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1pSA5. Modeling the acoustic absorption behaviour of polylactide open cell foams with bimodal structure for high acoustic absorption

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Through this research, highly porous open cell foams were fabricated with Polylactide (PLA) and Polyethylene glycol (PEG) using compression molding combined with particulate leaching technique. Salt and water soluble polymer were used as the particulates. The pore size of the foam structure is controlled by the particulates size and higher interconnectivity is achieved by the co-continuous blending morphology of PLA matrix with water-soluble PEG. As a result of employing water soluble polymer into PLA foams, a bimodal cell structure was fabricated. Both PLA and PEG are fully bio-based polymers derived from renewable resources. Therefore, the resulting acoustic foams are benign and environmentally friendly. As a result of the secondary porous structure formed into cell walls by the water soluble polymer, the overall absorption of fabricated PLA foams was increased to above 90% while the average absorption of the foams remained unchanged. Fabricated foams were characterized for cellular, acoustic and mechanical properties. Open porosities as high as 88% was achieved through this study. Johnson-Champoux-Allard model was used to investigate the effect of bimodal structure on acoustic properties of PLA open cell foams. Foams fabricated through this study have the potential to replace the petrochemical based foams currently used in acoustic applications.

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INTRODUCTION

Polymeric open cell foams are commonly used in various industries as sound absorbers. Besides the need to improve the acoustic absorption capabilities of the existing foams, disposal of these foams is a huge environmental concern considering the massive amounts of foams consumed each year. Since the existing polymeric foams are fabricated from petroleum based polymers, in the case of cross-linked polymer chains, the resulting foams will be non-recyclable and will be landfilled after their end of life. On the other hand, the recyclable foams such as polypropylene foams are often too expensive to recycle and even without considering the carbon dioxide generation from recycling operation in an incineration plant, there is no economic interest in reusing these foams. The solution to these problems is to fabricate open cell foams from Bio-based polymers for noise control applications. Bio-based polymers are fabricated from renewable resources such as plants and microorganisms, these polymers break down to water, carbon dioxide and biomass in contact with heat and humidity.

Bio-based polymers have been emerging in recent years and are now industrially produced in large amounts. Considering the rising oil price, the price of bio-based polymers will be comparable to petrochemical based material and therefore commercially affordable in near future.

Polylactic acid (PLA) is a thermoplastic polyester synthesized by fermenting the raw plant material that has been fixed inside the living plants by photosynthesis. Since it is derived from plants, it can be produced using small amounts of fossil fuel. In recent years the interest in bio-based polymers has increased and PLA has found numerous applications in different industries. PLA possesses properties that lie between those of polystyrene (PS) and polyethylene terephthalate (PET) [1], and as a bio-based alternative it is expected to replace these commodity polymers. Its production cost is expected to decrease further as the markets for the material expand.

In our previous study [2], we compared acoustic performance of PLA open cell foams fabricated using salt leaching method, with Polypropylene (PP) and Polyurethane (PU) open cell foams. PLA foams showed better acoustic performance at lower frequencies than PP and PU foams, but the overall acoustic absorption of PLA foams needs to be improved. If these foams are to replace existing polymeric foams, there is a need to find strategies to improve the acoustic absorption of PLA foams while also studying the relation between cell morphology and acoustic properties of polymeric open cell foams.

One possible strategy is to make the foam structure more open and therefore less resistive to air flow by using water soluble polymers. [3] Water soluble polymers are widely used to make highly porous scaffolds for tissue engineering purposes. [4, 5] Pertinent methods generally use solvents, particulate leaching, co-continuous blends (water soluble polymers), gas foaming or some combination of these. Kramschuster et al. [6] used PLA and polyvinyl alcohol (PVOH) with salt as the particulate and gas foaming with carbon dioxide to fabricate bio-based scaffolds. Kim et al. [7] fabricated polymeric membrane with a fine porous structure from polystyrene (PS), polyethylene glycol (PEG), and solvent solutions by exploiting the phase separation induced in the course of dry casting.

Fabrication method used in the present study is particulate leaching which is a relatively new method for open cell foaming. This method involves the addition of an inhomogeneous domain into the polymer matrix at the beginning. Solid particles, such as sodium chloride and potassium chloride crystals, are introduced into the polymer matrix at the beginning of the process. These particles will later be dissolved out of the matrix by a solvent, thereby resulting in a porous cellular network throughout the entire polymer matrix [3]. This technology has commonly been used in the production of highly porous bio-scaffolds [8]. Recent research has tried to apply the particulate leaching technique to non-continuous batch foaming processes. The incorporation of this technique into the rotational foam molding process has also been investigated in the literature. [9, 10]

In the present study, low molecular weight Polyethylene glycol (PEG) was used to fabricate highly porous open cell foams for the first time to study the effect of bimodal structures on sound absorption of PLA foams. Particulate leaching method makes it possible to closely control cell structure of the foams by changing fabrication parameters and therefore it is suitable for investigating the relation between cell morphology and acoustic properties of open cell foams. Different weight percentages of water soluble polymer were examined for two different amounts of PLA. Johnson-Champoux-Allard model was applied to fabricated foam samples to simulate the acoustic absorption and investigate the effect of the bimodal structure on acoustic properties of open cell foams.

Theoretical Modeling of the Acoustic Performance of PLA Open Cell Foams

In order to optimize the acoustic performance of porous membranes, the relation between different structure properties and acoustic behavior must be studied. The fabrication method used in this study makes it possible to closely control cellular structure of the resulting foam. Due to the regular cell shape and less complicated cell geometry it is possible to develop a microstructure based model to link cell morphology and acoustic properties of these foams. Using such model will help to better understand acoustic performance and optimize the resulting foam.

As the first step in simulating acoustic absorption behavior of PLA open cell foams, Johnson-Champoux-Allard model was applied to the fabricated foams. Allard developed a five parameter model for rigid frame porous media, based on the work of Johnson [11, 12], that works well at low and high frequencies. When excited by acoustic waves, the frame of the majority of porous materials can be approximated as acoustically rigid over a wide range of frequencies. The porous material can then be replaced on the macroscopic scale by an equivalent fluid of effective density $\tilde{\rho}(\omega)$ and effective bulk modulus $\widetilde{K}(\omega)$ that occupies a proportion $\emptyset$ of the volume of the porous material. In the equivalent fluid model developed by Johnson-Champoux-Allard [13] employed in this work, the values of these two quantities are determined by five macroscopic quantities of the porous medium, which are: the airflow resistivity $\sigma\ \left(\frac{N.s}{m^4}\right)$; porosity $\emptyset(\%)$; tortuosity α_∞; and viscous and thermal characteristic lengths $\Lambda(\mu m)$ and $\Lambda'(\mu m)$. The porosity represents the percentage of interconnected void space with respect to the bulk volume. Furthermore, the geometry and interconnection of the open cells govern one of the most important porous material properties of all: the airflow resistivity σ ($\sigma = \eta / K_0$ where η and K_0 are the dynamic viscosity of the saturating fluid and the permeability, respectively). The tortuosity α_∞, as identified by Johnson et al. [14], describes the complexity of the path followed by the acoustical wave inside the skeleton network. In other words, the way the open pores are oriented and interconnected determines the tortuosity of the material. As the path followed by the acoustic wave inside a porous material becomes more complex, tortuosity increases which yields better sound absorption in the porous material. [13] The characteristic lengths Λ and Λ' represent the average macroscopic dimensions of the pores with respect to the viscous and thermal losses, respectively. The thermal characteristic length Λ' reflects the pores of larger size, where thermal transferring surface is significant. By contrast, the viscous characteristic length Λ reflects the importance of airflow in the pores and thus represents the smaller pores due to the high air particle velocity at these locations. In other words, Λ' stands for the average radius of the largest pores, while Λ represents the average radius of the smallest pores. The ratio Λ'/Λ is always higher or equal to 1. [13] Based on the work of Johnson et al., it can be shown that Λ' is generally larger than Λ if the flow is considered laminar. [14]

The Johnson-Champoux-Allard model correlates the effective density and effective bulk modulus of the medium to the five macroscopic quantities described above [13]:

$$\tilde{\rho}(\omega) = \rho_0\alpha_\infty\left[1 + \frac{\sigma\emptyset}{j\omega\rho_0\alpha_\infty}G_J(\omega)\right] \tag{1}$$

$$\widetilde{K}(\omega) = \frac{\gamma P_0}{\left[\gamma - (\gamma-1)\left(1 + \frac{H'}{2j\omega}\acute{G}_J(\omega)\right)^{-1}\right]} \tag{2}$$

where P_0 is the atmospheric pressure, ρ_0 is the density of air, γ is the adiabatic constant. The tilde ($\sim$) indicates that the associated variable is complex and frequency dependent. $G_j(\omega)$ and $\acute{G}_J(\omega)$ are the viscous and thermal correlation factors and defined as:

$$G_J(\omega) = \sqrt{1 + j\frac{\omega}{H}} \tag{3}$$

$$\acute{G}_J(\omega) = \sqrt{1 + j\frac{\omega}{H'}} \tag{4}$$

With

$$H = \frac{\sigma^2\Lambda^2\emptyset^2}{4\alpha_\infty^2\eta\rho_0} \tag{5}$$

$$H' = \frac{16\eta}{B^2\Lambda'^2\rho_0} \tag{6}$$

where B^2 is Prandtl number, η is viscosity of the air saturating the porous material.

The characteristic lengths (Λ and Λ') and the parameter σ' ($\acute{\sigma} = \acute{c}\sigma$ where $\acute{c}$ is a coefficient) in the above equations can be related to the porosity (ϕ), airflow resistivity (σ), and tortuosity (α_∞) of the material by [13]:

$$\Lambda = \frac{1}{c}\sqrt{\frac{8\alpha_\infty\eta}{\emptyset\sigma}} \tag{7}$$

$$\Lambda' = \frac{1}{c'}\sqrt{\frac{8\alpha_\infty\eta}{\emptyset\sigma}} = \sqrt{\frac{8\alpha_\infty\eta}{\emptyset\acute{\sigma}}} \tag{8}$$

where c and c' are numerical coefficients related to the geometry of the pore section.

The viscous characteristic length (Λ) represents the dimension of the constriction sections in the pore network where viscous dissipation of sound energy is dominant. Similarly, the thermal characteristic length (Λ') corresponds to the dimension of the regions with larger surface areas within the pore network where thermal dissipation of sound energy is highly in favor. On the basis of this definition, Λ' will be larger than or equal to Λ. Hence, the value of c' will be less than or equal to c depending on the geometry of the pore network. [13]

When the porous medium is backed by an impervious wall and is subjected to acoustic wave excitation, with the characteristic impedance and wave number evaluated, the surface impedance (Z_s) at the air and porous material interface can be calculated:

$$Z_s = -j\frac{Z_c}{\emptyset}\cot(kd) \tag{9}$$

where Z_c is the characteristic impedance, k is the complex wave number and d is the thickness of the porous material. The characteristic impedance and wave-number can be related to the effective density, effective bulk modulus, and angular frequency (ω) of the sound waves [13]:

$$Z_c = \sqrt{\tilde{K}(\omega).\tilde{\rho}(\omega)} \tag{10}$$

$$k = \omega.\sqrt{\frac{\tilde{\rho}(\omega)}{\tilde{K}(\omega)}} \tag{11}$$

The reflection coefficient (R) as well as the absorption coefficient (α) of the material can then be evaluated using the following equations [13]:

$$R = \frac{Z_s - \rho_0 c_0}{Z_s + \rho_0 c_0} \tag{12}$$

$$\alpha = 1 - |R|^2 \tag{13}$$

Obviously cellular structure of any porous material (i.e., cell morphology, cell wall thickness, pore size, etc.) affects the macrostructure of the material. Therefore, the five macroscopic properties outlined by the Johnson-Champoux-Allard model, which are all responsible for the acoustic performance of a given material, are linked to the inner microstructure of the porous network. As sound propagates through a porous material, acoustical energy is dissipated by frictional effects at the cell walls, heat transfer through the skeleton, and the conversion of acoustical energy into internal energy. [13] The microstructure of a fabricated porous membrane is determined by the processing parameters during fabrication. Therefore, to optimize material's sound absorption effectiveness, the fabrication parameters that regulate the development of microstructure should be controlled appropriately. In order to do so, more research is needed to understand the relation between cell morphology and acoustic properties such as sound absorption coefficient. In the present study, Johnson-Champoux-Allard model will be applied to fabricated PLA foam samples with emphasis of rigid frame.

Experiment

Material Selection

Polylactic acid or polylactide (PLA) is a bio-based thermoplastic polymer produced from renewable resources, such as corn starch or sugarcanes. The PLA powder used in this study was provided by ICO Polymers. The powder has spherical particles which are 20 μm in diameter. The density of PLA is 1.25 g/cm^3 and its melting point is around 175°C.

Polyethylene Glycol (PEG) is a biodegradable water soluble polymer. PEG was manufactured by Wako Pure Chemical Industries, Ltd. Melt temperature of PEG is around 30°C and the molecular weight is approximately 2000. PEG was provided in the form of platelets and was grinded by mortar and pestle. PEG powder was then sieved to less than 250 μm particle size.Salt used in this study was commercially available sodium chloride salt. The salt particles have cubic shape and were sieved into 250–500 μm.

Fabrication Method

In this research, compression molding technique along with particulate leaching method was used in order to fabricate open-cell foams. This fabrication process consists of four main stages, dry blending salt and polymer, compression molding, leaching in water and drying the sample in oven to create an open cell material.

In order to study the effect of water soluble polymer on cellular structure and consequently the acoustic performance of the resulting foams, different amounts of water soluble polymer were compared while keeping the weight percent of the PLA constant. For PLA containing 10 percent of the total weight of the material, PEG to PLA weight ratios of 0, 0.1, 0.2 and 0.3 were examined. It was not possible to increase the amount of PEG to more than 30% of PLA weight due to lack of mechanical strength in the resulting foam.

Results and Discussion

Cellular Properties

Figure 1 shows the cell morphology of PLA foam samples with different amount of water soluble polymer. As seen in these images, by increasing the amount of PEG, the cracks in the cell structure increases. PEG melts during the molding process and blends with the melted PLA. Therefore, leaching out the PEG leaves micro pores and cracks inside the PLA matrix. By increasing the magnification of the SEM images, the cracks and micropores formed in the cell walls can be seen clearly. Figure 2 shows the SEM micrographs of the PLA foam samples with increased magnification. As illustrated by these images, as a result of increasing the amount of PEG, number of cracks and micropores increases and a secondary porous structure forms on the cell walls. Therefore, the fabricated PLA foams have a bimodal cell structure.

Cellular properties of samples such as cell density and average cell size were estimated from SEM images. The foam's cellular structure is created by dissolving the leachable particles, therefore the polymer mass ratio directly affects the number of cells and consequently the cell density increases. Average cell size on the other hand, is independent from polymer weight ratio and is close to average salt particle size respectively. Adding PEG doesn't show any considerable effect on cell density. Cell density of PLA foams was approximately 20×10^4 $cell/cm^3$. Average cell size for all foam samples was in the range of 350 μm.

Porosity of foam samples was measured by a gas pycnometer (Quantachrome Instrument UltraFoam 1000) in accordance with the ASTM D6226 standard. Increasing the PEG content doesn't affect porosity of PLA foams and average porosity was around 88%.

Acoustic Properties

The static airflow resistivity was measured according to the ASTM C522 standard. Results for static airflow resistivity at 0.1 Lit/min flow rate are shown in Figure 3. Adding the water soluble polymer decreases flow resistivity of PLA foams. Flow resistivity further decreases by increasing the PEG content to 20%. On the other hand, flow resistivity shows an increase for PEG/PLA weight ratio of 0.3. This observation can be explained by the fact that by increasing the amount of water soluble polymer, structural strength of the PLA foam decreases. As a result of weak structure, more walls break into cells which increases flow resistivity and is expected to have a negative effect on sound absorption as well.

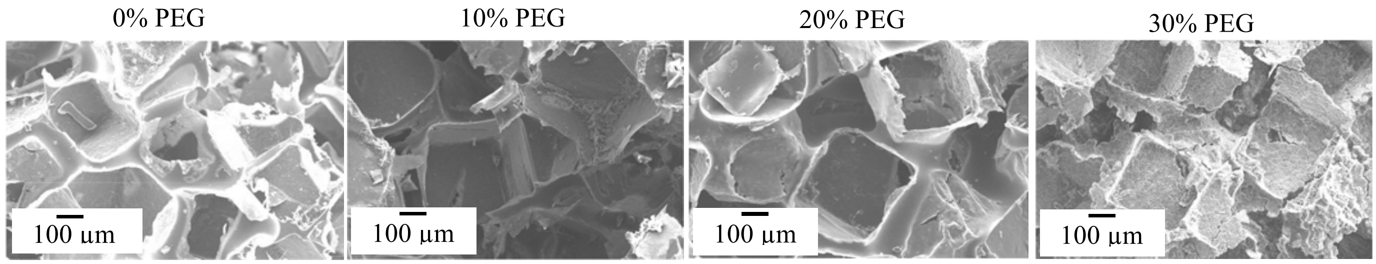

Figure 1: SEM micrographs of PLA foams with different PEG/PLA weight ratios.

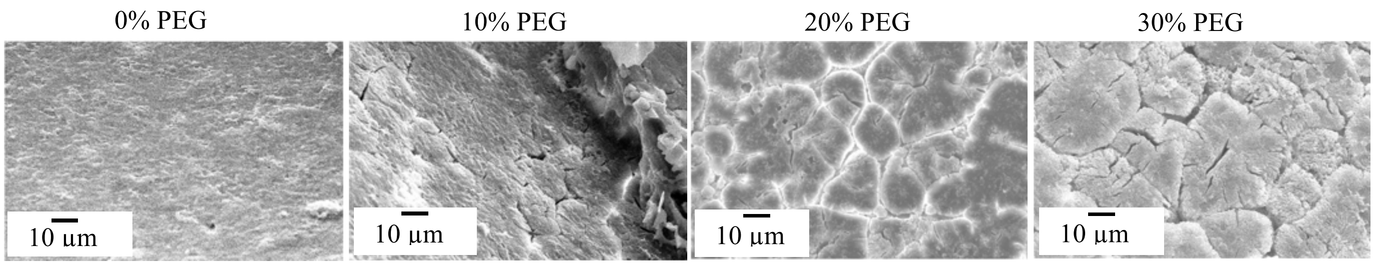

Figure 2: SEM micrographs of PLA foams with different PEG/PLA weight ratios.

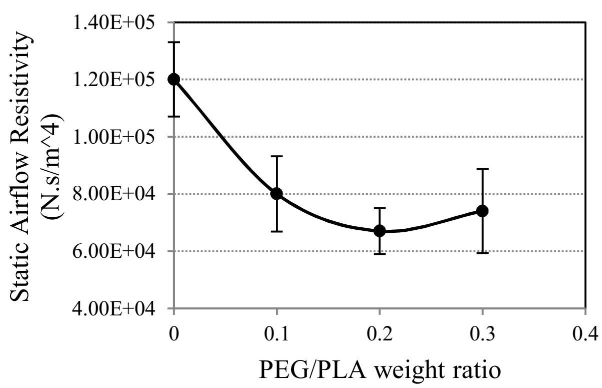

Figure 3: Static airflow resistivity for PLA foams with different PEG/PLA weight ratios.

The normal incident absorption coefficient was measured by impedance tube based on two microphone transfer function method and in accordance with the ASTM E1050 standard. The diameter of impedance tube is 29 mm with frequency range of 800-6300 Hz. Reported results are averaged and samples were sealed by Teflon tape to completely fit the impedance tube.

Figure 4 shows the acoustic absorption results of PLA foams with PEG/PLA weight ratios of 0, 0.1, 0.2 and 0.3 (or 0, 10%, 20% and 30% PEG). Adding water soluble polymer to PLA foams increases the maximum acoustic absorption coefficient and also increases the frequency of maximum acoustic absorption. This can be the result of the decrease in flow resistivity by making cell walls more open. Although the porosity is constant, acoustic absorption of the PLA foams improves as the amount of water soluble polymer is increased.

Absorption curve of foams with PEG/PLA weight ratio equal to 0.3 decreases compared to less amounts of PEG. It was also observed that the airflow resistivity was increased for this foam sample. This observation can be explained by the weaker foam structure as the amount of water soluble polymer increases. In other words, cell structure is very weak and brittle which results in deformed cell walls, higher flow resistivity and consequently lower acoustic absorption.

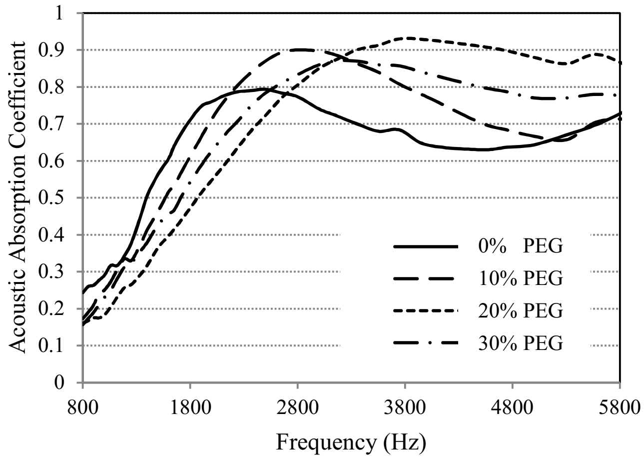

Figure 4: Sound absorption coefficient versus frequency for PLA foams with different PEG contents.

Theoretical Prediction of Acoustic Properties

Since analytical models are developed based on isotropic structures, to be able to apply analytical methods to a porous structure to predict the acoustic behavior, the structure must be homogeneous. Fabricated foam samples were tested in impedance tube for homogeneity and were found to be rather homogeneous. Acoustic results obtained from direct measurement for PLA foams are compared with acoustic inverse characterization in figure 5. As shown in these figures, the Johnson-Champoux-Allard model accurately predicts the absorption behavior of PLA open cell foams based on the characterizations.

Analytically estimated values for tortuosity, viscous and thermal lengths are listed in table 1. The value of tortuosity remains relatively constant by increasing the water soluble content. The secondly porous structure formed by water soluble polymer is not significant enough to affect tortuosity.

The values for thermal characteristic length increase by increasing the PEG content. This can be explained by increased surface area in the cell structure due to the cracks formed by water soluble polymer. Viscous characteristic length doesn't show any noticeable change for different amounts of water soluble polymer. Values of thermal characteristic length are generally larger than viscous characteristic length (Λ) as expected.

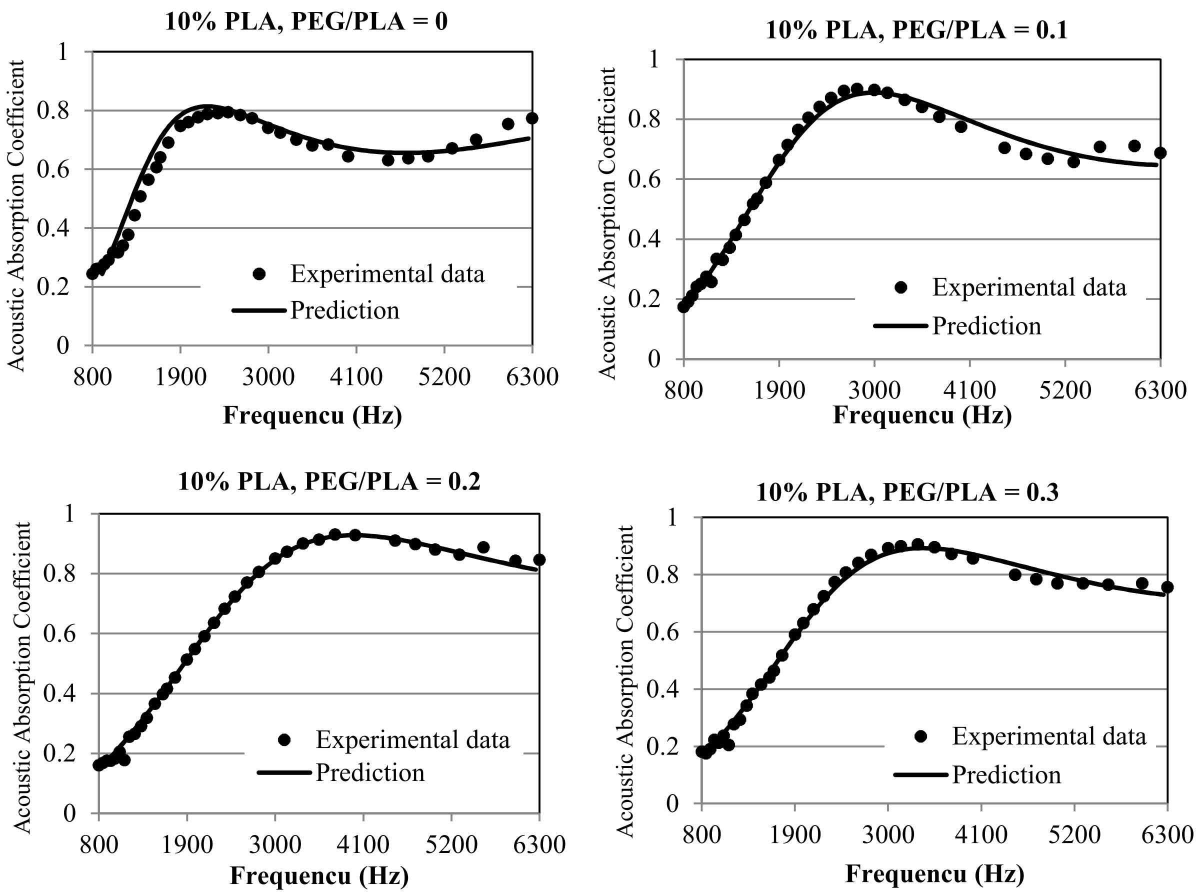

Figure 5. Acoustic modeling results of PLA foams with different PEG/PLA weight ratios. (Thickness: 10 mm)

Table 1: Analytically estimated acoustic properties of PLA foams with different PEG content.

Foam material	Tortuosity α_∞	Viscous length Λ (μm)	Thermal length Λ' (μm)
PEG/PLA=0	1.24 (±0.38)	5.9 (±1.5)	48.2 (±64.5)
PEG/PLA=0.1	1.23 (±0.15)	11.6 (±1.5)	140.1 (±60.9)
PEG/PLA=0.2	1.2 (±0.05)	11.8 (±2.5)	166.7 (±17.7)
PEG/PLA=0.3	1.22 (±0.25)	9.8 (±2.2)	164.6 (±33.3)

Conclusions

In this study, particulate leaching combined with compression molding was used to fabricate open cell foams from PLA. Water soluble polymer was used to form a secondary porous structure in the foam's cellular structure to study its effect on acoustic behavior of PLA foams. PEG/PLA weight ratios of 0, 0.1, 0.2 and 0.3 were examined.

Adding water soluble polymer decreasing the static air flow resistivity by forming cracks and micro pores in the cellular structure, and therefore, making a bimodal structure which in turn improves acoustic absorption of the PLA foam. Mechanical strength of the PLA foams on the other hand decrease as a result of adding water soluble polymer.

The fabrication method used in this study makes it possible to closely control cellular structure of the resulting foam. Due to the regular cell shape and less complicated cell geometry it is possible to develop a microstructure based model to link cell morphology and acoustic properties of these foams. Using such model will help to better understand acoustic performance and optimize the resulting foam.

Johnson-Champoux-Allard model was used to simulate the acoustic absorption of the fabricated PLA foams based on the characterization results. Predicted and experimental values of acoustic absorption are in good agreement and the Johnson-Champoux-Allard model accurately calculates the sound absorption behavior of PLA open cell foams.

It was observed that the bimodal structure formed by PEG, doesn't influence porosity, tortuosity or viscous characteristic length of the PLA foams while air flow resistivity and thermal characteristic length were affected by the formation of the secondary porous structure inside the primary foam matrix.

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