# High Density Polyethylene Foams. III. Tensile Properties

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**ABSTRACT:** The room temperature tensile properties of closed-cell polyethylene foams have been investigated. High density polyethylene (HDPE) foams of four different molecular weight were used to study the effect of molecular weight and foam density on mechanical properties during tension and at the break point. It was found that increasing the molecular weight changes the tensile behavior of polyethylene foams from brittle to ductile fractures. For brittle foams, the break strength follows a square power-law model and the break strain is independent of the volume fraction of the voids. For ductile foams, the normalized yield strength

# also follows a square power-law relation with normalized density, the yield strain is similar to the value of the solid polymer and remains constant for all void volume fractions, and the break strain increases with HDPE molecular weight. Finally, the toughness of the foams was found to increase with normalized density and HDPE molecular weight. © 2003 Wiley Periodicals, Inc. J Appl Polym Sci 90: 2130–2138, 2003

**Key words:** polyethylene; mechanical properties; modulus; toughness

#### INTRODUCTION

Plastic foams can be produced in a great variety of densities, ranging from about 1 kg/m<sup>3</sup> to solid polymer density. Since the density of a cellular material profoundly affects its mechanical properties, the end use of a foam is usually determined by its density.<sup>1</sup> Because the density of polymers varies from one polymer to another, the normalized density (the foam density divided by the polymer density) is used to eliminate the effect of the matrix. Polyethylene foams with normalized densities above 0.3 were first used for wire and cable insulation and structural purposes.<sup>2</sup> According to Gibson and Ashby,<sup>3</sup> at a normalized density of about 0.3, there is a transition from a cellular structure to one that is better described as a composite consisting of a matrix and isolated voids. In the first and second parts of this series of papers, the effect of high density polyethylene (HDPE) molecular weight on the morphology and the elastic modulus of HDPE foams was studied for normalized densities higher than 0.4.<sup>4,5</sup> In this case, HDPE foams can be considered as two phase composite materials because they have a closed-cell structure for the range of molecular weights and blowing agent concentrations studied. We also found that increasing the molecular weight results in a decrease of foaming grade and cell sizes.

In this third article, we report the tensile properties of HDPE foams in relation to their molecular weight and void volume fraction. The tensile properties will be compared with available mechanical models for composites and foams in order to determine which one best fits our measurements.

For low density cellular materials, Gibson and Ashby<sup>3</sup> developed some models for the tensile and compression properties of both open and closed-cell foams. For higher density foams, Moore and coworkers<sup>6,7</sup> experimentally observed that a square powerlaw relation between the normalized tensile strength and the normalized density produced a good representation of the data. Throne<sup>8</sup> also found this relation to be valid for several foams. On the other hand, Smith<sup>9</sup> developed a model for strain in two phase composites for which the matrix is filled with rigid spherical inclusions. Later, Nicolais and Narkis<sup>10</sup> developed a tensile strength model for particulate composites, neglecting the interaction between the matrix and the inclusions. Finally, Nielsen<sup>11</sup> also developed models for the strain and tensile strength of particulate composites.

Many reports are available on the tensile properties of cellular materials.<sup>1–3,6–8,12–17</sup> Unfortunately, no information could be found on the effect of the polymer molecular weight on the mechanical performance of the resulting foams.

#### EXPERIMENTAL

### Polymer and sample preparation

Four HDPE foams with different melt indices were used in this study. J60-1700-173, A60-70-162, and G60-110 were obtained from Solvay Polymers. HBW555-Ac is a high molecular weight polyethylene from Nova

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TABLE I Characteristics of HDPE Foams Used

HDPE	Manufacturer	$M_w^*$
J60-1700-173 A60-70-162 G60-110 HBW5555Ac	Solvay Polymers Solvay Polymers Solvay Polymers Nova Chemicals	$\begin{array}{c} 5.83 \times 10^{4} \\ 3.25 \times 10^{5} \\ 6.51 \times 10^{5} \\ 7.56 \times 10^{5} \end{array}$

\* From ref. 4.

Chemicals. The molecular weights and suppliers are shown in Table I. Azodicarbonamide (ACA, Sigma Chemicals) was used as a chemical blowing agent whose decomposition temperature range is  $190-240^{\circ}$ C and gas number is  $230-270 \text{ cm}^3/\text{g}$ . Concentrations between 1 and 3% were used based on total weight of all components. The foam plates ( $60 \times 60 \times 2.8-3.4$  mm) were produced by a compression molding method. More details on the polymers, the processing procedure, and foam properties can be found in the first part of this series.<sup>4</sup>

#### Mechanical measurements

Room temperature uniaxial tension mechanical properties were evaluated as a function of foam density and molecular weight using an Instron 5565 tester with a 500 N load cell. The samples were cut in a type IV format according to ASTM D-638. From each experiment, the modulus, tensile yield strength, tensile yield strain, break strength, break strain, and rupture energy were obtained. The initial length between the clamps was 25 mm, and a crosshead speed of 10 mm/min was used. Test procedures are described in the second part of this series.<sup>5</sup> A minimum of eight specimens were tested for each condition. In each case, the results are reported as the average value plus/ minus one standard deviation.

#### **RESULTS AND DISCUSSION**

#### Mechanical properties

#### Tensile behavior of foams

Typical tensile stress-strain curves of the four HDPE foams are shown in Figure 1. Foams with different ACA concentrations have similar behaviors. However, the lowest molecular weight HDPE foam (J60-1700-173) had a brittle behavior, while the others behaved like ductile tough solids, especially HBW555Ac. Because HBW555Ac has high molecular weight (7.56  $\times$  10<sup>5</sup>), its melt viscosity is higher than that of G60-110 and A60-70-162. An excessively high viscosity could prevent HBW555Ac from obtaining a high crystalline content compared to G60-110 and A60-70-162 in compression molding. This will affect the mechanical properties, such as yield stress and modulus. The un-

foamed HBW555Ac has a lower Young's modulus and tensile yield stress than G60-110 and A60-70-162. Therefore, HBW555Ac foam has a lower yield stress than G60-110 and A60-70-162 with similar density. It is known that the brittle-ductile transition of a polymer depends upon strain rate, chemical structure, and mostly temperature. Above a certain temperature at a set strain rate, the material is ductile. It is also known that molecular weight does not appear to have a direct effect on yield strength, but it does affect brittle strength.<sup>18,19</sup> Flory<sup>20</sup> proposed that the brittle fracture stress of a polymer is related to the number average molecular weight ( $M_n$ ) as

fracture stress = 
$$A - \frac{B}{M_n}$$
 (1)

where A and B are constants for a given polymer. Vicent<sup>21</sup> also gave evidence that this relationship holds as a rough approximation for the brittle strength of several polyethylenes. Eq. (1) implies that higher molecular weight polymers have higher brittle fracture stress for a constant chemical structure and strain rate. Equivalent to a brittle-ductile transition temperature, there seems to be a brittle-ductile transition molecular weight. For unfoamed samples, this molecular weight would be lower than  $5.83 \times 10^4$  for polyethylene under the conditions in our study. The addition of voids in the system will increase the brittleness and the brittle-ductile transition molecular weight will shift to higher values. This could explain why many ductile materials such as HDPE and poly(propylene) (PP) lose tensile characteristics, such as gross necking and elongation characteristic, after foaming and behave as brittle materials.8 From our results, this molecular weight would be between  $5.83 \times 10^4$  and 3.25 $\times$  10<sup>5</sup> for polyethylene under the conditions used in



Figure 1 Stress as a function of strain for HDPE foamed with 3% ACA.



J60-1700-173

A60-70-162

**Figure 2** Normalized modulus as a function of normalized density. Solid line represents Moore's empirical square power-law relation and differential scheme.

our study. Therefore high molecular weight is a very important factor to consider for polymer foams under tensile loads.

### Young modulus

The normalized modulus as a function of normalized density and molecular weight is shown in Figure 2. Foam moduli are discussed in the second part of this series.<sup>5</sup> It was found that all four HDPE foams follow the differential scheme very well:

$$\frac{E_f}{E_m} \approx (1 - f)^n \quad (1.93 \le n \le 2.01)$$
(2)

where  $E_f$  is the foam modulus,  $E_m$  is the matrix modulus, f is the volume fraction of voids, and n is the power index whose value is related to the Poisson ratio of the matrix, as given in Table II. In our case, a Poisson ratio of 0.34 for polyethylene has been used and eq. (3) becomes the well known empirical square power-law relation:<sup>7</sup>

$$\frac{E_f}{E_m} \approx (1 - f)^2 \tag{3}$$

This implies that the foam modulus is related to the matrix modulus in the same way for foams of similar polymers but different molecular weights. Because it is difficult to produce samples with exactly the same volume fraction of void and the same cell dimensions for different molecular weight polyethylenes, it is reasonable to define a new parameter as  $E_f/[E_m(1 - f)^2]$  to eliminate the effect of the void volume fraction. The effect of molecular weight on this parameter is shown in Table III. It can be seen that the values of  $E_f/[E_m(1 - f)^2]$  are equal to 1 within the error range. This means that molecular weight does not have an effect on this modified normalized modulus.

### Tensile stress at the yield point

The yield stress is an important mechanical property because it indicates the maximum load that the composite can sustain without experiencing plastic deformation. The relationship between the true and apparent stresses (or engineering stress) is

$$\sigma_{true} = (1 + \varepsilon_{Eng})\sigma_{Eng} \tag{4}$$

where  $\sigma_{true}$  is the load divided by the actual area, called actual stress;  $\sigma_{Eng}$  is the load divided by the initial area, called the apparent stress or engineering stress; and  $\varepsilon_{Eng}$  is the elongation per unit length, called engineering strain. Eq. (4) is valid before the yield point.

Yield stress can be defined as the true stress at the maximum observed load.<sup>19</sup> Because this stress is reached at a relatively low sample elongation, it is often adequate to use the engineering definition of yield stress as the maximum observed load divided by the initial cross-sectional area. The engineering definition of yield stress was adopted in our case.

The yield stress could be calculated only for our three high molecular weight HDPE foams. The curve of normalized yield stress as a function of normalized modulus is shown in Figure 3, where a linear dependence of the normalized stress with normalized modulus is obtained:

$$\frac{\sigma_{yf}}{\sigma_{ym}} \approx \frac{E_f}{E_m} \tag{5}$$

where  $\sigma_{yf}$  is the foam yield stress and  $\sigma_{ym}$  is the matrix yield stress. The insertion of eq. (2) into eq. (5) gives

$$\frac{\sigma_{yf}}{\sigma_{ym}} \approx \frac{E_f}{E_m} \approx (1 - f)^n \quad (1.93 \le n \le 2.01) \tag{6}$$

 TABLE II

 Relationship between Power Index of Eq. (2) and Poisson Ratio

			-		-			
$\overline{\nu_m}$	0.02	0.06	0.10	0.12	0.16	0.20	0.22	0.26
n	1.94	1.96	1.97	1.98	1.99	2.00	2.00	2.01
$\nu_m$	0.30	0.32	0.36	0.40	0.42	0.46	0.49	0.495
п	2.01	2.00	2.00	1.98	1.97	1.95	1.93	1.93

1.0

HD	PE			$E_f/[E_m(1-f)^2]$		
Name	$M_w$	1.0%	1.5%	2.0%	2.5%	3.0%
J60-1700-173 A60-70-162 G60-110 HBW5555Ac	$\begin{array}{c} 5.83 \times 10^{4} \\ 3.25 \times 10^{5} \\ 6.51 \times 10^{5} \\ 7.56 \times 10^{5} \end{array}$	$\begin{array}{c}$	$\begin{array}{c} 1.09 \pm 0.05 \\ 1.02 \pm 0.11 \\ 1.01 \pm 0.12 \\ 1.05 \pm 0.04 \end{array}$	$\begin{array}{c} 0.94 \pm 0.11 \\ 1.13 \pm 0.20 \\ 1.17 \pm 0.10 \\ 1.02 \pm 0.08 \end{array}$	$\begin{array}{c} 1.00 \pm 0.21 \\ 1.26 \pm 0.30 \\ 0.99 \pm 0.16 \\ 1.23 \pm 0.08 \end{array}$	$\begin{array}{c} 0.96 \pm 0.09 \\ 1.15 \pm 0.18 \\ 1.12 \pm 0.06 \\ 1.02 \pm 0.09 \end{array}$

 TABLE III

 Effect of Molecular Weight on Normalized Modulus for Different ACA Concentrations (%)

For polyethylene, a Poisson ratio of 0.34 can be used and eq. (6) becomes the square power-law model:<sup>7,8</sup>

$$\frac{\sigma_{yf}}{\sigma_{ym}} \approx (1 - f)^2 \tag{7}$$

Gibson and Ashby<sup>3</sup> assumed that a plastic foam yields in tension by the same mechanism and at essentially the same stress as in compression, but the post-yield behavior is different. For the collapse strength of closed-cells foams they obtained

$$\frac{\sigma_{yf}}{\sigma_{ym}} \approx 0.3 \left( f \frac{\rho_f}{\rho_m} \right)^{3/2} + (1 - f) \frac{\rho_f}{\rho_m} + \frac{P_0 - P_{atm}}{\sigma_{ym}} \quad (8)$$

where  $P_0$  is the pressure in closed-cells and  $P_{atm}$  is the atmospheric pressure. Usually the pressure difference is small and much less than the matrix yield strength, so the last term is negligible and eq. (8) reduces to:

$$\frac{\sigma_{yf}}{\sigma_{ym}} \approx 0.3 \left( f \frac{\rho_f}{\rho_m} \right)^{3/2} + (1 - f) \frac{\rho_f}{\rho_m} \tag{9}$$

Nicolais and Narkis<sup>10</sup> derived a yield stress model for polymer composites filled with spherical particles without interfacial adhesion. This means that no load



**Figure 3** Normalized yield stress as a function of normalized modulus.

transfer from the matrix to the particles is possible. The yield stress was given as

$$\sigma_{yc} = \sigma_{ym} [1 - (3/4)^{2/3} \pi^{1/3} f^{2/3}] = \sigma_{ym} (1 - 1.21 f^{2/3})$$
(10)

where  $\sigma_{yc}$  is the yield stress of the composite and  $\sigma_{ym}$  is the yield stress of the matrix. Nielsen<sup>11</sup> proposed that the yield stress in the case of no adhesion between polymer and filler should be given as:

$$\sigma_{yc}/\sigma_{ym} \approx (1 - f^{2/3})S \tag{11}$$

where *S* is a stress concentration factor with a value between 0 and 1. A value of 1.0 indicates that there is no stress concentration.

The curve of the normalized yield stress as a function of normalized density is shown in Figure 4 along with the predictions of eq. (12) for the case when *S* equals 1. It can be seen that the models give very close predictions to the experimental data except for eq. (11). The reason is that the contribution of the polymer is proportional to the average cross-section of the polymer in the composite. Actually, the minimum cross-section of the polymer in a sample should be



**Figure 4** Normalized yield stress as a function of normalized density. Different lines show predictions of different models: (---) Gibson-Ashby, (—) square power-law, (---) Nicolais-Narkis, and (---) Nielsen (S = 1) model.

 TABLE IV

 Effect of Molecular Weight on Normalized Yield Strength for Different ACA Concentrations (%)

HI	DPE		$\sigma_{yf}/[\sigma_{ym}(1-f)^2]$				
Name	$M_w$	1.0%	1.5%	2.0%	2.5%	3.0%	
A60-70-162 G60-110 HBW5555Ac	$\begin{array}{c} 3.25 \times 10^5 \\ 6.51 \times 10^5 \\ 7.56 \times 10^5 \end{array}$	$\begin{array}{c} 0.95 \pm 0.06 \\ 1.02 \pm 0.04 \\ 1.07 \pm 0.02 \end{array}$	$\begin{array}{c} 0.96 \pm 0.04 \\ 1.03 \pm 0.07 \\ 1.05 \pm 0.04 \end{array}$	$\begin{array}{c} 1.01 \pm 0.09 \\ 1.10 \pm 0.08 \\ 1.02 \pm 0.05 \end{array}$	$\begin{array}{c} 1.16 \pm 0.21 \\ 1.01 \pm 0.14 \\ 1.21 \pm 0.04 \end{array}$	$\begin{array}{c} 1.16 \pm 0.09 \\ 1.03 \pm 0.02 \\ 1.08 \pm 0.05 \end{array}$	

applied. This model underestimates the normalized yield stress in all volume fractions, even though a maximum value of S is taken. Because the Nicolais and Narkis model is based on spheres packed in a cube, the maximum filler load is 0.74 and the normalized yield strength is zero by definition at that point. Nevertheless, the model still gives reasonable predictions for normalized densities higher than 0.4. On the other hand, the model derived by Gibson and Ashby is based on cubic cells consisting of struts and walls disposed in a staggered way, so the corners of one cell rest upon the midpoints of adjacent cells. This structure does not correspond to the actual geometric characteristics of real foams. However, it reflects the deformation process of a cellular structure and gives reasonable predictions even for high density foams. Therefore, the square power-law and the Gibson-Ashby model are found to predict the yield stress of polyethylene foams when they have a yield point.

Once again, a relation in the form  $\sigma_{yf}/[\sigma_{ym}(1-f)^2]$  is used and the results are shown in Table IV. It can be seen that the values of  $\sigma_{yf}/[\sigma_{ym}(1-f)^2]$  are close to 1. This means that molecular weight does not have an effect on the normalized yield stress. Good agreement is again obtained for different densities and molecular weights.

### Yield strain

Very few papers report the dependence of yield strain on the composition of composites. Nevertheless, this property is useful because it indicates the transition from elastic to viscous behavior. Smith<sup>9</sup> developed a model for the yield strain as

$$\varepsilon_{yc} = \varepsilon_{ym} (1 - 1.105 f^{1/3})$$
 (12)

where  $\varepsilon_{yc}$  is the yield strain of the composites and  $\varepsilon_{ym}$  is the yield strain of the matrix. Nielsen<sup>11</sup> also proposed that the yield strain for perfect adhesion between filler and polymer is

$$\varepsilon_{yc}/\varepsilon_{ym} \approx (1 - f^{1/3}) \tag{13}$$

The curve of normalized yield strain as a function of normalized density is shown in Figure 5 for our HDPE foams. Eq. (13) is based on perfect adhesion between

polymer and filler. In our case, the foam is composed of polymer and voids and no real adhesion exists, and the model does not apply. The Smith model is based on composites with no adhesion between polymer and rigid spherical filler. This is better, but the inclusions are not rigid in foams (gas) and the model does not apply. For our HDPE foams, the yield strain is almost unaffected by the volume fraction. The foam yield strain was almost similar to the parent matrix yield strain. This implies that the voids volume fraction did not affect yield strain. It is only determined by the molecular structure of matrix. The effect of molecular weight on yield strain is shown in Table V. It can be seen that the normalized yield strain is around 1 for different molecular weights and different ACA concentrations. This could mean that for polyethylene foams with yield strain, the yield strain is solely determined by the molecular structure of the matrix.

#### Strength at break

The tensile strength at break depends on the fracture mechanism; whether the polymer is ductile or brittle. As shown in Figure 1, our HDPE foams exhibit different responses order tension. The lowest molecular

1.2 1.0 Normalized Yield Strain 0.8 A60-70-162 0.6 G60-110 HBW555A 0.4 02 0.0 0.4 0.6 0.8 1.0 Normalized Density

**Figure 5** Normalized yield strain as a function of normalized density. Different lines show predictions of different models: (----) Smith model, (—) Nielsen model.

HE	DPE		Normalized yield strain				
Name	$M_w$	1.0%	1.5%	2.0%	2.5%	3.0%	
A60-70-162 G60-110 HBW5555Ac	$3.25 \times 10^{5}$ $6.51 \times 10^{5}$ $7.56 \times 10^{5}$	$\begin{array}{c} 0.93 \pm 0.05 \\ 0.91 \pm 0.03 \\ 0.99 \pm 0.04 \end{array}$	$\begin{array}{c} 0.90 \pm 0.08 \\ 0.93 \pm 0.06 \\ 0.94 \pm 0.02 \end{array}$	$\begin{array}{c} 0.94 \pm 0.09 \\ 0.90 \pm 0.07 \\ 1.00 \pm 0.03 \end{array}$	$\begin{array}{c} 0.96 \pm 0.11 \\ 1.07 \pm 0.10 \\ 0.93 \pm 0.06 \end{array}$	$0.99 \pm 0.10$ $1.02 \pm 0.06$ $1.08 \pm 0.08$	

 TABLE V

 Effect of Molecular Weight on Normalized Yield Strain for Different ACA Concentrations (%)

weight foam has a brittle fracture behavior, while the others have ductile fracture behavior.

#### Brittle fracture

Under tension, brittle foams are linear-elastic up to fracture. In this case, tensile failure can be treated by the methods of linear-elastic fracture mechanics. Bueche and Berry<sup>22</sup> found a linear dependence of tensile strength on modulus for silicone elastomers containing various fillers. Because solid polymers have yield points and break during necking, it is reasonable to use the yield strength of a solid polymer to replace the break strength of a solid polymer for comparison with the brittle fracture of foams. In our study, the normalized brittle fracture strength is defined as the brittle strength divided by the yield stress of the matrix. The normalized brittle strength as a function of the normalized elastic modulus is shown in Figure 6. It can be seen that there is once again a linear relationship between the normalized fracture strength and the normalized modulus as

$$\frac{\sigma_{bf}}{\sigma_{ym}} \approx \frac{E_f}{E_m} \tag{14}$$

where  $\sigma_{bf}$  is the foam break stress and  $\sigma_{ym}$  is the matrix yield stress. Insertion of eq. (2) into eq. (14) gives the



Figure 6 Normalized rupture stress as a function of normalized modulus for J60-1700-173 foams.

brittle fracture strength as a function of the volume fraction of voids:

$$\frac{\sigma_{bf}}{\sigma_{ym}} \approx \frac{E_f}{E_m} \approx (1 - f)^n \quad (1.93 \le n \le 2.01) \quad (15)$$

For a Poisson ratio of 0.34, the power index is 2 and eq. (15) becomes

$$\frac{\sigma_{bf}}{\sigma_{ym}} \approx (1-f)^2 \tag{16}$$

Nielsen<sup>11</sup> also proposed a break stress model for two phase composites in the case of no adhesion between polymer and filler:

$$\sigma_{bc}/\sigma_{bm} \approx (1 - f^{2/3})S \tag{17}$$

where  $\sigma_{bc}$  is the break stress of composites and  $\sigma_{bm}$  is the break stress of the matrix. Here, we propose to modify eq. (17) by replacing the break strength of the matrix by the yield strength of the matrix to give:

$$\sigma_{bc}/\sigma_{um} \approx (1 - f^{2/3})S \tag{18}$$

The curve of normalized strength as a function of normalized density is shown in Figure 7. From Figure 7, it can be seen that eq. (18) with an *S* value of 1 does not apply to HDPE foams. Eq. (18) is based on composites with no adhesion between polymer and spherical rigid filler. This is more realistic for foams, but the inclusions (gas) are not rigid and the model does not apply to foams. It is seen that eq. (16) gives reasonable predictions for our experimental data.

#### **Ductile fracture**

Because polyethylene foams, except for J60-1700-173, break after their yield point, the actual cross-section of a specimen decreases substantially after the yield point. Foams of A60-70-162 break during load drop in a tensile experiment and have ductile failures. Foams of G60-110 and HBW555Ac break during necking propagation, in which the stress changes slowly but the strain changes rapidly. Because the break takes place after the yield point, plastic deformation also takes place at this time, and the resulting cross-section



**Figure 7** Normalized rupture stress as a function of normalized density for J60-1700-173 foams. Different lines show predictions of different models: (—) square power-law, (- - - -) Nielsen model with S = 1.

is much less than the initial cross-section. In that case, the engineering stress is much less than the true stress.

In Figure 8, we compare the break strength of different foams as a function of normalized density. From Figure 8, it can be seen that increasing the normalized density increases break strength and a similar behavior to the break strength/normalized density is recovered.

# Break strain

Each HDPE foam has a different break strain. The lowest molecular weight foam has a brittle behavior, and with increasing molecular weight the foams become ductile. Smith<sup>9</sup> developed a model for the break



**Figure 8** Rupture stress as a function of normalized density for high molecular weight HDPE foams: (- - -) A60-70, (--) G60-110, and (- - -) HBW555Ac.



**Figure 9** Normalized rupture strain as a function of normalized density for J60-1700-173 foam: Different lines show predictions of different models: (—) Smith, and (----) Nielsen model.

strain of composites when there is no adhesion between filler and polymer:

$$\varepsilon_{bc} = \varepsilon_{bm} (1 - 1.105 f^{1/3}) \tag{19}$$

Nielsen<sup>11</sup> also developed a semi-quantitative relationship between the elongation at break and the composite composition with perfect adhesion between filler and polymer as

$$\varepsilon_{bc}/\varepsilon_{bm} = 1 - f^{1/3} \tag{20}$$

# Brittle fracture strain

The curve of brittle fracture strain as function of normalized density for J60-1700-173 (lowest molecular weight) foam is shown in Figure 9. From Figure 9, it can be seen that both the Smith and the Nielsen model did a poor job of representing the data. We observed that the brittle fracture strain is almost independent of the volume fraction of voids. This means that the volume fraction of voids does not appreciably affect the brittle fracture strain, which can be determined by the sole molecular structure of matrix.

## Ductile fracture strain

The relationship between true and engineering strain is given by the following equation:

$$\varepsilon_{true} = \ln(\varepsilon_{Eng} + 1) \tag{21}$$

At small strains,  $\varepsilon_{Eng}$  is close to  $\varepsilon_{true}$ . However, at a large strain, this is not the case and it becomes necessary to use  $\varepsilon_{true}$  to analyze the data. The break strain as



**Figure 10** Real strain as a function of normalized density for high molecular weight HDPE foams.

a function of the normalized density is shown in Figure 10. Even though there is some scatter in the data for break strain, it can be seen that increasing the volume fraction of voids only slightly affects the break strain.

#### Effect of molecular weight

Looking at the results, we propose a linear relationship between the average break strain and the molecular weight:

$$\log \varepsilon = AM_w - B \tag{22}$$

where  $\varepsilon$  is the real strain of the sample,  $M_w$  is the molecular weight of the matrix, and A and B are the model constants. The effect of molecular weight on the average break strain is shown in Figure 11. It can be



**Figure 11** Average rupture strain as a function of molecular weight. Eq. (23) is graphed as a solid line.



Figure 12 Toughness as a function of normalized density.

seen that increasing the molecular weight increases the break strain of polyethylene foams:

$$\log \varepsilon = (1.49 \times 10^{-6})M_w - 1.37 \tag{23}$$

Toughness

Toughness is the amount of energy per unit volume that a material can absorb prior to fracture.<sup>23–24</sup> For an un-notched tensile bar, the energy at break can be calculated with the following equation:

toughness = 
$$\frac{\text{energy at break}}{\text{volume}} = \int_{0}^{l_{f}-l_{0}} F \, dl / V$$
 (24)

where  $l_f$  is the length of the sample between the clamps at break and  $l_0$  is the initial length of the sample between the clamps. F is the load applied to a sample of volume V. The toughness results are plotted against normalized density in Figure 12. For our polymers, structural properties such as stiffness and strength decreased with an increase in void fraction. This is also the case for toughness. For our lowest molecular weight HDPE, the foams behave as brittle materials (see Fig. 1) and toughness is low. Increasing molecular weight increases break strain, and above a certain molecular weight the foams behave as ductile materials. Figure 12 shows that toughness increases with molecular weight. For the moment, there is no clear relationship among toughness, density and molecular weight. This will be further studied in the future.

#### CONCLUSIONS

Tensile properties of closed-cell polyethylene foams were studied at room temperature using four different molecular weights of HDPE. Young's modulus was found to follow the square power-law relationship for all molecular weights using normalized properties.

For foams showing a brittle fracture behavior, the break strength also closely followed the square powerlaw model, and the break strain was independent of the volume fraction of voids in our experimental range (1 < f < 0.5). In the case of foams showing a ductile behavior, the normalized yield strength also followed a square power-law relation with the normalized density. The yield strain of foams was near that of the solid polymer and did not change appreciably over the range of void volume fractions studied. The engineering break stress seemed to have a similar tendency with the normalized density for different molecular weights. Increasing the molecular weight increased the break strain of the foam. Finally, foam toughness increased with density and polyethylene molecular weight.

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#### References

1. Klempner, D.; Frisch, K. C. Handbook of Polymeric Foams and Foam Technology; Carl Hanser Verlag: Munich, 1991.

- Chung, P. P. In Handbook of Polymeric Foams and Foam Technology; Klempner, D., Frisch, F. C., Eds.; Carl Hanser Verlag: Munich, 1991; Chapter 9.
- Gibson, L. J.; Ashby, M. F. Cellular Solids: Structure and Properties, 2nd ed.; Cambridge University Press: Cambridge, UK, 1997.
- 4. Zhang, Y.; Rodrigue, D.; Ait-Kadi, A. J Appl Polym Sci, submitted.
- Zhang, Y.; Rodrigue, D.; Ait-Kadi, A. J Appl Polym Sci, submitted.
- Moore, D. R.; Couzens, K. H.; Iremonger, M. J. J Cell Plast 1974, 10, 135.
- 7. Moore, D. R.; Iremonger, M. J. J Cell Plast 1974, 10, 230.
- Throne, J. L. Thermoplastic Foams; Sherwood Publishers: Hinckley, OH, 1996.
- 9. Smith, T. L. Trans Soc Rheol 1959, 3, 113.
- 10. Nicolais, L.; Narkis, M. Polym Eng Sci 1971, 11, 194.
- 11. Nielsen, L. E. J Appl Polym Sci 1966, 10, 97.
- Throne, J. L. In Engineering Guide to Structural Foams; Wendle, B. C., Ed.; Technomic Publishing Co: Westport, CT, 1976.
- Gibson, L. J.; Ashby, M. F. Proc Roy Soc London Ser A 1982, 382, 43.
- 14. Mehta, B. S.; Colombo, E. A. J Cell Plast 1976, 12(1), 59.
- 15. Wasserstrass, J. D.; Throne, J. L. J Cell Plast 1976, 12(2), 98.
- Shutov, F. A. Integral/Structural Polymer Foams-Technology Properties and Applications; Springer Verlag: Berlin, 1986.
- Goods, S. H.; Neuschwanger, C. L.; Henderson, C. C.; Skala, D. M. J Appl Polym Sci 1998, 68, 1045.
- Ward, I. M. Mechanical Properties of Solid Polymers; John Wiley & Sons Ltd: Hoboken, NJ, 1971.
- Ward, I. M. Mechanical Properties of Solid Polymers, 2nd ed.; John Wiley & Sons Ltd.: Hoboken, NJ, 1983.
- 20. Flory, P. J. J Amer Chem Soc 1945, 67, 2048.
- 21. Vicent, P. I. Polymer 1960, 1, 425.
- Bueche, A. M.; Berry, J. P. In Fracture; Averbach, B. L., et al., Eds.; Wiley: New York, 1959; p 265.
- Arges, K. P.; Palmer, A. E. Mechanics of Materials; McGraw-Hill Book Company, Inc.: New York, 1963; Chapter 2.
- Hertzberg, R. W. Deformation and Fracture Mechanics of Engineering Materials, 4th ed.; John Wiley & Sons, Inc.: Hoboken, NJ, 1996; Chapter 1.