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Fatigue behaviour in hybrid hollow microspheres/fibre reinforced composites

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Abstract This article presents the results of a current study concerning the influence of the addition of short fibres on the fatigue behaviour of syntactic foams. The material was obtained by vacuum-assisted resin transfer moulding adding hollow glass microspheres to an epoxy resin acting as binding matrix. Specimens with microsphere contents up to 50% and fibre reinforcement up to 1.2% in volume were tested at three-point bending at room temperature. Foams show significantly lower static and fatigue strength than an epoxy matrix. A significant decrease in the absolute strength with filler increase was observed, and even specific strength decreases for low filler contents and is nearly constant for the higher filler contents. Fatigue strength also decreases with the increase in filler content. The addition of glass fibre reinforcement produces only a slight improvement in flexure strength, while the addition of carbon fibres promotes an important improvement; a hybrid composite containing 0.9% carbon fibre is about 30% stronger than unreinforced foams. An improvement in fatigue strength more than 30% was obtained by the addition of small percentages of glass or carbon fibre.

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Introduction

Low-density sheet moulding compounds based on hollow glass micro-spheres are being increasingly used in the automotive industry, boats and as core materials. Compared to traditional metallic materials they exhibit advantages such as lower weight, no corrosion effects, more design freedom, etc. Also, they are able to reduce impact force and consequently could be potentially useful materials for applications with impact loads [1, 2]. As reported by Kim and Khamis [2], the addition of hollow microspheres tends to reduce the Young's modulus and ultimate strength. On the other hand, the specific impact force increases with the volume fraction of microspheres [2]. Wouterson et al. [3] conclude that the effect of the microsphere volume fraction on the specific tensile and flexural strength and stiffness depends not only on the filler content but also on the density of micro-spheres and thickness to radius ratio. When used as core materials in sandwich configurations, the compressive properties of syntactic foams also play an important role. Studies of syntactic foams' compressive properties can be found in the work of Rizzi et al. [4], Gupta et al. [5–8] and Kim and Plubrai [9].

Recent studies reported that using short fibre reinforced hybrid foams led to improvements in strength and fracture toughness. Wouterson et al. [10] observed a concomitant strengthening and toughening in foams manufactured with phenolic microspheres and epoxy resin binding, reinforced with 3 wt% content of short carbon fibres (3–10 mm length). Huang et al. [11] obtained important enhancements in compression and tensile strength by small additions of chopped carbon and aramid fibres in foams fabricated with amino microspheres and phenolic resin binding.

In spite of the abundant literature reporting the static mechanical behaviour of syntactic foam, few studies are reported on fatigue behaviour. However, Zenkert and Burman [12] verified a significant effect of the density on the fatigue strength under tension, compression and shear loading of high performance closed cell rigid polymer polymetacrylimide (PMI) foams. Fatigue strength was fitted using Basquin's law. Bezazi and Scarpa [13] investigated auxetic and conventional PU foams subjected to tensile fatigue tests concluding that the stiffness degradation and fatigue endurance of the auxetic foam were improved compared to the conventional parent phase. Bezazi and Scarpa [14] observed that for compression fatigue the auxetic foam shows dissipates significantly higher energy per unit volume than the conventional foam, at various loading levels. The fatigue behaviour of sandwich configurations incorporating core foam materials was studied among others by Shafiq and Quispitupa [15], Harte et al. [16] and Burman and Zenkert [17, 18].

Recent studies developed by Brown et al. [19] obtained significant improvement to fracture toughness on epoxy filled by urea–formaldehyde microcapsule using dicyclopentadiene healing agent. Fracture toughness was strongly dependent on the size and concentration of microcapsules. The concentration of microcapsules, at which the maximum value was reached, decreases with decrease in the diameter of microcapsule. The fracture toughening was observed only when good adhesion between the microcapsules and epoxy matrix was obtained. Brown et al. [20] studied also the fatigue crack propagation in same materials. The addition of microcapsules significantly decreased the fatigue crack growth rate and increased the fatigue life above a transition value of the stress intensity factor. Below that value, the fatigue behaviour was unaffected by the embedded microcapsules.

The present investigation aims to study the fatigue behaviour of syntactic foams manufactured with hollow glass micro-spheres and an epoxy resin binder. The influences of the following parameters on the fatigue strength and on the fatigue damage were studied: (i) the filler content on unreinforced foams, (ii) the reinforcement type and (iii) the weight fraction of the reinforcement.

Materials processing and testing

Three types of foams were produced: unreinforced, reinforced by short glass fibres and reinforced by short carbon fibres. K20 (3M™, St. Paul, Minnesota, USA) hollow glass microspheres, of which 50% have nominally a diameter less than 55 μm, were used as a lightweight filler. The resin used for binding the micro-spheres was 520 epoxy with 523 hardener supplied by Ashland Chemical Hispania (Benicarlo, Spain). Two types of reinforcement fibres were used: a general purpose chopped strand of E 3313 glass supplied by PPG with an average length of 3 mm and a continuous T300 carbon fibre thread supplied by Toray, which was also cut to 3 mm lengths. Resin and hardener were mixed in a pot and afterwards the microspheres (and fibre) were added while stirring the mixture. Vacuum-assisted resin transfer moulding was used to produce composite sheets in an aluminium mould with a rectangular parallelepiped cavity of 200 × 200 × 6 mm. The mould was cleaned using acetone, and hydroalcoholic solution was used as release agent (Polyvinyl alcohol, PVA), supplied by Nautica Escalada (Spain).

In order to separately analyse the influence of filler content, reinforcement fibre type and reinforcement content, 11 formulation plates were prepared. Table 1 summarizes the contents of each formulation and their densities obtained according to the Archimedes principle.

Test specimens were cut and machined from the moulded sheets to the dimensions of 65 × 12 × 6 mm^3 and subjected to three-point bending tests with a span of 48 mm. Static bending tests were performed at displacement rate of 1 mm/min, using a Shimadzu AG-10 10 kN universal testing machine, equipped with a 1-kN load cell and TRAPEZIUM X software.

The fatigue tests were carried out at room temperature in an electromechanical machine equipped with a 5-kN load cell to monitor the load. This machine allows such parameters as frequency and stress ratio to be varied. Constant amplitude displacement was applied with a sinusoidal wave form at a frequency of 10 Hz and a stress ratio of $R = 0.05$. During the tests, the load and the number of cycles were recorded in a computer file. Figure 1 shows a view of the specially designed three-point bending rig for the fatigue tests. As is known, in specimens of polymer matrix composites submitted to relatively high frequency cyclic loading, there can be a rise in temperature with consequent changes in material behaviour. In order to check this possibility, some specimens were instrumented with thermocouples at the surface in middle of the sample and the temperature was monitored during the tests. A negligible increase in temperature was observed during the continuous measurement procedure.

The nominal bending stress (σ) was calculated using:

$$\sigma = \frac{3PL}{2bh^2} \quad (1)$$

being P the load, L the span length, b the width and h the thickness of the specimen.

Table 1 Materials formulation and bending properties

Fibre reinforcement	Fibre reinforcement content, V_{fr} (vol.%)	Microballons content, V_{fm} (vol.%)	Density (g/cm^3)	Apparent modulus of elasticity (GPa)	Standard deviation (GPa)	Ultimate bending strength (MPa)	Standard deviation (MPa)	Fatigue strength for 1 million of cycles (MPa)	Standard deviation (log σ)
–	0	0	1.170	2.59	0.06	141.6	1.62	24.2	0.032
–	0	10	1.071	2.33	0.13	92.1	6.91	–	–
–	0	26	0.915	2.22	0.01	65.0	1.03	10.58	0.039
–	0	43	0.757	1.72	0.03	41.3	2.41	6.87	0.084
–	0	50	0.690	1.79	0.02	43.5	2.06	7.64	0.025
Glass	0.4	10	1.074	2.13	0.11	75.6	3.93	11.84	0.037
Glass	1.2	10	1.103	2.42	0.15	81.4	2.32	14.13	0.011
Glass	0.3	43	0.790	1.72	0.12	42.1	1.68	9.14	0.038
Glass	0.9	43	0.794	1.73	0.16	46.0	1.53	9.45	0.045
Carbon	0.4	43	0.792	2.08	0.19	50.1	9.80	9.14	0.039
Carbon	0.9	43	0.796	2.41	0.20	53.5	9.36	9.58	0.006

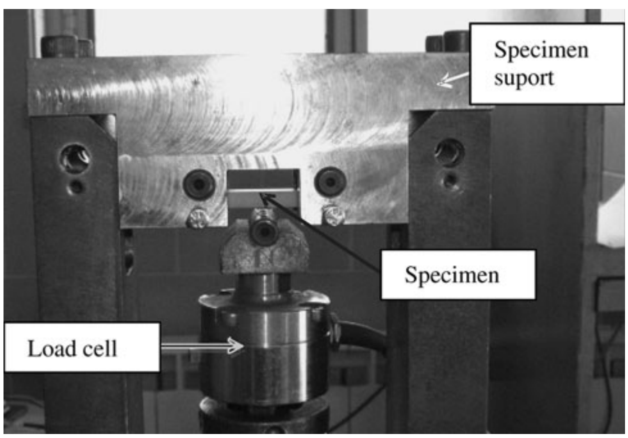

Fig. 1 View of 3 PB test rig

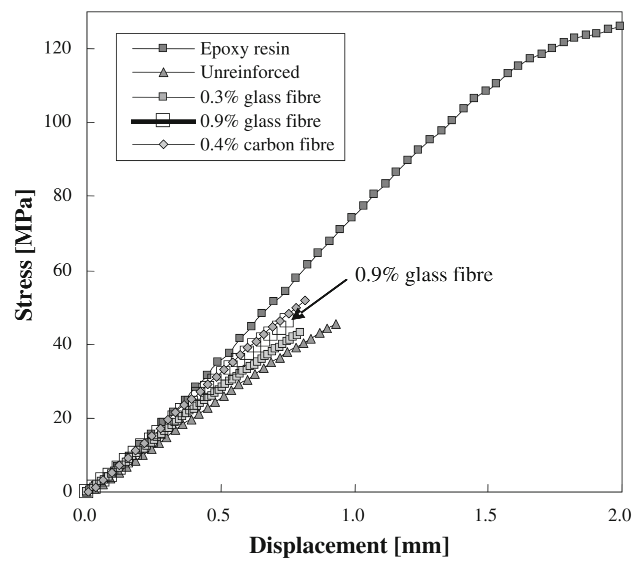

Fig. 2 Typical stress–displacement curves for different reinforcements. Foams filled 43% by volume

Results and discussion

Static strength

Figure 2 shows typical load–bending displacement curves for neat resin, unreinforced foams with 43% in volume of microspheres and for reinforced composites with a low percentage of glass (or carbon) fibre. Agreeing with Wouterson et al. [10], the figure shows an evident improvement in both stiffness and ultimate strength with the addition of a small amount of fibre reinforcement, particularly when carbon fibre is used, for which stiffness is very closed with neat resin. The displacement on fracture was much lower for all composites than for neat resin. Table 1 also summarizes the average values and standard deviation of both apparent modulus of elasticity and ultimate strength. Average values were calculated from the current values of at least five tests on each composite formulation. Bending strength was obtained using the peak load in Eq. 1, while the apparent modulus of elasticity was calculated from the linear elastic bending beam theory relationship:

$$E = \frac{\Delta P \cdot L^3}{48 \Delta u \cdot I} \tag{2}$$

where I is the inertia moment of the transverse section and ΔP and Δu are, respectively, the load range and flexural displacement range at middle span, calculated in the linear region of the load, versus displacement plot. The apparent modulus of elasticity value was calculated by linear regression on the stress versus strain record, ranging from zero to a defined strain value. The upper limit of strain, here used, was the maximum value that provides a correlation coefficient in linear regression of at least 99%.

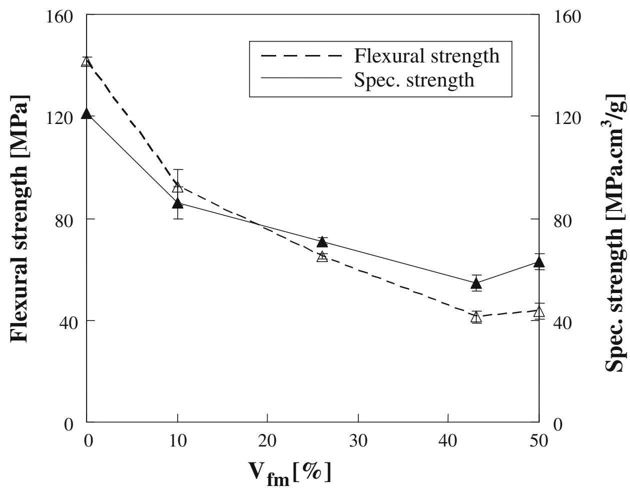

Fig. 3 Flexural strength versus microspheres volume content

Figure 3 shows average values and standard deviation of bending strength plotted against microsphere content of unreinforced foams. A large decrease in absolute strength with filler increase was observed, in accordance with Kim and Khamis [2]. Even specific strength decreases for low filler content and tends to stabilize only for high filler content. As is well known, strength and other mechanical properties of polymeric materials are highly sensitive to loading rate. The authors [21] obtained a significant increase in the strength with loading rate, reaching more than a 30% increase when the loading rate increases from 0.05 to 500 mm/min. The static strength sensitivity to the loading rate suggests that fatigue strength will be also influenced by frequency loading.

Figure 4 summarizes the results obtained for the bending strength against fibre reinforcement content. The addition of glass fibres produces only a slight improvement in bending strength for both 10 and 43% filler contents. However, the addition of a small percentage of carbon fibres produces an improvement of about 30% in bending strength when a hybrid composite containing 0.9% carbon fibre is compared to unreinforced foam. This trend is in accordance with the results obtained by Wouterson et al. from tensile testing [10].

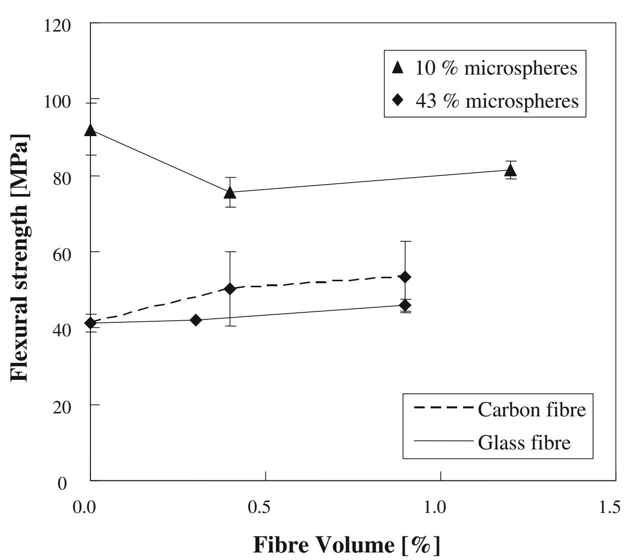

Fig. 4 Ultimate flexure strength against fibre volume fraction

Fatigue behaviour

Figures 5, 6 and 7 show fatigue life results obtained for unreinforced foams, glass fibre reinforced foams and carbon fibre reinforced foams, respectively. In these figures, it was plotted the stress range at the early fatigue cycles against the number of cycles to failure. Experimental results and medium stress range versus the number of cycles to failure (*S*–*N* curves) are superimposed.

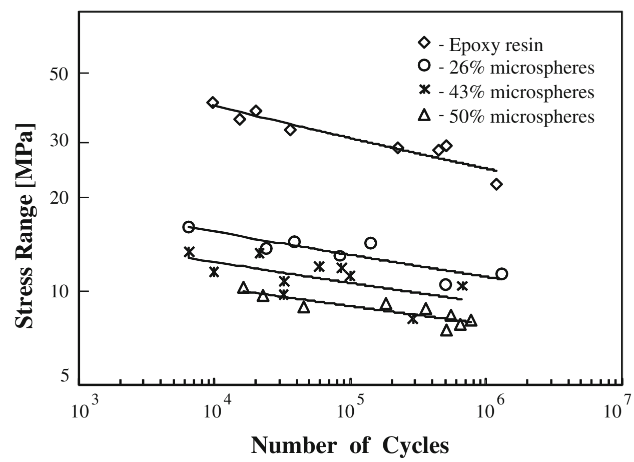

Fig. 5 Effect of filler content on fatigue endurance of unreinforced foams

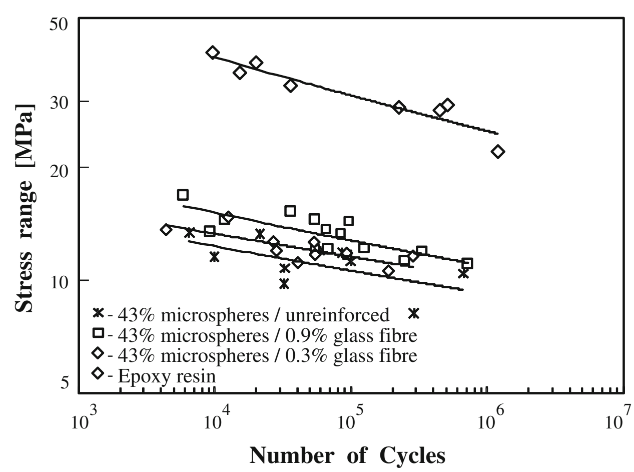

Fig. 6 Effect of glass fibre reinforcement on fatigue endurance

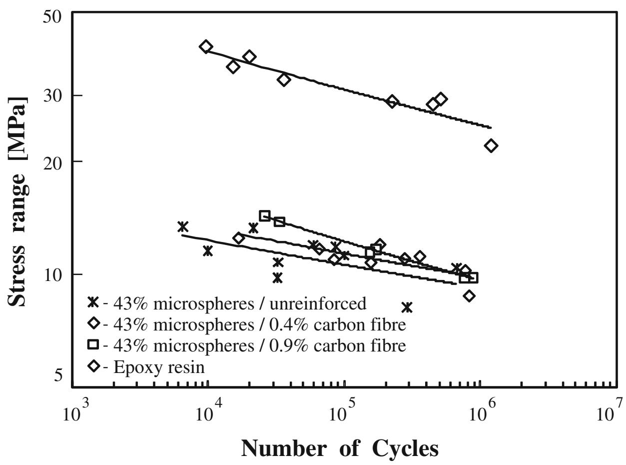

Fig. 7 Effect of carbon fibre reinforcement on fatigue endurance

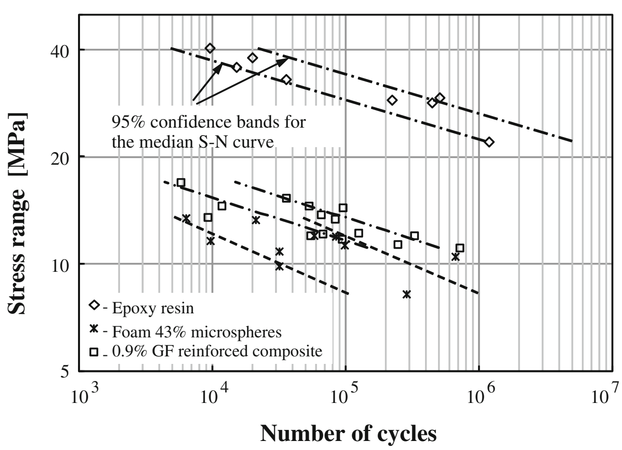

Fig. 8 Comparison of the dispersion bands for the neat resin, an unreinforced and a reinforced composite

Fatigue results have a significant spread, common in this type of materials. The results were statistically analyzed, according to ASTM E739 standard. Table 1 indicates the median stress range for 1 million of fatigue life and the standard deviation in terms of log σ. Figure 8 shows three examples of the 95% confidence bands for the median *S–N* curves for the epoxy resin, unreinforced syntactic foam with 43% microsphere and a glass fibre reinforced composite. According this figure and also Table 1, the dispersion bands are similar for hybrid composites and neat resin and tend to be lesser than for the foams.

According to Fig. 5, filled foams show a significant loss of fatigue strength compared to the epoxy matrix. Contrary to results of Brown et al. [20] that obtained improved resistance to fatigue crack propagation, attributed to toughening mechanisms induced by the embedded microcapsules as well as crack shielding due to the release of fluid as the capsules are ruptured, present results show that fatigue strength decreases with filler content, following the same trend previously observed for static tests. This loss in strength can be partially attributed by the lower resistance of microspheres and mainly caused by poor interface adherence, working in this case the microspheres as potential voids. For 1 million of cycles, foams with 26% microsphere content show about 56% loss of fatigue strength, in comparison with matrix resistance. If the filler content is increased to 50%, the loss of fatigue strength reaches about 68%.

Figure 6 shows fatigue results obtained from the glass fibre reinforced specimens with 43% in microspheres content and compares them with unreinforced foam and neat resin test series. The addition of glass fibres promotes a significant lengthening of the reinforced composites' fatigue life; increasing with the fibre reinforcement content. For 1 million of cycles, the addition of 0.3% fibre increases the fatigue strength about 33% in comparison with unreinforced foams. However, every composite remains fatigue strength much lower than epoxy neat resin.

The addition of small amounts of carbon fibre only promotes significant lengthening of fatigue life of reinforced composites in comparison with unreinforced foams, as shows Fig. 7 and as summarized in Table 1. Composites reinforced with 0.4% carbon fibre show fatigue strength improvement similar to 0.3% glass fibre reinforced composites, as indicated in Table 1 for 1 million of cycles. However, the increasing of fibre content from 0.4 to 0.9% improves the fatigue strength below expected.

Fracture surfaces were examined by scanning electronic microscopy (SEM). Figure 9 shows some pictures of these observations, which are apparently similar to ones obtained for static loads, except that in this case multiple microcracks can be observed. For low filler content in Fig. 9a, b show abundant step structure failure starting from microspheres and carbon or glass/epoxy interfaces appearing to be the dominant failure mechanism. Step structure failure is characterized as fracture in different planes that starts from a fibre or microparticle due to stress concentration or interface decohesion and eventually bridges between surrounding filler particles. This is in agreement with the work of Wouterson et al. [10] who also observed this mechanism to be dominant for static failures in carbon fibre/epoxy resin reinforced foams. The fibres contribute to the formation of additional step structures from which new fracture surfaces are formed. As observed in Fig. 9b, in many cases, for low microspheres content, microcracks initiate at interfaces and grow by fatigue striation into the resin matrix. For high filler content, fibre and microsphere debonding play apparently the most significant role on fatigue failure, as shown in Fig. 9c, d. Abundant microcracks initiated at microspheres and fibre interfaces grow first along the interfaces and by end towards the matrix.

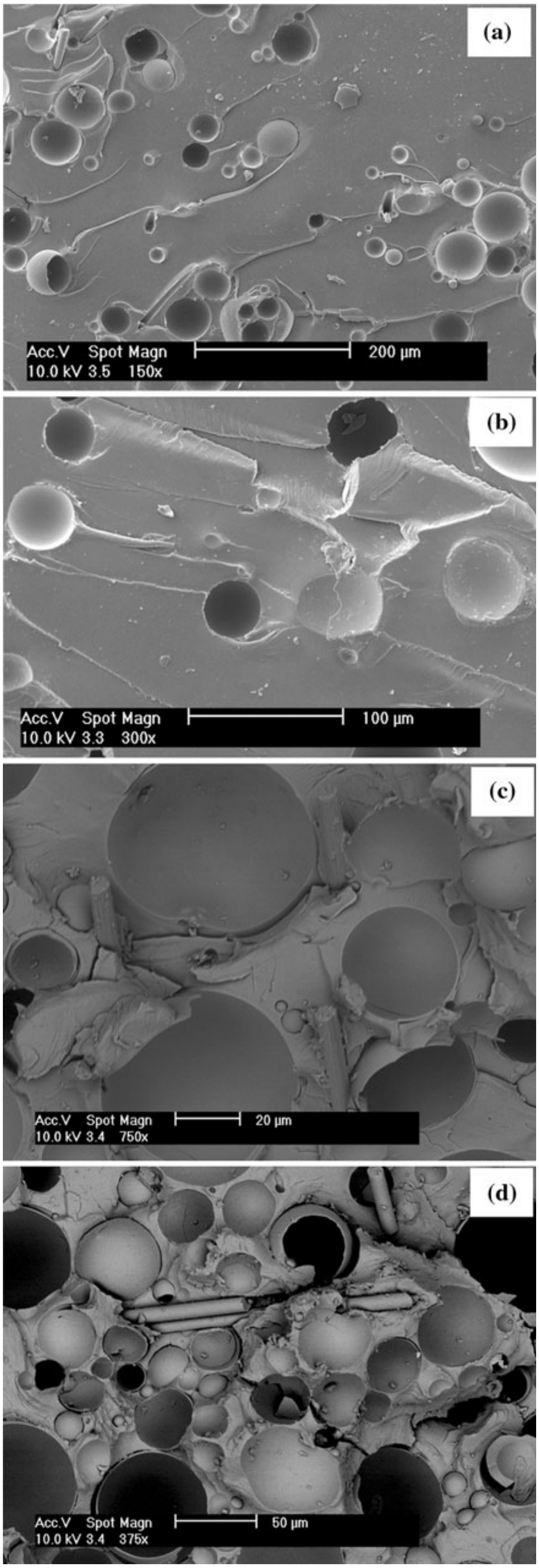

Fig. 9 SEM observations of failure surfaces: **a** $V_{fm} = 10\%/1\%$ glass fibre; **b** $V_{fm} = 10\%$/unreinforced; **c** $V_{fm} = 43\%/1\%$ carbon fibre; **d** $V_{fm} = 43\%/1\%$ glass fibre

Fatigue damage of composites synchronously builds up with the increasing number of loading cycles, leading to the change in mechanical properties, such as the decrease in strength or stiffness of the material. Fatigue damage, particularly for fibre composites, has been modelled using the evolution of the apparent modulus of elasticity. Generally, this is nonlinear; as was observed previously in [22] and in a recent study by Wu and Yao [23], which proposes a new damage model.

The apparent modulus of elasticity was measured during fatigue tests. Both maximum and minimum values of load cycle (P_{max} and P_{min}) were monitored together with corresponding displacements (u_{max} and u_{min}). These values were used to calculate the apparent modulus of elasticity (E) from Eq. 2, where $\Delta P = P_{max} - P_{min}$ and $\Delta u = u_{max} - u_{min}$. Figure 10 plots the evolution of E/E_0 versus N/N_f observed in some tests for different composite configurations, where E_0 is the initial value of E, N the current number of cycles and N_f the number of cycles to failure. Significant differences in the evolution of the non-dimensional apparent modulus were observed in Fig. 10a, corresponding to an unreinforced foam, and the other figures, corresponding to fibre reinforced composites. Figure 10a shows that for unreinforced foam stiffness remains nearly constant until failure, whereas for reinforced composites in the majority of tests the stiffness decreases continuously from an early stage of the fatigue process until it undergoes a sudden drop corresponding to final failure. This aspect of the non-dimensional modulus loss curves is apparently independent of both fibre content and fibre type (glass or carbon). Also, stress range load does not appear to play an important role in reducing stiffness since its effect is less than scatter. Figure 10a together with SEM observations suggest that for the unreinforced foams the fatigue initiates at microsphere interfaces, afterwards the microcracks grow within the interfaces without significant global stiffness change and at end grow quickly at neat resin up to final failure. On the other hand, in hybrid composites damage growing is longer corresponding to the development of macroscopic cracks at the fibre interfaces, promoting a decrease in the stiffness from earlier cycles of fatigue.

Conclusions

This article is concerned with both the static and fatigue bending strength of glass microspheres/epoxy resin binder syntactic foams. The following effects were assessed: (i) filler content in unreinforced foams, (ii) fibre reinforcement type and its volume fraction in reinforced foams.

An important decrease in the absolute strength was observed with increasing filler content, and even specific strength decreases for low filler content becoming nearly

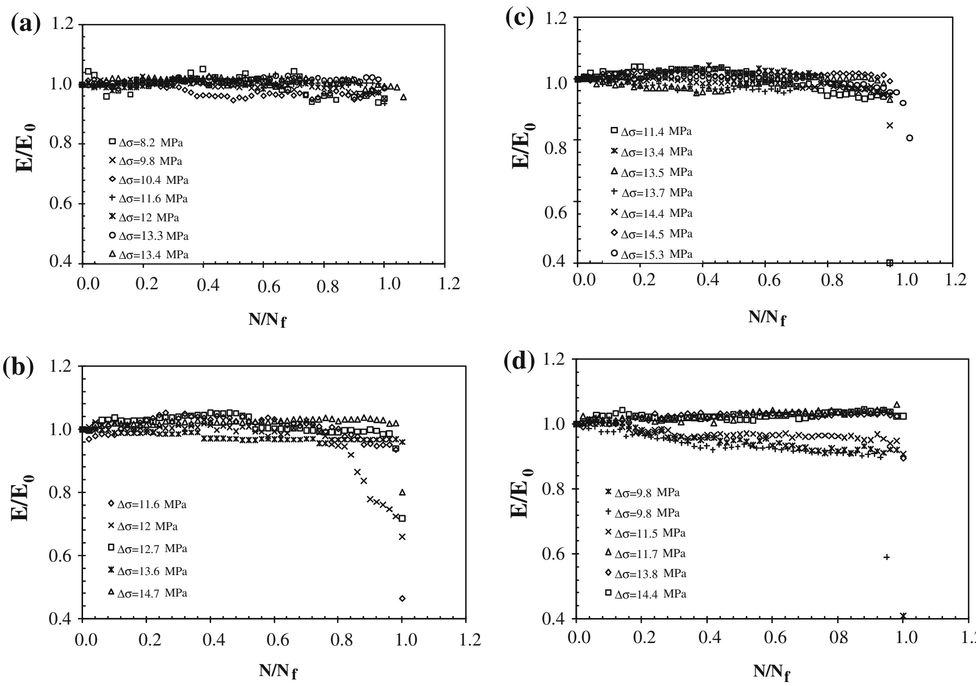

Fig. 10 Stiffness damage versus number of cycles evolutions with $V_{fm} = 43\%$: **a** unreinforced foams; **b** 0.3% glass reinforced foams; **c** 0.9% glass reinforced foams; **d** 0.9% carbon reinforced foams

constant for high filler content. A continuous decrease in the fatigue strength with increasing filler content was also observed. The addition of glass fibres produced only a slight improvement in bending strength when compared to unreinforced foam, whereas significant improvement in the bending strength was obtained for the hybrid composite containing 0.9% carbon fibre. An improvement in fatigue strength more than 30% was obtained by the addition of small percentages of glass or carbon fibre.

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