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Influence of processing and testing conditions on the mechanical behaviour of sheet-moulding compound laminates

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Studies have been carried out in order to optimize the compression-moulding cycle for two classes of commercial sheet-moulding compounds, standard and low-shrink prepregs. The laminates have been moulded at different temperature and pressure conditions, and afterwards their mechanical behaviour has been analysed by flexural and tensile tests at various temperatures and strain rates. Furthermore, dynamic-mechanical measurements have been used to correlate temperature-dependent viscoelastic properties, and the structure of the moulded materials.

1. Introduction

Sheet-moulding compound (SMC) technology has been used in manufacturing large composite automobile, aerospace and general industrial applications for more than two decades. The current trends of improving the surface appearance and mechanical properties can lead to even more applications both for exterior and structural parts [1].

The moulded SMC consists of a thermoset polymeric matrix and chopped glass fibres. The matrix is composed of a styrenated polyester resin including low-profile thermoplastic additives, large loadings of calcium carbonate filler, and small amounts of reactive chemicals such as thickening and curing agents.

One of the advantages of these materials is their easy manufacturing technique by compression moulding. The cure cycle of the material depends on its formulation, thickness and surface properties required [1–3]. During a moulding operation, the compound flows and solidifies into the desired shape under pressure and heat.

Many factors, such as the presence of low-shrink additives or air-release agents, contribute to surface properties variation [4]. Also, the processing conditions (moulding temperature and pressure, curing time and material flow during moulding) influence the concentration and compaction of SMC ingredients [2], which subsequently influence the mechanical properties of the material.

In the present work the mechanical properties of two different commercial materials, standard and low-shrink laminates, cured at several temperature and pressure-moulding processing conditions were investigated. The mechanical properties were analysed by flexural tests at various temperatures and strain rates, studying the modulus of elasticity, the flexural strength and the fracture strain, and also by tensile tests, in order to study the fracture behaviour at different strain rates. Finally, dynamic-mechanical measurements were used to evaluate temperature-dependent viscoelastic properties in flexure using a vertical clamp assembly.

2. Experimental procedure

2.1. Materials

A standard SMC APG 10-R, ST, and a low-shrink SMC APG 10-RS, LS (Astar S.A.) were studied. The formulations, as given by the manufacturer, are listed in Table I. The low-shrink material contained high-impact polystyrene (HIPS) as modifier.

2.2. SMC moulding

All compounds were moulded in a laboratory hydraulic press SATIM P.M.L.1, with pressure and temperature programming.

For the study of the optimum moulding processing conditions of each material, moulding temperatures from 125–145 °C under a 14.7 MPa moulding pressure over the material were used. In addition, other cycles from 10.9–16.4 MPa moulding pressure at 140 °C moulding temperature were carried out in order to determine the optimum moulding pressure.

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Furthermore, the influence of the test temperature and the strain rate during the test on the mechanical properties was investigated, by moulding laminates at 140 °C moulding temperature and a 14.7 MPa moulding pressure over the material.

The curing time and the amount of material used were the same for all the laminates.

2.3. Test specimens and conditions

Three-point loading flexural tests were carried out in a universal mechanical test machine Instron, model 4206, following the standard method of test for flexural properties of plastics ASTM D-790. The strain rate was 1.7 mm min^{-1} and the support span was 64 mm. Tensile test-pieces were machined following the sizes recommended in the standard method ASTM D 5083-90.

At least eight specimens were tested for each processing condition. They were cut with a diamond saw from the central part of the laminates with 80 mm × 10 mm × 4 mm dimensions.

Before testing each specimen, they were kept in a desiccator to avoid environmental variables influencing the testing process.

Tests between 20 and 100 °C were performed in order to investigate the influence of test temperature on the mechanical behaviour of both materials. Before testing, the specimens were kept for 3 h inside the thermal chamber of the Instron universal testing machine. In order to analyse the influence of the strain rate, flexural as well as tensile tests were carried out in the Instron testing machine within the following range of test rates: 0.1–20 mm min^{-1} in flexion and 0.5–20 mm min^{-1} in traction.

2.4. Dynamic-mechanical thermal analysis (DMTA)

Damping and complex modulus variations of the laminates can be studied by varying the frequency of application of a determined load or the test temperature. Changes in the internal molecular mobility of the material can be obtained.

In this study, temperature-dependent elastic modulus and damping, tan δ, were determined in flexure with a Metravib dynamic mechanical viscoanalyser using a vertical clamp assembly. A sinusoidal controlled strain was imposed at 10 Hz frequency. Rectangular specimens of 60 mm × 10 mm × 4 mm dimensions were cut from the laminates moulded at different temperature and pressure conditions for both materials. Data were acquired in the temperature range 20–250 °C at a heating rate of 3 °C min^{-1}.

3. Results and discussion

3.1. Influence of moulding conditions

Laminates of SMC cured under 14.7 MPa moulding pressure over the material, with a moulding temperature between 125 and 145 °C, were studied in flexion in order to analyse the influence of the moulding temperature in the mechanical behaviour of these laminates. The behaviour of the elastic modulus and the break strength with regard to the moulding temperature, is shown in Figs 1 and 2 for the standard and low-shrink laminates, respectively. For both materials, the laminates acquired an optimum value at a 135 °C moulding temperature and under 14.7 MPa moulding pressure over the material. For the higher moulding temperatures, the elastic modulus and the fracture strength decrease drastically, while for the lower moulding temperatures the decrease was not so severe.

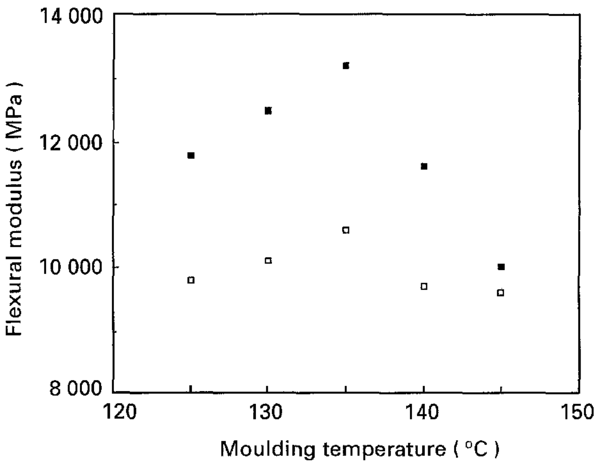

Figure 1 Elastic modulus variation as a function of moulding temperature. (□) Standard, (■) low-shrink laminates.

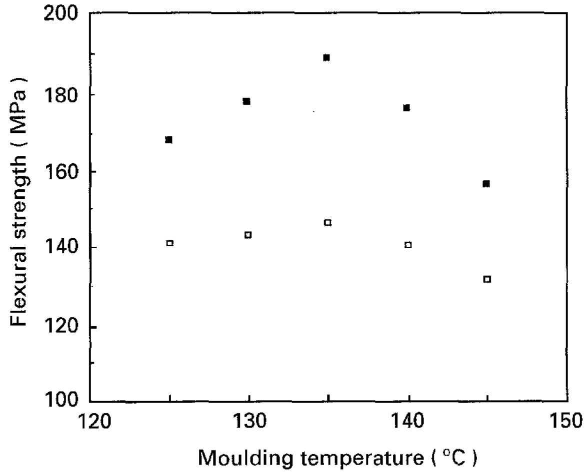

Figure 2 Strength of the laminates as a function of moulding temperature. For key, see Fig. 1.

Compression moulding of the material used supposes that the material flows at an earlier stage of the moulding process, filling the mould cavity at low temperature, and subsequently cure reactions take place as the material attains the cure temperature chosen. Therefore, the variation of elastic modulus and break strength of these materials should be a direct consequence of changes on the cure-reaction progress [1, 3] and not because of physical variations.

On the other hand, the values of the low-shrink laminates are higher than those for standard laminates at all the moulding temperatures used. As shown in Table I, the only obvious difference in the composition between the standard material and the low-shrink

TABLE I Composition of the SMCs used

Ingredient	APG 10-R (wt %)	APG 10-RS (wt %)
Ortophtalic resin	25	15
Low-shrink additive (high-impact polystyrene)	–	10
Catalyst	0.25	0.25
Zinc stearate	1.25	1.25
Inorganic fillers	42.5	42.5
Colorant pigments	4.5	4.5
Thickening agent, Mg (OH)$_2$	1.5	1.5
Glass fibre	25	25

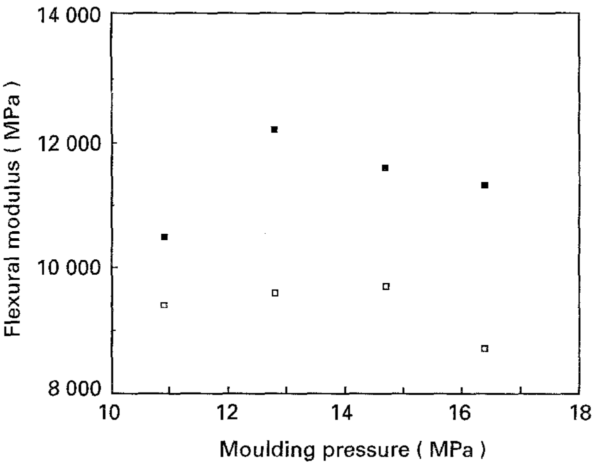

Figure 3 Elastic modulus variation as a function of moulding pressure. For key, see Fig. 1.

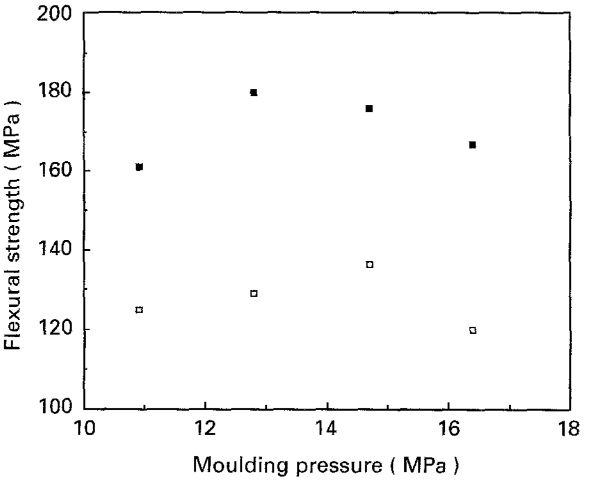

Figure 4 Strength of the laminates as a function of moulding pressure. For key, see Fig. 1.

material is the low-shrink additive (HIPS) that appears in the low-shrink material. By applying the rule of mixtures proposed by Nielsen [5], $E = \Sigma\phi_i E_i$, where E is the modulus of elasticity of the global material, ϕ the volume fraction and the subscript i refers to each component of the formulation, and taking into account that the modulus of elasticity of the polystyrene is higher than the elastic modulus of the polyester, the modulus value of the low-shrink material should be higher than that for the standard material. In addition, the difference in behaviour might also be related to differences in homogeneity of the material because, as has been shown [6, 7], the addition of a thermoplastic to polyester resins implies phase separation during curing that modifies the cure kinetics as well as the conversion on each phase with respect to that corresponding to the neat resin [8, 9]. On the other hand, the shrinkage of the polyester resin in the standard material could develop internal stresses in the matrix–fibre interphase, which possibly would imply a smaller adherence between both components. Ulterior considerations are presented below when dynamic-mechanical behaviour of these materials is analysed.

The behaviour of the flexural strength as a function of cure temperature, Fig. 2, was similar to that shown by the elastic modulus. This behaviour can be attributed, along with the other parameters mentioned above, to the action of the thermoplastic additive, which minimized, at the end of the curing process, the shrinkage effects that occurred in the standard material. Less internal stress is generated in the low-shrink material during the curing process and the cooling stage than in the standard material [10], possibly as a consequence of variations in the interphase between the resin and the fibre.

In order to analyse the influence of the moulding pressure on the mechanical behaviour of these laminates, a 140 °C moulding temperature was chosen under a set of 10.9, 12.8, 14.7 and 16.4 MPa moulding pressures over the material during the compression process. The stiffness variation of both SMC materials with regard to the used moulding pressure is shown in Fig. 3. While the optimum moulding pressure over the material appeared to be 14.7 MPa for the standard material, it was 12.8–14.7 MPa for the low-shrink material. As the flow of these materials is controlled at each cure temperature by the moulding pressure applied, as well as by the curing extent, the pressure applied over the material is a very important factor that may affect the fibre disposition. The behaviour of the flexural strength with regard to the moulding pressure over both materials is shown in Fig. 4. The maximum values obtained for break strength coincided with the moulding pressures and the corresponding maximum values of the modulus of elasticity. It seems that an excess moulding pressure drags the fibre away from the central parts of the mould, thus decreasing in an appreciable way the maximum strength level that the material can support [11]. The values of the standard SMC were lower at all the moulding pressure ranges than those for the low-shrink material, possibly as a consequence of higher internal stress. However, as it has recently been shown [12], the curing rate and the final degree of cure of unsaturated polyester resins may be affected by the pressure set, so modifying the mechanical properties.

When studying the strain properties, it is known that unlike metals and ductile plastics, SMC composites do not yield and so their strain at break is low (near 2%). For all the moulding pressures used, the strain for the standard material was 0.018 mm mm^{-1}, and 0.021 mm mm^{-1} for the low-shrink material, showing again the interest in adding high-impact polystyrene to the standard resin.

From the above results, it becomes evident that in the low-shrink SMC used, the added thermoplastic, used in the industry with the objective of improving the surface quality and decreasing the moulding shrinkage of the standard material, also increases the rigidity and the strength of the material in a way depending on the moulding conditions used. Moreover, the high-impact polystyrene added decreases slightly the fragility of these materials, probably as a consequence of a variation in the mechanism of the matrix deformation [4], as has been shown in epoxy systems modified with thermoplastics [13] or also by modification of the interphase between the glass fibres and the matrix. An investigation of the influence of these kinds of modifications with polyester matrix without fibre is now underway in our laboratory [14].

3.2. Influence of test conditions on the mechanical behaviour

Flexural as well as tensile tests have been carried out in order to determine the variation of mechanical properties of the laminates as a function of testing temperature and strain rate.

For the study of the influence of the temperature on the mechanical properties of both SMC materials, a 140 °C moulding temperature and a 14.7 MPa moulding pressure over the material, were chosen. Before studying the specimens by three-point loading flexural tests in a 20–100 °C range, samples were maintained in the Instron thermal chamber for 3 h at the test temperature selected.

As shown in Figs 5 and 6, the values of the mechanical properties corresponding to small deformations decreased remarkably as the test temperature increased, which agrees with other results in the literature [11, 12, 15]. Indeed, the stiffness of the material decreased by almost 50% for both SMC materials when the temperature changed from 20 °C to 100 °C. As shown below, a similar decrease, though in a lower magnitude, in the storage modulus, E', determined by dynamic flexural tests at 10 Hz, was also observed. This decrement in mechanical properties as a function of temperature is due to the variation of the matrix behaviour, and also to that for the matrix–fibre interphase in this temperature range [16]. As the temperature increased, the mobility of the resin chains became higher, therefore progressively softening the resin and so the material, in spite of the properties of the fibres not being affected substantially by the temperature increase analysed. At 100 °C, the stress–strain behaviour of both materials was quite different to that at room temperature. The elastic modulus and flexural strength decreased drastically and the fracture strain increased in an appreciable way with increasing temperature as a consequence of the higher chain mobility in the resin (Figs 5, 6 and 7).

On the other hand, the mechanical properties of low-shrink SMC laminates were higher than that for the standard ones at all temperatures used. This fact can be explained taking into account the Nielsen mixture rule for small deformation properties, as well as the variations in the internal structure of the cured resin due to kinetic modifications when the thermoplastic was added. Thus, values of elastic modulus became closer upon increasing the test temperature, because the glass transition temperature of the HIPS is

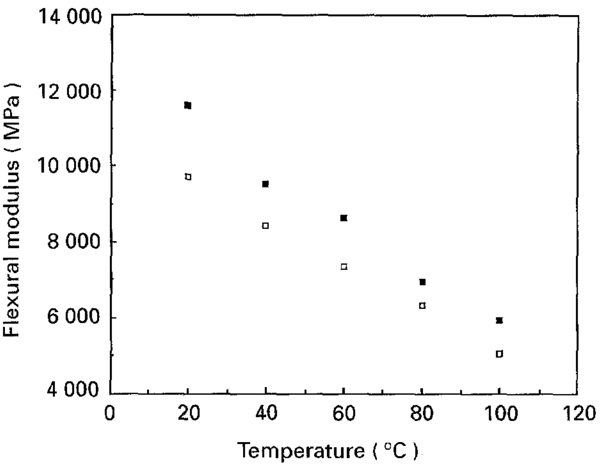

Figure 5 Elastic modulus variation as a function of test temperature. For key, see Fig. 1.

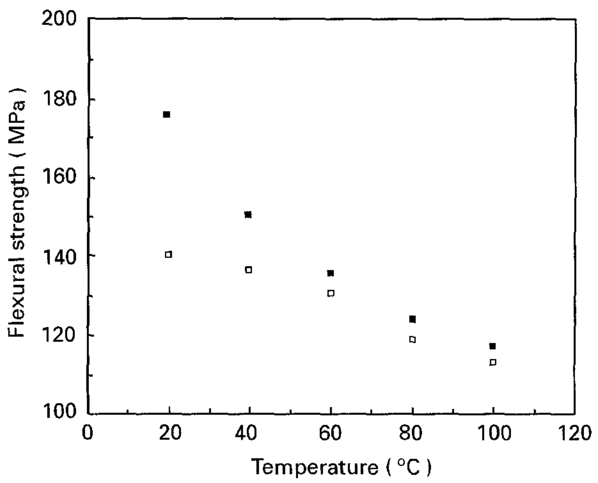

Figure 6 Strength of the laminates as a function of test temperature. For key, see Fig. 1.

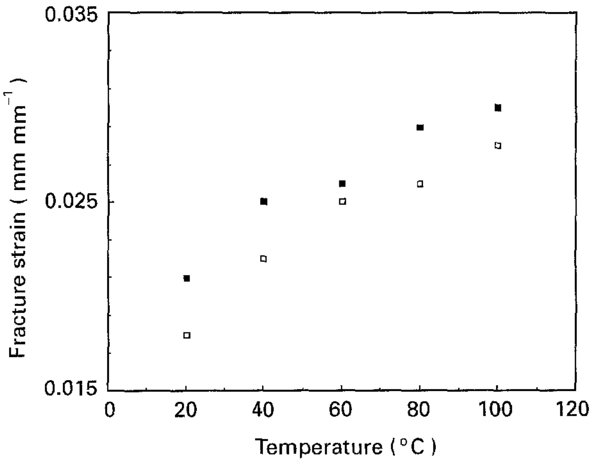

Figure 7 Deformability of the laminates as a function of test temperature. For key, see Fig. 1.

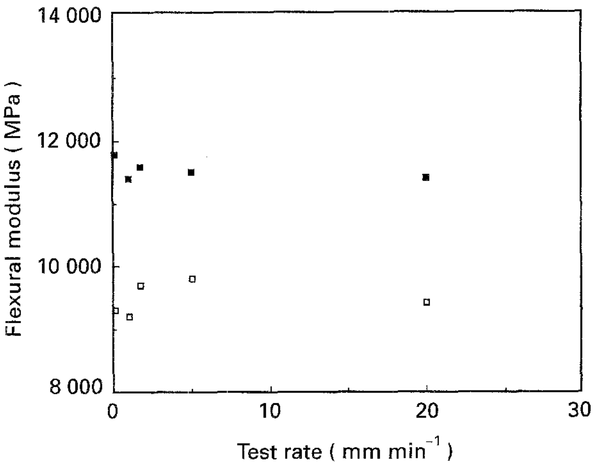

Figure 8 Elastic modulus variation as a function of test rate. For key, see Fig. 1.

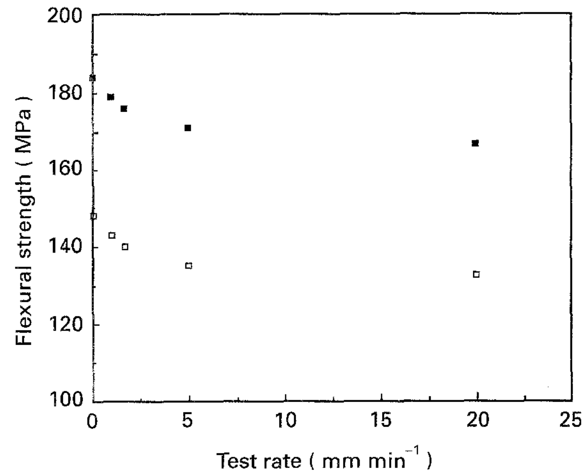

Figure 9 Strength of the laminates as a function of the test rate. For key, see Fig. 1.

lower than that for the resin. In addition, on increasing the temperature, the matrix relaxed, and so the strain at break increased as the test temperature was raised. Thus the action of the HIPS on the mechanical properties of the material was only significant at temperatures far away from its glass transition.

The influence of strain rate during the mechanical testing was analysed using standard and low-shrink laminates processed at 140 °C and 14.7 MPa. Flexural tests were carried out at room temperature at cross-head rates from 0.5–20 mm min^{-1}. The elastic modulus is plotted as a function of the test rate in Fig. 8. For both laminates, the values of elastic modulus did not show any significant variation at the test rates studied, higher values being obtained for the low-shrink laminate at all conditions used.

On the contrary, the flexural strength of both laminates showed an evident decrease when the strain rate was higher, as shown in Fig. 9. This decrease was possibly due to the increasing fragility of the laminates as the strain rate increased, Table II, because of the increasing stress in the matrix/fibre interphase. However, the explanation for this behaviour is complicated because of the different types of stress implied in a flexural test. The higher strength and deformation values, Fig. 9 and Table II, obtained for the laminates modified with HIPS, seem to indicate that the thermoplastic addition could modify the deformation mechanisms in the matrix and also in the interphase because, as has been found in our laboratories [17], the strength of neat polyesters containing no fibres can be decreased by the effect of thermoplastic addition.

TABLE II Deformability of SMCs as a function of test rate

Test rate (mm min^{-1})	Fracture strain (mm mm^{-1})	
	APG 10-R	APG 10-RS
0.1	0.240	0.410
1.0	0.031	0.038
1.7	0.018	0.022
5.0	0.005	0.007
20.0	0.001	0.002

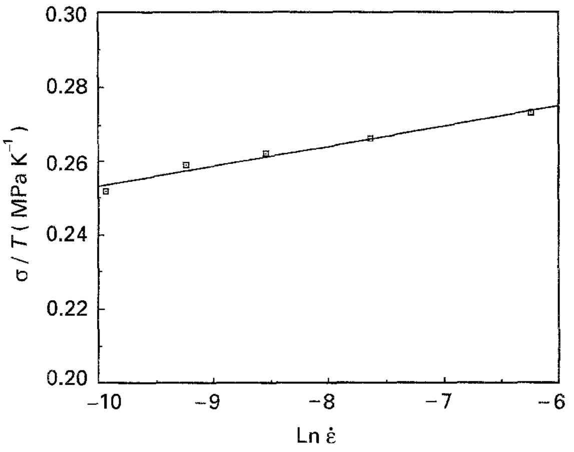

Figure 10 Ratio of the strength to temperature against the logarithm of the strain rate ($\dot{\varepsilon}$ in s^{-1}) for tensile tests.

In order to understand the strength behaviour, tensile tests conducted at room temperature and several strain rates, $\dot{\varepsilon}$, were also carried out on the standard laminate. The plot of σ_t/T as a function of ln $\dot{\varepsilon}$ is given in Fig. 10. An acceptable fit of the data was obtained by using an Eyring-type equation [18], $\sigma_t/T = A + B$ ln $\dot{\varepsilon}$, to simulate the flow resistance of the laminate.

3.3. Dynamic-mechanical behaviour

In an attempt to ascertain the influence of moulding temperature and pressure on the structure of these materials, their dynamic-mechanical behaviour has been investigated from 20–250 °C at a 10 Hz constant frequency. Mechanical spectra in flexion of these materials studied by means of the storage modulus, E', and loss factor (tan $\delta = E''/E'$) show complete mapping of the viscoelastic behaviour.

Results for the standard SMC moulded at several moulding temperatures under 14.7 MPa moulding pressure are presented in Fig. 11. At all moulding temperatures used, a very broad α relaxation was obtained. This behaviour has been attributed in neat polyester resins to the heterogeneity of the structure of the network formed in the curing process [19–21]. It

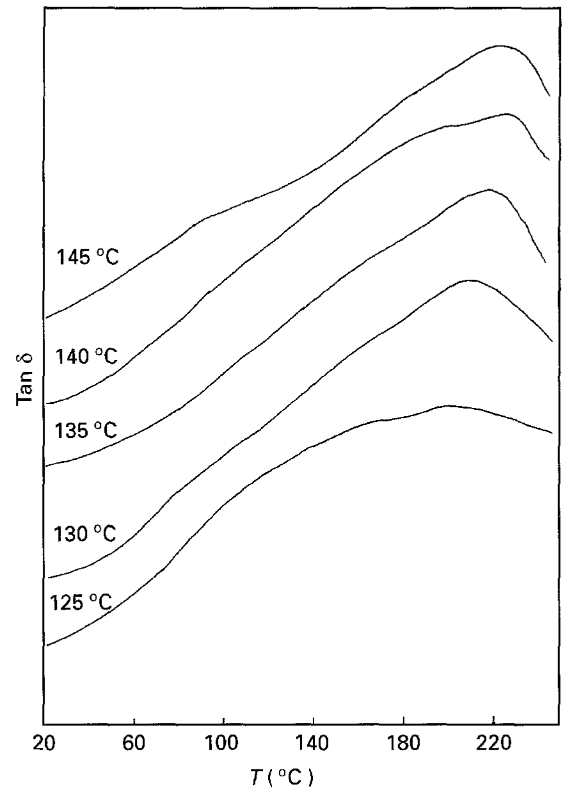

Figure 11 Damping curves of standard laminates moulded at 14.7 MPa.

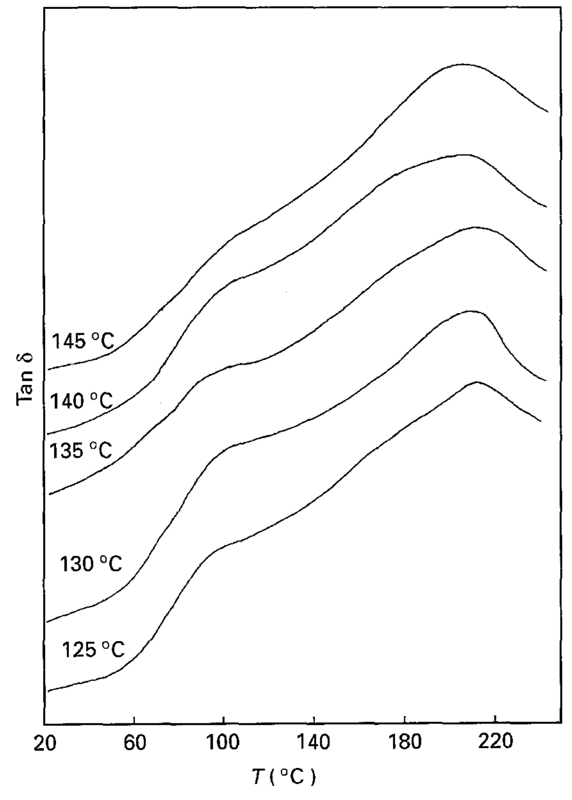

Figure 12 Damping curves of low-shrink laminates moulded at 14.7 MPa.

has been postulated [22], that the microstructure morphology of the network would consist basically of intramolecularly connected high cross-link microdomains, in which the styrene and polyester C=C bond conversions could vary as a function of curing conditions [12]. Moreover, the anisotropy introduced by the fibres would also contribute in some way to the increase in the broadness of the α relaxation. Also, it is well known [23] that employing fillers increases the broadness of the α relaxation in these materials. On the other hand, temperatures corresponding to the maximum value of the loss peak slightly increased with increasing moulding temperature, possibly as a consequence of variations on the conversion of styrene and polyester C=C bonds.

As shown in Fig. 12, the dynamic behaviour of low-shrink SMCs was even more complex than for the standard ones, because two α relaxations were observed at every moulding temperature used. In addition to the broad damping peak attributable to the heterogeneity of the cured polyester resin, the glass transition peak corresponding to the polystyrene of the low-shrink modifier, appeared as a shoulder around 100 °C. Both relaxations became closer as moulding temperature increased, showing a higher compatibility of the multiphase structure. When this behaviour is compared with that for the standard material, Figs 11 and 12, differences in the relaxation behaviour for both materials become evident, possibly because the introduction of the low-shrink additive modified the curing evolution in a way that permitted more homogeneous polyester networks to be obtained for the modified laminates, so improving the matrix/fibre interphase. This is possibly the reason, together with those cited above, why the mechanical properties of low-shrink materials were higher than those for the standard ones at all moulding temperatures.

On the other hand, the decrease in the elastic modulus and strength at high moulding temperatures could be due to some degradation occurring during the moulding cycle used. However, kinetic investigations on these materials must be carried out at the moulding conditions used in order to have a more complete knowledge of the variations shown in this study.

In addition, Figs 13 and 14 show the evolution of the storage modulus as a function of temperature for these materials. A slight decrease in the elastic modulus was observed at temperatures lower than 80 °C for both materials. On the other hand, a sharp decrease of elastic modulus, though lower than for the corresponding neat resin, was observed for both materials in the α relaxation region of the polyester resin. Moreover, the low-shrink laminates showed further lowering of the elastic modulus in the region in which the glass transition of polystyrene appeared, i.e. between 60 and 110 °C. This relaxation could be responsible for the closer values of mechanical properties of both laminates at the high temperatures used in the static tests, Figs 5 and 6, because the elastic modulus, which decreases for both materials as the temperature increased, showed a more evident variation in the region of the α relaxation of polystyrene.

In order to study the influence of moulding pressure on the dynamic behaviour, the damping corresponding to the standard and low-shrink laminates has been plotted against temperature in Figs 15 and 16, respec-

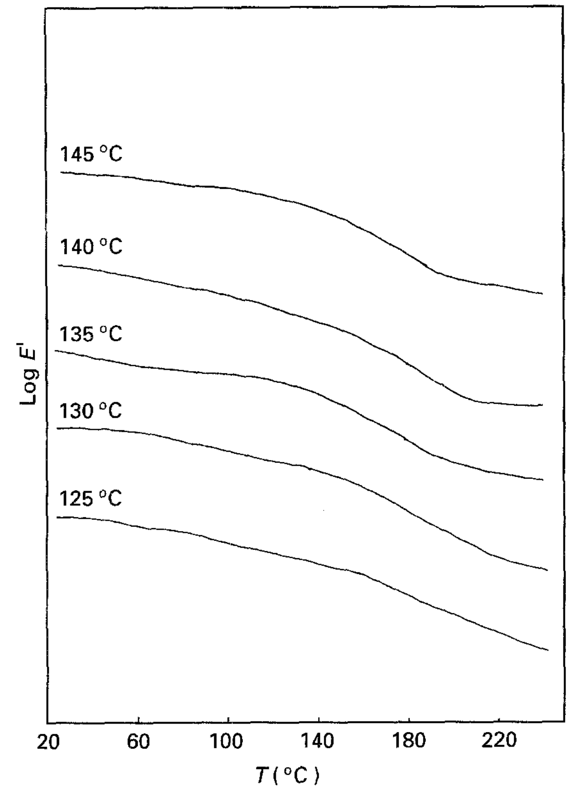

Figure 13 Storage modulus variation for standard laminates moulded at 14.7 MPa.

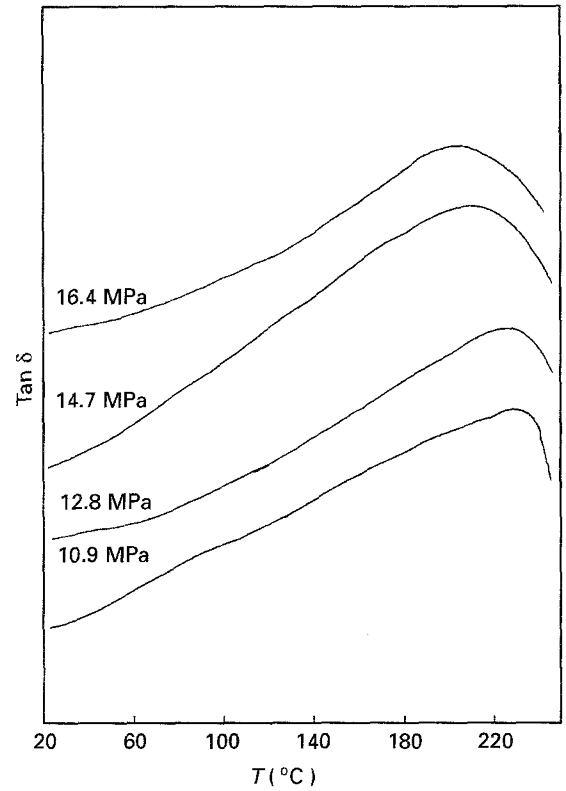

Figure 15 Damping curves of standard laminates moulded at 140 °C.

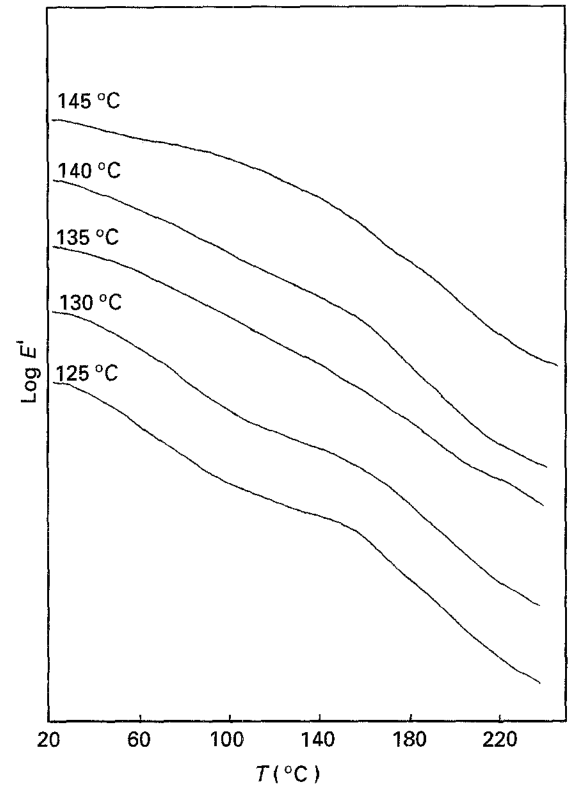

Figure 14 Storage modulus variation for low-shrink laminates moulded at 14.7 MPa.

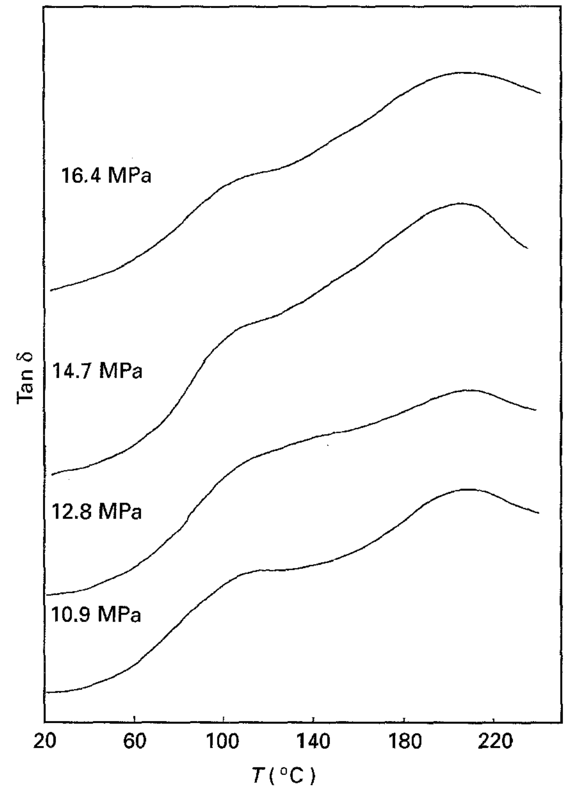

Figure 16 Damping curves of low-shrink laminates moulded at 140 °C

tively, for samples moulded at 140 °C and different pressures over the material. For both laminates, increasing pressure supposed a slight lowering in the damping peak temperature corresponding to the cured polyester resin, which is more evident in the standard laminate. This behaviour is possibly related to differences in the final conversion of the resin. Indeed, in a recent paper, Huang *et al.* [12] have shown that increasing pressure increases the styrene conversion more than the polyester C=C bond conversion by about 5%, and the final conversion decreases when high pressures are applied over the material.

On the other hand, as shown in Fig. 16, insignificant variations were observed in the α relaxation corresponding to the thermoplastic additive as moulding pressure varied. Going back to the static mechanical behaviour shown in Figs 1–4, the significant decrease in the elastic modulus and strength at high temperatures seems to be attributable to a certain degradation of the matrix in the matrix–fibre interphase. On the other hand, at the high moulding pressures used, with the exception of the low-shrink laminate moulded at 10.9 MPa, only slight variations of mechanical properties were observed, possibly because the variation of moulding pressure did not significantly affect the interphase between fibre and matrix.

4. Conclusions

Polyesters reinforced with glass fibres have been investigated in order to determine the influence of moulding parameters and test conditions on the mechanical properties of SMC materials. The conclusions obtained can be summarized as follows.

1. Both elastic modulus and flexural strength of the laminates analysed are a function of the temperature and pressure employed in the moulding process. Optimum values of these properties were obtained at 135 °C when laminates were moulded under 14.7 MPa moulding pressure. Although the influence of the moulding pressure on the mechanical properties was lower than that of the moulding temperature, an evident variation of elastic modulus and flexural strength appeared when the pressure was varied.
2. An interesting thermoplastic modification of the resin was evident because the mechanical properties of low-shrink laminates were higher than those for the standard ones at all moulding temperatures and pressures used, possibly as a consequence of variations in the fibre/matrix interphase and/or because of changes in the internal structure of the cured resin.
3. At temperatures lower than the glass transition of the cured resin, both elastic modulus and flexural strength showed an evident decrease when the test temperature was increased. However, both properties were higher for the low-shrink laminate than for the standard one at all temperatures tested.
4. The strain rate during testing has an evident influence in the mechanical behaviour of these materials despite the opposite trends observed in flexural and tensile tests. For the latter ones, the strength variation could be approximated by an Eyring-type equation.
5. Variations of the relaxational transitions as the moulding temperature changed were shown for both types of laminate, possibly because of differences in the internal structure of the matrix.
6. For both laminates, the α relaxation corresponding to the resin moved slightly to lower temperatures as moulding pressure increased.

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