

The thermal expansion of carbon fibre-reinforced plastics

Part 5 *The influence of matrix curing characteristics*

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The volume changes which occur during the cure of resins employed as matrices for carbon fibre-reinforced plastics have been monitored. A description of the apparatus is given and detailed results throughout the curing cycles of resins Derakane 411-48/MEKP, Epikote 828/MNA/BDMA and XB2878A/XB2878B are reported, together with results for the later stages of cure of resin DLS 351/BF₃400. Conclusions are drawn concerning: (i) the overall percentage volume contraction during cure, (ii) the shape and time-scale of the curing characteristics in relation to the state of completion of the curing process, (iii) the approximate location of the temperature below which no further shrinking due to polymerization or cross-linking is likely to occur.

1. Introduction

The prediction of the behaviour of carbon fibre-reinforced plastics (CFRP) structures in a variety of environments is a subject of considerable importance in their technology. The response of the dimensions of CFRP to temperature change depends upon: (i) the thermal and elastic properties of the constituents of the composite, (ii) stresses built into the structure during its preparation and (iii) residual stress relaxation and swelling stresses induced by moisture absorption [1]. Earlier investigations in these laboratories [2-5] have been concerned with the temperature dependence of the thermal expansion characteristics of some of the laminate configurations used in practical CFRP structures. The investigations have identified the principal characteristics which result from the properties mentioned in (i) above. That part of (ii) which relates to the resin must include

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the changes in volume, viscosity and adhesion which occur during the preparation of a composite.

This paper reports the volume changes which occur during the curing cycles of a representative selection of resins.

2. The resin systems

Three epoxy and one vinyl ester resin were selected for the study. These were:

- (i) Derakane 411-48/MEKP vinyl ester resin[†]
- (ii) Epikote 828/MNA/BDMA[‡]
- (iii) XB2878A/XB2878B[§]
- (iv) DLS351/BF₃400.

3. Apparatus

In order to accommodate liquid resin the dilatometer shown in Fig. 1 was constructed. The object was to contain the resin in the bulb E, the volume of which was typically 15 cm³, and to monitor its temperature with a thermocouple, the tip of which was located at the centre of the bulb within a re-entrant well which formed an extension of limb

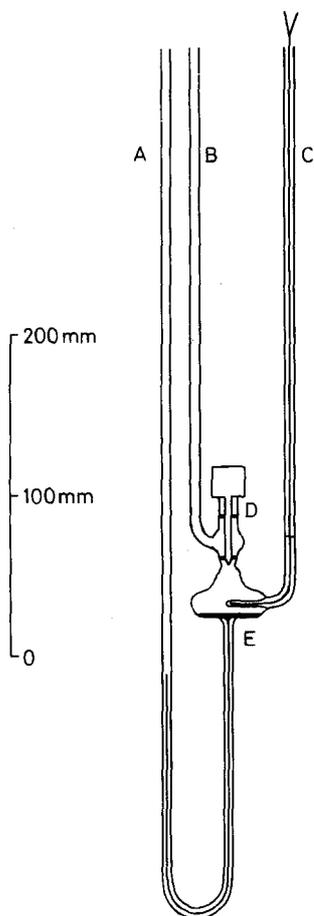


Figure 1 The manometer dilatometer. A, open limb of manometer; B, pumping tube; C, thermocouple sheath; D, tap; E, bulb containing resin.

C. The lower, sealed end of this limb contained silicone fluid in order to provide thermal communication between the thermocouple and the resin. The thermocouple voltage was recorded by a potentiometer and chart recorder. After chemically cleaning the bulb, its volume was determined by filling it with mercury, which was subsequently removed and weighed. The interior of the bulb was then coated with a parting agent (a 2% solution of Dow Corning silicone high vacuum grease in carbon tetrachloride). Mercury was introduced into limb A until the meniscus in the right-hand limb of the U-portion covered the base of the bulb E. In the cases of the first three resins the constituents were then weighed out, mixed, and the resulting liquid was allowed to stand for approximately 10 min, after which a vacuum was applied for approximately 10 min in order to remove dissolved gases. With the stopper of the tap D removed, the

resin was then poured into the bulb through the tap socket. After removing any bubbles of trapped air, the stopper was replaced and a vacuum was applied through B for approximately 10 min as a further precaution to remove dissolved gases. The stopper was screwed down and the dilatometer was supported within a viscometer bath containing silicone fluid and controllable within $\pm 0.01^\circ\text{C}$. Subsequent movement of the meniscus in limb A was monitored with the aid of a cathetometer sighted through a window in the wall of the bath.

Problems associated with adhesion during the later stages of the curing processes led to the adoption of alternative means of measuring resin shrinkage in these regions. The actual procedures finally adopted in the individual cases will be described in the sections which follow, without elaborating upon the exploratory work from which it was finally concluded that these problems had arisen from the use of the parting agent. Reference will be made in these later sections to the employment of an interferometer dilatometer and a dial-gauge dilatometer.

The original form of the interferometer dilatometer, which is described elsewhere [6], has been modified subsequently by the incorporation of an additional circular optically flat plate situated immediately below the prism of the original apparatus so that the two circular optical flats now constitute the interferometer. Additional improvements have been effected by re-siting the sensing thermocouple of the temperature controller and by enclosing the specimens in a more closely controlled environment, so that the limits between which temperatures may be held constant has been reduced.

The dial-gauge dilatometer took the form of a quartz tube, sealed at its lower end, in which rested a partially cured specimen in the form of a rod, 4 mm diameter and 50 mm long. The tube contained silicone fluid in order to achieve thermal contact between its wall, the specimen and a thermocouple used for temperature measurement. Temperature change was effected by the viscometer bath in which the tube was supported, and dimensional changes in the specimen were communicated to a sensitive dial gauge through a quartz push rod. Appropriate precautions were taken and corrections were applied for the uncompensated lengths of quartz. The reliance which could be placed upon subsequent measurements made with the

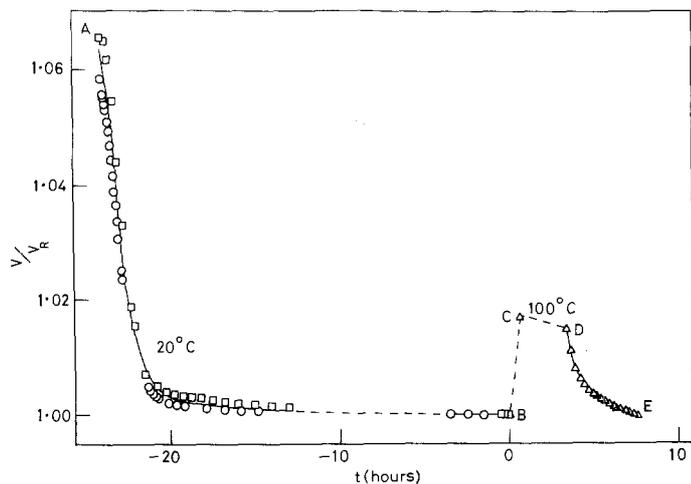


Figure 2 The time dependence of the reduced volume V/V_R of Derakane 411-48/MEKP resin during the curing process. The reference temperature, R , and the arbitrary zero of time have been taken at the point B, at which dilatometers were changed. \circ , manometer dilatometer results 1; \square , manometer dilatometer results 2; \triangle , interferometer dilatometer results; --- interpolated.

dilatometer was established by measuring the linear thermal expansion coefficient of a rod of copper over the temperature range covered in the later resin work. The results of these measurements agreed with those published by the National Bureau of Standards [7] within the combined experimental uncertainties.

4. Experimental procedure and results

4.1. Derakane 411-48/MEKP vinyl ester resin

1 g methyl ethyl ketone peroxide (MEKP) 50%, 0.5 g NL 63/ST amine and 0.5 g NL 49/ST cobalt naphthenate were added to 50 g Derakane 411-48. The resulting liquid was mixed thoroughly and placed in the manometer dilatometer within the viscometer bath, applying the preparatory treatment described earlier. Volume changes were recorded over the following 24 h, during which the temperature was maintained at 20°C. Specimens suitable for use in the interferometer dilatometer were prepared from a portion of the same batch of resin which had been pre-cured at approximately 20°C for several days. These were mounted in the interferometer dilatometer and their changes of length were measured as the temperature was raised to 100°C, during the following 3 h at 100°C and during the cooling period which followed. The combined results are displayed graphically in Fig. 2. In these and subsequent evaluations of the results obtained using the manometer dilatometer, corrections were applied in order to allow for dimensional changes of the vessel and the mercury.

Because of local arrangements concerning the relative dispositions of items of equipment required

during the early stages of the curing process, it was difficult to ensure the precise reproducibility of conditions and times preceding the first observation. After observing that the volume was much less critically dependent upon time towards the end of the 24 h period at 20°C than it was at the beginning, the end B of this period, denoted by AB in Fig. 2, was selected as the arbitrary origin of reference. Thus the reference temperature, R , employed in expressing the volumes of resin, V , in reduced form V/V_R , was 20°C in this instance. The results were not corrected in any way in order to achieve better agreement between the two sets in the region of AB where V/V_R is very dependent upon time. The difference between the two sets of results in this region provides a direct measure of the difficulty of reproducibility preceding the first observations, such as might be encountered in a typical commercial production.

The form of representation adopted in Fig. 2 is appropriate for the isothermal regions AB and CD, in which the dependence of reduced volume upon time provides an indication of the rate at which cross-linking is proceeding. The first rapid change in slope in the neighbourhood of $V/V_R = 1.005$ is very marked, but does not seem to be directly related to the gel time, which was measured as 23 min at 20°C in a separate experiment. On the basis of the results displayed in this graph taken in isolation, the fact that the slope of CD exceeds that of the latter part of AB leads one to conclude that the 20°C period might be reduced substantially without significant loss, while an extension of the period at 100°C would result in a more completely cured resin. The essentially time-

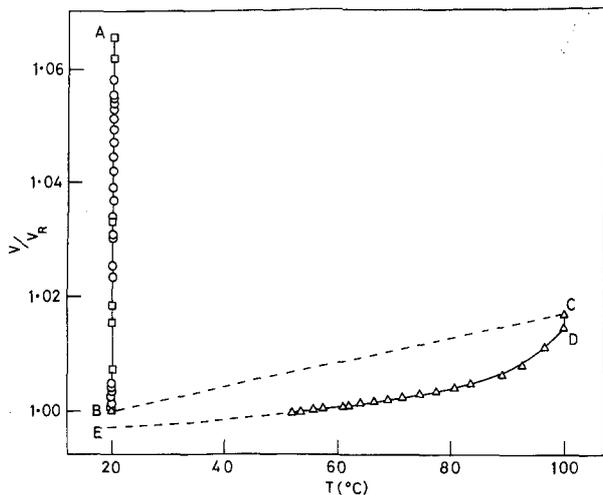


Figure 3 The temperature dependence of the reduced volume V/V_R of Derakane 411-48/MEKP resin during the curing process, in which the symbols A to E denote the same stages as in Fig. 2. The reference temperature, R , and the arbitrary zero of time are also as in Fig. 2. \circ , manometer dilatometer results 1; \square , manometer dilatometer results 2; \triangle , interferometer dilatometer results; --- interpolated.

independent character of AB in the neighbourhood of B also indicates the degree of latitude which is permissible in the time of pre-cure adopted for the resin from which specimens were prepared for the interferometer dilatometer. The region DE illustrates the return of the resin to ambient conditions from the end of the 100°C curing period. Because of the difficulty of recording interference fringe movements as functions of temperature and time simultaneously, the fringes were counted over separate intervals of 15 min duration in the cooling period, following which the temperature–time relationship was established in a cooling experiment conducted on a separate occasion, when the ambient temperature was the same as before. Appropriate transformations and integrations then allowed the dependence of specimen dimensions upon temperature to be established. Fig. 3 provides a more significant representation of the results corresponding to the region DE. It clearly shows the temperature dependence of the volume of the cured resin as it cools after the 100°C curing stage, but it does not depend upon the rate of cooling and is thus independent of the ambient temperature.

Concluding, three observations may be recorded at this stage:

(1) the overall contraction of the product exceeds 6%;

(2) The relative slopes of CD and the end B of region AB in Fig. 2 suggest that a reduction of the period at 20°C would probably have little effect upon the quality of the cured resin. (Additional evidence favouring this conclusion emerged later. In particular, it will be seen that of the resins for which results are reported, Derakane 411-48/MEKP

was the only one yielding specimens sufficiently stable at the end of the first curing period to permit their investigation using the interferometer dilatometer.) On the other hand, an extension of the 100°C region should produce a more completely cured resin.

(3) Stresses introduced during the curing of a resin influence the subsequent behaviour of the corresponding CFRP structure employing the resin as matrix. The data upon which the region DE is based may be employed in the assessment of residual stresses provided that a temperature can be recognized below which volumetric changes due to chemical causes no longer occur. Because isothermal volumetric changes are apparently not completed in region CD (see Fig. 2), it seems likely that the temperature below which further resin cure is unlikely to occur is to be found somewhere near to D in the region DE (Fig. 3).

4.2. Epikote 828/MNA/BDMA epoxy resin

52.2 g Epikote 828 resin was heated to 70°C in order to reduce its viscosity prior to the addition of 47.0 g methyl “nadic” anhydride and 0.8 g benzyl dimethyl amine. After mixing the constituents thoroughly the resulting liquid was placed in a manometer dilatometer by applying the pre-treatment described in Section 3, except that in this case the treatment was given at 70°C. The dilatometer was supported in the viscometer bath as before.

After raising the temperature to 125°C, volume changes were monitored over the following 1½ h, during which contraction occurred and the resin solidified. A number of attempts to monitor sub-

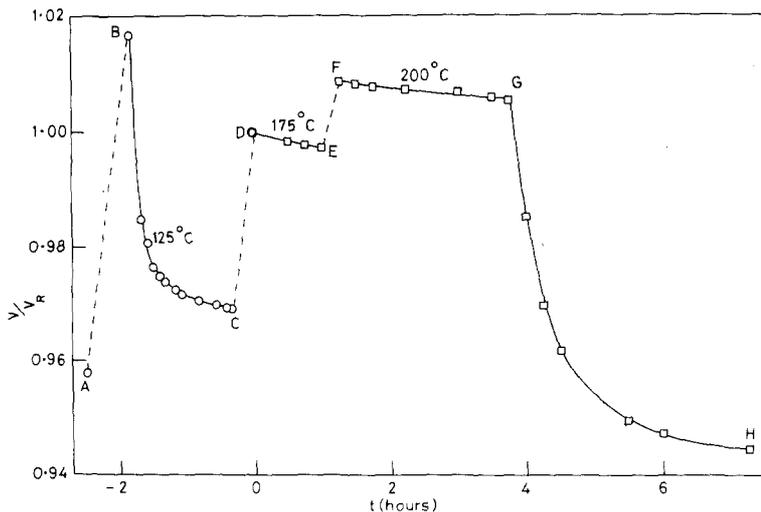


Figure 4 The time dependence of the reduced volume V/V_R of Epikote 828/MNA/BDMA resin during the curing process. The reference temperature, R , and the arbitrary zero of time have been taken as the point D, at which dilatometers were changed. \circ , manometer dilatometer results; \square , dial gauge dilatometer results; --- interpolated.

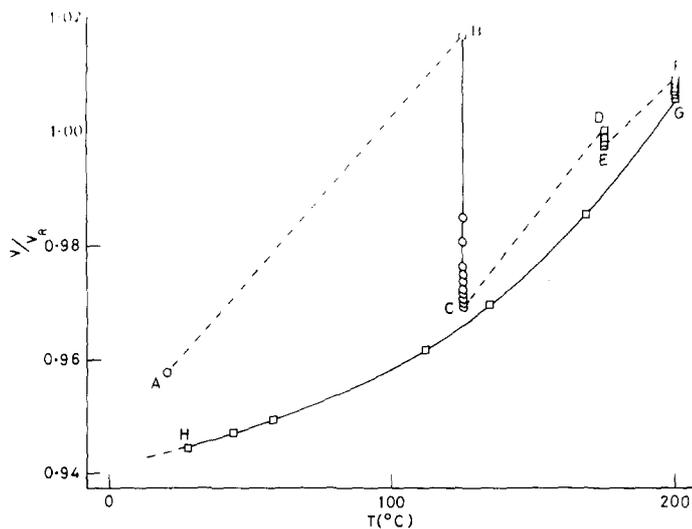


Figure 5 The temperature dependence of the reduced volume V/V_R of Epikote 828/MNA/BDMA resin during the curing process, in which the symbols A to H denote the same stages as in Fig. 4. The reference temperature, R , and the arbitrary zero of time are also as in Fig. 4. \circ , manometer dilatometer results; \square , dial gauge dilatometer results; --- interpolated.

sequent dimensional changes in the interferometer dilatometer over the following hour at 175°C, employing solid specimens prepared from resin cured at 125°C for 1½ h, were unsuccessful. This lack of success arose from the instability of the interference pattern, which was later attributed to softness of the specimens consequent upon an insufficiently advanced state of cure. Recourse was made to the dial-gauge dilatometer for the measurements required in the later stages of the curing process. The final results are displayed in Figs. 4 and 5, common letters in the two figures again identifying corresponding stages of the curing programme.

Because of the composite nature of Figs. 4 and 5, it was convenient to take the arbitrary zero of time at the point D, i.e. at the beginning of the 175°C curing stage. This meant that the reference

temperature, R , employed in expressing the volume, V , of resin in the reduced form V/V_R was 175°C in this instance. It may be mentioned that the selection, for reasons of convenience, of different values of R for different resins, produces no significant effect on V/V_R , which varies by only a few per cent during the whole curing process. The gel time, measured as 28 min at 125°C, bears no obvious relationship to the shape of the isothermal BC in Fig. 4.

Summarizing the main observations on these results:

- (1) the overall contraction of the resin during cure is less than 2%;
- (2) no suggestion concerning a significant reduction of curing period would be appropriate in the present case;
- (3) because the end G, of the region FG in Fig. 4

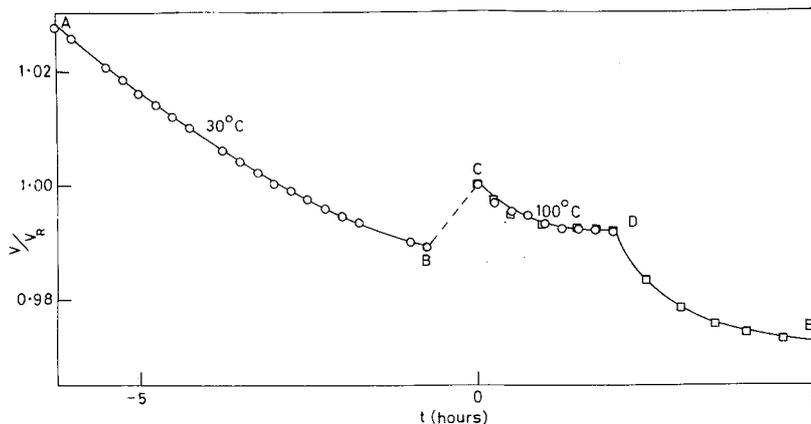


Figure 6 The time dependence of the reduced volume V/V_R of XB2878A/XB2878B resin during the curing process. The reference temperature R and the arbitrary zero of time have been taken at the point C , at which dilatometers were changed. \circ , manometer dilatometer results; \square , dial gauge dilatometer results; — — interpolated.

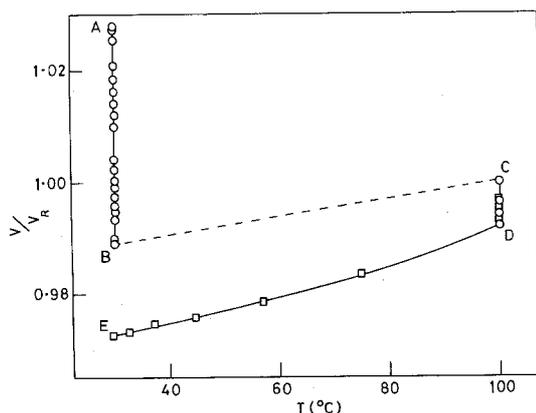


Figure 7 The temperature dependence of the reduced volume V/V_R of XB2878A/XB2878B resin during the curing process, in which the symbols A to E denote the same stages as in Fig. 6. The reference temperature, R , and the arbitrary zero of time are also as in Fig. 6. \circ , manometer dilatometer results; \square , dial gauge dilatometer results; — — interpolated.

is not parallel to the time axis, the temperature below which further resin cure is unlikely to occur in the present case would seem to be in the region GH.

4.3. XB2878A/XB2878B epoxy resin

36 g XB2878B hardener were added to 100 g XB2878A resin. The mixture was stirred thoroughly, pre-treated as in Section 3 and placed in a manometer dilatometer which was supported in the viscometer bath. In contrast to the work with the previous two resins, no parting agent was employed.

Volume changes were monitored in the manometer dilatometer during a 6 h curing period at 30°C , followed by a 2 h curing period at 100°C . In addition, a solid cylindrical specimen was prepared from resin which had been pre-cured at 30°C for 6 h. This was mounted in the dial-gauge dilatometer and the temperature was raised to 100°C . Length changes were monitored over the following 2 h and during the subsequent cool-down period. The results are displayed graphically in Figs. 6 and 7. It may be seen from Fig. 6 that the results taken with the two dilatometers agreed within the limits of experimental uncertainty in the 100°C region, from which it was concluded that troubles associated with the apparent expansions of resins Derakane 411-48/MEKP and Epikote 828/MNA/BDMA during advanced stages of the curing processes were probably associated with the parting agent allowing the solidifying resin to separate from the inner surface of the dilatometer, so producing voids in the system into which dilatometer fluid could not penetrate. It may be seen from Figs. 6 and 7 that 100°C was chosen for the reference temperature R . The gel time for this resin, measured in a separate experiment, was 52 min at 30°C ; Fig. 6 shows that, as in the cases of the previous two resins, this bears no obvious relationship to the form of the dependence of V/V_R upon time in the first stage of the curing process. Other features of the variation of volume with time and temperature are similar to those observed previously.

Summarizing the main features of these results; (1) the overall contraction of the resin during

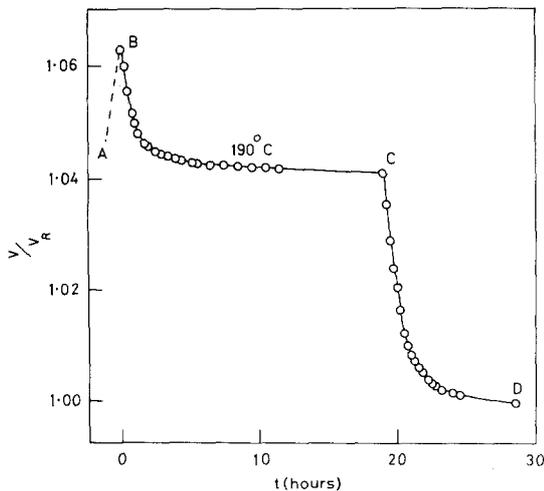


Figure 8 The time dependence of the reduced volume V/V_R of DLS 351/BF₃ 400 resin during the final stages of the curing process, in which the reference temperature, R , has been taken as 20° C. \circ , dial gauge dilatometer results; — — — interpolated.

cure approaches 6% over the temperature range covered in this investigation;

(2) the fact that region CD of Fig. 6 is almost independent of time towards the end suggests that the resin is in an almost completely cured state in the neighbourhood of D. This, in turn, suggests that the temperature below which further resin cure is unlikely to occur is probably located along DE, fairly close to D (Fig. 7).

4.4. DLS 351/BF₃ 400 epoxy resin

After heating to 100° C, 80 g DLS 351 resin and 1.2 g BF₃ 400 hardener were stirred together until they were thoroughly mixed. The high viscosity of the resulting liquid prevented its satisfactory entry

into a manometer dilatometer, and volumetric observations had to be restricted to those stages of the curing process over which the specimen was in a sufficiently solid form for examination using the dial-gauge dilatometer. For this reason the programme given below was followed.

The liquid resulting from the above mix was outgassed in a vacuum desiccator for about 30 min before being poured into a shallow bar mould, which was also held at a temperature of 100° C. The mould with its contents was placed in an oven for 1½ h at 100° C. The temperature was then raised to 150° C for a further 2 h, following which the semi-cured rod was extracted and quenched in an ice bath. The following day it was mounted in the dial-gauge dilatometer, which was supported in the viscometer bath as before. Length changes were monitored over the following 19 h, during which the temperature was held at 190° C, and over the final cooling period.

The results are displayed graphically in Figs. 8 and 9; the reference volume V_R has been taken as the final value at room temperature. The decrease in volume with time during the first part of the 190° C curing stage is considerably more marked than the corresponding parts of the final stages of the curing processes of the resins examined earlier. However, the time dependence of the volume along BC in the neighbourhood of C is only small, suggesting that the cure is well advanced at this stage. Although the results for this resin are less complete than those for the other resins, certain conclusions may be drawn as before:

- (1) the contraction over the restricted part of the process studied exceeds 4%;
- (2) the shape of region BC in Fig. 8 suggests

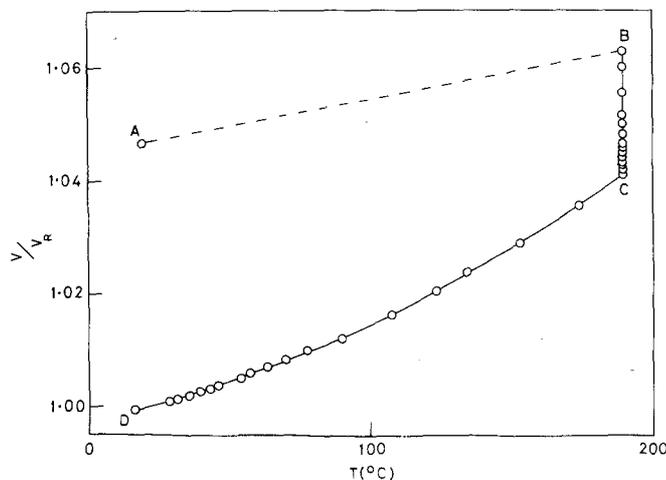


Figure 9 The temperature dependence of the reduced volume V/V_R of DLS 351/BF₃ 400 resin during the final stages of the curing process, in which the symbols A to D denote the same stages as in Fig. 8, and the reference temperature, R , has been taken as 20° C. \circ , dial gauge dilatometer results; — — — interpolated.

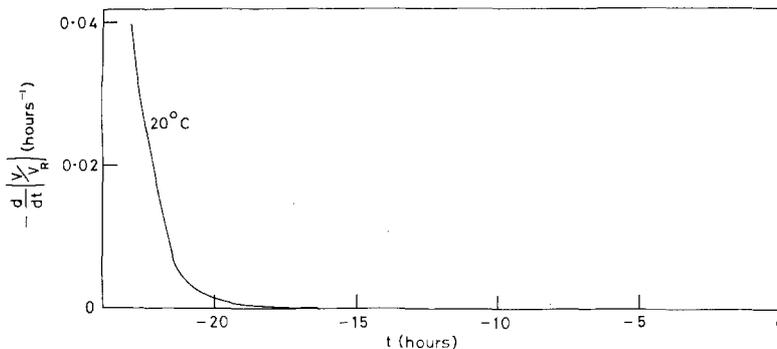


Figure 10 The time dependence of the shrinkage rate of Derakane 411-48/MEKP resin in the first stages of the curing process, in which the time scale is the same as that employed in Fig. 2.

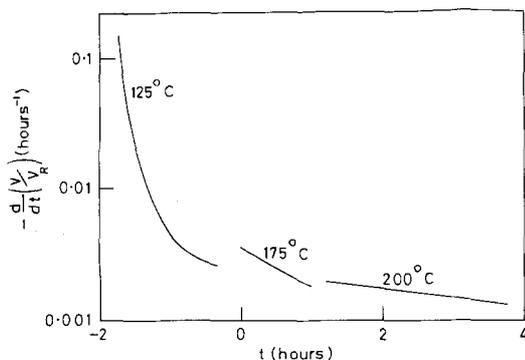


Figure 11 The time dependence of the shrinkage rate of Epikote 828/MNA/BDMA resin during the principal stages of the curing process, in which the time scale is the same as that employed in Fig. 4.

that the degree of cross-linking in a specimen cured for a few hours less than that studied here would probably not be very different from that of the present specimen;

(3) for similar reasons to those given earlier, the temperature below which further resin cure is unlikely to occur is probably somewhere near to C in the region CD (Fig. 9).

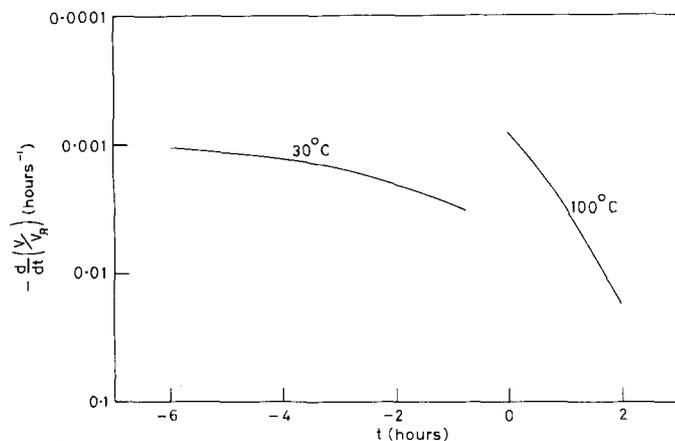


Figure 12 The time dependence of the shrinkage rate of XB2878A/XB2878B resin during the principal stages of the curing process, in which the time scale is the same as that employed in Fig. 6.

5. Discussion

Having established the principal features of the results, it is of interest to examine them quantitatively in connection with the progress of cure.

5.1. General features of the curing process

The rate of change of volume with time is clearly related to the rate of polymerization and cross-linking occurring during cure. The time dependence of this rate during the 20°C pre-cure of Derakane 411-48/MEKP is illustrated in Fig. 10, in which the same time scale has been employed as before. In the cases of Epikote 828/MNA/BDMA and XB2878A/XB2878B, illustrated in Figs. 11 and 12 respectively, logarithmic representations have been adopted in order to accentuate the changes of slope which occur from one stage to another. From these figures it will be seen that the effect of increasing the temperature between stages is to increase the rate of the volumetric reduction. This, in turn, is consistent with increasing the rate of polymerization and cross-linking. Fig. 13 illustrates the corresponding situation in the final stage of the curing of resin DLS 351/BF₃400. The extent

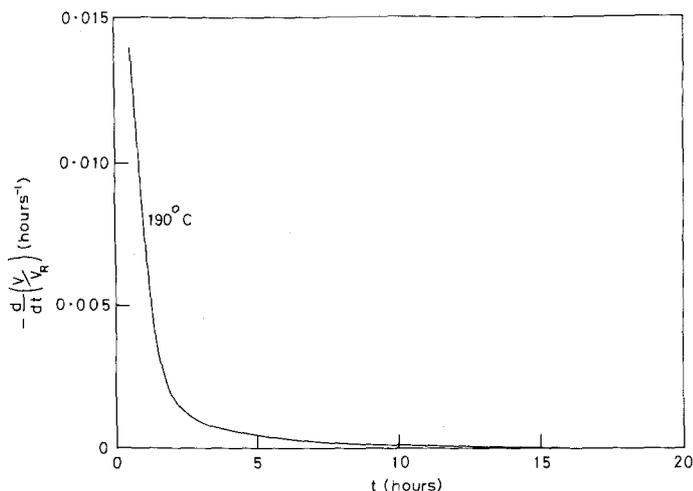


Figure 13 The time dependence of the shrinkage rate of DLS 351/BF₃ 400 resin in the final stage of the curing process, in which the time scale is the same as that employed in Fig. 8.

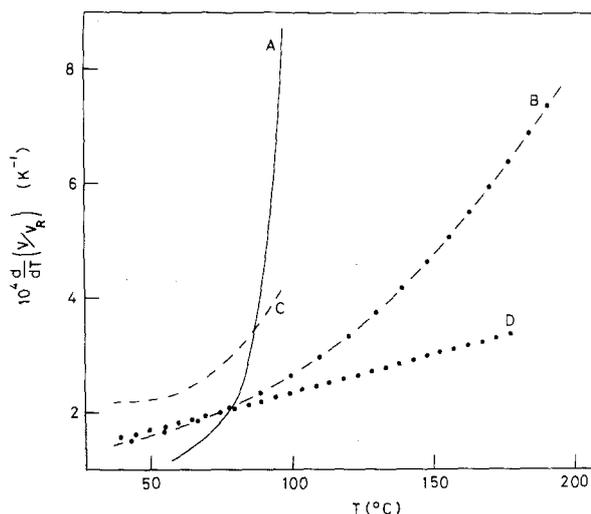


Figure 14 The temperature dependences of the reduced volumes of the four resins, displayed in smoothed form as functions of temperature during the cooling period which followed the last stages in the curing processes: — A, Derakane 411-48/MEKP resin; ·-·-· B, Epikote 828/MNA/BDMA resin; - - - C, XB2878A/XB2878B resin; ····· D, DLS 351/BF₃ 400 resin.

of the change of slope is somewhat surprising, but comments on this aspect of the variation would be more meaningful in the context of a comparative survey of a wider range of resins.

5.2. Early stages of the curing process

A striking feature of the first stage in the curing process of each of the first two resins is the shape of the graph of reduced volume versus time. Examination has revealed that this dependence is exponential over the greater part of the process. Representations of the variations illustrated in Figs. 2 and 4 are provided by the following empirical equations, in which t is the time in hours measured on the arbitrary time scales employed in the figures:

For Derakane 411-48/MEKP resin:

$$[2000 (V/V_R) - 1995] [12t + 280]^{0.922} = 397.2$$

For Epikote 828/MNA/BDMA resin:

$$[2000 (V/V_R) - 1925] [24t + 48]^{0.766} = 202.9.$$

It may be seen from Figs. 2, 4 and 6 that the percentage diminutions of V/V_R in the first stage of the curing process of each of the first three resins examined are of the same order. However, it is noticeable that the time over which this shrinkage was spread was considerably greater for XB2878A/XB2878B than it was for the other two resins. This seems to correlate with the gel times, that for XB2878A/XB2878B (52 min at 30°C) being considerably greater than that for Derakane 411-48/MEKP (23 min at 20°C).

5.3. Advanced stages of the curing process

The temperature dependences of the reduced volumes of the resins are compared with one another in Fig. 14. Particular attention was paid to

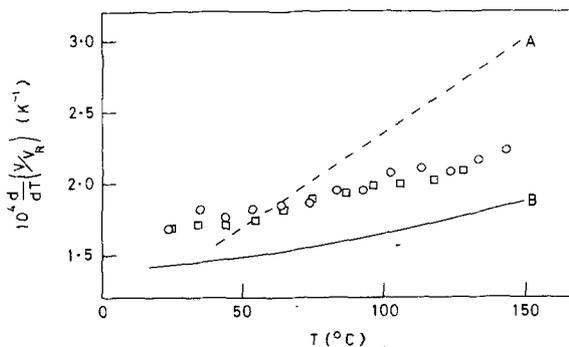


Figure 15 The temperature dependence of the reduced volume of resin DLS 351/BF₃400 determined in different ways: — — A, dial gauge dilatometer determination from the cooling curve of the present specimen (smoothed data); — B, interferometrically determined thermal expansion coefficient reported for an earlier specimen ([3], smoothed data); ○, dial gauge dilatometer determination during subsequent heating of the present rod specimen (primary data); □, interferometer dilatometer determination during the subsequent heating of specimens prepared from the present rod (primary data). Observations corresponding to — — B, ○ and □ were all made at steady temperatures.

the results for DLS 351/BF₃400 for which linear thermal expansion results were also available [3]. These results are displayed in Fig. 15, from which it may be seen that values of the apparent linear thermal expansion coefficient, derived from section CD of Fig. 9, lie above results for the linear thermal expansion coefficient of specimens prepared from an earlier bar, measured interferometrically [3]. This contrast led to further examinations of the temperature dependence of the dimensions of specimens prepared from the rod of resin which was cast during the present work. In the first set of additional measurements the rod was accommodated in the dial-gauge dilatometer and the linear thermal expansion coefficient was calculated from observations made at steady values of the temperature, as this was raised. These results are presented in primary form in Fig. 15, together with an additional set of results obtained for specimens prepared from the same rod and investigated with the interferometer dilatometer. The last two sets of results agree within the limits of experimental uncertainty. Although part of the difference between these results and those depicted by curve A may arise from different degrees of completion of cure on the different occasions, the greater part of the difference presumably arises from unavoidable temperature gradients and resin relaxation effects in the dynamic temperature process, which were absent in the measurements conducted under equilibrium conditions. An implication of the difference is that the slopes of cooling curves, such as those depicted in Fig. 14, cannot be used to determine thermal expansion coefficients with any accuracy. A comparison of the present equilibrium measurements with curve B reveals that although the thermal expansion coefficients of the two bars differ by approximately 16% in absolute terms, their temperature dependences are essentially similar. The explanation of

the absolute difference may lie in differences of detail in the curing programmes employed for the two bars. The programme for the present bar was given earlier in this section. In the case of the bar for which results were reported by Yates *et al.* [3], the programme consisted of a stage at 100°C for 2 h followed by 2 h at 150°C, followed by 15 h at 190°C, no break being experienced between stages. The present bar may, therefore, be marginally more completely cured and this would be consistent in sense with the difference between the two sets of results in question. It should also be recorded that whereas the measurements upon the present specimen were undertaken soon after its production, there was a break of approximately 4 months between the production and investigation of the earlier bar. It is, therefore, possible that the earlier bar had a higher moisture content than the later bar, since it had been exposed to prevailing ambient conditions between production and investigation. In a series of investigations which included an examination of the influence of moisture content upon the dimensional characteristics of resins, Freeman and Campbell [8] presented results which were consistent with the thermal expansion coefficients of Hercules 3002 and TRW P13N resins increasing as the moisture content was reduced. The sense of the difference between the two sets of results currently under consideration is entirely consistent with these earlier findings, if moisture is the cause.

6. Conclusions

As part of a pilot study directed towards an evaluation of the importance of resin shrinkage which occurs during the processing of carbon fibre-reinforced plastics, variations of volume with time during different stages of the curing cycles of four resins have been monitored. In addition to producing results which should be of direct

interest in connection with component moulding technology, it has been suggested that this approach might be applied directly to assess the progress of the various stages of resin cure. In particular, the results indicate which stages of accepted cure cycles could with advantage be shortened and which could be lengthened, but no direct relationship has been found between gel time and resin shrinkage. In the three cases in which measurements were possible over the entire curing cycle it has been established that the major shrinkage occurs during the early stages of solidification, although the shrinkage characteristics vary in detail from one resin to another. This suggests, for example, that transverse cracking in multi-layer cross-ply laminates during the cool-down period after moulding is more likely to be caused by thermal expansion and contraction effects than by the small amount of resin shrinkage occurring during the final stage of cure. It has also been established that in connection with residual stress evaluation there is a need for more detailed examinations of the influence of thermal and environmental history on the thermal expansion behaviour of resins.

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