Modelling hollow thermoplastic syntactic foams under high-strain compressive loading

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Abstract

The mechanical responses of syntactic foams comprising hollow thermoplastic microspheres (HTMs) embedded in a polyurethane matrix were experimentally examined under uniaxial compressive strain. Phenomenological strain energy models were subsequently developed which capture both the axial stress-strain and transverse strain response of the foams. HTM syntactic foams were found to exhibit increased small-strain stiffness with reduced density, revealing a highly-tuneable and extremely lightweight syntactic foam blend for mechanical applications. The foams were also found to possess strong compressibility ($J \approx 0.75$) and a high threshold for plastic deformation, making them a robust alternative to hollow glass microsphere (HGM) syntactic foams. The non-standard transverse strain relationship exhibited by HTM syntactic foams at high filling fractions was captured by advanced strain energy models.

Keywords: syntactic foams, thermoplastic, microspheres, strain energy, transverse strain behaviour

1. Introduction

Syntactic foams are composite materials comprising a suspension of gas-filled microspheres (microballoons) within a polymer matrix \cite{1}. These foams are well-known

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for their enhanced mechanical performance, which has motivated their widespread use in the automotive, marine, and aerospace industries, primarily as lightweight cores in sandwich panels [2, 3]. Syntactic foams are also widely used in other applications, particularly in thermal management and for vibration isolation [1, 4, 5]. In mechanical applications, the vast majority of syntactic foams comprise hollow glass microspheres (HGM) as a filler [6–15], as they increase the small-strain stiffness and compressive yield strengths of the matrix [14]. However, HGM syntactic foams are often heavier than the matrix material and possess poor mechanical recoverability.

An emerging class of syntactic foam comprises hollow thermoplastic microspheres (HTMs) embedded in a polymer matrix, which have demonstrated increased small-strain stiffness, lower densities, and strong recovery at large deformations [5, 16–19]. The property of strong recoverability is attributed to the buckling response of individual copolymer shells [20], in contrast to glass microspheres that typically crack under large strains. Earlier works on the mechanical properties of HTM foams with nonlinear (elastomeric) matrix materials [16, 17] have examined only low-filling fraction samples, subsequently missing key features of these foams.

In this work, the mechanical performance of elastomeric HTM syntactic foams is investigated, revealing increased small-strain stiffness and reduced density with increasing filling fraction. The syntactic foams are also found to possess strong recoverability to large peak strains. Strain energy models are constructed to describe the foam response [21], which are qualitatively well-described by simplified Ogden models [21, 22] at low filling fractions and well-described by advanced Ogden models at large filling fraction [23, 24]. When modelling HTM syntactic foams, a significant challenge lies in finding strain energy ansatzes that capture both the axial stress-strain and transverse strain response of the syntactic foam, requiring an appropriate compressibility condition $f(J)$ in the strain energy [21, 24].
2. Material Fabrication and Mechanical Testing

2.1. Materials and sample preparation

The HTM syntactic foams were made by blending hollow copolymer microspheres (Expancel 920 DE supplied by Expancel AzkoNobel) into a polyurethane matrix made from a blend of Polytetramethylene Ether Glycol (Terathane 1000 supplied by INVISTA Textile (UK) Ltd), Trimethylolpropane (supplied by Tokyo Chemical Industry) with Methylene diphenyl diisocyanate (Isonate M143 supplied by Dow Chemicals) as a curing agent. To ensure that the microspheres were distributed uniformly, fumed silica (Aerosil 200 supplied by Evonik Inc.) was used as a thixotropic additive. A summary of the microsphere properties is given in Table 1. Scanning-electron microscope (SEM) images of the spheres are given in Fig. 1, both in suspension and resting on imaging surface. After blending, the mixture was cured in open trays at 55°C and then machined into cylinders (diameters of 29 mm and heights of 12.5 mm).

2.2. Mechanical testing methods

Transversally unconfined uniaxial compression testing of the samples was conducted on an Instron testing machine (see Fig. 2) at a strain rate of 10 mm/min, following BS ISO 7743-2011. The top and bottom platens were sprayed with WD-40 to minimise barrelling of the samples. The relationship between the radial and axial stretch under loading was determined from video recordings of the samples under compression. Further details are given in the Supplementary Material.

3. Experimental results and discussion

In all figures, results are given for the initial loading curve at room temperature averaged over three samples. For reference, the raw and toe-compensated experimental data generated in connection with this work is freely available on FigShare [25].
3.1. Axial stress-strain response

Fig. 3 shows the axial stress-strain diagram for HTM syntactic foams at the filling fractions $\phi = 0\%$ (unfilled), 2%, 10%, and 40%, up to compressive strains of 50%. At dilute filling fractions $\phi \leq 10\%$, the small-strain stiffness increases with increasing filling fraction with the samples exhibiting nonlinear elastic behaviour [21]. At $\phi = 40\%$ the material instead exhibits conventional syntactic foam behaviour [26]: the emergence of a linear region at small strains, a graded plateau region at medium strains, and strong densification at large strains. Fig. 3(b) presents a zoom-in of Fig. 3(a) up to 5% strain, clearly demonstrating small-strain stiffening effects. The results for $\phi = 40\%$ suggests that microsphere buckling significantly influences the macroscale foam response.

3.2. Transverse-to-axial strain response

The corresponding transverse-to-axial strain response of these materials under compression is given in Fig. 4(a). Results are expressed in terms of stretches $\lambda$, defined via the engineering strain $\epsilon_E = \lambda - 1$, where $\lambda_r$ and $\lambda_z$ denote the radial and axial stretch, respectively. Fig. 4(a) presents test data for $\lambda_r$ as a function of $\lambda_z$ where dilute $\phi$ foams respond comparably to an incompressible material (green curve). For $\phi = 40\%$, a considerable change in the response was observed; the emergence of a linear region at small strains, and a 15% reduction in $\lambda_r$ at 50% strain relative to the unfilled material. The highly compressible response of the foam is attributed to both the strong compressibility of the enclosed gas and the buckling response of the microsphere shells.

3.3. Volume ratio response

Fig. 4(b) gives the corresponding volume ratio $J = \lambda_r^2 \lambda_z$, where low filling fraction samples correspond to weakly compressible media ($J \approx 1$). However for $\phi = 40\%$, a significant compression to $J \approx 0.75$ at 50% strain was observed, in addition to the emergence of linear and large-strain response regions as discussed above.
3.4. Small-strain materials constants

Table 2 presents the small-strain properties of the syntactic foams, namely the ground-state Young’s modulus $Y_0$ (calculated at 2% strain), and the ground-state Poisson ratio $ν_0$ (calculated at 3-5% strain), following the procedure discussed in Sec. 2. The density $ρ$ and specific stiffness $Y_0/ρ$ of these materials is also included, revealing an enhancement in the specific stiffness of the matrix by a factor of 2.5 at $φ = 40\%$, and revealing an extremely lightweight alternative to other syntactic foams (i.e., [6]).

3.5. Poisson’s ratio

Fig. 5(a) presents the corresponding Poisson’s ratio for the syntactic foams as defined by the Hencky ratio [27]

$$ν_0 ≈ −\log(λ_r)/\log(λ_z),$$

which implicitly assumes the relationship $λ_r ≈ λ_z^{−ν_0}$ for all strains. This relationship holds for dilute $φ$, as shown by the straight line fits for $ν_0$ (red curves). However for $φ = 40\%$, the Poisson ratio is a function of strain, for example

$$ν(λ_z) ≈ κ_3(λ_z − 1)^3 + κ_2(λ_z − 1)^2 + κ_1(λ_z − 1) + κ_0,$$

where $λ_r ≈ λ_z^{−ν(λ_z)}$, (2)

with $κ_0 ≈ ν_0$. Fig. 5(b) examines these models for $φ = 40\%$ foams, superposing the test data (blue line) with Eq. (2) where $(κ_3, κ_2, κ_1, κ_0) = (−0.4973, 0.1009, 0.3518, 0.34)$ (green dashed line). Reference curves for the Hencky ratio [1] at small- and large-strains ($ν_0 ≈ 0.34$ and $ν_L ≈ 0.25$, respectively) are also superposed.

3.6. Recoverability properties

Fig. 6(a) presents test data for the axial stress-strain curves to a maximum strain of 25% (blue curves), and stress-strain results for the same samples, to the same peak strain level, after several days of recovery (green curves). Generally excellent recoverability
was observed for all filling fractions \( \phi \) to this strain level. Fig. 6(b) shows test data for an identical experiment up to a peak strain of 50%, where strong recoverability was observed to high filling fractions. For comparison, HGM syntactic foams would typically fracture at such high strain levels \[28\] leaving them structurally compromised.

4. Mathematical results and discussion

4.1. Phenomenological strain energy modelling

For each filling fraction \( \phi \), a strain energy function \[21, 24\] was sought in the form

\[
W = \Phi(\lambda_1, \lambda_2, \lambda_3) + f(J),
\]

where \( \Phi(\lambda_1, \lambda_2, \lambda_3) \) is a function of principal stretches \( \lambda_j \) and \( f(J) \) is the compressibility condition. Through an appropriate choice of \( \Phi(\lambda_1, \lambda_2, \lambda_3) \) and \( f(J) \), an accurate recovery of both the axial stress-strain and transverse strain behaviour of the syntactic foams is possible after differentiation of (3).

Following Figs. 4(a) and 5(a), compressible neo-Hookean and compressible Ogden (type-I) strain energy models \[21, 29\], were considered, as their associated \( f(J) \) forms capture the Hencky response at low filling fractions. For \( \phi = 40\% \) foams, Ogden (type-II) models \[21, 29\] were investigated. Results were obtained using ABAQUS \[30\] and tables of coefficients are given in the Supplementary Material.

4.2. Compressible neo-Hookean

In this model, the strain energy takes the form

\[
W = C_{10}(\lambda_1^2 + \lambda_2^2 + \lambda_3^2 - 3) + \frac{1}{D_1}(J - 1)^2,
\]

(4a)
where \( \bar{\lambda}_j = J^{-1/3} \lambda_j \), \( J = \lambda_1 \lambda_2 \lambda_3 \), and \( C_{10} \) and \( D_1 \) denote real constants. Under uniaxial compression (\( \lambda_3 = \lambda_z \), \( \lambda_1 = \lambda_2 = \lambda_r \)), the engineering stress takes the form

\[
\sigma_E = \frac{\partial W}{\partial \lambda_z} = \frac{4 C_{10} J^{-2/3}}{3 \lambda_z} \left( \lambda_z^2 - \frac{J}{\lambda_z} \right) + \frac{2 J}{D_1 \lambda_z} (J - 1),
\]

(4b)

where \( J \) is obtained by solving the **compressibility relation**, given by imposing zero transverse stress conditions (i.e., \( \sigma_1 = 0 \)) and takes the form

\[
\frac{2 C_{10} J^{-5/3}}{3} \left[ \frac{J}{\lambda_z} - \lambda_z^2 \right] + \frac{2}{D_1} (J - 1) = 0.
\]

(4c)

Fig. 7 presents the stress-strain and transverse stretch response for CnH fitted models (4a), for all filling fractions (test data superposed in blue). These figures reveal that this strain energy form only qualitatively describes dilute syntactic foams. As anticipated, CnH models were unable to accurately describe either response for \( \phi = 40\% \).

### 4.3. Compressible Ogden type-I

In this model, the strain energy ansatz takes the form

\[
W = \sum_{j=1}^{N} \left\{ \frac{2 \mu_j}{\alpha_j^2} \left[ \bar{\lambda}_1^{\alpha_j} + \bar{\lambda}_2^{\alpha_j} + \bar{\lambda}_3^{\alpha_j} - 3 \right] + \frac{1}{D_j} (J - 1)^{2j} \right\},
\]

(5a)

where \( \mu_j, \alpha_j, \) and \( D_j \) are real-valued constants. Under uniaxial compression

\[
\sigma_E = \frac{\partial W}{\partial \lambda_z} = \sum_{k=1}^{N} \left\{ \frac{4 \mu_k}{3 \alpha_k \lambda_z} \left[ (J^{-1/3} \lambda_z)^{\alpha_k} - (J^{-1/3} \lambda_z)^{-\alpha_k/2} \right] + \frac{2 k J}{D_k \lambda_z} (J - 1)^{2k-1} \right\},
\]

(5b)

where \( J \) is obtained by solving the compressibility relation

\[
\sum_{k=1}^{N} \left\{ \frac{2 \mu_k}{3 \alpha_k J} \left[ (J^{-1/3} \lambda_z)^{-\alpha_k/2} - (J^{-1/3} \lambda_z)^{\alpha_k} \right] + \frac{2 k}{D_k} (J - 1)^{2k-1} \right\} = 0.
\]

(5c)
Fig. 8 presents results for OgI fitted models for $\phi = 0\%, 2\%$, and $10\%$, under different truncation values $N$. In these instances, the axial stress-strain and transverse strain data were qualitatively well-modelled, even in the simplest setting of $N = 1$. However, Fig. 9 reveals that OgI strain energy models do not possess an appropriate $f(J)$ to accurately recover the transverse strain response for $\phi = 40\%$.

4.4. Compressible Ogden type-II

In this model, the strain energy ansatz takes the form

$$W = \sum_{k=1}^{N} \frac{2\mu_k}{\alpha_k^2} \left[ \lambda_1^{\alpha_k} + \lambda_2^{\alpha_k} + \lambda_3^{\alpha_k} + \frac{1}{\beta_k} (J^{-\alpha_k \beta_k} - 1) \right],$$

(6a)

where $\alpha_k$, $\beta_k$, and $\mu_k$ are real-valued constants. Under uniaxial compression

$$\sigma_E = \frac{\partial W}{\partial \lambda_z} = 2 \frac{\mu_k}{\lambda_z} \sum_{k=1}^{N} \frac{\mu_k}{\alpha_k} \left[ \lambda_2^{\alpha_k} - J^{-\alpha_k \beta_k} \right],$$

(6b)

where $J = \lambda_t^2 \lambda_z$ is obtained by solving the compressibility relation

$$\sum_{k=1}^{N} \frac{2\mu_k}{\alpha_k} \left[ \lambda_2^{\alpha_k} - (\lambda_t^2 \lambda_z)^{-\alpha_k \beta_k} \right] = 0.$$  

(6c)

Fig. 10 presents results for an OgII strain energy model at $\phi = 40\%$ (other filling fractions were not examined in order to avoid numerical instability issues with modelling weakly compressible syntactic foams with highly compressible $f(J)$ ansatzes). For the truncation $N = 4$, an accurate description of both the axial stress-strain and transverse strain behaviours was finally recovered. Consequently, OgII strain energy ansatzes were found to be appropriate forms for describing the compressive performance of high filling fraction HTM syntactic foams.
4.5. Small-strain accuracy of fitted models

Using the coefficients of the above fitted models, the associated small-strain material constants were computed to provide a quantitative measure for goodness-of-fit at small strains (details and Tabels given in Supplementary Material). The CnH models were found to be within a 5% relative error for $\phi = 0\%$ and $\phi = 2\%$ foams, however, for $\phi = 10\%$ and $\phi = 40\%$ foams, the deviation in the Young’s modulus was sufficiently large as to preclude the CnH model from quantitatively describing HTM foams at small strains. In contrast, OgI models with $N \geq 2$ recover the ground state constants for all filling fractions to high accuracy. Also, OgII models for $\phi = 40\%$ recover constants within a relative error of 10% for all $N$.

4.6. Large-strain accuracy of fitted models

Using the coefficients of the above fitted models, the stress-strain response was computed to a higher strain level of 70% and compared against test data. Fig. 11(a) presents results for $\phi = 0\%$, showing that all fitted models were stable to 70% strain, with CnH and OgI ($N = 1$) being the most accurate. Fig. 11(b) presents a similar picture for $\phi = 2\%$, where CnH and OgI ($N = 1, 3$) were accurate and stable, but that OgI ($N = 2, 4$) truncations were unstable, due to the range in fitted coefficient values. Fig. 11(c) shows that all models were accurate to 70% strain for $\phi = 10\%$, with the exception of OgI ($N = 2$). For $\phi = 40\%$, Fig. 11(d) reveals that all CnH and OgI models were either inaccurate or unstable at higher strains, and Fig. 11(e) shows that only OgII ($N = 1$) is able to qualitatively describe the response in the extended strain regime, which may well correspond to the plastic deformation region.

5. Conclusions

A combined theoretical and experimental investigation into the compressive performance of hollow thermoplastic microsphere (HTM) syntactic foams revealed increased...
stiffness with increasing filling fraction $\phi$, strong recoverability, and a non-trivial transverse stretch relationship for large $\phi$. Strain energy models were constructed to describe both the stress-strain and transverse strain response of the foams under uniaxial compression, where it was found that OgI ($N = 3$) models achieve the best fits for $\phi = 0\%$, 2\%, and 10\% foams, up to strains of 50\%, as they recover accurate ground state constants, accurate qualitative descriptions up to 50\% strain, and were extendable to higher strains. In contrast, only OgII ($N = 4$) strain energy models were able to describe high filling fraction ($\phi = 40\%$) foams, due to the general form of the compressibility condition $f(J)$. The strain energy descriptions obtained are anticipated to prove useful for industrial applications, the development of future strain energy models, and for the validation of micromechanical-based models. The recoverability properties of these foams show considerable promise when coupled with their small-strain stiffening response.

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Interest Statement

No competing interests were reported by the authors.

Appendix A: Supplementary Material

Supplemental Material related to this article is given at www.dx.doi.com/xx.xxxx/j.compscitech.xxxx.xx.xxx.
Figure 1: (Caption in list of figures)

Figure 2: (Caption in list of figures)
Figure 3: (Caption in list of figures)

Figure 4: (Caption in list of figures)
Figure 5: (Caption in list of figures)

Figure 6: (Caption in list of figures)
Figure 7: (Caption in list of figures)

(a)

(b)

Figure 8: (Caption in list of figures)
Figure 9: (Caption in list of figures)

Figure 10: (Caption in list of figures)
Figure 11: (Caption in list of figures)
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| Properties | Value          |
|------------|---------------|
| Particle diameter | 55-85 µm   |
| Shell thickness  | 0.35 µm     |
| Bulk density    | 30 ± 3 kg/m³ |

Table 1: Properties of the hollow thermoplastic microspheres

| Y₀ [MPa] | ν₀   | ρ [kg/m³] | Y₀/ρ [×10³ m²/s²] |
|----------|------|-----------|-------------------|
| 0%       | 7.08 | 0.49      | 1081              |
| 2%       | 7.10 | 0.48      | 1061              |
| 10%      | 7.53 | 0.46      | 980               |
| 40%      | 10.9 | 0.34      | 670               |

Table 2: Test values for small-strain bulk properties of HTM syntactic foams: Young’s modulus Y₀, Poisson’s ratio ν₀, density ρ, and specific stiffness Y₀/ρ.