THERMAL ELLIPSOMETRY: A TOOL APPLIED FOR IN-DEPTH RESOLVED

CHARACTERIZATION OF FIBRE ORIENTATION IN COMPOSITES

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INTRODUCTION

Need for a NDE Technique for Fibre Orientation Evaluation in Composites

Composites with a thermoplastic resin reinforced with short fibres or platelets are commonly processed by injection molding. As far as the fibre orientation is concerned, a stratification generally appears during this operation: near the mould surfaces the fibres are oriented in the direction of the flow, whereas in the middle they are oriented perpendicularly to this direction. The flow parameters obviously determine the relative thickness of this core region and consequently the longitudinal and transverse properties of the composite material. The moulding process has therefore to be optimized so that these anisotropy variations contribute the composite to fulfil the mechanical performance specifications. The in-depth resolved characterization of the filler local orientation as well as its control are thus of prime importance.

When the fibres have substantially higher thermal conductivity than the matrix (i.e. carbon or metallic fibres in polymer matrix) a well ordered orientation can induce a strong thermal anisotropy. One can thus expect that, in that case, thermal anisotropy monitoring can provide valuable information on fibre orientation and possibly on internal orientation changes.

Principle of Thermal Ellipsometry

More than one century ago, De Senarmont applied a thermal approach to find out the principal orientations in crystal plates [1]: he covered them with a thin layer of wax, heated them over a small spot and monitored the isotherm shape revealed by the solid/liquid transition contour appearing in the wax layer. The isotherm proved to be elliptical and its aspect ratio is equal to the square root of the principal conductivities in the surface plane.

This method, to which we will refer as "Thermal Ellipsometry", was later used for various applications (with, of course, up-to-date experimental equipment, as for example thermography). It was applied on polymer materials to establish a correlation between their draw ratio and the induced thermal anisotropy [2-5]. It was also used to evaluate the fibre orientation in the case of composite materials containing short or long carbon fibres. For the latter problem, the thermal anisotropy measurements were performed either in a "reflection" configuration [5-7], or both in "reflection" and "transmission" configurations [8]. The former was thought to give an information about the fibre orientation prevailing in the surface layer, whereas the latter was thought to provide a through-depth

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averaged result. Several temporal regimes were considered: the stady-state or quasi-steady-state regime [2-4, 6-8]; the transient regime pertaining to step heating [9]; the modulated regime associated with radiometric detection [5], or with mirage detection [10].

Theoretical results showing how in-depth change of anisotropy main directions is reflected on the surface temperature field were first published in 1987 [11]. The analysis was made for the transient regime corresponding to step heating. Experimental results were obtained on bilayer laminates made of carbon/epoxy plies or short steel fibre in polypropylene layers. A more detailed theoretical analysis was later undertaken through an analytical treatment of thermal diffusion in laminates made of orthotropic layers assuming the surface is submitted to concentrated heating [12, 13]. Three temporal regimes were considered: steady-state regime, transient regime (as obtained during step heating), and modulated regime (in order to analyze how the so-called thermal waves "propagate" in orthotropic laminates). Experiments were performed on carbon-epoxy laminates for all three regimes. Let us mention that some results in thermal ellipsometry through lockin thermography were recently published by a team involved in the same research project [14, 15].

This paper will first present some results of an analysis about the joint influence of a series of parameters in the case of a three-layer structure. A thermal inversion method will then be described. Its purpose is to infer the thickness of the skin and core layers. Inversion results as obtained on experimental thermal images of carbon/epoxy laminates will finally be presented.

THEORY

Surface Temperature Field in the Case of a Three-Layer Structure

Fig. 1 (a) shows the geometry of the considered model. Taking into account the morphology which generally prevails in short fibre injection moulded composites one is allowed to make the following assumptions: the skin and core layers are homogeneous, orthotropic, and have the same thermal properties except that the in-plane main directions of the skin layers are at 90° relatively to those of the core layer. The stacking upper surface is heated by a gaussian laser beam (radius: ω_s) and the temperature field is assumed to be monitored by a infrared detector which integrates the IR intensity over a gaussian pattern (radius: ω_d - actually, these two parameters intervene through a sole parameter: the "resolution radius" $(\omega_s^2 + \omega_d^2)^{1/2}$). An analytic expression involving Fourier and Laplace transforms was found for the transient response in the case of step heating and in the presence of linear heat losses. Then, by setting proper values to the Laplace variable one easily obtained the thermal response prevailing to the modulated regime (sinusoïdally modulated input) and to the steady-state regime (constant heating).

Fig. 1 (b to d) presents some results for the latter case. The temperature field analysis was restricted to the surface principal directions. The anisotropy coefficient of the different layers was set to 6.4, a value which corresponds to the carbon/epoxy samples with continuous fibres that we tested. If the skin layer were semi-infinite and isolated, and if the resolution radius was 0, the signal would be as 1/r. Actually, due to the finite resolution radius, the signal is levelled off at the centre, and due to heat leaks the signal decrease accelerates far from the centre. In between, the signal distribution depends on the skin/core relative thickness (Fig. 1.b and 1.c). Near the centre, the signal is higher along the direction along which the skin has highest conductivity. The isotherms are thus elongated in this direction. Further from the centre, the signal can become higher in the other direction if the core relative thickness is higher than 0.5 (Fig. 1.c). In that case, the wide isotherms are rotated at 90° relatively to the central ones. Furthermore, there is an intermediate isotherm with the same dimensions along the two principal directions. In the particular case of a 2/3 core thickness, this pseudo-circular isotherm has a radius corresponding to roughly two times the stacking thickness . For each iso-signal curve, one can define an aspect ratio (> 1 if the curve is oriented in the skin highest conductivity direction; <1 otherwise). This parameter is plotted on Fig. 1.d vs. the distance from centre for a core relative thickness of 1/3 and 2/3. One can deduce from this figure that the information on internal fibre orientation changes is contained in iso-signal curves which radius is higher than about 0.3
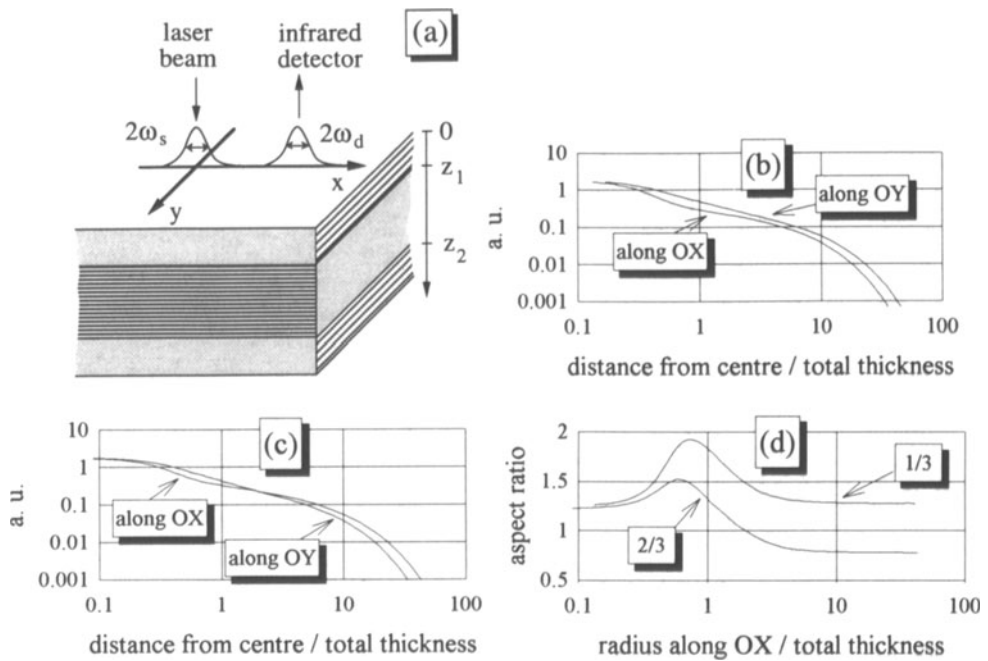
Fig. 1: Theoretical results in the case of a 3-layer stacking. (a): 3-D model used for simulations. IR signal distribution along the principal directions when the core layer relative thickness is 1/3 (b), and 2/3 (c). Aspect ratio of the iso-signal curves (d).

times the composite thickness. Away from about 4 times this thickness, all iso-signal curves have reached the same aspect ratio. This faraway distribution indicates how the core is thick, but doesn't help to locate it inside the stacking. The latter information can only be obtained by analyzing the intermediate distribution (from about 0.8 to 4 times the total thickness).

The modulated regime presents the advantage that the phase angle distribution is relatively immune to surface emissivity spurious variations [14, 15]. However, when internal orientation changes occur, the interpretation of the phase angle image proves to be far more complicated than for the amplitude image obtained in steady-state regime. As the latter one already provides in-depth resolved information on orientation changes we thus restricted the inversion to this particular regime.

Definition of an Inversion Procedure

We had to develop an inversion method which would, from a surface temperature pattern, provide a description of the orthotropic layers stacking sequence. We first restricted to the case of a three-layer stacking. The problem was thus reduced to identify the depth of the two skin/core interfaces.

As the direct calculation of the temperature profiles and then of the isotherm aspect ratio distribution takes several minutes for each configuration, their use in an inversion routine was discarded. We thus had to devise a method to rapidly compute an approximate theoretical isotherm shape distribution and make it progressively fit to the experimentally observed one. The adopted procedure is as follows: by taking the geometric mean of the isotherm principal radius s as the size variable; by taking the in-depth orientation function of the stacking $O(z)$ as a 0/1 function with a 0 value if the fibres are in the same direction as those of the upper skin, and 1 otherwise, we actually assume that the isotherm shape

function is related to the orientation function through a linear integral relation of the following kind:

$$\text{Isotherm Shape Function}(s) = \int_{z=0}^{z=\text{total thickness}} O(z)\, H(s,z)\, dz \qquad (1)$$

where $H(s, z)$ is a particular kernel.

For the isotherm shape function we chose:

$$c(s) = \frac{A - F(s)^2}{(A - 1)(F(s)^2 + 1)} \qquad (2)$$

where $F(s)$ is the aspect ratio of the isotherm and A the anisotropy coefficient of the individual layers (common to all of them).

It is easy to verify that, by applying thermal homogeneization, and by taking for $F(s)$ the far field isotherm aspect ratio, one obtains the proportion of fibres which are misorientated relatively to the skin layer fibres, i.e. the core relative thickness. This asymptotic relation was then formally extended to the whole isotherm field. The question which first appears deals with sensitivity: is the $c_3(s)$ function, which pertains to a 3-layer stacking, only sensitive to the core thickness or also - as it is desired - to its depth ? Will the deep boundary of the core layer be more difficult to localize than the shallow one ?

In order to provide quantitative answers to these questions, we will focus on the particular case of a symetric stacking containing a core layer of 0.5 relative thickness. Fig. 2 illustrates the $c_3(s)$ function and its sensitivities to the interfaces depths. The $c_3(s)$ function rises from 0 for central isotherms (on which the core layer has practically no influence), to 0.5 for wide isotherms (i.e. the actual core relative thickness). Its sensitivity to the first interface obviously rises earlier than its sensitivity to the second one. At large distance these sensitivities are respectively equal to -1 and +1: actually, $c_3(s)$ becomes merely sensitive to core thickness and no more to its depth. Anyway, the sensitivities to the interfaces depth are independant; these depths can thus be identified through the use of the $c_3(s)$ function. Nevertheless, due to lower values for the sensitivity to the deepest interface location, its characterization is expected to be less precise.

One of the main advantages for the use of the approximation described in (1), is that it induces the following additive property:

$$c_3(s, z_1, z_2) = c_2(s, z_1) - c_2(s, z_2) \qquad (3)$$

where $c_2(s, z_1)$ and $c_2(s, z_2)$ are the shape functions pertaining to two-layer stackings in which the second layer extents respectively from z_1 and z_2 down to the composite bottom. Thanks to this relation, the inversion problem which relies on a least-square minimization between the experimental and a theoretical $c_3(s)$ function reduces to the search of two unknowns to be taken from the $c_2(s)$ functions space. Furthermore we approximated this space by interpolation over a limited series of particular $c_2(s)$ functions which were calculated once for all. The inversion process was thus finally reduced to a few seconds.

As the identification method relies on several approximations, one had to assess their influence on the precision of the results. The first point to check was the validity of the additive relation (3). We again took the particular case of a symetric three-layer stacking with a 0.5 thick core layer. We reported on Fig. 3 the actual $c_3(s)$ function pertaining to this stacking and the approximated $c_3(s)$ function as obtained through relation (3) where z_1 and z_2 were assigned to their actual values. A small difference can be observed between the two curves, putting thus in the wrong the additivity expressed in (3). Nevertheless, if the actual $c_3(s)$ function is used as a pseudo-experimental result in the inversion process, one obtains as an answer that the core layer extents between 0.268 and 0.765 in relative depth, instead of 0.25 and 0.75. The approximated $c_3(s)$ function which fits the true one at best was also reported on Fig. 3. The error in core layer characterization is less than 2% and affects mainly its depth and hardly its thickness.
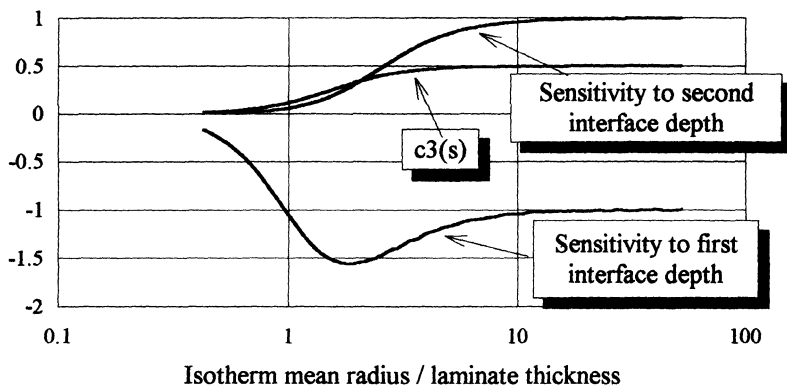
Fig. 2: $c_3(s)$ function in the case of a 0.5 thick symetric core layer and its sensitivities to first and second interface depths.
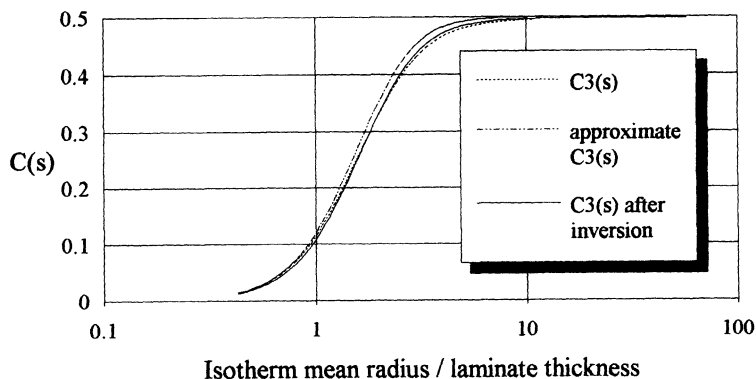

Fig. 3: $c_3(s)$ function for the previous 3-layer stacking obtained either exactly or approximately (through use of eq. (3)). Plain curve: approximate $c_3(s)$ function which fits at best to the exact one.

A few other tests were performed: a shallow core layer extending from 0.1 to 0.25 was identified between 0.109 and 0.243, which again shows that the inversion procedure is precise enough for practical use. Nevertheless, the results get worse when the core layer is deep into the composite: when it extends from 0.75 to 0.9, the inversion process identified 0.623 and 0.773 for the interfaces depth. Again, the error mainly affects the core mean depth, and it corresponds to a 13% underestimation. One can admit that the introduced approximations lead to an error of this range in the case of deep core layers. One has however to notice that, if the experiment could be performed from both sides of the composite plate, this bias would be considerably diminished.

EXPERIMENTS

In order to illustrate the performance of the inversion procedure for the core layer identification, we will present some experimental results obtained with a series of academic samples. These samples were prepared by ISRIM, Terni, Italy. They are carbon/epoxy samples, 2.2 mm thick, made of continuous fibre prepregs which were laid so as to realize 0/90/0 configurations with different core relative thicknesses (0.167, 0.333, 0.5, and 0.667). An other sample, 1.5 mm thick, was prepared in the symetric 0/90 configuration.

Each sample was heated for a few minutes with a 0.2 W Argon laser beam, about 0.8 mm in diameter. The experimental configuration was as depicted on Fig. 4. The samples were hold horizontally in order to prevent the temperature field from convection distortions. An infrared camera monitored the temperature distribution around the heated spot and the recorded isotherms were then fitted to ellipses. The resulting fields obtained for the four 3-layer composites are reported on Fig. 5. It is obvious on this figure that, as the core layer grows, the aspect ratio of the wider isotherms is progressively modified.

The ellipse aspect ratio distributions were inverted by the procedure previously presented. The nominal anisotropy coefficient of the layers had however to be known. It was measured on a unidirectional laminate, and we found about 6.4. An other parameter was still missing to perform the calculations: the ratio between the conductivities perpendicularly to the fibres, i.e. in the composite plane, and through the thickness. The data obtained with the sample having a 0.33 thick core were used to indirectly determine this parameter. One actually had to set it to about 2 in order to find, after inversion, a symetric stacking. This value can be explained by the composite fabrication process (compression moulding) which induced resin-rich sub-layers between the prepregs, leading thus to a lower mean conductivity through the thickness. This parameter was to be determined because of the particular morphology of the academic samples. It is expected that, in the case of injection moulded samples, it would be very close to 1.

The inversion was performed for the remaining samples. We reported on Fig. 6 the experimental $c_3(s)$ distribution as well as the theoretical fit in the case of the 0.5 thick core
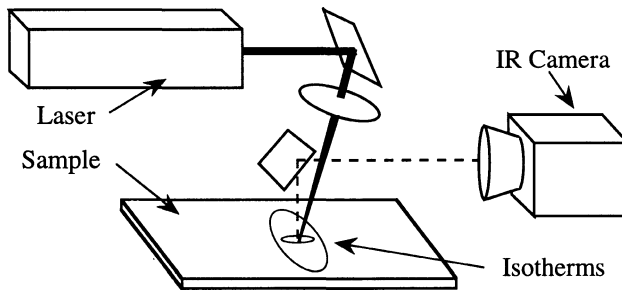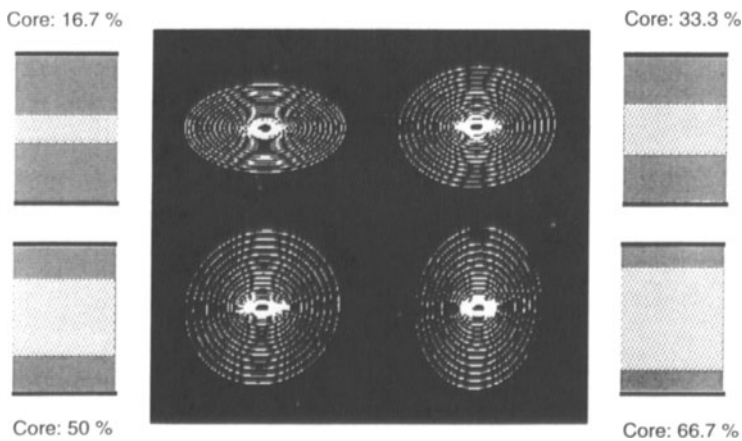

Fig. 4: Experimental set-up.

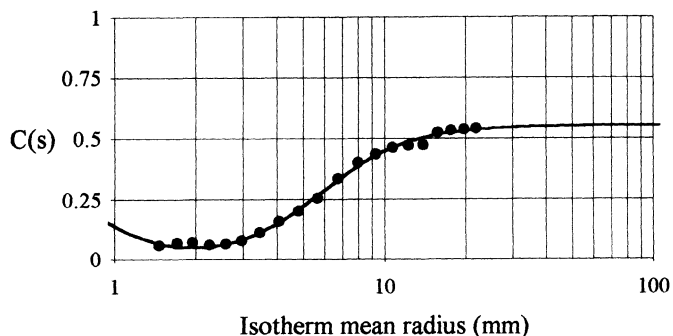Fig. 5: Ellipses which fit at best to the isotherms obtained on four different C/epoxy 3-layer stackings.

Fig. 6: c(s) function, i.e. apparent concentration of misoriented fibre, in the case of the 0.5 thick core layer. Experimental data (dots) and fitted curve as obtained after inversion.

layer. The identified depth of the boundaries are 0.30 +/- 0.01 and 0.85 +/- 0.02 instead of 0.25 and 0.75 (the covariance matrix was used to evaluate the confidence intervals at 95% probability). For this particular stacking, the error on core depth was of 7% and the one on core thickness was of 5%. These errors were respectively 6% and 3% for the 0.167 thick core composite, and finally 3% and 3% for the 0.667 thick core composite.

As far as the symetric 0/90 two-layer composite is concerned, we found two minima during the inversion: one solution nearly corresponded to the actual configuration (interfaces depth found at 0.49 and 0.99), whereas the other gave interface depths at 0.4 and 0.88, which essentially corresponds to a 11% underestimation of the "core" mean depth. Again, this spurious solution would have been discarded if a second measurement on the other side of the sample had been performed.

The confidence intervals at 95% were about +/- 3 to 4 % wide for core depth and thickness, except in the case of the thin core layer case (0.167 thick) where the width of the confidence interval for the depth reached 7 %. Finally, the error on core thickness was always less than 5%, and the error on its depth was at worst 7% (11% if one takes into account the spurious solution in the case of the 2-layer configuration).

CONCLUSION

Thermal ellipsometry refers to a thermal anisotropy measurement method which consists in analyzing the shape of the isotherms which develop around a heated spot. Initially restricted to homogeneous materials, it was recently extended to multilayer composites. A model of a three-layer stacking was devised in order to simulate the thermal behaviour of short fibre injection moulded composites. It helped for the build-up of an inversion procedure devoted to thickness and depth characterization of the core layer found in such materials. A first assessment of the inversion performances was experimentally undertaken with laminates made of continuous fibre prepregs. The core layer was successfully characterized for most configurations. In any case the error could still have been reduced by performing the measurements on both sides of the samples.

In this study we only considered stackings sequences with either parallel or perpendicular orientations. It is however to mention that the inversion in the case of anisotropic skin layers and an isotropic core layer would be straightforward.

Only three layers were considered in the inversion, but it would be easy to formally extend the identification procedure to more layers. Nevertheless it is unlikely that a description of the fibre orientation stratification into more than three layers would be

precise (as one increases the number of layers to characterize, the sensitivities of the isotherm shape function to the different interface depths progressively become dependant, preventing thus the inversion to remain stable). This limitation is not so dramatic as far as injection moulded composites are concerned, but even with a 3-layer inversion, the results are expected to be less precise than with the presented C/epoxy samples. First reason is that in such composites the transition between the orientation strata is smoother. Secondly, the anisotropy coefficient of individual strata is less important, thus reducing the aspect ratio of the observed isotherms.

ACKNOWLEDGEMENTS

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