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## Sustainable materials for sound absorption and insulation: Paper ICA2016-865

### Biobased porous acoustical absorber made from polyurethane and waste tires particles

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#### Abstract

The production of flexible polyurethane foams (FPF) incorporating bio/recycled raw materials is an interesting alternative to conventional acoustic absorbent materials. In this sense, bio-based polyols like glycerol or hydroxylated methyl esters derived from tung oil as multifunctional polyols, and waste tires particles as fillers with capability for acoustical absorption and low thermal conductivity, are prospective feedstocks for FPF preparation. In this work, FPF were prepared by adding different amounts of these components to a formulation based on a commercial polyether polyol. Results of normal sound absorption coefficient measurements at different frequencies, scanning electron microscopy analysis and compression tests are presented and discussed. The addition of waste tires particles or glycerol to the commercial formulation gives good performance as acoustic absorbers from 400 500 Hz, with NRC and SAA values near and above 50%. Moreover, the absorption coefficient reaches high values mostly at the highest evaluated frequencies (~62-89% at 2000 Hz and ~70-91% at 5000 Hz), for 30 mm thickness samples. On the other hand, the obtained FPF presented enhanced both the modulus and yield stress and in all the cases, a high recovery of the strain (>90%) applied in compression tests was attained after 24 hours. Scanning electron microscopy micrographs revealed that the obtained foams present a combination of open and closed cell structures and both, the modifiers and particles, tend to decrease the cell size. Based on acoustical and mechanical performance, and morphological analysis, the results show that these new kind of materials could be innovative sound absorbers, cheaper and environmentally more convenient in comparison with other available materials.

**Keywords:** acoustic absorbers, biobased foams, waste tires particles

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## 1 Introduction

Flexible polyurethane foams (FPFs) belong to a well-known class of polymeric materials that offers the possibility to be used in thermal and acoustic absorption, among others interesting technological applications. FPFs are porous materials that can absorb the sound energy, making them very useful for the control of noise. When a porous material is exposed to incident sound waves, the air molecules at the surface of the material and within the pores are forced to vibrate and lose some of their original energy. This is because part of the energy of the air molecules is converted into heat due to thermal and viscous losses at the walls of the interior pores and tunnels within the material [1].

Nowadays, the production of FPFs is based on petrochemical feedstock and the use of natural raw materials and/or waste products in their formulation is a big challenge with environmental and economic advantages [2]. In this sense, the chemical modification of vegetable oils is a promising alternative in the production of green polyols to be reacted with the isocyanate component to form the required polyurethane [2,3]. On the other hand, the use of recycled rubber in the production of sound absorbers can help to solve the existing problems of both waste disposal and noise pollution [4]. In this work, hydroxylated methyl esters derived from tung oil (HMETO) and glycerol (GLY) as the by-product of the biodiesel production were added to the formulation of flexible foams made from a commercial polyether polyol and a polyisocyanate. Both (HMETO and GLY) are multi-functional polyols that would allow modifying the crosslinking density and consequently the morphological, physical, acoustic and mechanical properties of the foams [3]. Furthermore, the foams were modified by the addition of particles of waste tires (WTP) as fillers with capability for acoustical absorption and low thermal conductivity.

## 2 Materials and methods

### 2.1 Preparation of the foams

The materials used to prepared the FPU were: Polyether polyol (JEFFOL G31-35), GLY or HMETO (10% respect to JEFFOL), pMDI (Rubinate 5005, Hunstman), blowing agent (water, 4%), surfactant agent (Tergostab B8404 Hunstman, 1.5%), 3% of n,n-dimethylbenzyl amine and 1.5% of dibutyltin dilaureate (both from Aldrich). The reactive mixture (index = 1.1) was blended with different contents of WTP (average diameter < 1 mm), placed in an open container and allowed to free rise at room temperature.

The preparation and characterization of the biobased polyols (HMETO and GLY) were described in detail in a previous work [3]. The names of the foams indicate the mass percentage of commercial polyether polyol replaced by the bio-derived one: for example 10GLY 10HMETO applies for the foam made with 10 wt.% glycerol plus 10 wt.% hydroxylated methyl ester derived from tung oil and 80 wt.% commercial polyether polyol; the reference foam (100 wt.%

commercial polyether polyol) is named as Ref. For composite foams, the number preceding the acronyms WTP indicate the mass content of particles of waste tires.

## 2.2 Characterization of the foams

The tests that were carried out in order to characterize the foams were: normal sound absorption coefficient measurements, apparent densities, scanning electron microscopy and compression tests.

### 2.2.1 Normal sound absorption coefficient ( $\alpha_N$ ) measurements:

Acoustic tests were performed using a standing wave tube Brüel & Kjaer 4002 (Denmark) to determine the acoustic property of the foams.

Sound absorption coefficients were measured for normal incidence sound generating pure tones matching centre frequencies of thirds octave bands between 100 and 5000 Hz, divided in two ranges test: from 100 to 1600 Hz, and from 800 to 5000 Hz.

Test specimens had a cylindrical shape of 100 mm in diameter for the range of frequencies between 100 y 1600 Hz, and 30 mm in diameter for the range of frequencies between 800 y 5000 Hz. In both cases, the height of the cylindrical samples was 30 mm. For each foam, the Noise Reduction Coefficient (NRC) and the Sound Absorption Average (SAA), according to ASTM C423-02a [5] were calculated.

### 2.2.2 Apparent densities:

Apparent densities were calculated as the ratio between the mass and the volume; the average values are reported.

### 2.2.3 Scanning electron microscopy (SEM):

The surfaces of the foams were analyzed using a scanning electron microscope (JEOL, model JSM-6460 LV). Small specimens were cut from the middle of the foams in the direction of growth. The pieces were coated with gold before being observed under the microscope.

### 2.2.4 Compression tests:

The specimens were tested at room temperature in an INSTRON 8501 Universal Testing Machine, according to the ASTM D3574-08 standard. The compression force was applied in the foam rise direction.

## 3 Results and discussion

### 3.1 Sound absorption, density and mechanical properties

Table 1 and Figure 1 shows the acoustic absorption coefficient ( $\alpha_N\%$ ) versus frequency for some of the tested FPFs. Figure 1a denotes that the replacement of the synthetic polyol by GLY and/or HMETO does not change the general feature of variation of absorption coefficient as a function of the frequency.

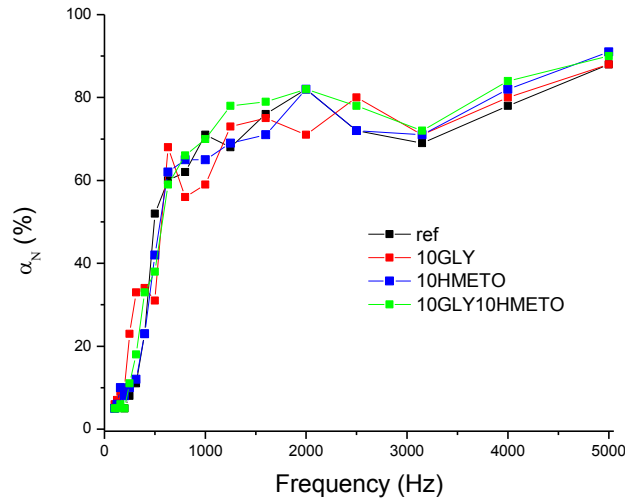
The number, size and type of pores are the important factors that should be considered while studying sound absorption mechanisms in porous materials. Large size porous cells or interconnected cells favoured the low frequency sound absorption.

The foams with GLY and HMETO clearly present higher density values (Table 2) and this fact is directly associated with the smaller sizes of pores (as was observed by SEM, Figure 2), however these pores seem to be more interconnected improving the sound absorption due to the increase in tortuous paths. Evidently in this case these two contrary effects compensate, resulting in acoustic absorption coefficients that are not significantly affected in all the analyzed frequency range.

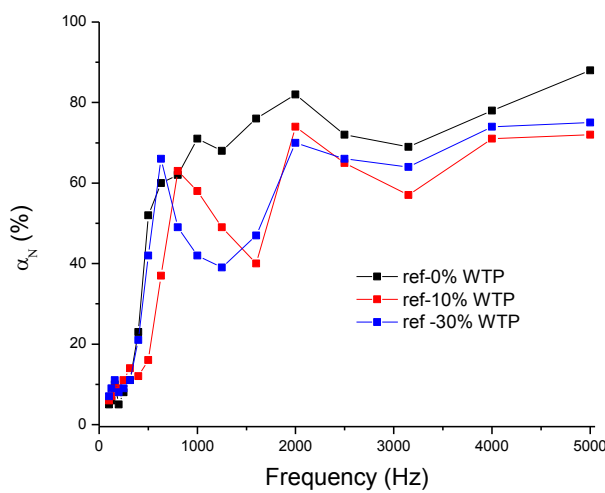
Table 1. Acoustic absorption coefficient ( $\alpha_N$ ) versus frequency for selected FPFs

Freq [Hz]	0% WTP				Reference			10GLY		
	Ref	10GLY	10HMETO	10GLY 10HMETO	0% WTP	10% WTP	30% WTP	0% WTP	10% WTP	30% WTP
100	5	6	5	5	5	6	7	6	6	5
125	5	7	6	5	5	7	9	7	7	6
160	8	9	10	6	8	9	11	9	11	7
200	4	9	8	5	4	9	8	9	10	6
250	9	19	10	11	9	11	9	19	21	11
315	12	28	12	18	12	14	11	28	35	15
400	20	38	23	33	20	12	21	38	32	17
500	41	49	42	38	41	16	42	49	24	29
630	62	71	62	59	62	37	66	71	45	46
800	67	60	65	66	67	63	49	60	66	72
1000	77	56	65	70	77	58	42	56	55	77
1250	73	61	69	78	73	49	39	61	48	61
1600	79	63	71	79	79	40	47	63	51	56
2000	85	69	82	82	85	74	70	69	63	52
2500	76	76	72	78	76	65	66	76	75	72
3150	72	73	71	72	72	57	64	73	80	73
4000	78	77	82	84	78	71	74	77	82	77
5000	89	82	91	90	89	72	75	82	86	75
<b>NRC %</b>	<b>55</b>	<b>50</b>	<b>50</b>	<b>55</b>	<b>55</b>	<b>40</b>	<b>45</b>	<b>50</b>	<b>45</b>	<b>45</b>
<b>SAA %</b>	<b>50</b>	<b>50</b>	<b>48</b>	<b>51</b>	<b>50</b>	<b>37</b>	<b>39</b>	<b>50</b>	<b>43</b>	<b>43</b>

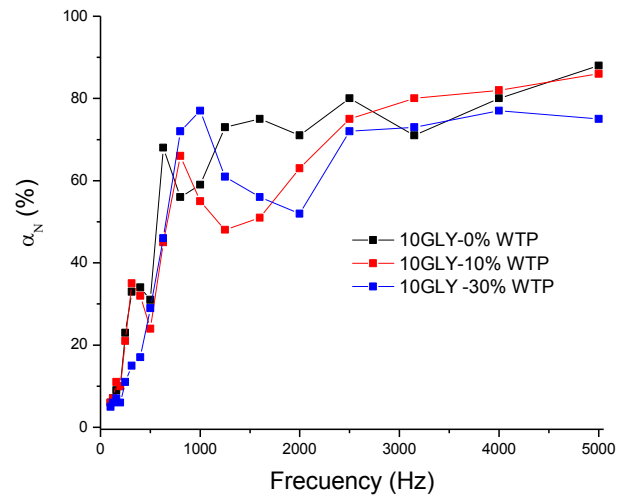
Figure 1b indicates that the incorporation of WTP shifts the absorption coefficient to lower values in the system based on the synthetic polyol. The WTP has the ability of improving the acoustic damping of the PU foam [4]. However, the increase in the composite foam density as WTP is added (Table 2) appears to be the most important effect affecting the  $\alpha_N$  values. Higher density leads to less empty spaces to dissipate sound as thermal energy.



(a)



(b)



(c)

**Figure 1. Sound absorption coefficients of the foams: a) Unfilled foams, b) filled foams based on the reference polyurethane, c) filled foams based on the reference polyurethane modified with 10% of GLY**

The foams modified with 10% GLY present an increase in the sound absorption with WTP content in the 500-1000 Hz zone indicating that recycled rubber particles are advantageous to be applied as sound absorber in the middle-high frequency region [4]. The more interconnected cells in the foams with 10% of GLY (associated with their compact structure) reduce the size and volume fraction of air voids and make the air passages more tortuous and much narrower. Therefore, in a compact structure the sound wave travels a longer distance and this leads to a reduction of the sound energy [6].



On the other hand, the obtained FPU's presented NRC and SAA values near and above 50% denoting good performance as acoustic absorbers from 400-500 Hz.

Table 2 shows the density values and mechanical properties of the different foams. The average density of the unfilled foams increases with the addition of 10% of glycerol (65.4 kg/m<sup>3</sup>) with respect to the reference foam (31.9 kg/m<sup>3</sup>) due to the higher reactivity of the GLY that decreases the volume expansion of the foams and increases the crosslinking density [3]. The addition of HMETO also increases to some degree the density of the foam (36.6 kg/m<sup>3</sup>), which was related with the higher functionality of the HMETO in comparison with that of the synthetic polyol (although it is lower than that of the GLY). As was expected, the foam prepared with both GLY and HMETO presents an intermediate density between that of the 10% HMETO and 10% GLY (62.8 kg/m<sup>3</sup>).

On the other hand, the addition of GLY significantly increased the modulus and yield stress of the reference foam, as a result of the higher apparent and crosslinking densities of the modified foam. The addition of HMETO produces a small increase in these properties and the foam with 10GLY and 10HMETO shows modulus and yield stress values not significantly modified with respect to those of the 10GLY foam.

The addition of WTP to the reference polyurethane (prepared with the synthetic polyol) clearly leads to an increase in the density of the foams. This can be associated to the fact that the WTP in the reactive mixture delay the expansion of foams during curing.

The effect of the WTP in the foams modified with 10 wt.% of GLY is less significant probably due to that the high reactivity of the reactive modifier masks the WTP effect in terms of decreasing the growth of the foam. The same result was observed in the foam modified with GLY and HMETO when WTP were added. However, the differences in the density values of these foams are not statistically significant.

Table 2 also presents the compressive properties of foams filled with WTP. The modulus of the samples prepared with the commercial polyol shows a significant increase with the addition of 10 wt% of WTP. Higher contents of WTP do not produce additional changes in the rigidity of the foams. This behavior is associated with the higher density of the filled foams respect to that of the unfilled one, as was discussed above.

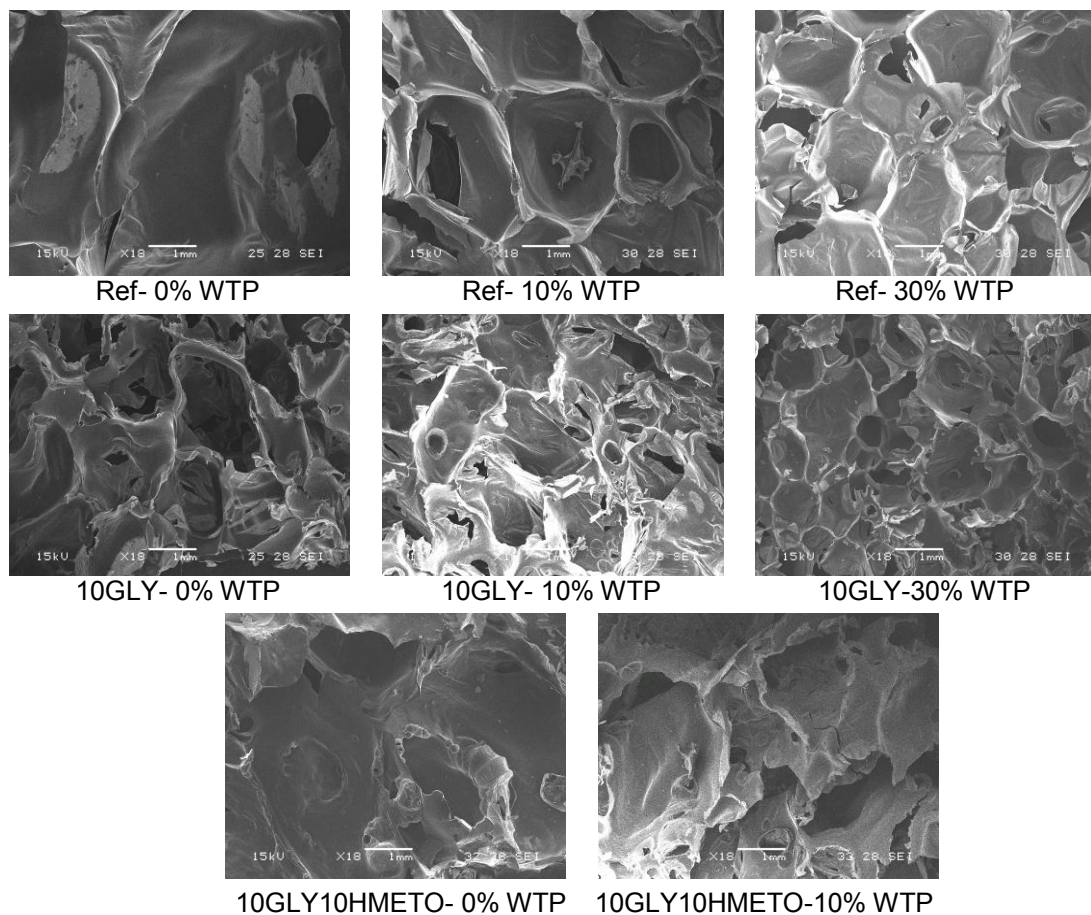
**Table 2. Densities and compression properties of the foams**

Sample	Density (kg/m <sup>3</sup> )	Modulus (kPa)	Yield stress (kPa)
Ref-0% WTP	31.9 ± 2.3	84.9 ± 19.7	14.8 ± 2.7
Ref-10% WTP	39.3 ± 1.3	180.4 ± 43.0	21.7 ± 1.6
Ref-30% WTP	50.4 ± 6.3	174.4 ± 51.6	20.3 ± 3.0
10GLY-0% WTP	65.4 ± 3.0	845.9 ± 182.6	67.3 ± 5.5
10GLY-10% WTP	52.9 ± 6.6	656.0 ± 139.4	74.9 ± 9.8
10GLY-30% WTP	66.0 ± 8.6	564.8 ± 90.4	58.0 ± 12.9
0GLY-10HMETO	36.6 ± 4.7	90.7 ± 26.6	5.6 ± 2.0
10GLY10HMETO-0% WTP	62.8 ± 10.3	530.1 ± 39.4	93.9 ± 15.3
10GLY10HMETO-10% WTP	62.1 ± 5.3	565.1 ± 110.1	68.1 ± 4.9

Contrary, the addition of WTP to the polyurethane system with 10% of GLY as replacement of the synthetic polyol leads to a decrease in the modulus. In foamed systems, the change in density is one of not the most important factors for explaining the variations in mechanical properties. However, the disruption of the foam cell walls due to the addition of particles is also significant and in this case is the responsible for the decrease in the mechanical properties.

### 3.2 Scanning electron microscopy (SEM)

Figure 2 shows SEM micrographs of samples cut from the middle of the foams, in the direction of growth. A mix between close and open cellular structure was observed for all the foams. The images shows that the addition of GLY and HMETO leads to reduced cell's sizes. The foams prepared with 10GLY plus 10HMETO present a more distorted cell structure than those based on GLY.



**Figure 2: SEM micrographs of selected foams (x18 magnification)**

The figure also shows the micrographs of the foams filled with WTP. In general, the presence of larger amounts of WTP leads to smaller cells with narrow distribution of sizes, mainly in the foams prepared with just the synthetic polyol, in agreement with their higher densities, which is associated to the restricted expansion of the foams. During the development of the foam, the

particles can act as nucleation sites promoting the formation of a larger quantity of bubbles, which tends to increase with the filler content. At the same time, growth of the cells is hindered by the increase in viscosity of the polyurethane matrix, as was also noted by other researchers [7]. In our case, the cells are allowed to expand less easily in the foams containing WTP and, therefore, an increase of the final foam density is observed [7]. The series of foams with 10% GLY seems to present a more interconnected (open) structure than the others.

## 4 Conclusions

The present work presented a route for the production of more environmentally friendly filled flexible PU foams through the replacement of part of the synthetic polyol by biobased ones and by the addition of waste tires particles. Promising acoustic absorption properties were observed in a wide range of frequencies.

## Acknowledgments

Soto, Marcovich and Mosiewicki gratefully acknowledge the financial support provided by the National Research Council of Argentina (CONICET), the Science and Technology National Promotion Agency (ANPCyT) and the National University of Mar del Plata.

Vechiatti, Iasi and Armas gratefully acknowledge the Acoustics and Lightning Laboratory of the Scientific Research Commission of Buenos Aires Province (LAL-CIC).

## References

- [1] Arenas, J. P.; Crocker, M. J. Recent Trends in Porous Sound-Absorbing Materials. *Sound & vibration*, Vol 44(7), 2010, pp 12-17.
- [2] Caillol S, Desroches M, Boutevin G, Loubat C, Auvergne R, Boutevin B. Synthesis of new polyester polyols from epoxidized vegetable oils and biobased acids. *Eur. J. Lipid Sci. Technol.* Vol 114(12), 2012, pp 1447-1459.
- [3] Soto, G. D.; Marcovich, N.E.; Mosiewicki, M. A.. Flexible polyurethane foams modified with bio-based polyols: synthesis and physical/chemical characterization. In press, *Journal of Applied Polymer Science*, 2016.
- [4] Hong, Z.; Bo, L.; Guangsu, H.; Jia, H. A novel composite sound absorber with recycled rubber particles. *Journal of Sound and Vibration*. Vol 304, 2007, pp 400–406.
- [5] ASTM C423-02a: Standard test method for sound absorption and sound absorption coefficients by the reverberation room method
- [6] Elammaran Jayamani, E.; Sinin Hamdan, Md Rezaur Rahman, Kok Heng Soon, Muhammad Khusairy Bin Bakri. Processing and Characterization of Epoxy/Luffa Composites: Investigation on Chemical Treatment of Fibers on Mechanical and Acoustical Properties. *Bioresources*. Vol 9 (3), 2014, pp 5542-5556.
- [7] Silva, V.R.; Mosiewicki, M.A.; Yoshida, M.I.; Silva, M.C.; Stefani, P.M., Marcovich, N.E. Polyurethane foams based on modified tung oil and reinforced with rice husk ashes I: synthesis and physical chemical characterization. *Polymer Testing* 32 (2), 2013, pp 438-445.
- [8] ASTM D3574 – 08: Standard Test Methods for Flexible Cellular Materials—Slab, Bonded, and Molded Urethane Foams