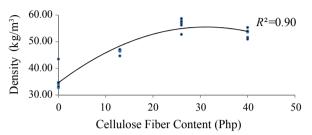


Figure 2
Polynomial Fitted Lineplot of Density as a Function of Fiber Content

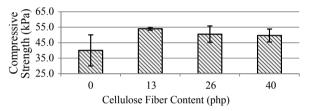
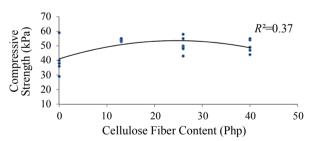


Figure 3
Effect of Fiber Content on Compressive Strength of the Foam



Polynomial Fitted Line Plot of Compressive Strength as a Function of Fiber Content

When a load is applied on a foam material, it will gradually be compressed, and the stress-strain curve experiences three regions of deformation-linear elasticity, plateau and densification^[12]. The behavior is linear elastic under 5% of strain and as the load is increased, the foam cell walls begin to collapse (plateau region), until the opposing walls in the cells meet one another to elevate stress rapidly (densification region). Under the load, the urethane soft segments stretch, while the urethane hard segmentscarry the load and release it when the stress is removed. Therefore, it is important to have sufficient amount of urethane linkages that contain both segments within the network structure of PU foam.

The compressive strength of the composite foam system showed improvement with incorporation of cellulose fiber. It can be explained by the fact that cellulose fiber containing a large number of hydroxyl groups could form hydrogen bonds or other chemical bonds with the matrix, contributing to the foam enhancement. There were three free hydroxyl groups available in each monomer of cellulose chain which could easily interact with isocyanate active groups at the interfaces, and increase cross-linking within the foam network. As illustrated in Figure 5a & 5b of the SEM images, fiber was well embedded in the cell wall and interacted firmly with the matrix. This suggested that high strength of cellulose fiber, along with efficient interaction between matrix and the fiber, enhanced the composite foam compressive strength. However, as fiber concentration was increased above 26 php, dispersion became a problem. The hydrophilic cellulose fiber was agglomerated and resulted in less number of available hydroxyl groups for fibre-matrix interactions.

Figure 6 represents FTIR spectra of neat (without fiber) and composite foams. The absorption peaks of urethane at 1,740 and urea at 1,660 indicated a good balance between the hard and soft segment production during foaming process. It seemed that for every urea link, an equal amount of urethane was generated to link them together and eliminate the phase separation^[13]. As a result a strong network structure was formed which could carry more load capacity.

Table 2
Mechanical and Thermal Properties of Open-Cell Spray PU Foam Insulation*

Foam type	Density (kg/m³)	Compressive strength (kPa)	Modulus of elasticity (kPa)	Water vapor transmission (g/h·m²)	Thermal resistance (K·m²/W)	R-value (US) (h·ft²·F/Btu)
Neat Foam	35 (±4)	40 (±10)	700	3.77	0.61	3.43
CF 13 php	46 (±1)	54 (±1)	908	2.15	0.65	3.69
CF 26 php	57 (±2)	50 (±5)	861	1.63	0.57	3.19
CF 40 php	54 (±2)	50 (±4)	846	1.30	N/A	N/A

Note. *Number of samples tested for water vapor transmission was 1 and for thermal resistance were 2.

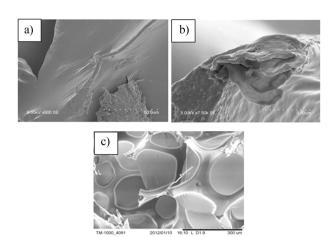


Figure 5 SEM Cellular Foam Images: a) and b) Fiber Compatibility, c) Free Fiber Detection

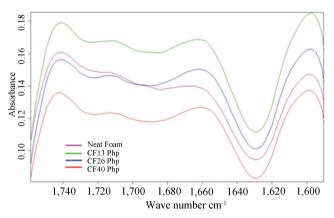


Figure 6 FTIR Spectrum Peaks of Urethane Carbonyl Groups at 1,740 cm⁻¹ and Urea Carbonyl Groups at 1,660 cm⁻¹

6.2.1 Statistical Analysis

Properties of bio foam insulation vary with fiber content. To establish a relationship between variable Y (property) and variable X (fiber content) and to assess the validity of that relationship, the regression analysis has been performed. The observed data were plotted with their best fitted lines. R^2 value for density of cellulose fiber was 0.90, which was statistically significant at 95% confidence level ($\alpha = .05$), representing inter-dependence between both variables. The regression fitted line indicated that density would increase with addition of fiber in foaming. On the other hand, the R^2 value of compressive strength was found to be 0.37 which was statistically insignificant and could not be explained by the model. However, the trend line showed that addition of cellulose fiber slightly improved the compressive strength of the composite foam system.

6.3 Thermal Resistance and R-Value

Thermal resistance of a material is its resistance to heat flow. Foam insulations are rated in terms of their resistance to heat flow or *R*-value. The higher the

resistance, the greater is the R-value and insulating effectiveness. The open-cell foam samples were analyzed by heat flow apparatus in terms of their heat conductivity and heat resistance, and their R-values determined. This is summarized in Table 2. Results revealed that thermal resistance and R-value of the composite foam system have been enhanced at 13 php fiber content, and then reduced at 26 php. Thermal resistance of the foam would be greatly enhanced if less air could get into the system and be replaced by the escaped carbon dioxide^[14]. The above observation could be explained by the fact that at lower concentration the fiber dispersed uniformly throughout the matrix and increased the adhesion between them, allowing morediffused gas to be trapped in the composite foam, enhancing its R-value. As fiber content increased in foaming, dispersion became more difficult, resulting in fiber agglomerations due to strong intermolecular hydrogen bonding between the fibers. This lack of fiber dispersion resulted in free passages throughout the matrixthrough which the diffused gas could escape quicker from the composite foam to the atmosphere^[15] and be replaced by air permeation into the foam. Since thermal resistance of air is lower than CO₂, the R-value of the composite foam at higher fiber concentration had not been improved. However, both thermal resistance and R-value measurements of the neat and composite foams were above the minimum requirements specified by CAN/ULC-S706-09.

6.4 Moisture Permeability

Open-cell spray polyurethane foam is an airtight foam insulation applied *in-situ* by a spray gun to walls of both residential and commercial buildings. Once it is cured, it acts as a good moisture barrier layer. However, condensation occurs when water vapor moves through the foam insulation from warmer to a cooler area and affects the thermal efficiency of the insulation. Therefore, moisture permeability becomes an important property of the foam insulation and can be measured by the rate of water vapor flowing throughthe foam material of unit area and thickness under particular temperature and humidity conditions^[16].

Water vapor transmission test was carried out to measure moisture permeability of the foam system. An apparatus was built and the permeability of water vapor through the foam material was recorded during different time intervals. It was observed that the passage of water vapor through the composite foam materials was significantly reduced compared to the neat one (Table 2), suggesting that cellulose fiber restricted the flow of water vapor. The moisture permeability was proportionally decreased with the increase in fiber concentration. Figure 7 illustrates slope of the lines representing rate of water vapor transmission for the foam system.

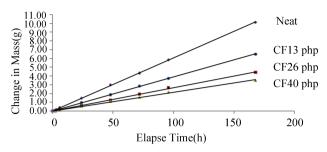


Figure 7
Rate of Water Vapor Transmission for the Foam System

This reduction in moisture permeability could be attributed to two factors. Firstly, the primary drawback of using cellulose fiber as reinforcement was its high moisture absorption because of hydrogen bonding betweenhydroxyl groups in fiber cell wall and water molecules. This led to fiber swelling or moisture build-up in fiber, and also in the fiber-matrix interface^[17]. As water vapors were passed through the composite foam, they were captured by the fiber, allowing less water vapor to pass through. Secondly, incorporation of fiber increased the cell density of the composite foam compared to the neat one as discussed earlier, and delayed the water vapor flow.

7. CHARACTERIZATION OF COMPOSITE FOAM SYSTEM

The knowledge of foam cellular structure aids in improving itsthermal and mechanical properties. Addition of fiber requires morphological analysis to observe the effects of fiber in foam insulation.

7.1 Morphological Characterization of Composite Foam

The fiber used in PU foam must havea good compatibility with the selected resin in order to provide a good reinforcement. Synthetic glass and nylon fibers are synthesized from petro-chemical substances and are mostly hydrophobic and strong, making them compatible with the resin and improving the strength of the matrix. But natural cellulose and wood fibers compatibility have always raised question amongst researchers. Almost all resins are hydrophobic and not compatible withhydrophilic character of natural fibers. This creates weakness in adhesion and wettability between the fiber and the polymer, and results in poor mechanical properties of the composite products.

Fiber size and dispersion of fiber within the matrix significantly affect the strength of the composite foam as well. A small fiber size with low aspect ratio holds a larger surface area which could be non-reactive to the matrix, creating more stress points and resulting in weak material strength. While long fibers provide more reinforcement than short fibers, the latter have less impact on the viscosity of the mixture. Thus a greater amount of

short fiber could be incorporated in the mixture at a given viscosity to achieve higher reinforcement. Short fibers also provide a better dispersion within the matrix than long fibers. The confocal microscopy image (Figure 8) shows thatalthough the fiber was distributed fairly within the composite foam, the dispersion was not uniform. The agglomeration of fiber (dark areas) could be clearly seen.

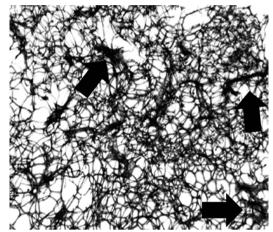


Figure 8
Confocal Microscopy Image of Fiber Dispersion

It is well known that hydrogen bonding contributes a great deal to the strength and modulus of PU foam network. The foam molecules react with cellulose fiber through the reaction between the isocyanate -NCO groups and the -H groups on the surface of the fibril and generate additional cross-links. This is a positive effect. However, it could also not participate at all in the reaction, interfering with the hydrogen bonding between urethane molecules, causing a negative effect. As observed in SEM image in Figure 5c, some fibers did not interact with the resin. The overall performance of composite foam depends on the competition between the positive effect of reinforcing polymer, and the negative effect on hydrogen bonding in foam structure. The morphological observations and enhancement in compressive strength proved thatthe positive effect dominated here, enhancing cross-linking within the fiber foam system network. This effect was also demonstrated by Xia Cao et al. [6], in trying to incorporate clay in foaming. This interference may be illustrated below in Figure 9.

Figure 9 Hydrogen Bonding Between Urea and Urethane Linkages

FTIR results (Figure 6) revealed another impact of cellulose fiber in foaming. The urea carbonyl group absorption of fiber foam was higher than that of no-fiber suggesting slightly more formation of urea linkages within the composite foam system network. It is likely that increased viscosity affected foaming process and retarded the polymerization reaction promoting the gel process. This is assumed that there is a good chemical interaction between the urethane and urea linkages to form a strong network matrix within the composite foam.

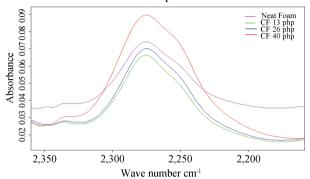


Figure 10 FTIR Spectra of Free-NCO Absorption Peaks at 2,275 cm⁻¹

The reaction between isocyanate active groups and the hydroxyl groups of cellulose fiber could also be demonstrated by FTIR spectra changes as shown in Figure 10. The free-NCO absorption peaksat 2,270 cm⁻¹ were lowered in composite foam with 13 php and 26 php fiber contents compared to the neat foam without fiber, indicating reactivity between the fiber and free-NCO groups. The unreacted isocyanateamount was increased at 40 php fiber concentration as viscosity of the matrix was elevated significantly and interfered with foaming process.

CONCLUSION

Spray foam insulation was produced using sustainable soy-based polyol and water as blowing agent. Cellulose fiber was incorporated in foaming up to 40 php in three stages and its impact was investigated in terms of mechanical and thermal properties as well as foam characterization. It was demonstrated that presence of fiber in foaming process increased the bulk density due to nucleation process and increased cell density. Good adhesion between resin and fiber at interfaces enhanced the compressive strength of the composite foam system. The interaction of fiber and polymer was efficient at lower fiber concentration and slightly improved the thermal resistance of the insulation foam, but at higher fiber concentration, agglomeration of fiber (due to its hydrophilic nature) provided open channels, allowing more diffused gas to escape from the material, leading to slightly reduced thermal resistance of the composite foam. Pre-treatment of fiber wouldhelp to separate fibers from each other, and eliminate hydrogen bonds holding them together. As fiber was embedded in the matrix, the viscosity was increased and affected the foaming process by delaying ease of flow. Although the impact was not significant, thegel time was increased and rising height was reduced. This led to slight volume reduction in composite foam systemas compared to the foam without fiber. The polyol viscosity was significantly increased with incorporation of fiber above 26 php.

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REFERENCES

- Zimmerman, R. L., Humbert, H. H., & Walker, M. (1997). Low-odour amine blowing catalysis in flexible polyether foam. Paper presented at the Seminar Organized by Rapra Technology Ltd., U.K.
- [2] Sekkar, V., Gopalakrishnan S., & Ambika Devi, K. (2003). Studies on allophanate-urethane networks based on hydroxyl terminated polybutadiene: Effect of isocyanate type on the network characteristic. *European Polymer Journal*, 39(6), 1281-1290.
- [3] Bagdi, K., Molnar, K., Sajo, I., & Pukanszky, B. (2011). Specific interactions, structure and properties in segmented polyurethane elastomers. eXpress Polymer Letters, 5(5), 417-427.
- [4] The Essential Chemical Industry Online. (2014, August 15). *Polyurethanes* [Online forum comment]. Retrieved from http://www.essentialchemicalindustry.org/polymers/polyurethane.html
- [5] Irle, C., Roschu, R., Luehmann, E., & Feng, S. (2002). Waterborne parquet coatings based on crosslinked polyurethane dispersions. *Paint & Coatings Industry*, 18(2), 28.
- [6] Szycher, M. (1999). Szycher's handbook of polyurethanes. New York, USA: CRC Press.
- [7] ASTM International. (2012). ASTM D883-12 Standard Terminology Relating to Plastics. West Conshohocken, USA: ASTM.
- [8] Xia, C., Lee, J. L., Widya, T., & Macosko, C. (2005). Polyurethane/clay nanocomposites foams: Processing, structure and properties. *Polymer*, 46, 775-783.
- [9] Ema, Y., Ikeya, M., & Okamoto, M. (2006). Foam processing and cellular structure of polylactide-based nanocomposites. *Polymer*, 47, 5350-5359.
- [10] Ting, L., Liang, M., Fuwei, L., Wuzhou, J., Zhaobo, H., & Pengfei, F. (2011). Preparation, structure, and properties of flexible polyurethane foams filled with fumed silica. *Journal of Natural Sciences*, 16(1), 29-32.

- [11] Gayathri, R., Vasanthakumari, R., & Padmanabhan, C. (2013). Sound absorption, thermal and mechanical behavior of polyurethane foam modified with nano silica, nano clay and crumb rubber fillers. *International Journal of Scientific & Engineering Research*, 4(5), 301-308.
- [12] Vladimir, G. (2010). Testing and application of new phenomenological material model for foam materials [Online forum comment]. Retrived from http://www.posterus.sk/?p=3923
- [13]Xiao, C., & Fan, Q. (2008). Effects of crosslinking density on structure and properties of interpenetrating polymer networks from polyurethane and nitroguar gum. *Polymer Composites*, 29(7), 758-767.
- [14]Federation of European Rigid Polyurethane Foam Association. (2006). *Thermal Insulation Materials Made of Rigid Polyurethane Foam* [Online forum comment]. Retrived from http://www.excellence-in-insulation.eu/site/fileadmin/user_upload/PDF/Thermal_insulation_materials_made of rigid polyurethane foam.pdf

- [15] Matuana, L. M., Park, C. B., & Balatinecz, J. J. (1998). Cell morphology and property relationships of microcellular foamed PVC/Wood-fiber composites. *Polym. Eng. Sci.*, 38(11), 1862-1872.
- [16] Mukhopadhyaya, P., Kumaran, K., Lackey, J., & Reenen, D. (2007). Water vapor transmission measurement and significance of corrections. *Journal of ASTM International*, 8(4), 1-12.
- [17]Bismarck, A., Aranberri-Askargorta, I., & Springer, J. (2002). Surface characterization of flax, hemp and cellulose fibers; surface properties and the water uptake behavior. *Polymer Composites*, 23(5), 872-894.