Polystyrene Foams. II. Structure–Impact Properties Relationships

Saeed Doroudiani,* Mark T. Kortschot

Department of Chemical Engineering and Applied Chemistry, University of Toronto, Toronto, Ontario, Canada M5S 3E5

Received 21 June 2002; accepted 11 February 2003

ABSTRACT: In the first part of this series of articles, the relations between the foaming conditions and the microstructure of expanded polystyrene (EPS) were explored. In this part, the effects of the foaming conditions and the microstructure of EPS on impact properties are discussed. Regression analysis was conducted on the data and expressions were developed to quantify these relationships. Moreover, the importance of the individual structural parameters was determined. Statistical analysis of the data showed that *foaming time* was the most important factor determining the impact strength, while *foaming temperature* was the most important factor controlling the specific impact strength. The deformation of cells at the crack tip, as a result of bending and/or buckling of cell walls, can increase the failure strain, which leads to an increase in failure energy. In expanded polymers, the majority of the absorbed energy during impact loading is dissipated as plastic work. The transition of plane stress conditions to plane strain conditions, due to expansion, can be considered as another source of toughening in EPS. © 2003 Wiley Periodicals, Inc. J Appl Polym Sci 90: 1421–1426, 2003

Key words: polystyrene; structure-property relations; impact resistance

INTRODUCTION

The first part of this series explored the relation between the foaming conditions and the structure of expanded polystyrene (EPS) made using a physical blowing agent.[1] It was shown that by controlling the foaming conditions a wide range of cellular structures and expanded samples having the same foam density and different cell sizes could be produced. Foaming time was found to be the most important factor determining foam density, followed by foaming temperature and saturation pressure. Moreover, saturation pressure was the most important factor determining cell size and cell density, followed by foaming time and foaming temperature.

There are a number of articles on the mechanical properties of cellular materials and conventional foams.[2–4] Waldman[5] studied the impact strength of microcellular EPS and found a greater impact strength for microcellular EPS compared with that of an unfoamed polymer. Collias and Baird[6] investigated the impact behavior of microcellular EPS, the styrene–acrylonitrile copolymer, and polycarbonate. They found no improvement in the impact strength of expanded samples compared to neat polymers.

This article presents a systematic study of the effect of the foaming conditions and structural parameters of EPS on the impact strength. In this study, a three-stage batch-foaming process using CO_2 as a blowing agent was utilized. Samples of polystyrene (PS) sheets were saturated with CO_2 at room temperature and expanded by heating the saturated samples after rapidly releasing pressure. Standard samples were cut from expanded sheets, and after notching, their impact strengths were examined. The impact strengths of the expanded sheets were as much as twice those of neat PS sheets.

EXPERIMENTAL

Details of the materials, foaming process, and sample preparation were described in Part I of this series.[1] Specimens for impact testing were cut from compression-molded sheets of PS. The impact strength of the notched specimens was determined using a Tinius Olsen Model 92T impact tester at room temperature, according to ASTM standard method D-256, the standard Izod impact test. The dimensions of the specimens were $60.30 \times 12.60 \times 3.20$ mm^3. Before testing, specimens were notched using a milling machine and were conditioned at room temperature and humidity for at least 2 weeks. The average values from at least five tests were reported.

In the experimental design, *saturation pressure*, *foaming temperature*, and *foaming time* served as the input

Correspondence to: M. T. Kortschot (kortsch@chem-eng.utoronto.ca).

**Current address:* Department of Chemical Engineering, Kyoto University, Sakyo-ku, Kyoto, 606-8501 Japan.

Journal of Applied Polymer Science, Vol. 90, 1421–1426 (2003)

TABLE I
Design Matrix for Three Variables at Three Levels Based on Central Composite and Data (Replicates Are Designated by an Asterisk)

Sample no.	Pressure (MPa)	Temperature (°C)	Time (se)	Relative density	Cell size (μm)	Impact strength (J/m)	Specific impact strength (J cm^3 m^{-1} g^{-1})
13	3	105	10	0.5663	92	22.87	40.38
17*	3	105	10	0.5547	81	22.15	39.94
6	3	105	30	0.2414	163	8.62	35.71
9	3	110	20	0.2107	118	12.63	59.94
14	3	120	10	0.2424	27	17.64	72.77
12	3	120	30	0.1275	341	10.98	86.12
7	4.5	105	20	0.2911	74	13.00	44.66
5	4.5	110	10	0.3809	77	14.17	37.20
3	4.5	110	20	0.1836	112	9.83	53.54
16*	4.5	110	20	0.1745	114	8.87	50.83
8	4.5	110	30	0.1098	132	7.23	65.85
19*	4.5	110	30	0.1133	148	5.35	47.22
2	4.5	120	20	0.1212	145	6.09	50.25
4*	4.5	120	20	0.128	133	7.18	56.09
1	6	105	10	0.3361	3	18.07	53.76
15	6	105	30	0.0952	29	8.30	87.18
18	6	110	20	0.1004	15	6.30	62.75
11	6	120	10	0.1927	23	12.96	67.25
10	6	120	30	0.0318	57	4.84	152.20

parameters. The large number of samples required for a classical factorial design suggested the use of a central composite design plan.

Table I presents the design for three variables at three levels. Four replicates were used to estimate the experimental error. The regression model relating the response y (representing impact strength or specific impact strength) to x_1 (representing *pressure*), x_2 (representing *foaming temperature*), and x_3 (representing *foaming time*) that is supported by this design is

$$y = \beta_0 + \beta_1 x_1 + \beta_2 x_2 + \beta_3 x_3 + \beta_{12} x_1 x_2 + \beta_{13} x_1 x_3 + \beta_{23} x_2 x_3 + \beta_{11} x_1^2 + \beta_{22} x_2^2 + \beta_{33} x_3^2 + \varepsilon$$

RESULTS AND DISCUSSION

In this study, the Izod impact strengths of notched EPS were examined. First, the values of the impact strengths and specific impact strengths (strength divided by density) were analyzed as a function of the foaming conditions. Next, the relationship between the impact strength and structural parameters (foam density and cell size) are discussed.

The impact data were statistically analyzed. The relationships between the impact strength and the foaming conditions was determined from the experimental design analysis, which led to eq. (1):

$$I = 51.243 - 1.574P - 0.226T - 0.492t + 3.245\bar{t}^2 \quad (1)$$

where $\bar{t} = (t - 20)/10$ is the scaled (coded) form of t. Model term ranking analysis indicated the importance (effectiveness) of the model terms in the order of foaming time, saturation pressure, and foaming temperature. The relative model term effects for t, P, and T were 1, 0.50, and 0.41, respectively. The term t^2 was also found to be significant. While useful for making design predictions, eq. (1) does not give detailed insight into the mechanisms governing the process. The analysis of variance data were presented in detail in ref. 7.

Since density is of major importance in controlling the load-bearing capacity of the foam, an attempt was been to determine the mechanical properties independent of the density of the foams. Cost reduction is one of the main motivations for using expanded materials. For this reason, properties computed on the basis of the equivalent mass are commercially important. In this study, a *specific property* was calculated by dividing the property in question by the sample density. The specific properties are used to compare samples on an equal mass basis.

A statistical analysis was also performed on the specific impact strength data and eq. (2) was obtained:

$$I_{sp} = -255.727 + 8.732P + 2.11T + 1.461t + 22.46\bar{P}^2 + 13.425\overline{Pt} \quad (2)$$

where $\bar{P} = (P - 4.5)/1.5$ and $\bar{t} = (t - 20)/10$ are scaled (coded) forms of P and t, respectively. It was found that the foaming temperature was the most important parameter in determining the specific impact strength. The levels of importance of the foaming time and saturation pressure were less than those of the foaming temperature, but their effects were still significant.

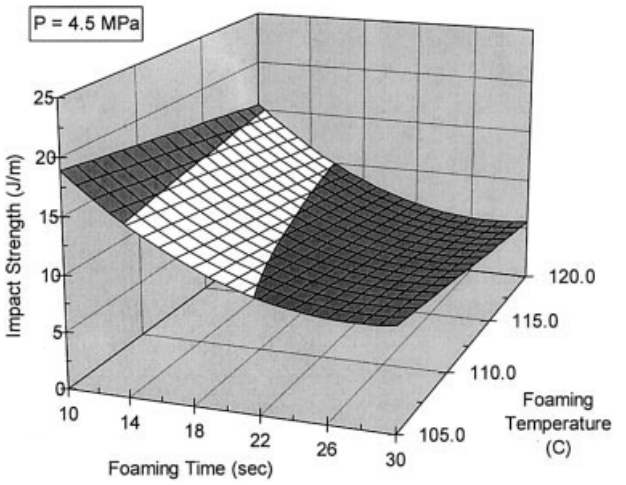

Figure 1 Dependence of Izod impact strengths of notched EPS samples on foaming conditions. The plotted surface represents the fitted regression equation [eq. (1)].

The effects of the foaming conditions on the impact strength and specific impact strength of EPS are shown in Figures 1 and 2, respectively. The impact strength increased greatly (as high as double that of the neat PS) as a result of foaming. Although the impact strength decreased with an increasing foaming temperature, foaming time, and saturation pressure (Fig. 1), the specific impact strength increased with respect to the foaming temperature and foaming time (Fig. 2). Interestingly, the specific impact strength of EPS exhibited an improvement of about six to eight times, relative to that of PS. The impact strengths and specific impact strength of the PS were measured as 8.5 J/m and 8.2 J cm^3 g^{-1} m^{-1}, respectively.

The variation of the impact properties of EPS can be explained by considering the structure produced by

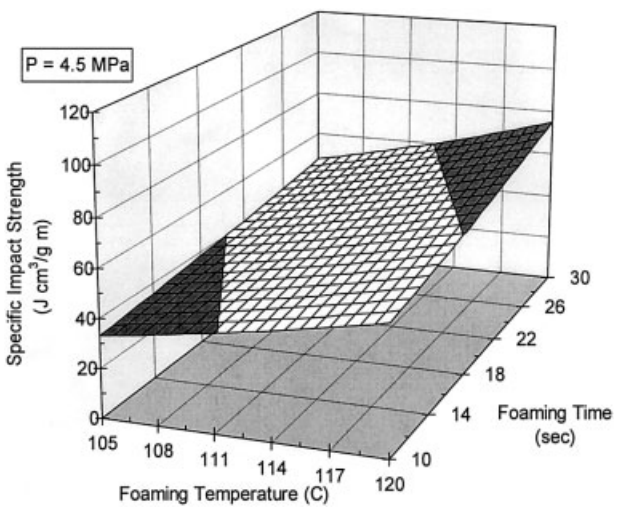

Figure 2 Dependence of Izod specific impact strengths of notched EPS samples on foaming conditions. The plotted surface represents the fitted regression equation [eq. (2)].

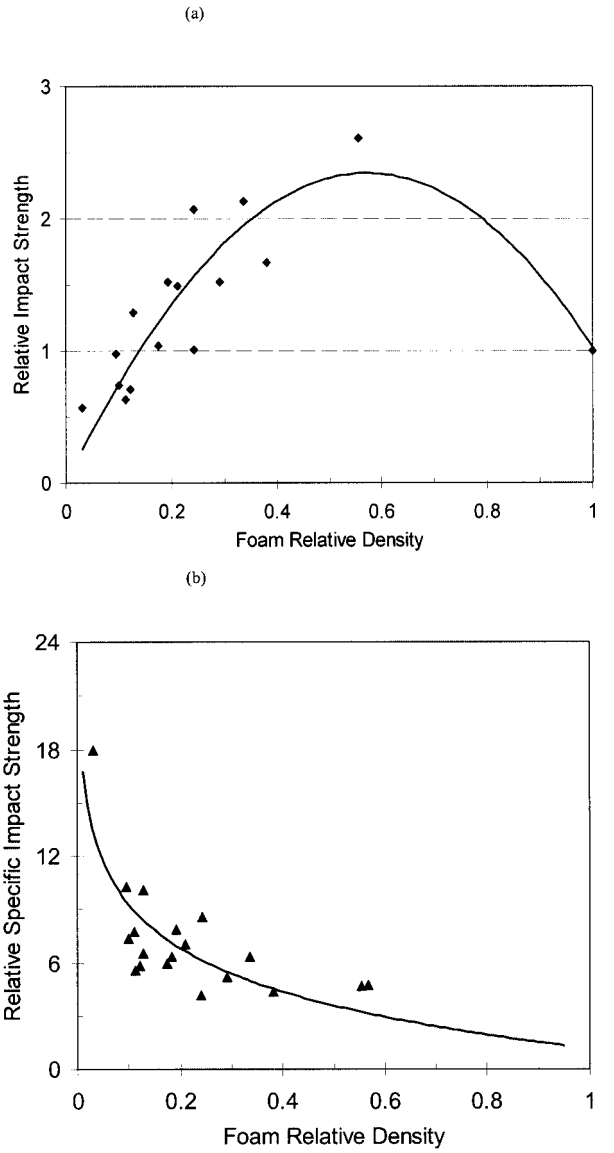

Figure 3 (a) Relative and (b) relative specific impact strengths of notched (Izod) samples of EPS versus relative density. The curves represent eqs. (3) and (4).

foaming. The Izod impact strength plotted in Figure 3(a) increased with an increasing relative density. A quadratic expression fit the data quite well, with R^2 equal to 0.736. The impact strength of the foam exceeded that of the neat polymer at a relative density of about 0.15 and reached its maximum at a relative density of about 0.6. Hence, an impact strength equal to that of the unfoamed polymer can be achieved with just 15% of the material provided a suitable expanded structure is used.

$$I_r = -7.17\rho_r^2 + 8.20\rho_r \quad (3)$$

The trend of the variation of the specific values of the impact strength versus the relative density was

completely different from that of the impact strength. A logarithmic expression with R^2 equal to 0.666 could satisfactorily represent these data:

$$I_{r,sp} = -3.54 \ln \rho_r + 1.12 \quad (4)$$

Figure 3(b) clearly demonstrates that PS absorbs more energy per unit mass when it is distributed in thin membranes in the highly expanded structure. An empirical equation was used to relate the impact resistance of foams to their relative density and thickness, as follows[8]:

$$I_r = \rho^m \tau^e \quad (5)$$

The effect of cell size on the impact strength is shown in Figure 4. Recall from first part of this series of articles[1] that variations in saturation pressure (and foaming time and temperature, with lesser effect) led to variations in cell size. It appears that neither the impact strength [Fig. 4(a)] nor the specific impact strength [Fig. 4(b)] depend on the cell size in a consistent way.

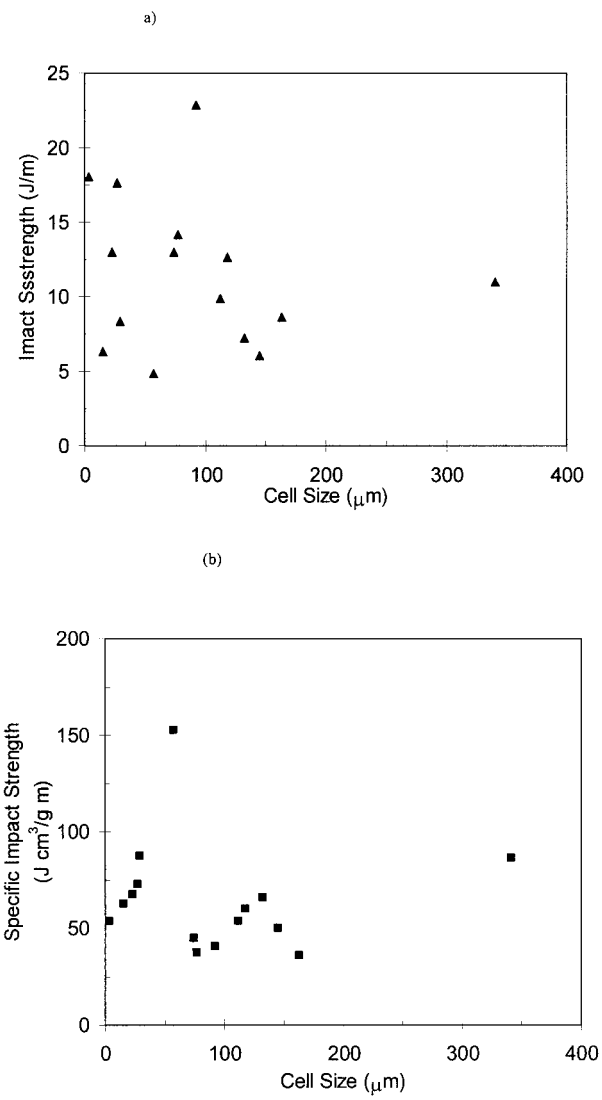

Figure 4 (a) Izod impact and (b) specific impact strengths of EPS as a function of cell size.

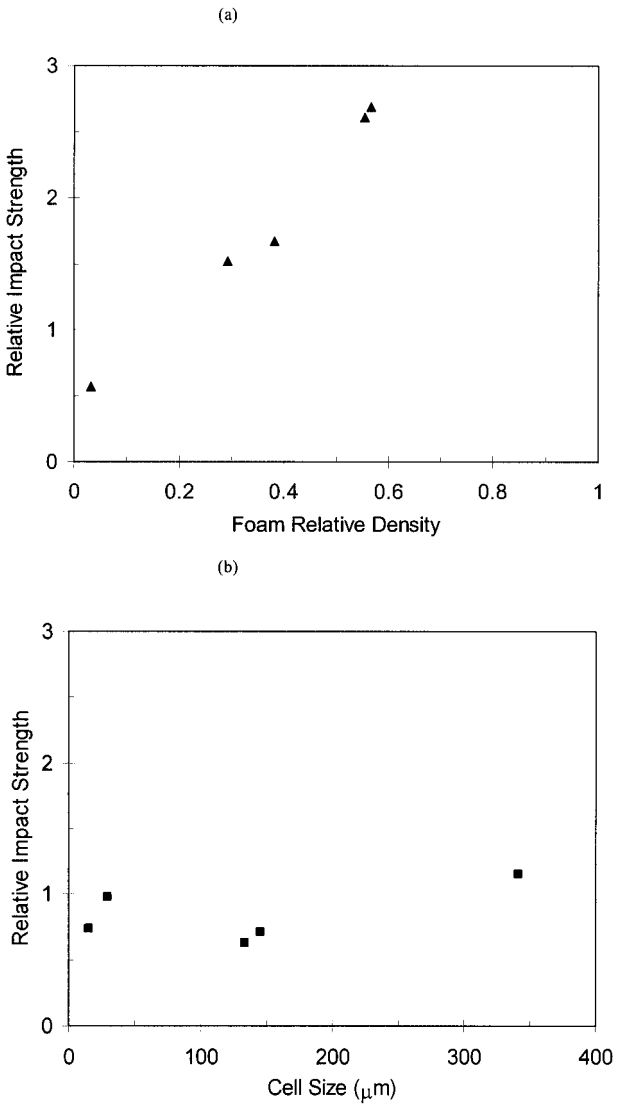

Figure 5 Relative impact strengths of EPS (a) versus density for similar cell sizes and (b) versus cell size for similar densities.

To isolate the effects of cell size and foam density, two sets of results were extracted from Table I. In Figure 5(a), the relative impact strengths of samples with similar cell sizes but differing density (sample numbers 5, 7, 10, 13, and 17) are plotted. The data show a trend similar to that for the whole data set [Fig. 3(a)]. At very low densities, the relative impact strength crosses the value 1 as less solid material is available to absorb energy. In Figure 5(b), the impact strength for a series of samples with similar densities but differing cell size is reported (sample numbers 2, 8, 15, 18, and 19). The data indicate that, over this range of cell sizes, cell size does not affect the impact

strength. The relative densities of the samples in Figure 5(b) were in the range of 0.100–0.120.

Figure 6 shows schematically the propagation of a crack through a low-density cellular polymer. The deformation of cells at the crack tip as a result of bending extension and even buckling of cell walls and all of these mechanisms can contribute to the overall energy absorption in an impact test. Viscous deformation of the fluid within the cells can also contribute to the energy-absorption process; however, this mechanism is less important in rigid plastic foams such as EPS than it would be in elastomeric foams, where the energy dissipated by the material is smaller.

Plastic deformation of the cell walls is an important source of energy dissipation and toughness. Polymers will yield at some stress level, and this deformation is irreversible and leads to energy dissipation. Yielding at the tip of a crack in a thick material is constrained by "plane strain" conditions that exist, and this reduces the energy needed to drive the crack and, hence, reduces the measured toughness.[9] In a foamed polymer, the material is distributed in relatively thin membranes, in which yield is facilitated due to the absence of constraint from the surrounding material. This "plane stress" condition leads to an increase in plastic flow and, hence, to an increase in toughness.

Some similarities exist between deformation of foams and filled polymers. One way to understand the behavior of expanded polymers might be to treat them as multiphase systems, in which cell cavities are

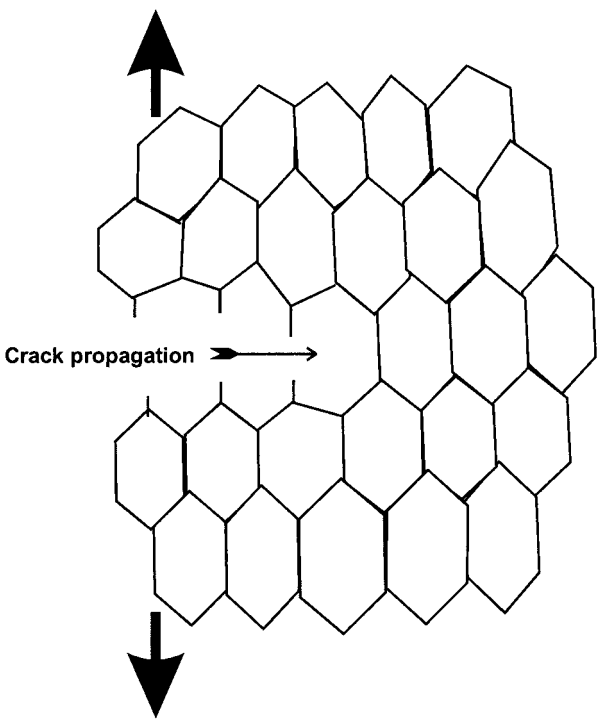

Figure 6 Propagation of a crack in a low-density cellular polymer.

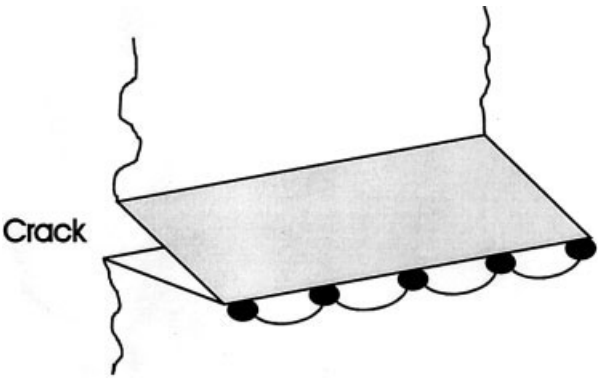

Figure 7 Toughening by crack pinning around second phase.

treated as a second phase. In this way, the theories established for other multiphase systems can be applied to expanded polymers, where the cavity is treated as a filler of zero modulus.

It is well known that incorporating ductile or rubbery particles can toughen polymers. In these filled polymers, toughening is often modeled using the concept of a crack-pinning mechanism, in which the crack front changes in length as it interacts with particles or bubbles (Fig. 7). Various models have been developed to relate toughening to the size of the particles. The models presented by Lang,[10] Evan,[11] and Rose[12] all favor smaller particles as toughening agents. In our studies, cell size was not found to have a significant influence on the toughness [see Fig. 5(b)], suggesting that the concept of crack pinning may not be applicable to microcellular foams, at least in the range of cell sizes and for the relative density examined here. Note that we were only able to isolate a subset of foams of constant relative density in the range of about 10–15% and that our results may be specific to this particular range. The results obtained in this work are in agreement with previous work on vitreous carbon foam. Brezny and Green[13] found the fracture toughness and elastic modulus of vitreous carbon foam to be independent of cell size.

CONCLUSIONS

PS samples were expanded after saturating with CO_2. The impact properties of the EPS samples were measured. A significant improvement was observed in the Izod impact strength of EPS, compared to that of neat PS. It was found that cell size, at least in the range of this study, does not affect the impact strength significantly. Statistical analysis of the data showed that *foaming time* plays the most important role in controlling the impact strength, and this is because it is the principal factor affecting foam density. It was found that the impact strength increased with an increasing relative density up to a relative density of at least 0.6, where the impact strength was approximately double that of the un-

foamed resin. At a relative density of 0.15, the impact strength of EPS equaled that of unfoamed PS. Plastic and viscoelastic deformation in the cell walls is the major source of energy dissipation in these materials and this is enhanced by lowering the density of the foam.

Partial funding for this research was provided by the University of Toronto. The support is gratefully acknowledged by the authors. The authors also thank Nova Chemicals (Calgary, Canada) for providing materials and Mr. Shiang Law for technical assistance.

References

1. Doroudiani, S.; Kortschot, M. T. J Appl Polym Sci 2003, 90, 1412.
2. Gibson, L. J.; Ashby, M. F. Cellular Solids: Structure and Properties; Pergamon: New York, 1997.
3. Hilyard, N. C. Mechanics of Cellular Plastics; Applied Science: London, 1982.
4. Hilyard, N. C.; Cunningham, A. Low Density Cellular Plastics: Physical Basis of Behavior, 1st ed.; Chapman & Hall: London, 1994.
5. Waldman, F. M.S. Thesis, Massachusetts Institute of Technology, Cambridge, MA, 1982.
6. Collias, D. I.; Baird, D. G. Polym Eng Sci 1995, 35, 1178.
7. Doroudiani, S. Ph.D. Thesis, Department of Chemical Engineering and Applied Chemistry, University of Toronto, 1998.
8. Thron, J. L.; Progelhof, R. C. SPE Tech Pap 43th ANTEC 1985, 31, 442.
9. Knott, J. F. Fundamentals of Fracture Mechanics; Butterworth: London, 1973.
10. Lang, F. F. Philos Mag 1970, 22, 983.
11. Evans, A. G. Philos Mag 1972, 26, 1327.
12. Rose, L. R. F. Mech Mater 1987, 8, 11.
13. Brezny, R.; Green, D. J. Acta Metal Mater 1990, 38, 2517.