Mechanical Behaviour at Low Strains of LDPE Foams with Cell Sizes in the Microcellular Range: Advantages of Using These Materials in Structural Elements

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ABSTRACT

This paper presents the production method and the compressive mechanical response at low strains for a collection of polyethylene foams with high densities and cell sizes in the microcellular range. The materials were produced using an improved compression moulding technique that allows and independent control of density and cell size.

The materials had a relative density between 0.27 and 0.92, an homogeneous and multi-structured cellular structure with dense skin and foamed core and cell sizes in the range 30 to 100 microns. The Young´s modulus decreased with density. For relative densities higher than 0.7, the reduced Young's modulus of the foams was higher than that of the solid. In addition, it has been proved that variations in the cell size at constant density did not influence the Young's modulus. The advantages of using these materials for the production of plastic pipes have been analysed. In comparison with a solid pipe a reduction of the weight of foamed pipes loaded in compression of up to 40% can be reached.

1. INTRODUCTION

Microcellular plastics are foamed polymers characterised by cell sizes averaging 100 microns or less, typically between 5 and 50 microns(1). It has been proved

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that these materials exhibit high Charpy impact strength, high toughness, high fatigue life, high thermal stability and low thermal conductivity[2] than solid polymers. Because of these unique properties, a large number of innovative applications can be imagined. For instance, food packaging with reduced materials costs, pipes or panels with improved strength to weight ratio, airplane and automotive parts with improved strength and acoustic dampening, sporting equipment with reduced weight and high energy absorption, etc. Due to these potential applications, over the last two decades substantial research and development has been conducted on the topic of microcellular processing and characterisation of microcellular products[3-10].

Microcellular plastics were initially produced at Massachusetts Institute of Technology (MIT) using a batch process[11]. This process has been mainly used to produce microcellular foams of amorphous materials with cells sizes around 10 microns[12].

The previous concept has been extended to the production of microcellular foams by injection moulding and extrusion[1,2,13-15], and although considerable progress has been reached, there is still a need for further research in several areas. For instance, in injection moulding there is a poor surface quality of the produced products in conventional processing[1,15], three is a limitation in the maximum size of the produced parts[1], and a limited weight reduction of the parts, not higher than 30-35%[1]. In extrusion, most of the studies have been performed at a laboratory scale or at a semi-industrial scale and as far as we know microcellular foams of semicrystalline polymers are not produced industrially by extrusion. Only polystyrene has been successfully commercialised[12].

Another open aspect is connected with the physical and mechanical characterisation of these products. Although several investigations have analysed the mechanical properties[1,4,7-10] there is still a need to know, for several polymer systems as polyolefins, the effect of cell size on the elastic response, the effect of the presence of a thick skin on the strength, the influence of the open cell content and the properties of these materials in comparison with foams produced from conventional technologies, etc.

Finally, another open aspect of interest is the identification of industrial areas in which these materials can play a key role[12]. One area of potential interest is plastic pipes of higher stiffness and lower weight with the idea of replacing the solid material. As it is expected that microcellular foams could have better mechanical properties than conventional foams this approach seems to be an interesting way of improving the properties of conventional pipes.

Bearing the previous ideas in mind, this paper focus on the following aspects: first a novel route, improved compression moulding technique or Pucel Technique, to produce foams of semicrystalline polymers with cell sizes in the microcellular range is explained, second the elastic properties of these foams are characterised and third the potential of these materials for the production of plastics pipes is evaluated by performing finite element modelling. The aims of the paper are to gain knowledge on both the production, the structure-property and the potential applicability of high density foams with cell sizes in the microcellular range.

2. MATERIALS

A low density polyethylene (PE008) provided by Repsol Quimica was used as matrix polymer (MFI = 4 g/10 min at 190 ºC, density 920 kg/m^3). Azodicarbonamide with an average particle size of 4.9 μm provided by Urquinsa (Spain) was used as blowing agent. Typical amounts of stearic acid (Stearic Acid 301 supplied by Renichen S.L) and zinc oxide (Silox Active grade provided by Safin Alcan Spain) were used as processing aid and catalyser of the decomposition reaction of the blowing agent respectively[16-18]. No crosslinking agents were used; therefore non crosslinked cellular materials were fabricated.

Foams were produced by an improved compression moulding technique[19]. In this process, first a precursor solid material that incorporates all the chemical compounds needed was produced by melt blending and compression moulding. Then the precursor material was foamed as follows.

The precursor material that contains the blowing agent was inserted in a mould with an internal diameter equal to that of the precursor material (**Figure 1**). During the heating of the precursor material up to the decomposition temperature of the blowing agent (180 ºC in this paper), which yields the blowing agent decomposition, a pressure high enough (around 200 bars) to avoid the foam expansion was applied by using a piston connected to a hydraulic press. When the entire blowing agent was decomposed, pressure was released and the piston moved to its final position. Foaming took place during the piston displacement. After this the mould was cooled and the stabilised foam was extracted from the mould.

In this process, the foams grow under pressure, and the expansion ratio, and as consequence the density, is controlled by the displacement of the piston that applies the pressure. This has a significant advantage over the conventional

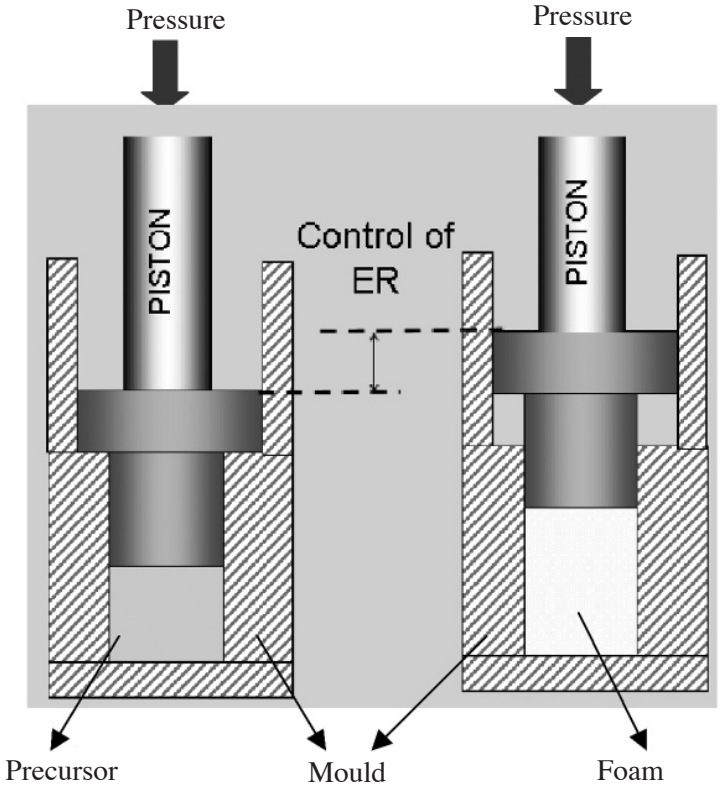


Figure 1. Schematic diagram showing the last step of the improved compression moulding technique. The expansion ratio (ER) is controlled by the piston displacement

compression moulding method[16-18] in which pressure is reduced to atmospheric pressure during decompression and density is controlled by formulation and processing parameters. In this improved compression moulding method density can be easily controlled and it is independent on the formulation. Due to this reason density and cell size can be controlled in an independent way.

Densities of the produced foams ranged between 250 kg/m^3 and 750 kg/m^3, i.e. relative densities in the range 0.27-0.92. Cylindrical samples of 22.8 mm in diameter and 17.5 mm in height were produced.

3. EXPERIMENTAL

Density Measurements

Density measurements were performed by Archimedes principle using the density determination kit for a AT261 Mettler balance.

Scanning Electron Microscopy (SEM)

Quantitative analysis was used to characterize the cellular structure. For this purpose, sections parallel to the cylinders height were microtomed at low temperature to provide a smooth surface that was vacuum coated with gold and examined by SEM using a JEOL JSM 820. Cell size distributions were measured and used to determine the average cell size. Cell density was calculated as the number of cells per unit volume of the unfoamed material using the equation (2):

$$N = \left(\frac{n_b}{A}\right)^{3/2}\left(\frac{\rho_s}{\rho_f}\right) \qquad (1)$$

where n_b is the number of cells in a defined area A, ρ_s is the density of the unfoamed solid and ρ_f is the density of the foam.

Compression Experiments

Compressive stress(s) strain (ε) curves were measured using an Instron machine (model 5500R6025) at room temperature and at a displacement rate of 1 mm/min. The maximum static strain was approximately 75% for all the experiments. These experiments were used to determine the foam Young's modulus[20,21].

Finite Element Modelling (FEM)

The size of the modelled solid pipes was nominal diameter D = 315 mm, thickness 8 mm, modulus of elasticity of the solid material E = 200 MPa, length of the pipe L = 300 mm. The pipe thickness was used as an adjustable parameter in the variational analysis. The compressive applied load was selected in order to have a defection near to 3% of the nominal radius, i.e. 9.45 mm. The simulation was done in three phases.

Initially, the quasi static structural simulation of the configurations was carried out for a 8 mm thick LDPE pipe. The analysis used a normal Lagrange formulation to consider the change in the contact zone . The mesh was generated using Brick 8node 185 and Mapped Face Meshing elements.

In a second step the simulation was performed using the same type of approximations and meshes but in this case foam pipes were simulated. In order to do that, the relationship between Young's modulus and density of the foam was introduced in each model.

In a third step, a variational analysis (Goal Driven Optimization) was performed in order to select the density and thickness of the pipe with optimum performance (similar stiffness than that of the dense pipe and minimum weight). The analysis was carried used a multi-objective genetic algorithm (MOGA) which is able to optimize problems with continuous input parameters.

In all cases, the selected input parameters were the foam density, the Young's modulus and the pipe thickness. The response parameters were the maximum pipe deformation, the maximum stress and the pipe weight. The objective was to find out the pipe (density and thickness) with a similar stiffness to that of the dense pipe and with a minimum weight.

4. RESULTS

Cellular Structure

Figure 2a shows the typical cellular structure of the inner part of one of the foams (density 550 kg/m^3). It can be observed that the foam has a very homogeneous closed cell cellular structure. Cells with diameters lower than 50 microns are observed. In addition, the foams presented skin-core morphology as it can be appreciated in **Figure 2b** for the same material. The skin has a higher density than the core, giving materials with a spatial mass distribution which in general terms optimises mechanical properties and surface quality.

Figures 3a and **3b** summarises several general trends of the average cell size, and cell density for some of the foams under study. As expected for foams produced using chemical blowing agents[22] cell size is reduced and cell density is increased when density is increased. Appreciable values of cell density (higher than 10^8 cells/cm^3) were obtained for high density foams. These values strongly decreased for the lower density foams, which seems to be due to a strong coalescence when the density of the foams was reduced.

By controlling the expansion ratio during the process (**Figure 1**) and the formulation of the precursor materials it was possible to produce foams with relative densities in the range 0.27-0.92 and with average cell sizes in the range 25-100 microns, as it is depicted in **Figure 3c**. The possibility offered

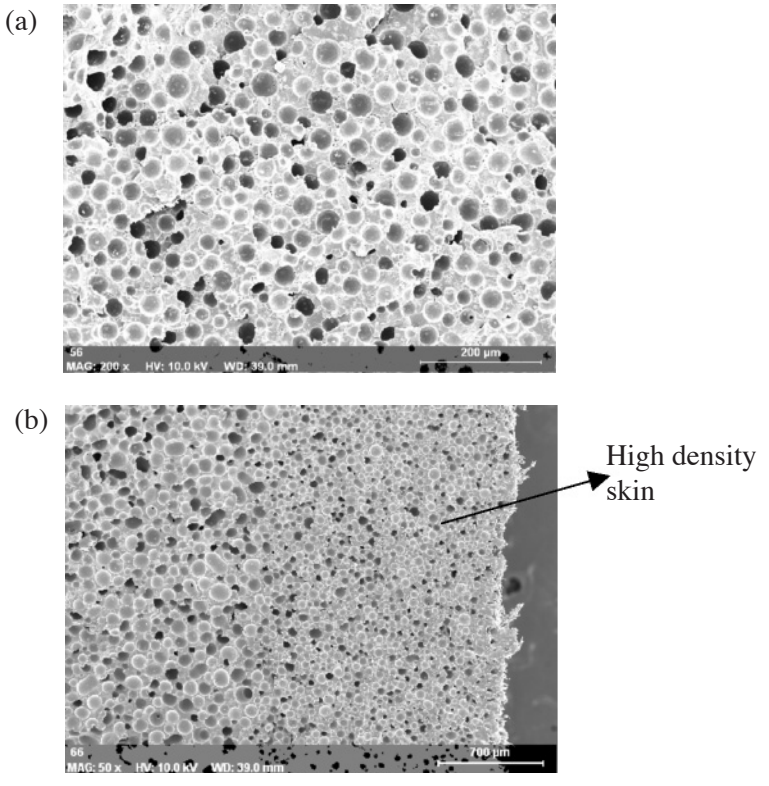


Figure 2. a) SEM micrograph of the inert part of one of the produced foams (density 550 kg/m^3); b) skin of the same material

by the improved compression moulding technique of fabricating foams with a similar density but with a different cellular structures made possible analysing independently the effect on both parameters in the mechanical response as it is explained in the next section.

Mechanical Properties

Figure 4 shows the Young's modulus of the foams as a function of cell size for a given relative density (0.60 in this case). Similar results were found for other densities. It can be observed that Young's modulus showed constant values in the cell size range between 30 and 100 microns. This result that was also found for low density foams[23], has a significant practical importance because indicates that for materials working in the elastic regime controlling the cell size is not the key objective. Other structural parameters such skin

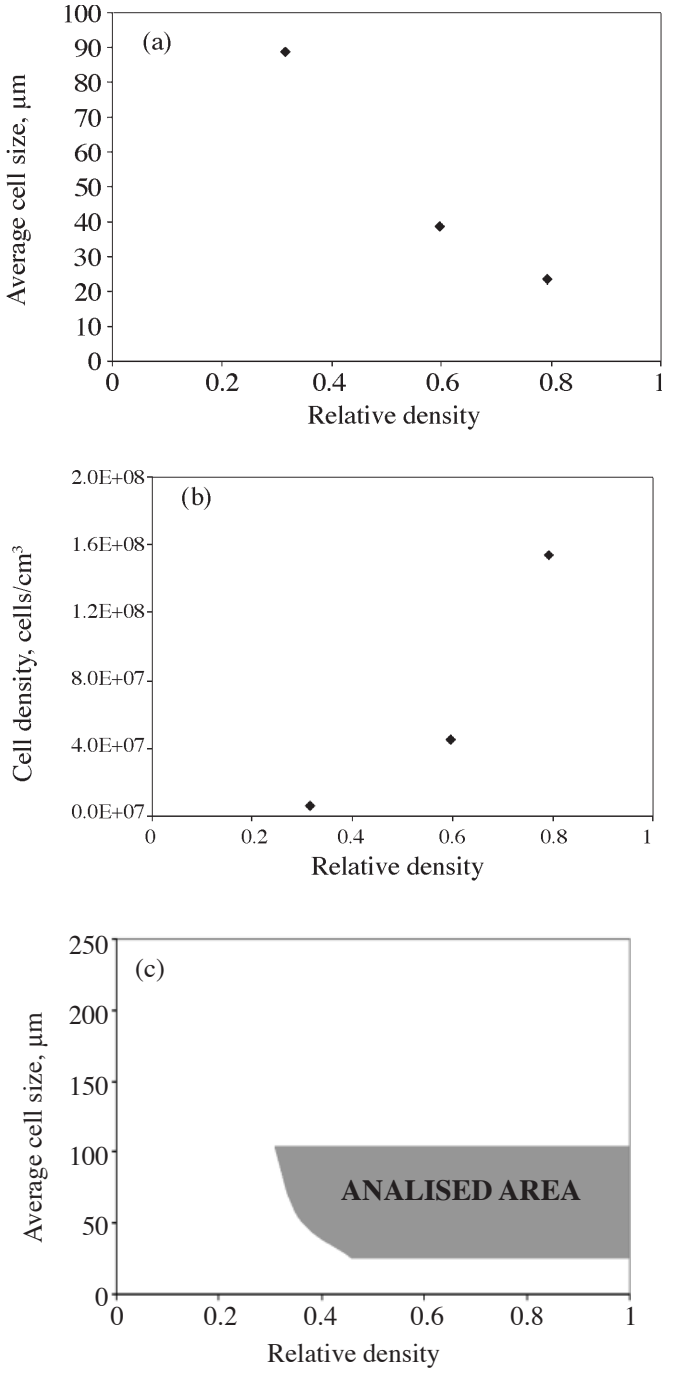


Figure 3. a) Average cell size *vs*. relative density, b) Cell density *vs*. relative density c) Density and cell size of the analysed foams

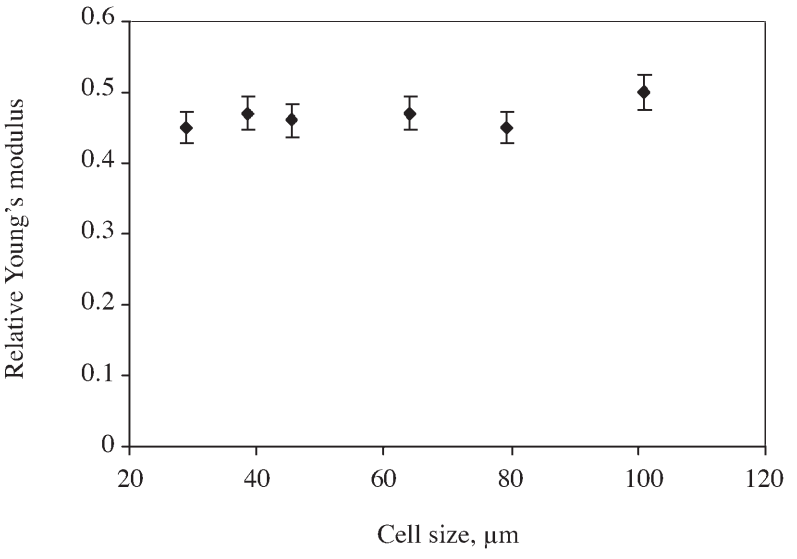


Figure 4. Relative Young's modulus as a function of cell size for foams with a relative density of 0.6

thickness, open cell content, homogenous cellular structure, etc seems to be more important.

As mechanical properties did not showed a dependency with cell size the average values of the Young's modulus for foams of a given density were obtained and analysed as a function of the foams density. **Figure 5a** shows the experimental results compared with two theoretical estimations that were obtained using a potential law of relative Young's modulus versus relative density (equation 2) with exponent n = 1 and exponent n = 2. C was assumed to be 1 in these plots[20].

$$\frac{E_f}{E_s} = C\left(\frac{\rho_f}{\rho_s}\right)^n \qquad (2)$$

where E_f and ρ_f are the foam Young's modulus and the foam density and E_s and ρ_s are the modulus and density of the solid material.

Values of n between 1 and 2 are typical for a wide amount of cellular materials[20], therefore it is expected that most foams, including the ones in this paper, would have properties between these two trends.

The materials under study followed an approximately linear trend given by equation 3.

$$\frac{E}{E_s} = \left[1.292\,\frac{\rho}{\rho_s} - 0.285\right] \qquad (3)$$

The equation has an intercept of -0,285 (the expected value should be close to zero) indicating that the behaviour at lower densities should be different, (i.e. this equation is not able to predict the behaviour of these materials in the whole relative density range; it is only valid for relative densities above 0.3).

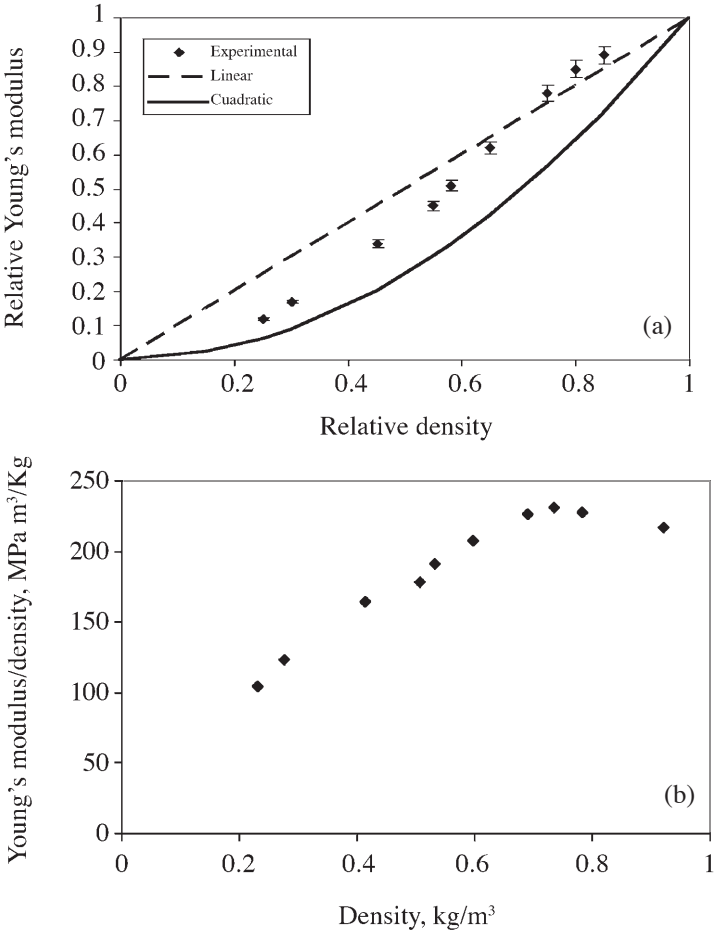
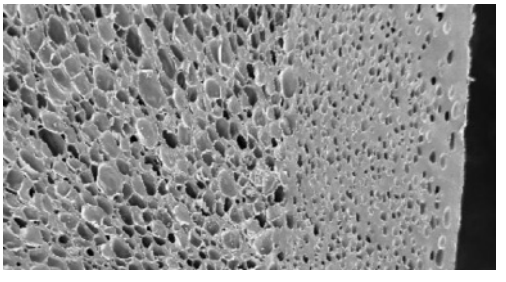


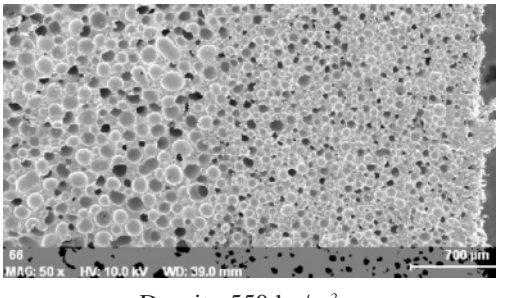
Figure 5. a) Relative Young's modulus as a function of density; b) Reduced Young's modulus as a function of density

A detailed analysis of **Figure 5b** shows different trends depending on the foam density. At high relative densities the mechanical properties of the produced foams are slightly above the linear trend. This shows that in this density range the foams have very good mechanical properties. In fact, it can be observed in **Figure 4b**, that the reduced Young's modulus (modulus divided by density) was higher for these foams than for the solid from which the materials were produced. Two different contributions are the source of this behaviour. First, the solid material (relative density 1) did not include in the formulation the blowing agent; as the blowing agent residues (that are included in the foam) could as a reinforcement, a slightly improvement of the mechanical properties of the solid matrix is expected in the foams in comparison with the solid precursor. In addition, as it is explained in the next paragraph, the foams with higher densities present a solid thick skin which performs as a reinforcement improving the mechanical behaviour.

For relative densities below 0.7 the properties are between the linear trend and the parabolic one. For densities below 0.5 the properties are closer to those of a potential law with exponent 2. Two different structural sources can be the origin of this non-unique trend with foam density. First reason is related with a different thickness of the skin when the foam density was modified. **Figure 6**



Density 730 kg/m^3



Density 550 kg/m^3

Figure 6. Presence of a more dense skin in samples with a higher density

shows that at low densities the skin seems to have a smaller density, which would reduce the mechanical properties. Second reason would be related with the open cell content in the foams. As the low density polyethylene of this study was not crosslinked, it is expected an increase of the open cell content in the materials when the density was reduced, it is well known that an increase of the open cell content will also reduce the foams stiffness[24].

FEM of the Foams Behaviour in Plastic Pipes

Table 1 shows the results for the finite element modelling comparing the behaviour of a solid pipe with that of the optimum pipe for this application (using a selected material from those produced in the paper). Both pipes were selected with dimensions to assess an equal annular stiffness (the same deformation for a similar compressive load). The pipe produced from the solid material had a density of 920 kg/m^3 and a thickness of 8 mm. The optimum foam had a density of 312 kg/m^3 and a thickness of 14.3 mm. It was calculated (last column) that the pipe produced with the foam has a smaller weight (1.26 kg in spite of 2.1 kg), therefore a 40% reduction in weight was obtained for a similar annular stiffness.

In addition, another advantage is that the maximum stress on the foamed pipe was much smaller (**Table 1** and **Figure 7**). In this case the reduction was much higher, from 1.37 MPa in the solid pipe to 0.42 MPa in the foamed pipe (a 69% reduction).

Finally, a map for pipes with equivalent bending stiffness was obtained (**Figure 8**). In this diagram the mass of the pipe is presented as a function of the foam density, three types of materials are considered. First, ideal materials, following a power law relationship between modulus and density with exponent equal to one, second typical foams that it is known[20] follow a power law relationships between modulus and density with exponent equal to two, and third the materials analysed in this paper that follow the relationship given in equation 3.

Several interesting conclusions are obtained from this figure. On the one hand, it can be concluded than for pipes loaded in compression, the minimum

Table 1.

Case	Thickness (mm)	Density (kg/m^3)	Deformation (mm)	Stress (MPa)	Mass (kg)
Solid Pipe	8	910	9.43	1.37	2.10
Foam Pipe	14.3	312	9.34	0.42	1.26

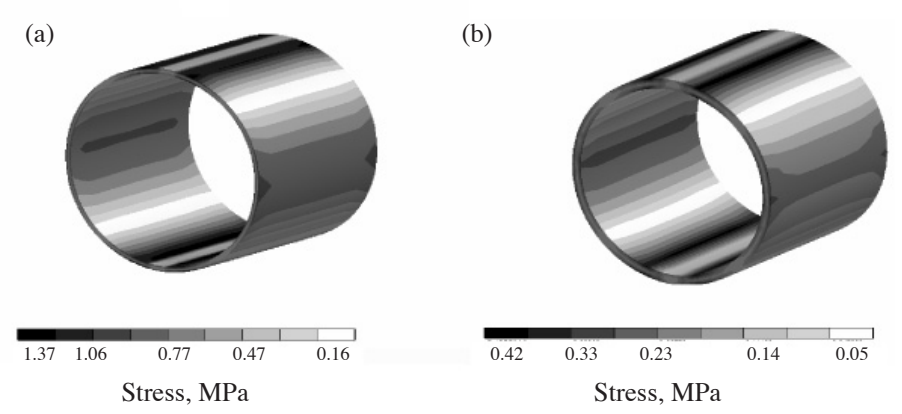


Figure 7. Equivalent stress distribution of a) solid polymer b) foam of 310 kg/m^3 in density. Values in the scale bar are in MPa.

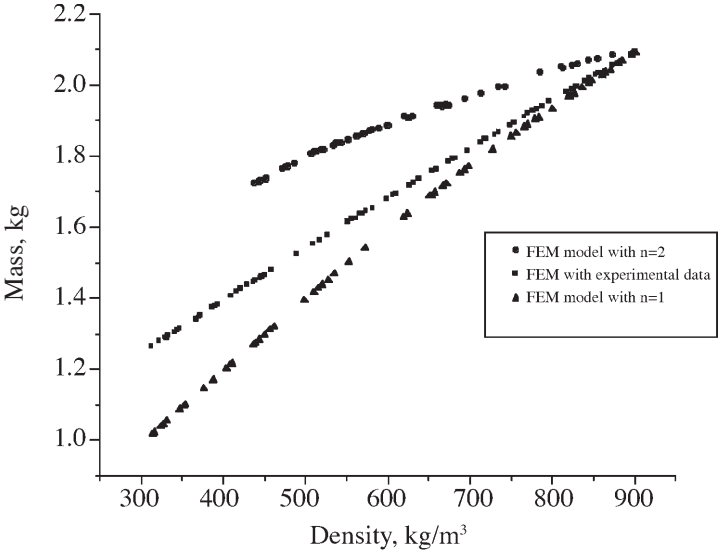


Figure 8. Mass of *vs* foam density for pipes with equal stiffness. The three curves correspond to: (linear) materials following a power law relationships with exponent equal to one, (quadratic) materials following a power law relationships with exponent equal to two, and (experimental data) the materials analysed in this paper that follow the relationships given in equation 3

weight for a given stiffness is reached by using low density foams; in other words it can be said that for this geometry (pipes loaded in compression) foams are stiffer materials than solids at equivalent weight. On the other hand, the maximum weight reduction (minimum panel weight) is obtained for materials with a power law with exponent equal to one, followed by the materials in this study and the materials following a quadratic trend with density. The weights reductions for a relative density of 0.5 are: linear law 38%, experimental data 28%, quadratic law 18%. These data clearly point out the importance of producing foams in which the mechanical properties at low strains take values close to the linear relationship. As it can be observed in the data, the materials produced in this paper give very close weight reductions to the ideal trend for high density foams and intermediate values between the optimum values and the ones obtained using the quadratic trend for mediums and lower density foams.

5. SUMMARY AND CONCLUSIONS

A collection of high density polyethylene foams with cell sizes in the microcellular range have been produced using a novel procedure based on an improved compression moulding technique. This process allows controlling the foam density and the cell size in an independent way.

The cellular structure of the foams was very homogeneous with cell sizes around 50 microns, having the materials a skin-core morphology. The cell size increased and the cell density decreased with a reduction of the density as it is expected for materials produced from a chemical blowing agent. Materials with different cell sizes for a given density were produced, but no improvement of the Young's modulus and the collapse stress was obtained using this strategy. The Young's modulus followed a linear trend with reduced values higher than those of the solid material at high densities. Using finite element modelling it has been demonstrated that the materials can be used to produce light weight structural pipes. Weight reductions up to 40% can be reached using the foams produced in this paper with densities in the range of 300 kg/m^3.

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