Microstructure and Physical Properties of Open-Cell Polyolefin Foams

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ABSTRACT: The cellular structure, physical properties, and structure–property relationships of novel open-cell polyolefin foams produced by compression molding and based on blends of an ethylene/vinyl acetate copolymer and a low-density polyethylene have been studied and compared with those of closed-cell polyolefin foams of similar chemical compositions and densities and with those of open-cell polyurethane foams. Properties such as the elastic modulus, collapse stress, energy absorbed in mechanical tests, thermal expansion, dynamic mechanical response, and acoustic absorption have been measured. The experimental results show that the cellular structure of the analyzed materials has interconnected cells due to the presence of large and small holes in the cell walls, and this structure is clearly different from the typical structure of open-cell polyurethane foams. The open-cell polyolefin foams under study, in comparison with closed-cell foams of similar densities and chemical compositions, are good acoustic absorbers; they have a significant loss factor and lower compressive strength and thermal stability. The physical reasons for this macroscopic behavior are analyzed. © 2009 Wiley Periodicals, Inc. J Appl Polym Sci 114: 1176–1186, 2009

Key words: mechanical properties; polyethylene (PE); polyolefins; thermal properties

INTRODUCTION

Polymer foams are two-phase materials in which a gas is dispersed in a continuous macromolecular phase.[1–3] These materials are important items in the economy, and for commercial, technical, and environmental reasons, they play an important role in our society. Polyolefin (PO) foams are an important group of polymer-based foams. From a technical point of view, a variety of properties, such as light weight, buoyancy, chemical resistance, friendliness to skin, no water absorption, cushioning performance, energy absorption, and thermal insulation, have ensured the success of these PO-based foams.[4]

Several processes are used nowadays to produce crosslinked PO-based foams.[4–7] All these well-known technologies give rise to foams with closed cells. One of the primary applications of closed-cell foams is thermal insulation; more foam is used for thermal insulation than for any other purpose.[1,8,9] These materials have the lowest thermal conductivity of any conventional nonvacuum insulator. It is generally accepted that a closed cellular structure improves the mechanical properties,[1] making the foam suitable for applications in which the absorption of energy is required (e.g., packaging). In addition, this kind of structure allows the foam to have excellent buoyancy and permits the use of the material as a sealant against air and dust.

However, because of the low acoustic absorption and poor recovery after creep of closed-cell foams, open-cell PO-based foams have been recently introduced into the market.[10–12] These materials are produced by a compression-molding technology in which a last step, consisting of the mechanical deformation of the foam block, is included to fully open the cells. This particular foaming route results in cellular structures that are very different from those of typical open-cell polyurethane (PU) foams[13] and are, of course, different from those of closed PO foams.

The change in the type of cellular structure (from closed cells to open cells) introduces a dramatic modification of the physical mechanisms controlling the physical properties of this type of materials. However, as far as we know, these possible modifications have not been studied in detail in open-cell PO foams, and in fact, the physical mechanisms determining the macroscopic behavior of these new materials are not obvious.

This article presents the structure and several properties of novel open-cell PO foams. The main aim is to establish the structure–property relationships for these materials through a detailed analysis of the physical mechanisms controlling each property. As a result, new knowledge about the structure–property relationships of foams is established.

EXPERIMENTAL

Materials

The open-cell foams studied were manufactured with the two-stage molding procedure[5–7] with azodicarbonamide as the foaming agent, dicumyl peroxide as the crosslinking agent, and calcium carbonate as the filler. After the foam blocks were produced, the cells were opened by mechanical deformation; this procedure[10–12] allowed a 100% open-cell PO-based foam to be obtained. The samples of this study were cut from the central part of the blocks to avoid possible effects related to the typical inhomogeneous density and cellular structure of blocks produced with this technology.[14,15] The thickness of the analyzed samples was 10 mm.

The foams were blends of an ethylene/vinyl acetate copolymer (EVA; vinyl acetate content = 18%) and a low-density polyethylene (LDPE). The chemical composition of the materials was 40% LDPE, 40% EVA, and 12% $CaCO_3$. All the foams presented a proportion of foaming agent residues of approximately 7%. The remaining 1% contained the activator for the blowing agent (zinc oxide) and processing aids. The gel content was determined in xylene at 140°C during 24 h according to standard procedures for POs. The measured values were constant for all the materials under study, the average value being 59 ± 3%.

To obtain a clearer understanding of the physical behavior of the foams, the structure and properties were compared with those of standard foams. The selected materials were a standard open-cell flexible PU foam with a density of 24 kg/m^3 (used for mattresses and supplied by Recticel, Brussels, Belgium) and two conventional closed-cell PO foams, one based on LDPE and the other based on EVA. The closed-cell PO foams were also produced by compression molding, that is, the same technology, but in these materials, the last step of compression to open the cells was not included. The gel content for these two materials was 52 ± 3%. Table I shows the main characteristics of the foams under study. The open-cell foams were kindly supplied by Sanwa Kako Co., Ltda. (Kyoto, Japan), and the closed-cell foams were supplied by Microcel S.A. (Burgos, Spain).

Although both types of PO foams were produced with the same technology (compression molding), there were several aspects in which the processes were different.

First and very important, in the production of the open-cell materials, a last step of mechanical compression was included to complete cell opening; this last step was not used in the closed-cell materials.

Second, in the manufacturing of closed-cell foams, the crosslinking process was activated before the expansion of the foams[5–7]; therefore, when the foam expanded, the cell walls had a melt strength high enough for deformation without breaking, which resulted in a small rate of cell rupture. As a result, the open-cell content should be low for these materials. However, it is important to take into account that this open-cell content is a function of several parameters, such as the base polymer, the expansion ratio, the crosslinking grade reached just before the foam is expanded, the cell wall thickness before expansion, and the cell wall thinning during foaming, and some degree of cell connectivity should be expected for high expansion ratios.

For the open-cell materials, the main strategy used to partially open the cells was to produce the foam expansion before the matrix polymer reached a significant degree of crosslinking[10–12]; this modification of the main process order (foaming before crosslinking) allowed increasing the degree of broken cell walls, and because of this, a partial opening of the cells during foaming was obtained. To complete cell opening, as previously mentioned, the final foam blocks were compressed.

Differential scanning calorimetry (DSC)

The thermal properties of the foams were studied with a Mettler DSC30 (Zurich, Switzerland) previously calibrated with indium, zinc, and lead. The weights of the samples were approximately 2.5 mg. The temperature program was between −40 and 200°C at a rate of 10°C/min. The crystallinity was calculated from the area of the DSC peak, with the heat of fusion divided by the heat of fusion of 100% crystalline material (288 J/g for 100% crystalline polyethylene). The inorganic content was taken into account to determine the crystallinity of the polymeric phase. The 95% confidence interval of these three measurements was approximately ±8% of the average value for the crystallinity.

Scanning electron microscopy (SEM)

SEM was used to characterize the cell size and fraction of cell edges. Micrographs were taken with a JEOL JSM-820 (Tokyo, Japan) microscope.

TABLE I
Main Characteristics of the Foams Under Study

Foam	Foam density (kg/m^3)	Average cell size (µm)	Fraction of mass in the edges	Open-cell content	Tortuosity
CA23	23.1	3294	0.51	0.97	11.1
CA26	25.9	630	0.55	0.98	25.4
CA33	33.2	1390	0.57	0.99	19.8
CA44	43.6	970	0.58	0.99	17.3
CA49	48.7	1011	0.53	0.98	14.6
CA64	63.7	3017	0.51	0.99	18.6
CCP30	25.4	255	0.31	0.07	—
CCO30	28.7	247	0.3	0.63	—
CAPU	24.4	356	1.0	0.99	1.7

The following designations are used for the foam samples: CA for open-cell foams, CCP30 for the closed-cell foam based on LDPE, CCO30 for the closed-cell foam based on EVA, and CAPU for the open-cell PU foam.

The mean cell size in each foam direction (x, y, z) was estimated with the intersection method; this consisted of measuring the number of cells that intersected several reference lines and dividing the appropriate reference length by the number of cells. The average cell size was computed as a mean value of the cell sizes in the different directions. All the foams were isotropic, so it was not necessary to analyze the anisotropy coefficient. Taking account the relationship between the mean measured length of the randomly truncated cells and the real diameter of the cell, we multiplied the previous result by 1.623.[16] The fraction of mass in the edges was obtained with the method suggested by Kuhn et al.[17] The 95% confidence interval for the mean cell size in each direction was estimated to be approximately ±7% of the average value. The confidence interval for the fraction of mass in the edges was 12%.

Density

The density was obtained as the ratio of the mass and volume of the sample. The estimated accuracy was 5%.

Open-cell content

The test was conducted according to ASTM Standard D 2856-94 in an air pycnometer (model 08.69) from Eijkelkamp (Giesbeek, The Netherlands). The air pycnometer measured the volume of the sample that was not gas (V_q), that is, the solid volume plus the gas volume of the unconnected cells. This volume could be calculated with the following equation:

$$V_q = V_g(1 - f) + V_s \qquad (1)$$

where f is the open-cell content and V_g and V_s are the gas and solid volumes in the sample, respectively. The latter are defined as follows:

$$V_g = V_f\left(1 - \frac{\rho}{\rho_s}\right) \qquad (2)$$

$$V_s = V_f\left(\frac{\rho}{\rho_s}\right) \qquad (3)$$

where V_f is the volume of the tested sample, ρ is the foam density, and ρ_s is the density of the solid phase.

Using eqs. (1)–(3), we found that f could be calculated with the following equation:

$$f = \frac{V_f - V_q}{V_f\left(1 - \frac{\rho}{\rho_s}\right)} \qquad (4)$$

Samples of $3 \times 3 \times 1$ cm^3 were used for these measurements. The 95% confidence interval for the open-cell content was estimated to be approximately ±7% of the average value.

Tortuosity

The tortuosity can be measured with a simple device if the frame material is an electrical insulator.[18] In this device, the air in the pores of the sample is replaced by an electrical, conducting liquid, and the electrical resistivity of the sample (r_s) is measured with an electrical circuit. The value obtained is then compared with the electrical resistivity of the liquid (r_l). The tortuosity (T) is calculated with the following equation[18]:

$$T = f\frac{r_l}{r_s} \qquad (5)$$

In our case, the auxiliary liquid was a 0.4 molar solution of CuSO$_4$. The samples were immersed in the liquid for 12 h before the electrical measurements. This cellular structure characteristic was measured for the open-cell foams, in which it was possible to substitute the air in the cells with the liquid.

The 95% confidence interval for the tortuosity was estimated to be approximately ±10% of the average value.

Thermal expansion coefficient

The linear thermal expansion coefficient (α) is the property that determines the dimensional behavior with temperature:

$$\alpha = \frac{1}{l_0}\left(\frac{dl}{dT}\right) \quad (6)$$

where l_0 is the initial length at the reference temperature and dl/dT is the change in length over a temperature range.

A PerkinElmer TMA7 (Waltham, MA) testing apparatus was used. The experiments were carried out with a parallel-plate measurement system 15 mm in diameter, and the samples were prepared in a cylindrical form with a diameter of 10 ± 0.1 mm. The static stress applied to avoid sample deformation and to ensure that the probe remained in contact with the sample was 13 Pa.

The temperature ramp used to characterize the thermal behavior was from -40 to $120°C$ at $5°C/$min. Each material was measured three times, and the standard deviation was approximately 4% of the mean value.

Mechanical properties at low strain rates

The experiments were carried out with an Instron model 5500 R6025 machine with a strain rate of $0.1\ s^{-1}$ up to 75% strain. The samples had a diameter of 10 ± 0.1 cm and a thickness of 10 ± 0.1 mm. Every test was performed at $23 \pm 2°C$ with a relative humidity of $50 \pm 5°C$. Each material was measured three times. The following mechanical properties were obtained from the previous experiments:

1. The elastic modulus (E) of the foam was taken as the slope in the initial zone of the stress strain curve (<3% of strain).
2. The collapse stress was measured directly in the strain–stress curve as the intersection points of two straight lines: the first one was a tangent to the initial region (elastic region), and the second one was a tangent to the plateau region (between 20 and 60% strain).
3. The absorbed energy was computed as the area under the curve up to 75% strain.

The standard deviation of these measurements was 5%.

Dynamical mechanical properties

For this purpose, a PerkinElmer DMA7 testing apparatus was used. The experiments were preformed with a parallel-plate measurement system (15 mm in diameter) at a frequency of 1 Hz. The samples were prepared with a cylindrical shape with a diameter and thickness of 10 ± 0.1 mm. The measurements were carried out as a function of temperature between -30 and $80°C$ with a heating rate of $5°C/$min. All the foams were subjected to the same strain conditions: 2% static strain and 0.11% dynamic strain.[19–21] Each material was measured three times; the mean standard deviation was 7% for the storage modulus and 2% for the loss factor (tan δ).

Acoustic absorption coefficient

The two-microphone or transfer-function method was used to measure the acoustic absorption coefficient. A Brüel & Kjaer (Naerum, Denmark) model 4206 impedance tube, in accordance with ASTM E 1050 and ISO 10534-2 methods, was used. In this method, a sound source (loudspeaker) is mounted at one end of the impedance tube (a cylindrical tube), and a sample of the material is placed at the other end. The loudspeaker generates broad-band, stationary, random sound waves, which propagate as plane waves in the tube, hit the sample, and reflect. The propagation, contact, and reflection result in a standing-wave interference pattern due to the superposition of forward and backward traveling waves inside the tube. By the measurement of the sound pressure at two fixed locations inside the tube and the calculation of the complex transfer function with a two-channel digital frequency analyzer, it is possible to determine the sound absorption of the material. The usable frequency range depends on the diameter of the tube and the spacing between the microphone positions. In our case, the measurements were made in the 500–6400-Hz frequency range. The samples were 29 mm in diameter and 10 ± 0.1 mm thick. Each material was measured six times, and the maximum standard deviation obtained in the acoustic absorption values was 7% of the mean value.

RESULTS AND DISCUSSION

Structural characterization

The degree of crystallinity is constant for the open-cell PO foams under study. The average value is $27 \pm 3\%$, which is intermediate between that for the closed-cell LDPE foam ($36 \pm 2\%$) and that for the closed-cell EVA foam ($21 \pm 2\%$). This is an expected result if we take into account that the base polymer of the open-cell PO foams is a blend of LDPE and
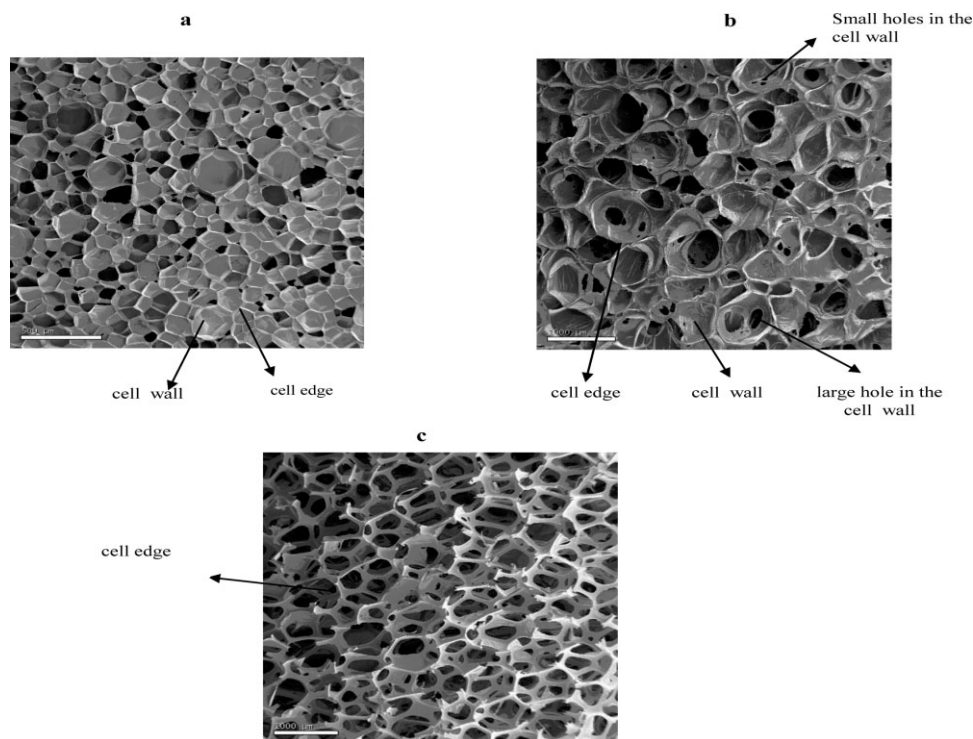
Figure 1 SEM micrographs of (a) a conventional closed-cell, LDPE-based foam (CCP30), (b) a novel open-cell PO foam (CA49), and (c) a standard open-cell PU foam (CAPU).

EVA, and for this reason, an intermediate crystallinity between those of these two polymers should be found. The PU foam is an amorphous material.

Table I shows the foam densities and average cell sizes of the foams under study; the density ranges between 23 and 64 kg/m^3, and this means porosities in the range of 97–93%.

Figure 1 shows examples of the microstructures of the different types of foams considered. The closed-cell foam presents a structure built from polyhedral cells [Fig. 1(a)], the cells being separated by a solid face (cell wall) and the cell edges being located in the intersections of several cell walls. In the open-cell foam [Fig. 1(b)], the elements of the cellular structure are similar. There are also edges and faces; however, most of the cell walls present holes, which can be large (of a size similar to the cell wall size) or very small. These holes allow the gas to be a continuous phase, and therefore this phase would have some degree of mobility through the foam. From this point of view, these materials can be considered open-cell foams. In fact, the open-cell content of all these materials is 100% (Table I). This kind of cellular structure is clearly different from the typical structure of an open-cell flexible PU foam [Fig. 1(c)]. For these materials, the cellular structure is built from edges; that is, there are no cell faces including holes.

From the previous description, it seems that the structure of the materials under study has several characteristics of the closed-cell material (the

presence of faces in the cells) and several other characteristics of the open-cell material (the gas phase is continuous, and the open-cell content is near 1). It can be concluded that the cellular structure of the open-cell foams is intermediate between that of closed-cell PO foams and that of open-cell PU foams. In fact, the values for the fraction of material in the edges are between 0.5 and 0.6 (Table I), that is, between that for the closed-cell PO foams (0.3; Table I)[22–25] and that for the open-cell PU foams (fraction of mass in the edges ≈ 1).

Cell sizes for the open-cell foams range from 630 to 3300 μm; these cell sizes are much higher than those of the closed-cell foams selected (250 μm) and that of the open-cell PU foam (350 μm). The cell sizes for typical closed-cell foams produced by compression molding[22–25] are in the same range as those of the foams selected in this article. In fact, it has been shown that compression-molding technology is able to produce PO foams with smaller cell sizes in comparison with other foaming technologies.[26]

The differences in cell size for the PO foams are a result of the manufacturing process. As explained previously, in the manufacturing of closed-cell foams, the crosslinking process is activated before the expansion.[5–7] The high degree of crosslinking of the matrix during foaming results in foams with a low average cell size.[3] However, in the foaming of open-cell materials, the crosslinking degree when the foam is expanded is much lower (this is the
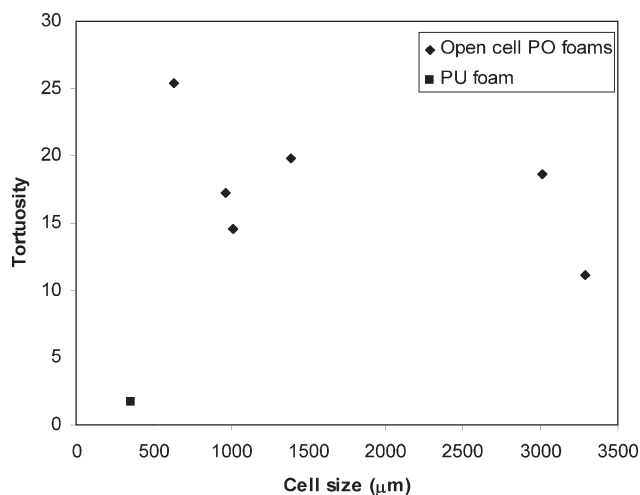
Figure 2 Tortuosity of the open-cell foams as a function of the cell size.

main strategy to open the cells); therefore, as the cells grow in a matrix with a low degree of crosslinking, the foams have larger cell sizes.[3]

As already mentioned, the open-cell PO foams (CAs) and the open-cell PU foam (CAPU) present 99% interconnected cells; the closed-cell LDPE-based foam (CCP30) has a value of 7%, whereas the closed-cell EVA-based foam (CCO30) has 63% connectivity. The unexpected high value of the open-cell content presented in the CCO30 foam (which was processed without mechanical deformation after foaming) suggests that the EVA component makes cell opening easier without the action of postprocessing mechanical compression or without special care in inverting the crosslinking reaction and the foam expansion.

As explained in the previous section, the open-cell content obtained in a process in which crosslinking is produced before foaming (a process to produce closed-cell materials) depends on many factors.
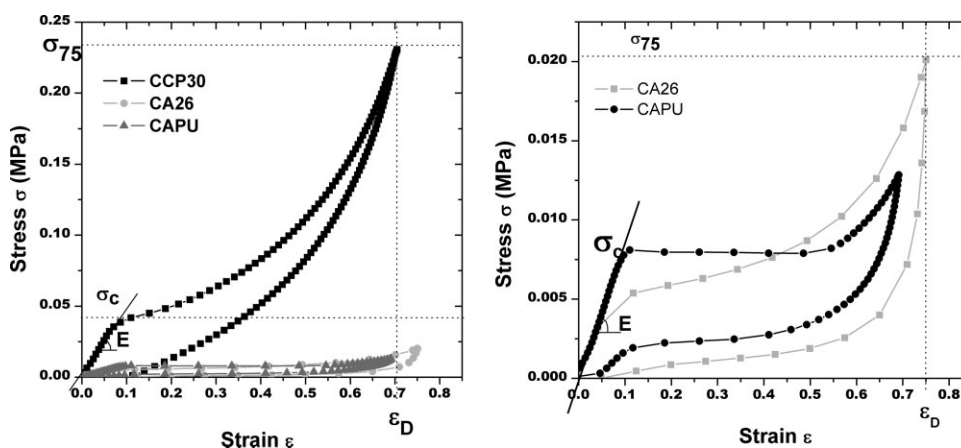
Some of the most important are the final expansion ratio, the crosslinking grade reached just before the foam is expanded, the cell wall thickness, and the cell wall thinning during foaming. The result observed for the EVA foam of this study seems to indicate that foams based on EVAs are more susceptible to cell opening during conventional compression molding. The explanation of this behavior of compression-molded EVA foams is not a simple task, and an additional scientific study of the expansion of these materials is needed. From the point of view of this study, which is mainly focused on the structure–property relationships of open-cell PO foams, to have a foam with an intermediate open-cell content is interesting because this material will allow additional information to be obtained about the foam behavior (e.g., see the acoustic behavior analysis)

The values for the cellular structure tortuosity are given in Table I and in Figure 2 as a function of the cell size. The extremely high tortuosity of the open-cell foams under study (10 times higher than that of the PU foam) is remarkable. This result can be understood if we take into account that the connection between cells in these materials is through very few small or large holes in the cell faces, whereas in the PU foam, there are no faces, so the gas is easily connected. This difference will lead to, as a first outcome, a significant air flow resistance for these materials and then important effects on the physical properties, as explained later. Also, the tortuosity seems to decrease when the cell size increases for the open-cell foams under study.

Physical properties

Mechanical properties

It is well known that the typical stress–strain behavior of foams can be divided into three regions.[1] At

Figure 3 Stress–strain curves for the foams under study: (a) foams with similar densities (26 kg/m^3) and (b) an open-cell PO foam and a PU foam with similar densities. E is the Young's modulus, σ_c is the collapse stress, σ_{75} is the stress at 75% strain and ε_D is the densification deformation.

low stresses, the behavior is linear (linear zone), and it is controlled by cell edges bending in open-cell materials and by cell edges bending and cell faces stretching in closed-cell materials. This region finishes when the structure collapses and a new zone begins (plateau zone). In open-cell foams, this region is characterized by an almost constant stress when the strain increases. However, for closed-cell foams of low density, the stress increases when the strain does because of an additional contribution of the compression of the gas.[1,27,28] Finally, at higher strains (ca. 65% for low density foams), opposite cell walls touch each other, producing a sudden increase in stress; this is known as the densification region.[1]

Figure 3(a,b) shows examples of the stress–strain curves for three foams of similar densities and different structures. Figure 3(a) clearly shows that it is necessary to apply a higher stress (ca. 10 times higher) to produce the same strain in the closed-cell LDPE foam. The expected decrease in the mechanical behavior for the open-cell foams is due both to the presence of holes in the walls that make the structure weaker and to the plastic strain originating during the last stage of the foaming process. It is also interesting to compare the behavior of the open-cell PU foam and that of an open-cell PO foam of a similar density [Fig. 3(b)]. Both curves present stress values of the same order of magnitude; however, although the open-cell PU presents a clear plateau region with constant stress (between 10 and 60% strain), the open-cell PO foam has a constant increase of stress versus strain after collapse and before densification. This interesting fact seems to be related to a very different air flow resistance for both types of materials. It has been shown that the tortuosity and as a result the air flow resistance is much higher for the open-cell PO foams (Table I and Fig. 2), and then during compression, it is difficult for the gas to escape. This resistance of the gas to escape from the foam contributes to the mechanical strength in these materials and is the reason for the nonzero slope in the plateau region.

Figure 4(a) shows the results for the elastic modulus. As previously mentioned, closed-cell foams present the highest values. Even when the polymeric matrix of the closed-cell foam is more flexible than that of the open-cell materials (the closed-cell EVA foam has a lower crystallinity than the open-cell materials), the modulus is 6 times higher. It can also be observed that the PU foam has a slightly higher elastic modulus than open-cell PO foams of similar densities. Finally, an increase in the elastic modulus versus the density has been detected for the open-cell PO foams. The increase can be fitted with good accuracy to a potential law with an exponent close to 2; this result is in agreement with the typical theoretical predictions for open-cell materials [1]
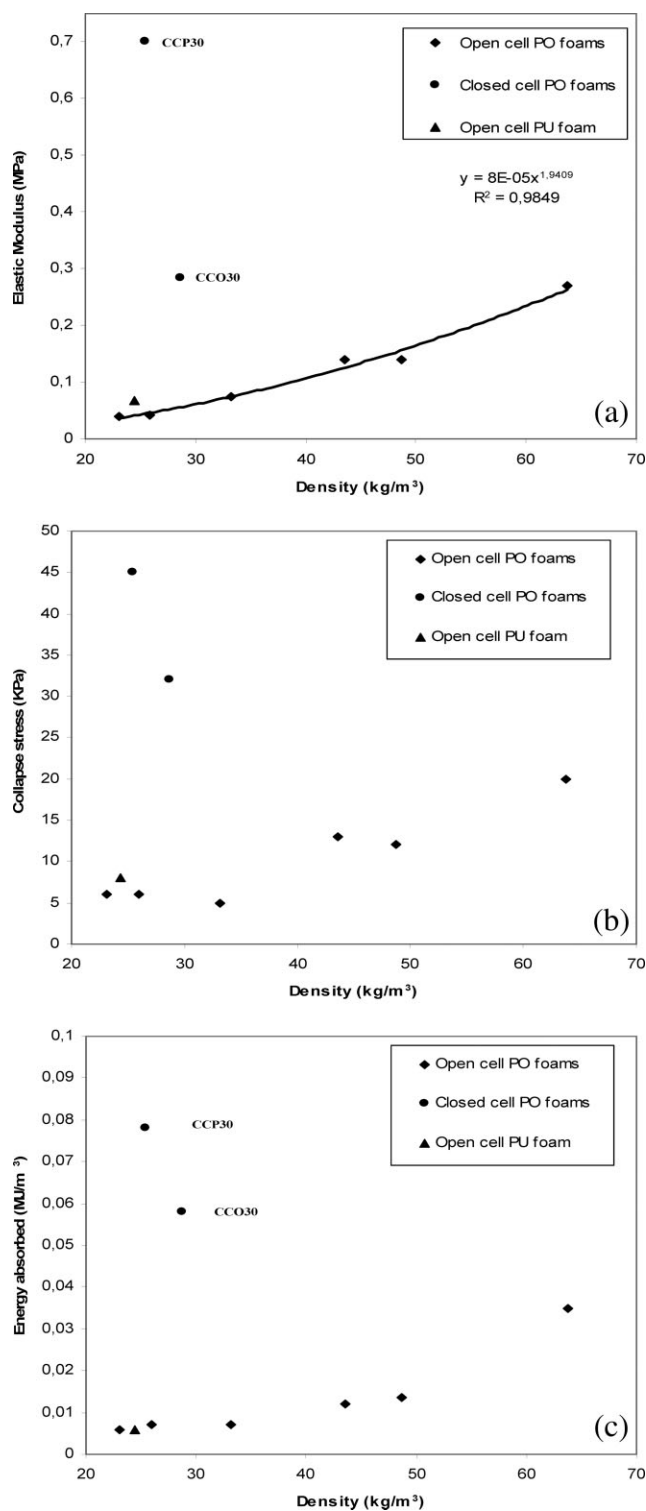
Figure 4 (a) Elastic modulus, (b) collapse stress, and (c) energy absorbed up to 75% strain as a function of density.

The data for the collapse stress and energy absorbed during compression are given in Figure 4(b,c). Trends similar to those observed for the modulus are detected, that is, increases in both characteristics with density, much higher values for
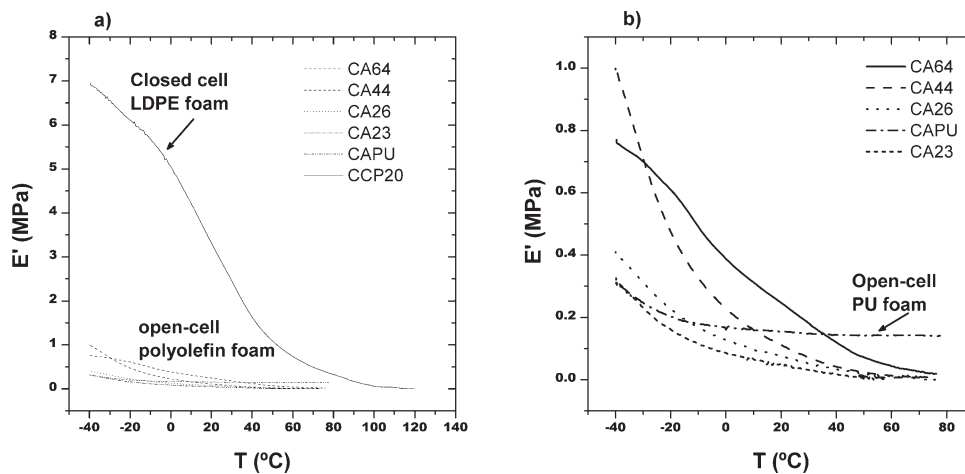
Figure 5 (a) Storage modulus (E') as a function of temperature (T) for some of the foams under study and (b) detail of panel a.

the closed-cell foams, and values similar for open cell foams and PU foam.

Dynamic mechanical behavior and thermal expansion

Figure 5 shows the storage modulus as a function of temperature for some of the materials under study. As expected for thermoplastic-based materials (PO foams), the modulus decreases when the temperature rises. The storage modulus in an open-cell foam of a high density decreases from 0.7 to 0 MPa in the temperature range of −30 to 80°C. In the closed-cell LDPE foam, the decrease is from 6.5 to approximately 0 MPa. A different behavior is shown in the PU foam; in this case, the modulus is 0.25 MPa at −30°C, and from 20°C, the values are almost constant at 0.2 MPa [Fig. 5(b)]. The numerical values show a trend as a function of the density and type of cell similar to that of the elastic modulus (see the previous section); that is, the closed-cell PO foam shows much higher values, the values increase with density for the open-cell PO foams, and the PU foams show values of the same order as those of the open-cell PO foams of a similar density.

Tan δ is a parameter commonly used to characterize the ability of a given material to damp mechanical vibrations; examples of applications in which this factor is taken into account are floating floors in the construction industry and damping layers used to isolate vibrations from engines in the automotive industry. It has been previously demonstrated[19,21,29] that for experiments performed at low strains (like the ones presented in this investigation), the response of closed-cell foams is similar to that of the solid polymer from which the foams are produced. The same relaxations are detected at similar temperatures and with very similar intensities; that is, the gas phase does not play a significant role in the foam damping.
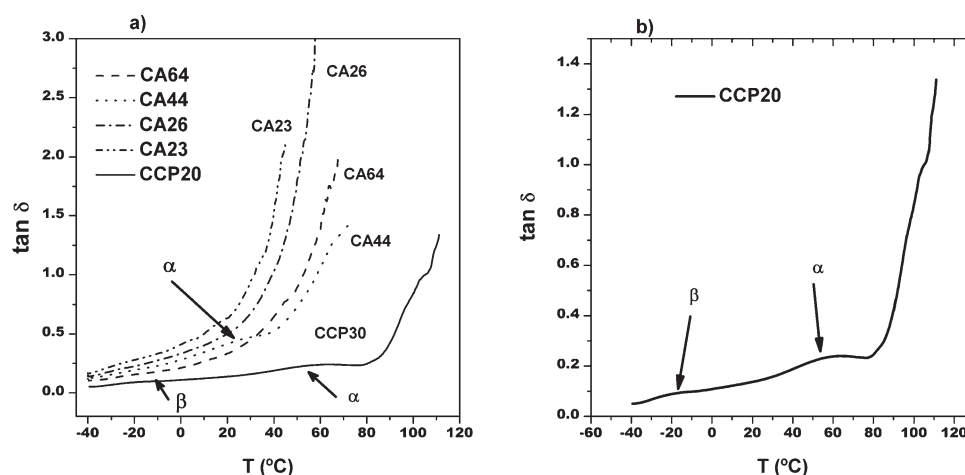

Figure 6 (a) Tan δ as a function of temperature (T) for PO foams and (b) tan δ as a function of T for a closed-cell polyethylene foam (CCP30). The α- and β-relaxations are shown.

TABLE II
Values of Tan δ at 20°C for the Foams Under Study

Foam	CA23	CA26	CA33	CA44	CA49	CA64	CCP30	CCO30	CAPU
Tan δ	0.55	0.48	0.47	0.40	0.32	0.24	0.12	0.16	0.22

Figure 6 shows this parameter as a function of temperature for open- and closed-cell PO foams. Comparing both materials, we can clearly observe that open-cell foams show high values of tan δ in the whole temperature range under study. This significant increase (a doubling of the values at room temperature) seems to indicate that an additional mechanism of damping is taking place in the open-cell materials. This additional mechanism should be related to the connectivity of the gas phase and in particular to the possibility of air flowing through the cellular structure during mechanical cycling. The importance of this mechanism was suggested by Hilyard[30] previously for open-cell PU foams; however, the low tortuosity of those PU materials means that its contribution is much smaller than that observed in the open-cell materials of this study.

Another feature shown in these figures is that the open-cell foams do not exhibit clearly the thermal relaxations typical of the solid polymer (β-relaxation at temperatures around 20°C and α-relaxation at temperatures near 60°C); these relaxations are clearly shown by the closed-cell LDPE foam [Fig. 6(b)]. This fact is another indication of the importance of the air flow mechanism in the damping response. The damping due to the solid matrix is masked by this additional mechanism, which takes place only in the open-cell materials.

In addition, it can be observed in Figure 6(a) and Table II that the tan δ value seems to increase when the foam density is reduced; this can be understood if we take into account that a reduction of the foam density is connected to an increase in the gas phase content. The tan δ value for the PU foam is smaller than that for the open-cell PO foams of similar densities, and this is a logical result when we take into account the lower tortuosity of this material.

Another important aspect that can be detected in Figure 6 is that the maximum temperature of use of the open-cell materials (70°) is clearly lower than that of the closed-cell ones (90°C). Two reasons may be the source of these differences. First, the different chemical composition of the open-cell materials and the presence of EVA should reduce the melting temperature and as a result the thermal stability. The second reason is connected to the presence of an enclosed gas within the structure. It is known[24,31] that the macromolecules composing the cell walls and edges of PO-based foams are oriented. With an increase in temperature, these macromolecules

have a tendency to reach an isotropic configuration, which in foams implies a reduction of cell size and a collapse of the structure. For open-cell materials, there are no additional mechanisms competing with the previous natural behavior of the macromolecular materials; however, in closed-cell ones, the gas inside the cells has a pressure above atmospheric pressure[32] at temperatures above 60°C. This pressure makes more difficult the collapse of the foam at temperatures below the ones at which considerable gas diffusion through the cell wall membranes takes place. As a result of these two competing mechanism (a tendency to isotropy of the macromolecules and gas pressure inside the cells), the closed-cell foams have higher thermal stability than the open-cell ones.

This result was confirmed by measuring the thermal expansion (Fig. 7). At temperatures below 40°C, the open-cell foams have lower thermal expansion than the closed-cell ones; this is due to the gas contribution in the expansion of the closed-cell materials. In addition, the collapse of the open-cell material takes place at a lower temperature than that of the closed-cell material based on EVA. The thermal expansion of the material from which the open-cell foams were produced (preform) has also been included in this figure. In this material, the reduction of thermal expansion occurs at higher temperatures because this is a nonfoamed material and therefore the mechanism of collapse of the cellular
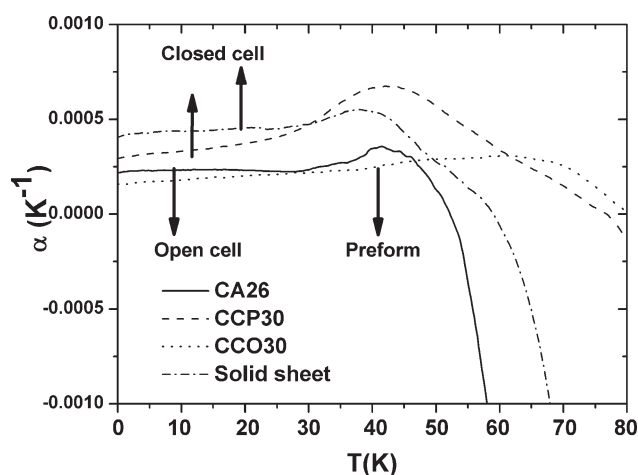

Figure 7 Thermal expansion as a function of temperature (T) for some of the materials under study.

structure observed and discussed for the foams is not taking place.

Acoustic properties

The acoustic absorption for some of the materials under study is shown in Figure 8. The absorption coefficient in open-cell PO foams starts to increase for frequencies above 1000 Hz, reaching maximum absorption values around 0.7 (in the frequency range between 1500 and 2000 Hz; Fig. 8). Once the maximum absorption is reached, the curve does not fall abruptly but maintains a constant value around 0.6 up to frequencies of 6000 Hz. All the open-cell foams show this characteristic behavior. The PU foam shows a very different behavior with a continuous increase of the absorption coefficient, reaching a maximum value of 0.9 at frequencies close to 4000 Hz. Finally, the closed-cell foams have almost zero absorption up to frequencies near 4000 Hz, and then these materials show an absorption peak around 4500 Hz with a value of 0.55.

To analyze the results in more detail, the normalized absorption coefficient (α_n) has been obtained with the following equation:

$$\alpha_n = \frac{\int_{f_1}^{f_2} \alpha(f)df}{f_2 - f_1} \qquad (7)$$

where f_2 and f_1 are the limits in which the absorption coefficient is measured (500 and 6400 Hz).

Figure 9 shows the values of the normalized acoustic absorption coefficient as a function of the open-cell content. It can be observed that a higher open-cell content results in an increase of the absorption coefficient. The optimum material for this property is the open-cell PU foam, although some of the open-cell PO foams have very close values of the
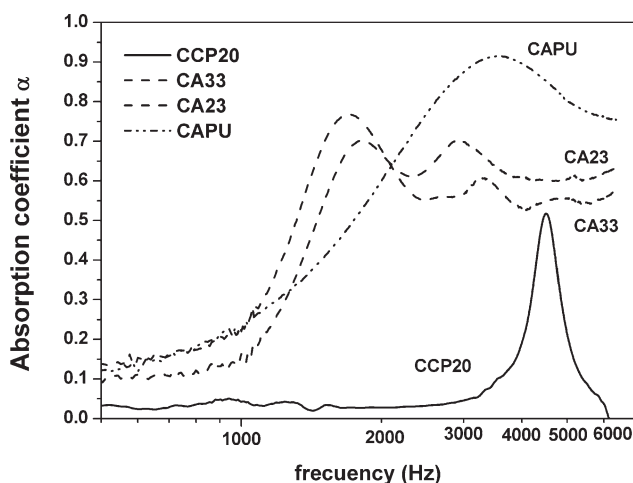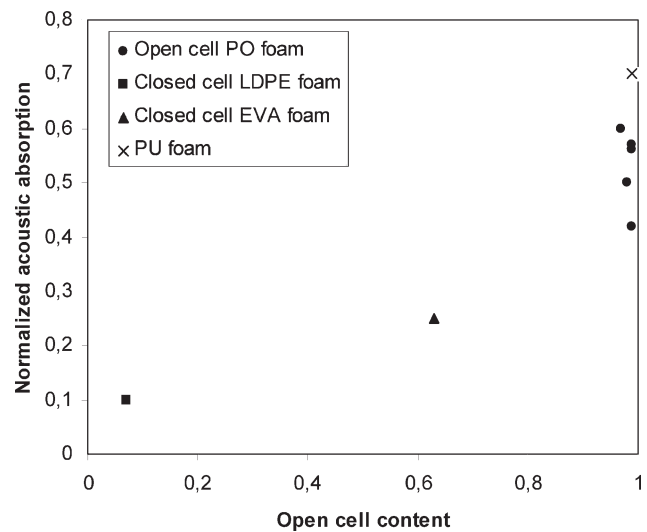

Figure 9 Normalized absorption coefficient as a function of the open-cell content.

normalized absorption coefficient. As expected, the material with an intermediate fraction of open cells (the CCO30 foam) also has an intermediate value of the absorption coefficient.

The opening of the cell walls results in a completely different behavior of the foams as acoustic absorbers. The open-cell PO foams can be considered acceptable acoustic absorbers with values even higher than those of a typical acoustic absorber, such as the PU foam, in the frequency range of 1500–2000 Hz.

CONCLUSIONS

The structure and physical properties of new open-cell PO foams have been studied. It has been found that these new materials present an intermediate structure between that of the standard closed-cell PO foams and that of flexible open-cell PU foams; in others words, they exhibit cell walls, struts, and holes in the walls. It has also been found that the structures of these materials have extremely high tortuosity.

The mechanical property study has revealed that the new open-cell foam presents poor mechanical properties in comparison with closed-cell materials because of the presence of holes in the walls and the plastic strain caused in the manufacturing process (mechanical rupture of the cells). However, the properties are similar to those of open-cell PU foams. On the other hand, these materials present high values of tan δ; that is, the new open-cell PO foams are more suitable for the damping of low-frequency vibrations. The physical reason explaining this fact is the air mobility inside a cellular structure with a very high tortuosity.

Figure 8 Absorption coefficient versus the frequency for some of the analyzed foams.

The open-cell foams have lower thermal stability, which is due to the absence of trapped gas. This gas makes difficult an early collapse of the structure in closed-cell foams.

Finally, the open-cell PO foams have better acoustic absorption than the closed-cell materials. This result was expected because of structural features. In addition, between 1000 and 2000 Hz, these new foams exhibit better absorption than the conventional PU foams.

The completely different properties of open-cell PO foams versus closed-cell materials increase the potential applications of this type of material. In view of the obtained results, they can be considered potential substitutes of flexible PU foams in acoustic absorption and cushioning. In addition, these materials are interesting for the damping of low-frequency vibrations. Finally, it has been shown that one of the key limitations of these novel open-cell materials seems to be related to poor thermal stability.

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