COB-2021-0946
NEW BIO-BASED POLYURETHANE FOAM WITH ALUMINA TRIHYDRATE: SOUND TRANSMISSION LOSS PROPERTIES

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Abstract. Rigid polyurethane foams (PUFs) are among the most consumed polymeric materials worldwide. They show a varied range of applications, such as insulation, furniture and the automobile industry. The main concerns regarding PUF application are its high flammability and oil-based raw materials. Those are the reasons for the recent increasing demand over new bio-based PUFs and mechanisms for decreasing their flammability without harming their properties and ecological friendly appeal. Aluminum hydroxide (ATH) is a halogen-free non-toxic additive flame retardant that perfectly suits the needs. Therefore, in this study, a bio-based PUF was synthesized with a 1.2/1 (isocyanate/green-polyol) ratio with the addition of 10%, 20%, 30%, 40%, and 50% of ATH mass fraction to evaluate the different responses in the sound transmission loss (STL) as ATH was added inside the PUF matrix. The STL tests were carried out in two impedance tubes manufactured by Brüel & Kjær, model 4206-T. These two tubes follow the ASTM E2611 standard, which defines the methodology for STL tests in frequencies before and after the resonance region. Therefore, the test required specimens with two different diameters, 100 mm and 29 mm, to fit the low and high-frequency tubes respectively. The specimens were polymerized inside a mold designed specifically for these tests, therefore the samples showed good dimensional standardization. The STL test results showed an increasing number for the stiffness sensitive region (100 mm tube) as the amount of ATH increased, going from 34.7±1.6 dB and 14.5±4.5 dB at 500 Hz and 1000 Hz respectively, for the pure PUF, up to 41.9±0.4 dB and 25.0±1.1 for the 50% ATH added foam. This result was acceptable since the stiffness of the polymer should increase with the addition of the ceramic flame retardant, moreover, the amount of ATH that exhibited these numbers is high enough to gain on good fire extinguishment properties. At higher frequencies (29 mm tube), where the STL numbers are sensitive to mass, the samples also showed better results as the amount of ATH increased, going from 58.7±1.6 dB and 47.3±2.1 dB at 1000 Hz and 5000 Hz, respectively, for the pure PUF, up to 61.9±1.2 dB and 50.0±2.4 for the 50% ATH added foam at the same frequencies. Therefore, the addition of ATH would increase the flame resistance of the polymeric foam with a decrease in the intensity of the sound waves crossing the material.

Keywords: composite materials, cellular materials, polyurethane foam, sound transmission loss.

1. INTRODUCTION

Polyurethane foams (PUFs) are widely spread among different applications, such as furniture, automobile, aerospace industries, biomedical industry, sandwich structures, and building constructions (Choe *et al.,* 2020). This vast number of application fields can be associated with some of the PUF's most valuable characteristics, such as lightweight, inexpensive cost, excellent thermal and acoustic insulation, and simple manufacturability (Mohammadpour and Sadeghi, 2020).

PUFs are defined as solid polyurethane (PU) filled with gases. Therefore, it can be separated into two parts, the first one being the PU, which makes the structure of the foam and is the main responsible for the material's mechanical properties. The second part is constituted by the gases that are stuck inside the PU in the foaming reaction process. The gases are mainly responsible for the PUF's excellent thermal and acoustic insulation properties (Szycher, 2013).

The PU structure often called cell walls, is made by macromolecules built of urethane linkages, which are formed based on the reaction of isocyanate groups with hydroxyl groups (Pillai *et al.*, 2016). The reaction between the isocyanate and a foaming agent (water for example) releases carbon dioxide gas. The carbon dioxide gas gets trapped inside the PU as the mixture viscosity increases. Thus, the gas inside the PU is responsible for cell creation inside the polymer. These cells can be categorized as open or closed cells according to their structure (Wan *et al.*, 2006).

The most significant properties of the PUF are highly dependent on its density. Higher densities provide better mechanical properties. Also, they provide poor insulation properties than foams with smaller densities. This follows the assumption that the higher is the foam density, the smaller is the amount of gas inside the material.

Furthermore, in several applications, the main undesirable characteristic of the PUF is its high flammability (Liu *et al.*, 2021). Therefore, the addition of a flame retardant should help to reduce this issue (Kairytė *et al.*, 2018). Another important undesirable characteristic of the PUF is its oil-based raw materials. However, since the demand for eco-friendly materials has grown in the past years, green substitutes for the PUF raw materials have been developed.

Alumina trihydrate or aluminum hydroxide (ATH) is an additive halogen-free flame retardant, often used in polymeric foams (Marques *et al.*, 2018), and it is responsible for half of all the flame retardants used around the world (Gooch, 2007). It presents three main flame retardancy mechanisms, an endothermic reaction that produces aluminum oxide and water, the water dilution in the air, and the creation of an insulation layer of aluminum oxide that protects the foam. The mechanisms' contribution to the flame retardancy are listed respectively (Kind and Hull, 2012). Silva *et al.* (2021) showed that 40% of ATH mass fraction added into foam is enough to provide a good flammability resistance to a green PUF.

To evaluate how much sound crosses a material, the sound transmission loss (STL) test is an easy and effective analysis that shows the decay in decibels (dB) of the sound intensity after it goes through a material. Since one of the main application fields of the PUF is the insulation industry, the evaluation of acoustic properties of a new green foam is essential to assure that it can replace the petroleum-based foam properly (Berger *et al.*, 2003).

Therefore, the aim of this work is to evaluate the changes in STL shown by the PUF when different mass fractions of ATH was added into it as flame retardant.

2. MATERIALS AND METHODS

The bio-based raw materials (KT1106 – diisocyanate/KT1106-R – polyol) were kindly supplied by Kehl Company. The ATH was generously offered by Alcoa Corporation. Thus, more details about the materials can be obtained directly from their cited companies.

In order to synthesize the PUF with the addition of ATH, it is mandatory to homogenize the polyol and the ATH first, due to the strong reaction between the isocyanate and the hydroxyl groups present in the ATH. Therefore, the isocyanate is poured into the mixture after the first polyol and ATH homogenization. Then, a second homogenization takes place. After the second homogenization, the mixture was poured inside a mold. The mold was designed to keep the desired shape and mass in the specimens. Thus, controlling their density and assuring their accordance to the test standard.

Figure 1 (a) shows the first mixture of polyol and ATH, (b) shows the moment after the second homogenization where it is possible to see that the foaming reaction has already started, and (c) shows the closed mold.

Figure 1. (a) polyol+ATH, (b) poliol+ATH+isocyanate, (c) 29 mm mold for STL samples.

Visual inspection is done through the sample looking for white stains as a first indication of the quality of the homogenization. If any sign of a non-uniform homogenization and density standardization was observed, the samples were discarded. Figure 2 shows a flowchart that illustrate the PUF manufacture process.
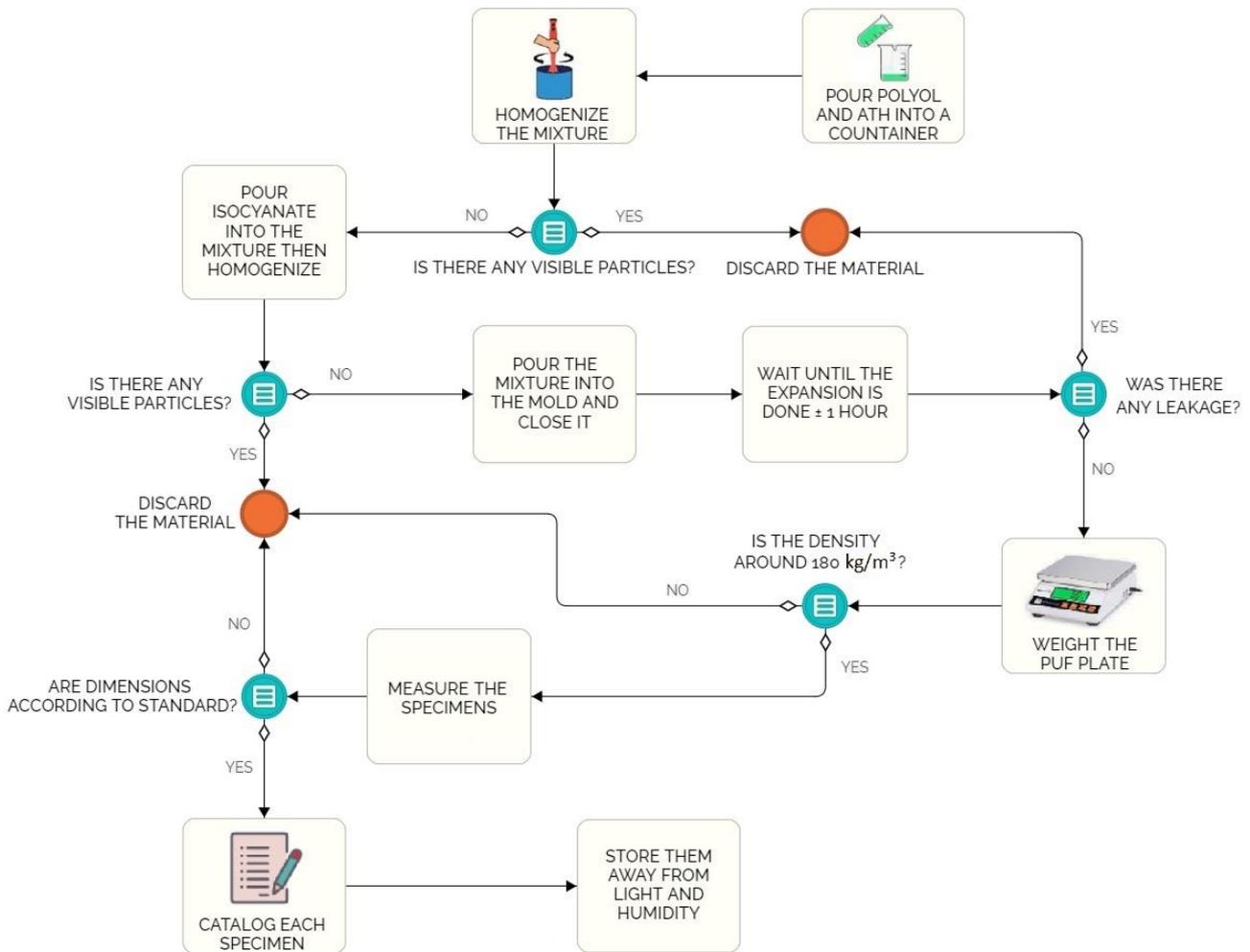

Figure 2. PUF manufacturing process.

The density chosen for the pure PUF was 180 kg/m³, which was the smallest density that showed no superficial visible pores to the naked eye. The foam densities with ATH increased as higher was the amount of ATH added into the mixture, although always keeping the same amount of polyol and isocyanate. Table 1 shows the raw material masses added into the mixture when synthesizing the foam, as well as the amount of mass poured into the mold.

Table 1. Raw material masses used in the PUF synthesis and the mass of mixture poured into the molds.

Material	Polyol (g)	Isocyanate (g)	ATH (g)	29 mm mold (g)	100 mm mold (g)
PUF	8.3	10	0	1.2	14.1
PUF+10%ATH	8.3	10	1.8	1.3	15.6
PUF+20%ATH	8.3	10	3.7	1.4	17.0
PUF+30%ATH	8.3	10	5.5	1.5	18.4
PUF+40%ATH	8.3	10	7.3	1.7	19.8
PUF+50%ATH	8.3	10	9.2	1.8	21.2

According to the mass of polyol and isocyanate used in the synthesis process, the mass of ATH was set. Three test specimens for each percentage (0%, 10%, 20%, 30%, 40%, and 50%) of ATH were manufactured.

The STL tests were made in two impedance tubes, models 4206-T, manufactured by Brüel & Kjær. The tubes follow the ASTM E2611-19 standard, which defines the methodology and frequency limits for sound transmission loss tests. The tests were carried out in the Vibrations and Lightweight Structures Laboratory (LaVEL) of the Santa Catarina State University (UDESC). According to the standard, the tubes present four microphones and a frequency measurement digital

analysis system to detect the acoustic transfer matrix. The Brüel & Kjær equipment possesses a sound source (speaker) on one side of the impedance tube. The test subject is placed in a sample holder in the center of the tube. The sound waves hit the test sample and part of them are reflected, part of them is absorbed by the material, and the other part passes through the sample. The sound pressure is measured in four regions, two before the specimen and two after it. As shown in Figure 3, the sound wave pressures A (incident) and B (reflected) are measured by microphones 1 and 2 and the sound wave pressures C (passing through) and D (reflected by the termination) are measured by the microphones 3 and 4. As it is also shown by this image, the microphone positions (x_1, x_2, x_3, and x_4) must be fixed and the specimen thickness (d) must be measured.
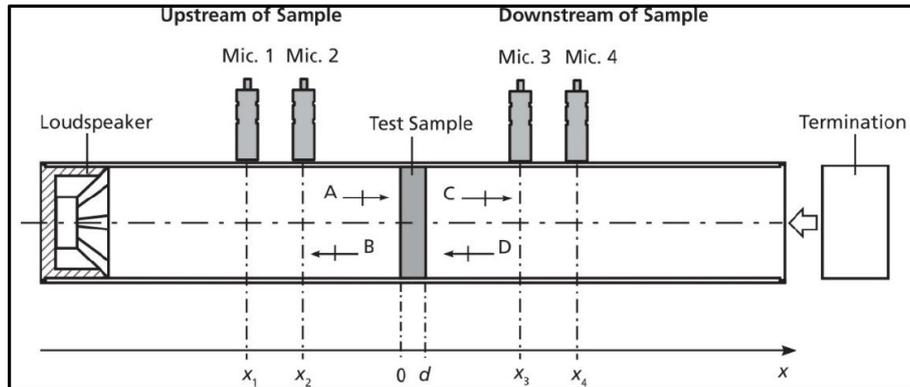

Figure 3. STL test schematic procedure.

Also, according to the ASTM E2611-19, the inner diameter (D) of the impedance tube depends on the maximum sound frequency (f_u) hitting the material. Eq. (1) defines the maximum diameter for a specific value of f_u as

$$D = 1000 \frac{K * c}{f_u} \ , \tag{1}$$

where K is a constant equal to 0.586 and c is the speed of sound in the air, which is equal to 343.24 m/s at room temperature of 20 °C. Therefore, the Brüel & Kjær tubes are in accordance to the standard, since the first tube with a diameter of 100 mm is applied for smaller frequencies (up to 1600 Hz) and the second tube, with a diameter of 29 mm, is applied for frequencies up to 6400 Hz.

3. RESULTS

According to Chandra *et al*. (2014), the interpretation of the STL-frequency curve can be divided into three parts. The first one is the stiffness sensitive part, where the higher is the stiffness of the material, the higher will be the value of the STL. The second part is a damping sensitive part and the third one is the mass-controlled region.

The stiffness-sensitive part corresponds to the portion of the curve from zero to the first resonance frequency, moreover, the damping-controlled region corresponds to a narrowband frequency, close to resonance frequencies, whereas the portion of the curve after the first resonance frequency corresponds to the mass-controlled region. In this region, the STL curve is proportional to mass (Chandra *et al*., 2014).

Therefore, the impedance tubes used in this work can provide STL data for before (100 mm tube) and after (29 mm tube) the first resonance region up to 6400 Hz. Figure 4 shows the results for three specimens of pure PUF in the (a) 100 mm tube and (b) 29 mm tube. It is quite noticeable that the manufacturing process presented a good standardization of the specimens where all three present similar STL-frequency curves for both impedance tubes. Moreover, according to Berger *et al*. (2003), STL values for a 100 mm concrete wall show similar STL values to the PUF investigated in this study. For 250 Hz and 2000 Hz, the concrete STL is 35 dB and 44 dB, respectively, whereas the PUF is 38.5±1.7 dB and 58.7±1.4 Hz. It is worth mentioning that the foam is much lighter and thinner than the concrete mentioned by Berger. Therefore, 10 mm of PUF would stop the same amount, or more, of sound pressure as 100 mm of concrete in most of the frequencies.
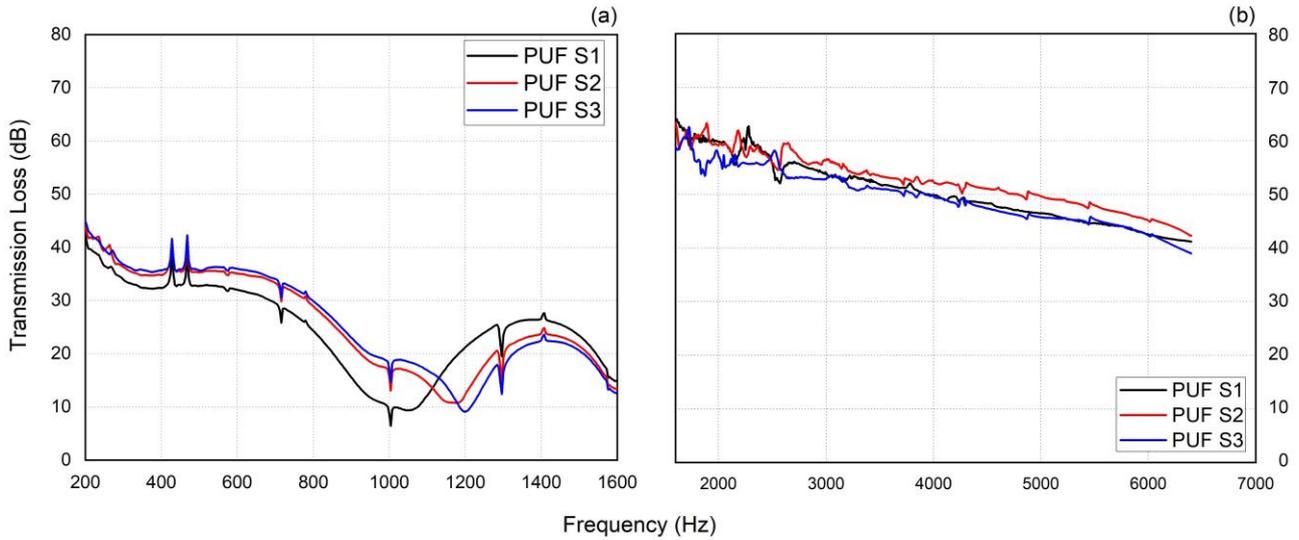
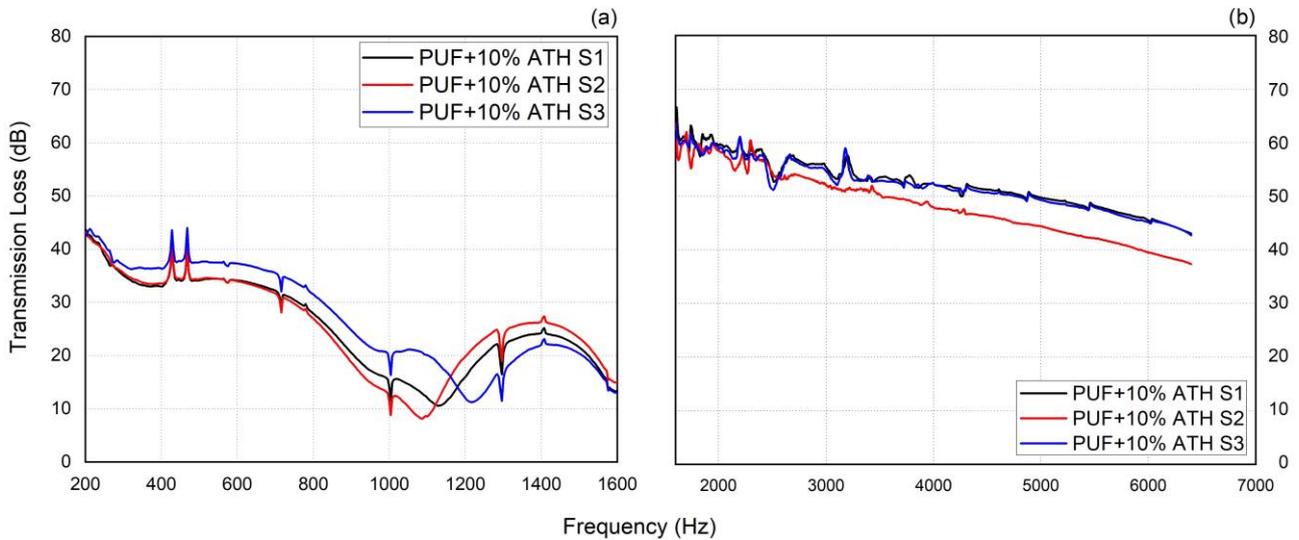Figure 4. Pure PUF STL results for the (a) 100 mm and (b) 29 mm tube.

Figure 5 shows the STL results for the PUF+10%ATH in the (a) 100 mm tube and (b) 29 mm tube. There was no noteworthy difference between the pure PUF and the PUF+10%ATH in both stiffness and mass-sensitive regions. For example, at 400 Hz, the PUF and PUF+10%ATH STL values are 34.3±1.7 dB and 34.3±1.8 dB, respectively. This is understandable since a small addition of ATH is not able to drastically change the stiffness and the mass of the foam (Silva *et al.*, 2021).

Figure 5. PUF+10%ATH STL results for the (a) 100 mm and (b) 29 mm tube.

Figure 6 exhibits the STL results for the PUF+20%ATH in the (a) 100 mm tube and (b) 29 mm tube. With the increasing amount of ATH, it is possible to assume that the stiffness of the foam shall increase with it. This higher stiffness should increase the value of STL in the stiffness-sensitive region. As shown by the PUF+20%ATH S2, there are higher values for STL in frequencies up to 1200 Hz, which is probably near a dumping sensitive strip region. For example, at 600 Hz, the PUF+20%ATH S1 and PUF+20%ATH S2 STL values are 36.0 dB and 39.8 dB, respectively. Therefore, the 20% added ATH samples showed higher mean values of STL than the ones with lesser amounts of ATH in the stiffness-sensitive region. There was no explicit difference among the PUF+20%ATH and the prior ones regarding the mass-controlled region, as the mean values for the pure PUF and the PUF+20%ATH at 3000 Hz are 54.3±1.9 dB and 56.9±2.5 dB, respectively.
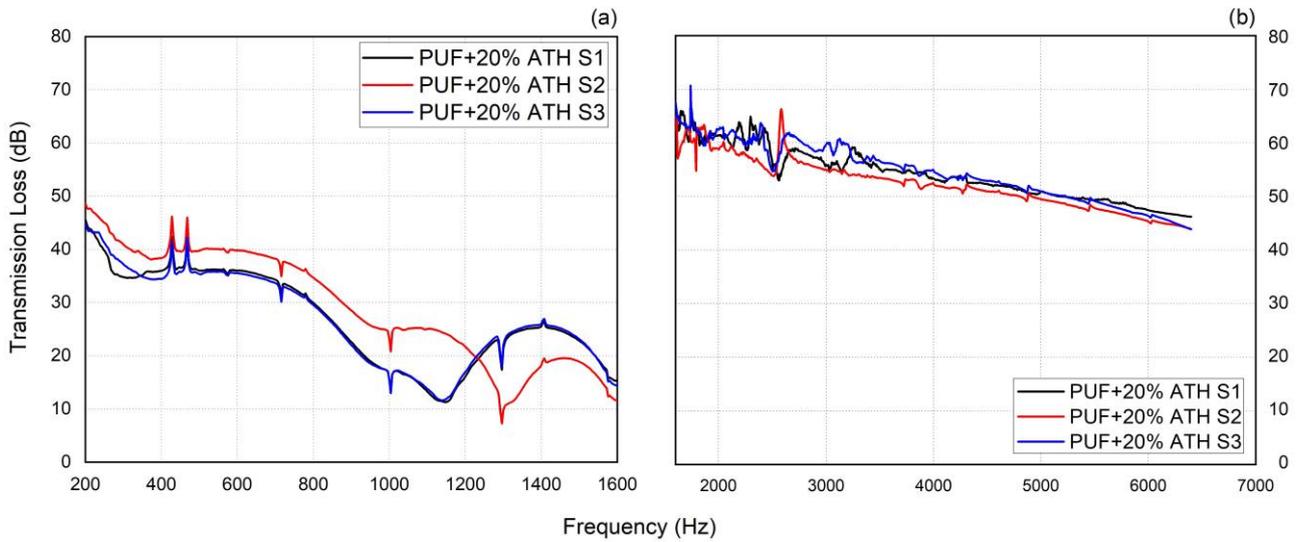
Figure 6. PUF+20%ATH STL results for the (a) 100 mm and (b) 29 mm tube.

Figure 7 presents the STL results for the PUF+30%ATH in the (a) 100 mm tube and (b) 29 mm tube. Now it is possible to see that the results for the 30% ATH added foam look more like the PUF+20%ATH S2 (Fig. 6a) in the stiffness sensitive region. This implies that the increased stiffness of the PUF+30%ATH is now enough to show a clear improvement in STL values in that region. Furthermore, a damping sensitive strip is now evident at 1300 Hz. The mass-controlled region in Fig. (7b) shows slightly higher values when compared to the pure PUF, especially from 3000 Hz to 4000 Hz, where the increment was in the order of 5 dB. These numbers are acceptable since Chandra *et al.* (2014) presented a material that increased in 6 dB the STL when doubling the mass of the samples.
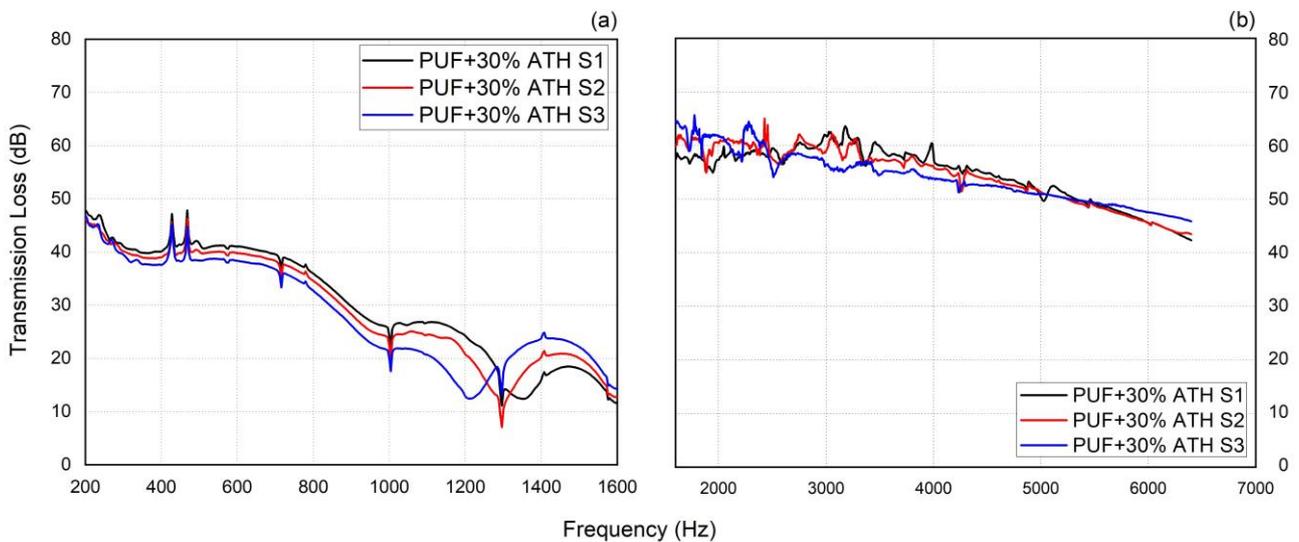

Figure 7. PUF+30%ATH STL results for the (a) 100 mm and (b) 29 mm tube.

Figure 8 exhibits the STL results for the PUF+40%ATH in the (a) 100 mm tube and (b) 29 mm tube. In the stiffness-sensitive region, the results for the 40% ATH added PUF were similar to the PUF+30%ATH, with STL values very similar up to 1600 Hz. This indicates that the addition of up to 20% of ATH presents slightly higher or even the same STL values of the pure PUF. When there was only one specimen of PUF+20%ATH that showed values similar to the ones with higher amounts of ATH. However, when more than 30% of ATH is added into the foam, the increasing stiffness is high enough to contain a higher amount of sound wave pressure passing through the material. The mass-sensitive region showed a higher number of STL at 2000 Hz, although, for higher frequencies, there were no noteworthy changes in this region. For example, at 5000 Hz, the PUF+30%ATH and PUF+40%ATH STL mean values are 50.9±0.5 dB and 51.4±1.1 dB.
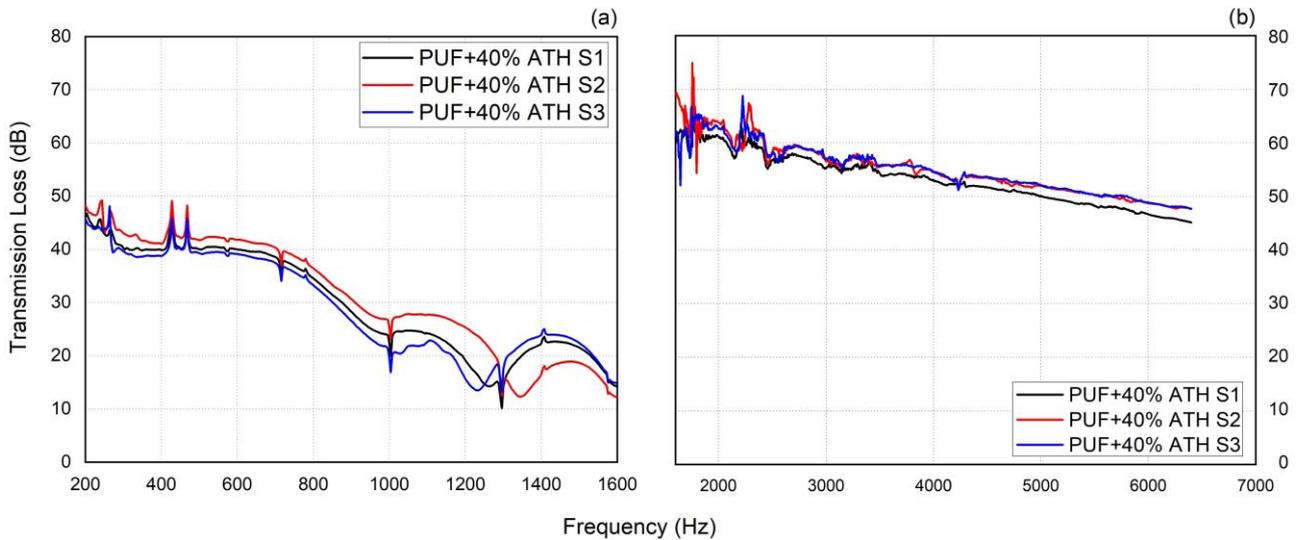
Figure 8. PUF+40%ATH STL results for the (a) 100 mm and (b) 29 mm tube.

Figure 9 shows the STL results for the PUF+50%ATH in the (a) 100 mm tube and (b) 29 mm tube. The results for the 50% ATH added foam in the stiffness-sensitive region were quite similar to the ones shown in Fig. (8a) and Fig. (7a). Therefore, it is possible to assure that after 30% of ATH, the addition of more flame retardant does not improve significantly the STL for minor frequencies. Furthermore, as shown by Silva *et al.* (2021), a proper flame extinguishment response is achieved with 40% of ATH inside the foam. Therefore, the addition of 50% of ATH does not improve the STL of the material, only making it denser, whereas the usage of 40% of ATH can be justified, instead of the PUF+30%ATH, by its lesser flammability, even though presenting practically the same STL values of the 30% one.
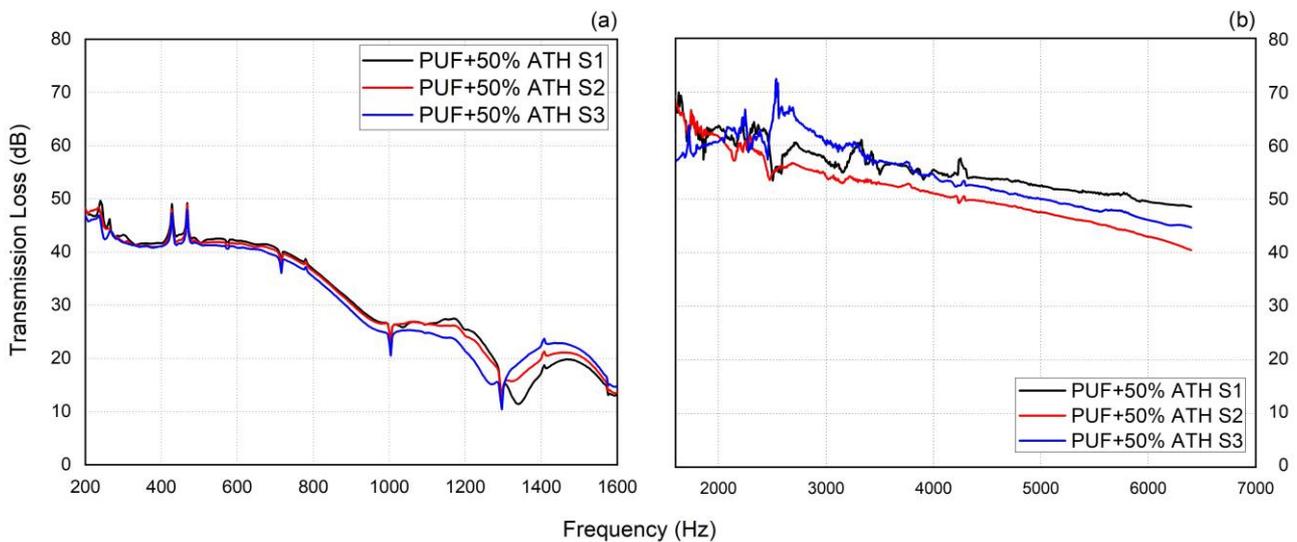

Figure 9. PUF+50%ATH STL results for the (a) 100 mm and (b) 29 mm tube.

Figure 10 presents the average curves for each tested material. It shows the increased STL values for higher mass percentages of ATH in both stiffness-sensitive and mass-sensitive regions. The variation between the pure foam and the 10% ATH added one was almost imperceptible. The same behavior was shown for the 30%, 40%, and 50% ATH added materials. The PUF+20%ATH showed a mean value among the tested materials and by its three characteristic curves it can be considered a transitory STL behavior between the tested materials.
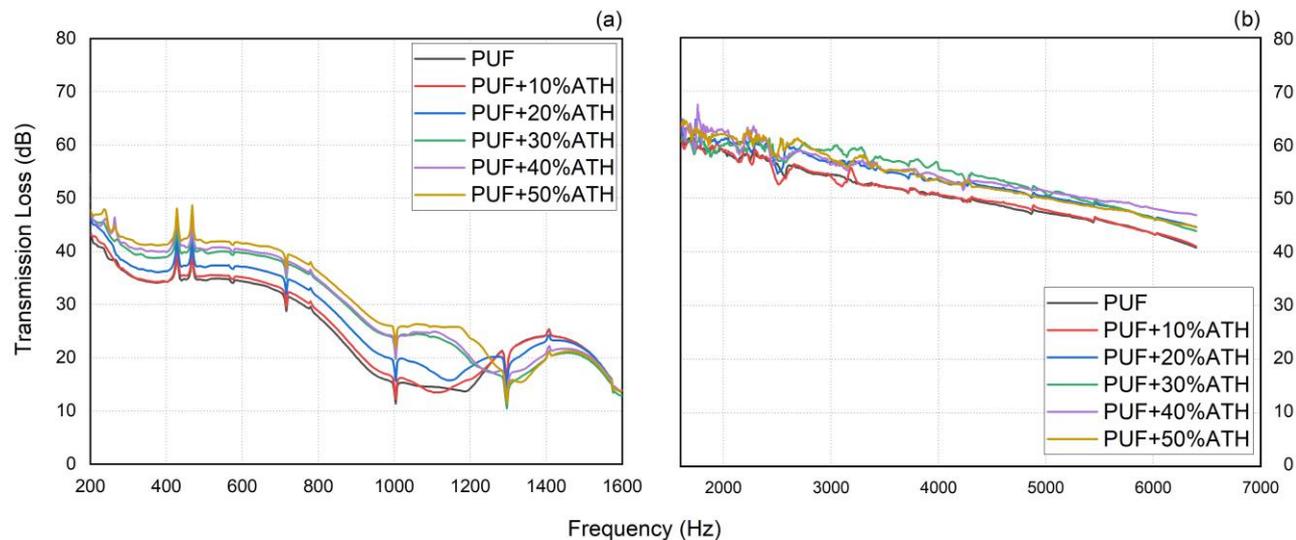
Figure 10. Average STL results for the (a) 100 mm and (b) 29 mm tube.

4. CONCLUSIONS

The acoustic properties of foams are among the most relevant themes for researches around the world. Therefore, proving that a green foam can be suitable to replace petroleum-based foams is essential in a world where the importance of sustainability is higher every day. Furthermore, improving some bad characteristics, such as their high flammability, is essential to improve the range of application for such kind of material. This paper investigated the changes in sound transmission loss for a bio-based polyurethane foam with alumina trihydrate added as a flame retardant in different mass fractions. The tests followed the ASTM E2611-19 in two different impedance tubes (29 mm and 100 mm). The STL-frequency curves were divided into three phases as described in the literature, the first one is the stiffness sensitive region and occurs in minor frequencies. Since ATH is a ceramic particle, it acts as a stiffness reinforcement and increased the STL values in this region for ATH additions of 30% or higher. The other region that was influenced in this study was the mass-controlled region, which happens in higher frequencies. Since the addition of flame retardant increases the mass of the material, it was possible to observe increasing STL values for materials with more than 30% of ATH added into them. Therefore, this study showed that besides improving the fire resistance of the material, the ATH also decreases the amount of sound passing through the material. This is an important data for many of the PUF requests and may increase the range of application for this bio-based foam. However, sound absorption tests could complement the data presented in this paper, since the STL also takes into consideration the reflected sound waves, which can be a problem in certain applications.

5. ACKNOWLEDGEMENTS

6. REFERENCES

ASTM E2611-19. 2019. *Standard Test Method For Normal Incidence Determination Of Porous Material Acoustical Properties Based On The Transfer Matrix Method*. ASTM - American Standard Test Method, West Conshohocken.

Berger, E., Royster, L.H., Driscoll, D.P., Royster, J.D., Layne, M., Johnson, D.L., 2003. *The Noise Manual*. Fairfax: American Industrial Hygiene Association, North Carolina.

Chandra, N., Raja, S., Gopal, K.V.N., 2014. "Vibro-Acoustic Response And Sound Transmission Loss Analysis Of Functionally Graded Plates". *Journal Of Sound And Vibration*, Vol. 333, No. 22, pp. 5786–5802.

Choe, H., Lee, J.H., Kim, J.H., 2020. "Polyurethane Composite Foams Including Caco3 Fillers For Enhanced Sound Absorption And Compression Properties". *Composites Science And Technology*, Vol. 194, pp. 108153.

Gooch, J.W. (Ed.), 2007. *Alumina Trihydrate. In: Encyclopedic Dictionary Of Polymers.* Springer, New York.

Kairytė, A., Kirpluks, M., Ivdre, A., Cabulis, U., Vaitkus, S., Pundienė, I., 2018. "Cleaner Production Of Polyurethane Foam: Replacement Of Conventional Raw Materials, Assessment Of Fire Resistance And Environmental Impact". *Journal Of Cleaner Production*, Vol. 183, No. 10, pp. 760–771.

Kind, D.J., Hull, T.R., 2012. "A Review Of Candidate Fire Retardants For Polyisoprene". *Polymer Degradation And Stability*, Vol. 97, No. 3, pp. 201–213.

Liu, Q., Gao, S., Zhao, Y., Tao, W., Yu, X., Zhi, M., 2021. "Review of layer-by-layer self-assembly technology for fire protection of flexible polyurethane foam". *Journal of Materials Science*, Vol. 56, No. 16, pp. 9605–9643.

Marques, D.V., Barcelos, R.L., Silva, H.R.T., Egert, P., Parma, G.O.C., Girotto, E., Consoni, D., Benavides, R., Silva, L., Magnago, R.F., 2018. "Recycled polyethylene terephthalate-based boards for thermal-acoustic insulation". *Journal of Cleaner Production*, Vol. 189, pp. 251-262.

Mohammadpour, R., Sadeghi, G.M.M., 2020. "Effect Of Liquefied Lignin Content On Synthesis Of Bio-Based Polyurethane Foam For Oil Adsorption Application". *Journal Of Polymers And The Environment*, Vol. 28, No. 3, pp. 892–905.

Pillai, P.K.S., Li, S., Bouzidi, L., Narine, S.S., 2016. "Metathesized Palm Oil Polyol For The Preparation Of Improved Bio-Based Rigid And Flexible Polyurethane Foams". *Industrial Crops And Products*, Vol. 83, pp. 568–576.

Silva, E.H.P., Souza, G.S.C., Janes, D.B., Waldow, G., Sales, F.C.P., Guedes, R.M., Tita,V., Costa, R.R.C., 2021. "Flexural And Flammability Evaluation of a New Bio-Based Polyurethane Foam with Alumina Trihydrate". *Proceedings of the Institution of Mechanical Engineers, Part L: Journal of Materials: Design and Applications*, Vol. 235, No. 5, pp. 1160–1171.

Szycher, M., 2013. *Szycher's Handbook Of Polyurethanes*. 2. Ed. Taylor & Francis Group, Boca Raton.

Wan, Y.Z., Hong, L., Jia, S.R., Huang, Y., Zhu, Y., Wang, Y.L., Jiang, H.J., 2006. "Synthesis And Characterization Of Hydroxyapatite–Bacterial Cellulose Nanocomposites". *Composites Science And Technology*, Vol. 66, No. 11–12, pp. 1825–1832.

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