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The thermal conductivity of Kevlar fibre-reinforced composites

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The thermal conductivities of a series of blocks consisting of Shell DX210/BF$_3$400 resin reinforced with Kevlar 49 fibre are reported in the approximate temperature range 180–270 K. The results are used to calculate the thermal conductivities of the fibres in directions parallel and perpendicular to their length. Varying the angle between the principal fibre directions of bidirectional laminates produced in-plane results that varied in a manner which was quantitatively consistent with expectation. The out-of-plane results proved to be independent of fibre orientation, as expected. In-plane and out-of-plane results for a Kevlar 49 fabric reinforced laminate proved to be essentially similar to results for a laminate reinforced with unwoven fibres of the same type, arranged in a 90° cross-plied disposition at the same fibre volume density.

1. Introduction

The principal advantages and disadvantages of glass and carbon for the fibrous reinforcement of epoxy resins are well known. Less common but very important are high modulus polyamide fibres produced by E. I. Du Pont de Nemours and Co Inc, marketed under the trade name Kevlar. Briefly, Kevlar fibre has a tensile strength comparable with that of carbon fibre, a modulus between those of glass and carbon fibres and a lower density than both. Its weakness lies in its compressive strength, but for applications where this is not important and in cases where the fibre may be used in association with carbon within a hybrid assembly, Kevlar has established a firm place among the fibres which are now in regular use within modern composite technology.

Kevlar is believed to be essentially poly (*p*-phenylene tetraphthalamide), having a crystal structure which conforms with the monoclinic (pseudo-orthorhombic) unit cell [1, 2]. One form of the material is suitable for rubber reinforcement, Kevlar 29 is employed in specialized industrial applications and in armour jackets and Kevlar 49, a very high modulus form, is used in plastics reinforcement. This is the form of Kevlar employed exclusively in the present investigation. Detailed characteristics of the fine structures of the fibres have been identified [3, 4] and aspects of structure have been associated with observed mechanical properties.

The fuel savings to be gained in rail and road transport by replacing metal components with lighter weight fibre-reinforced plastics having a high stiffness and strength are very attractive. A knowledge of the thermal conductivity of these materials enhances their commercial value in such applications and the present investigation has been undertaken against this background, both to provide technologically useful data and to gain a measure of understanding of the performance of Kevlar fibre-reinforced plastics in analytical terms.

2. The specimens

2.1. Constituent materials

The resin system adopted for the present study was one employed in an earlier investigation [5] of the thermal conductivity of carbon fibre-reinforced

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composites. This was Shell DX210, utilized in association with hardener BF_3400. 100 m lengths of Kevlar 49 fibre were supplied by Fothergill and Harvey Ltd in the form of tows, each of which had a diameter of approximately 0.3 mm and contained approximately 10^4 fibres. The suppliers also produced a quantity of cloth which had been manufactured from tows of the same fibre in a satin weave, having nominally identical crimp in the warp and weft directions.

2.2. Reinforced composite blocks

In order to safeguard against subsequent degradation, all the fibre employed in the investigation was held at a temperature of 101° C for 16 h in order to remove moisture, before being pre-impregnated with resin within an hour of removal from the oven. The actual process of pre-impregnation of the unwoven fibre was achieved with the aid of a large rotating drum, having an approximately elliptical cross-section, which had release paper attached to its surface. The drum was fed with tow from a bobbin, the speed of traverse of which was geared to the rate of revolution of the drum so that this was uniformly coated with tow. This layer of fibre was then painted with resin, left to dry overnight and then cut with a knife along two lines running parallel to the axis of the drum, thus producing two sheets of pre-impregnated fibre (prepreg), which were coated with tissue paper to facilitate handling. Pre-impregnation of sheets of woven fabric was achieved by laying sheets of the cloth on release paper and painting these with resin. After production, the prepreg was stored in an airtight refrigerator until required.

Steel moulds having cross-sections 110 mm × 55 mm and 110 mm × 110 mm were produced for the unwoven and woven prepreg, respectively. Sheets of unwoven prepreg cut slightly undersize were held at 80° C for approximately 7 min in a controlled flow of hot air to remove excess solvent from the resin before being placed in the mould. Knowing the diameter of the fibres, the number of sheets of prepreg required within the predetermined thickness of the block (55 mm) to achieve a fibre volume fraction of 60% was calculated. The appropriate number of sheets was laid up in the mould, rolling between the addition of successive layers to remove air and applying pressure at intervals to compact the assembly. The block of composite formed in this way was removed from the mould, which was then pre-heated to 170° C in a press, after being sprayed with silicone release agent. The temperature of the composite block was meanwhile raised to 130° C over a period of approximately an hour in an air controlled oven, where it remained for a further 15 min before being transferred to the mould. The main purpose of this preliminary heat-treatment was to minimize temperature gradients with a view to producing uniformly cured blocks. The mould was then mounted in the heated press, where the block was cured under closing pressure at a temperature of 170° C over the following 80 min. The block was then removed from the mould and post-cured for a further 3 h at 170° C.

All the blocks containing unwoven fibre were produced in the above manner. That containing fabric, in which the warp tows were aligned parallel to one another and the weft tows were parallel to one another, was produced in a similar manner, except that this somewhat larger block was allowed $1\frac{1}{2}$ h in the oven at 130° C before being transferred to the mould.

2.3. Experimental details

The magnitude of thermal conductivities encountered led to the use of the disc apparatus described earlier [5]. The apparatus required specimens in the form of circular discs and this requirement dictated the dimensions of the composite blocks. Two cylinders were machined from each of the three blocks containing unwoven fibre. In the case of the block containing unidirectional fibres the principal axis of each individual cylinder was arranged to be parallel or perpendicular to the fibres. In the case of the blocks containing fibres arranged with inter-ply angles of 45° and 90°, the axis of one cylinder bisected the inter-ply angle while the axis of the other cylinder was directed at right angles to the planes of the constituent laminae. In the case of the block containing woven fibre three cylinders were required, the axes of which were directed parallel to the warp, parallel to the weft and at right angles to the plane of the fabric. It was explained earlier [5] how the use of three discs of each material, each of which had a different thickness, allowed end effects to be eliminated. Nominal thicknesses of 2.5, 5.0 and 6.5 mm proved to be convenient in the present investigation and discs having these thicknesses and a common diameter of 35 mm were cut from the cylinders with the aid of a Macrotome 2 rotary pre-

TABLE I The specimens

Block number	Specimen designations	Angle between fibre directions (deg)	Direction of thermal conductivity measurements	Orientation of direction of measurement with respect to plane of fibres	Fibre volume (%)
1	1	0	Parallel to fibres	In-plane	64.4
1	2	0	Perpendicular to fibres	Out-of-plane	64.4
2	3	45	Bisecting the 45° angle	In-plane	64.9
2	4	45	Perpendicular to fibres	Out-of-plane	64.9
3	5	90	Bisecting the 90° angle	In-plane	57.8
3	6	90	Perpendicular to fibres	Out-of-plane	57.8
4	7	90	Parallel to warp	In-plane	67.6
4	8	90	Parallel to weft	In-plane	67.6
4	9	90	Perpendicular to fibres	Out-of-plane	67.6

cision saw. A combination of a low cutting rate (between 6 and 24 h per slice) and the use of cooling fluid gave a good surface finish to the discs and prevented any significant temperature rise. The discs so produced were ultrasonically c-scanned for voids. Because the scanner available had been calibrated for carbon fibre-reinforced plastics only, accurate figures for the void distribution could not be derived. However, no serious voidy regions were detected in the discs, the void content of which was believed to be generally below 1%. The fibre volume fractions of the blocks were taken as the averages of values for two volumes of waste material taken from each block and analysed by acid digestion. Descriptions of the blocks and specimens prepared from them are collected in Table I.

3. Results

3.1. In-plane thermal conductivities

Values read at rounded temperatures from smooth lines drawn by eye through the primary experimental data for directions in the plane of the fibres are collected in Table II for convenience of application. A detailed consideration revealed experimental uncertainties in the primary data amounting to approximately ±3%. From Table I and Fig. 1, in which the results are compared graphically, one may draw the following qualitative conclusions.

(i) Comparing curves A and B in Fig. 1 and noting the magnitude of the results displayed in Fig. 2, it is clear that the thermal conductivity of the fibre is greater in the longitudinal direction than in the transverse direction. The lower fibre volume fraction of specimen 5 compared with specimens 1 and 3 makes an interpretation of the comparison of results for this specimen with the others more difficult, though such deduction as is possible is clearly not in conflict with the above conclusion.

(ii) Since the in-plane thermal conductivity of a balanced 90° cross-ply of unwoven fibres is independent of direction, any difference between the results depicted in curves C and D must result from the different fibre volume fractions of blocks 3 and 4 and from the fact that one set of fibres is woven while the other set is unwoven. The comparatively low fibre volume fraction of specimen 5 will reduce the conductivity below the value for a hypothetical 90° cross-ply with a fibre volume

TABLE II Smoothed values of the thermal conductivity, K, of the pure resin [5] and of composite specimens corresponding to in-plane directions of blocks 1 to 4, described in Table I

T (K)	K ($W m^{-1} K^{-1}$) for the specimens listed below					
	Pure resin	1	3	5	7	8
180	0.196	1.11	1.00	0.750	0.722	0.729
190	0.198	1.13	1.02	0.758	0.732	0.738
200	0.201	1.15	1.04	0.769	0.742	0.749
210	0.204	1.16	1.05	0.779	0.751	0.758
220	0.207	1.18	1.06	0.788	0.760	0.768
230	0.210	1.20	1.07	0.797	0.768	0.776
240	0.214	1.21	1.08	0.806	0.776	0.781
250	0.218	1.22	1.09	0.815	0.785	0.788
260	0.222	1.24	1.10	0.823	0.792	0.793
270	0.226	1.25	1.11	0.828	0.799	0.800

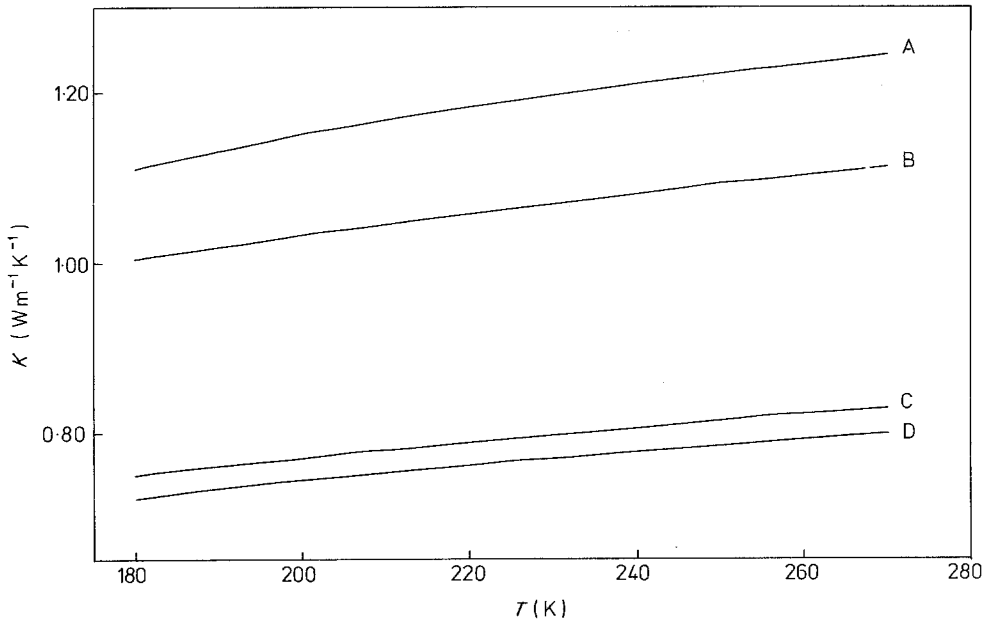

Figure 1 Graph comparing the thermal conductivities, K, of specimens 1 (curve A), 3 (curve B), 5 (curve C), and 7/8 (curve D), from which it may be deduced that the longitudinal thermal conductivity of Kevlar 49 fibre exceeds its transverse thermal conductivity.

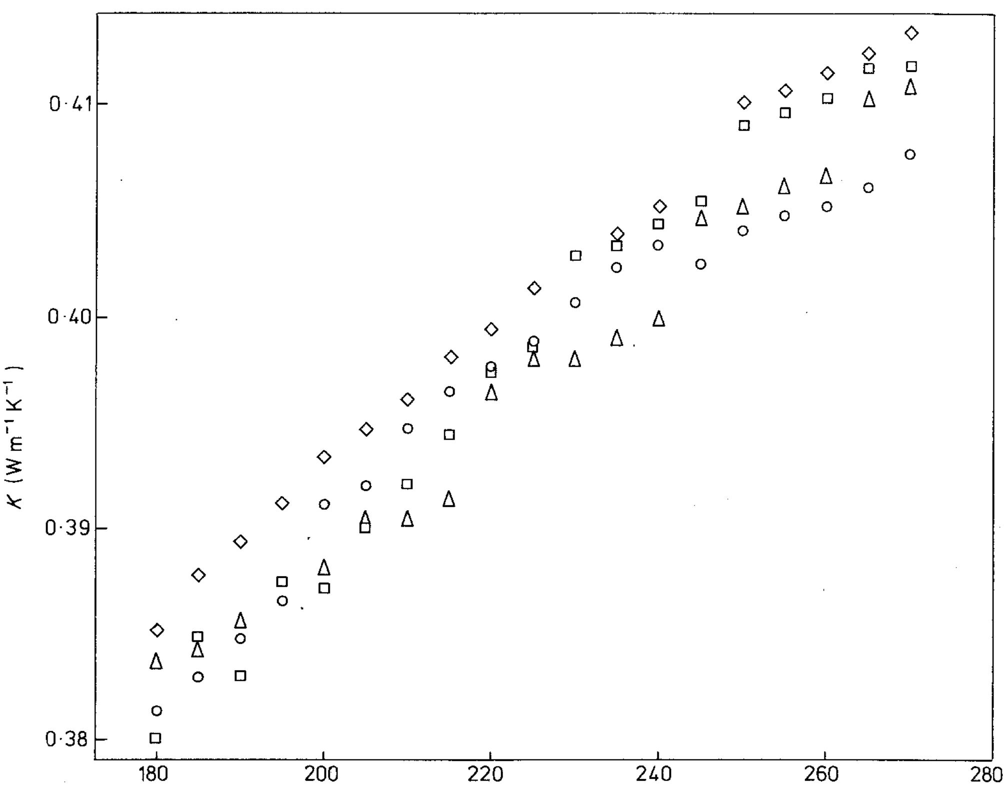

Figure 2 Comparison of the out-of-plane thermal conductivities, K, of specimens 2 (○), 4 (□), 6 (△), and 9 (◇).

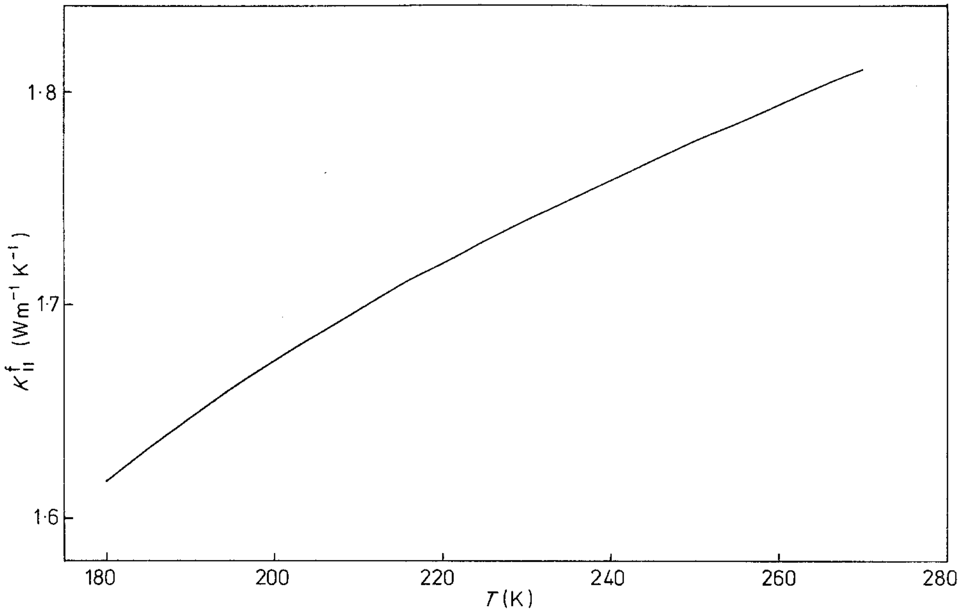

Figure 3 The longitudinal thermal conductivity $K^{f}_{\parallel}$ of Kevlar 49 fibre.

fraction similar to specimens 1 and 3. One may note further that accepting the above deduction that the transverse fibre conductivity is lower than its longitudinal conductivity, the effect of introducing crimp into the fibres of a 90° cross-ply will be to reduce the in-plane thermal conductivity to a value below that of a hypothetical 90° cross-ply having the same volume fraction of unwoven fibre. From the fact that curve D lies below curve C one can thus deduce that the reduction of the in-plane conductivity caused by crimping the fibres exceeds that resulting from a 10% reduction in fibre volume content.

3.2. Out-of-plane thermal conductivities

The out-of-plane conductivities are compared in Fig. 2. Setting aside the results for specimen 9, the only major differences between the constructions of the remaining specimens for which results are presented concern the angle between the principal fibre directions and the somewhat low fibre volume content of specimen 6 compared with specimens 2 and 4. This last variable is likely to be less significant in the out-of-plane case that it was in the in-plane case because of the closer similarity between the transverse thermal conductivity of the fibres and the thermal conductivity of the resin than exists between the longitudinal fibre thermal conductivity and the thermal conductivity of the resin. It is clear that increasing the interply angle from 0° to 90° produces no change in the transverse thermal conductivity of the composites. This independence of the transverse thermal conductivity of bidirectional composites upon interply angle contrasts with a dependence of the transverse thermal expansion of bidirectional composites upon interply angle, which has been verified elsewhere [6]. Both results conform with theoretical expectations.

The fact that the results for specimen 9 stand higher than the others is consistent with the influence of the longitudinal conductivity of the fibres in the woven case.

4. Analysis of data

4.1. The longitudinal thermal conductivity of Kevlar 49 fibre

The longitudinal thermal conductivity of the fibre, $K^{f}_{\parallel}$, was calculated by applying the equation

$$K^{c}_{11} = K^{f}_{\parallel}v_{f} + K^{r}(1 - v_{f}) \qquad (1)$$

to the results K^{c}_{11} for specimen 1. In this equation K^{r} is the thermal conductivity of the resin and v_{f} is the fibre volume fraction. The result of this calculation is displayed in Fig. 3.

4.2. The transverse thermal conductivity of Kevlar 49 fibre

From an appraisal [5] of models upon which assessments of the transverse thermal conductivity K^{c}_{22} of unidirectionally reinforced composites might be based, it was concluded that analogies in elasticity

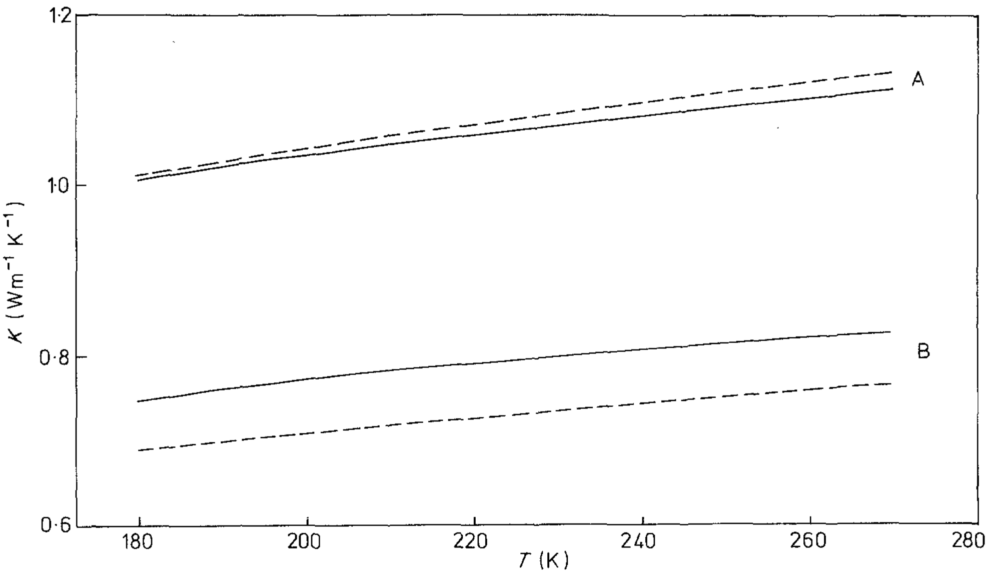

Figure 4 The thermal conductivities, K, of specimens 3 and 5. A: specimen 3 ——— measured, - - - calculated; B: specimen 5 ——— measured, - - - calculated.

provided a reasonable degree of agreement with experiment. The equation corresponding to this analogy, i.e.

$$K_{\perp}^{f} = \frac{K^{r}(1-v_{f}) - K_{22}^{c}(1+v_{f})}{(K_{22}^{c}/K^{r})(1-v_{f}) - (1+v_{f})}, \quad (2)$$

has been applied to the present results. The influence of the relative rates at which the thermal conductivities in the transverse direction of specimen 2 and in the pure resin increase with temperature, upon the calculated thermal conductivity of the fibres in a direction perpendicular to their length, is particularly critical. For the present purpose it was decided to take an average value from the results calculated for the transverse thermal conductivity of the fibres and to take no account of the influence of temperature. The value so calculated was $0.6\,W\,m^{-1}\,K^{-1}$. It is recognized that other models would yield alternative results and for this reason a detailed assessment of the uncertainty in this figure has not been undertaken.

4.3. Quantitative self-consistency of the in-plane thermal conductivities

The form taken by the basic transformation equation in the case of a symmetrically balanced laminate is

$$K^{c} = K_{11}^{c}\cos^{2}\theta + K_{22}^{c}\sin^{2}\theta. \quad (3)$$

In this equation K_{11}^{c} and K_{22}^{c} represent the conductivities parallel and perpendicular to the fibres of a unidirectionally reinforced lamina having the same fibre volume fraction as the bidirectionally reinforced laminate, in which K^{c} is the conductivity in a direction bisecting the angle 2θ between the principal fibre directions. Employing the results for $K_{\parallel}^{f}$ and $K_{\perp}^{f}$ derived above in association with values of K^{r} quoted in Table II, values of K_{11}^{c} and K_{22}^{c} were calculated for fibre volume fractions of 64.9% and 57.8%, corresponding to blocks 2 and 3, with the aid of Equations 1 and 2. The results corresponding to specimen 3, i.e. $v_{f} = 64.9\%$ and $\theta = 22.5°$, were then substituted into Equation 3 to calculate the result which is compared with the experimental result for this specimen as case A in Fig. 4. Case B in Fig. 4 covers the corresponding comparison of measured and calculated results for specimen 5. Bearing in mind: (i) limitations in the model employed to calculate $K_{\perp}^{f}$; (ii) the neglect of any temperature dependence of this quantity; (iii) the increasing importance of $K_{\perp}^{f}$ as θ increases; (iv) structural imperfections in the specimens, e.g. the presence of voids, the non-uniform distribution of fibres and associated uncertainties concerning the geometrical generality of the values of v_{f} employed, the agreement between measured and calculated results, i.e. to within a few per cent, must be regarded as being as satisfactory as can reasonably be expected.

4.4. Edge effects

It was explained earlier that in order to eliminate end effects from the results, sets of three specimens were employed throughout the investigation and the term "specimen" used in the above account actually refers to the average of results for such sets of three specimens. In cases where the fibres within the specimens ran parallel or perpendicular to the faces of the specimens, deviations from the assumption of linear heat flow in the vicinity of the edges of specimens were minimal. However, for other fibre orientations this was not the case and computer simulation techniques were applied to assess the magnitude of the problem of edge effects. In the case of the dimensions employed in this investigation it was concluded that percentage uncertainties associated with these effects were less than experimental uncertainties and no further account was taken of them.

5. Conclusions

In assessing the standing of theoretical models which predict the thermal conductivity of a composite in terms of properties of its constituents, it is frequently difficult to discriminate between the validity of assumptions made in the models and constructional imperfections within the specimens. Simultaneous unintentional variations in fibre content provide an additional complication and the problems are enhanced in fabric reinforced blocks, where the accurate alignment of picks and ends is sometimes difficult to maintain in commercially available materials. With these reservations in mind, the in-plane results of the present investigation fall into a clearly defined pattern. A detailed quantitative appraisal of the out-of-plane results, which are closely similar to one another, was prevented by uncertainties surrounding the validity of the theoretical model employed, constructional limitations and experimental uncertainties. In the long term, the analytical treatment of the transverse thermal conductivity of a fabric reinforced laminate would be of considerable interest if this contained quantitative allowance for crimp, particularly when more nearly perfectly constructed composites become available.

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