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REMOTE NON-DESTRUCTIVE MONITORING OF SPATIAL FIBRE ORIENTATION IN POLYMER COMPOSITES.
SYNTHESIS REPORT – substitute the previous version

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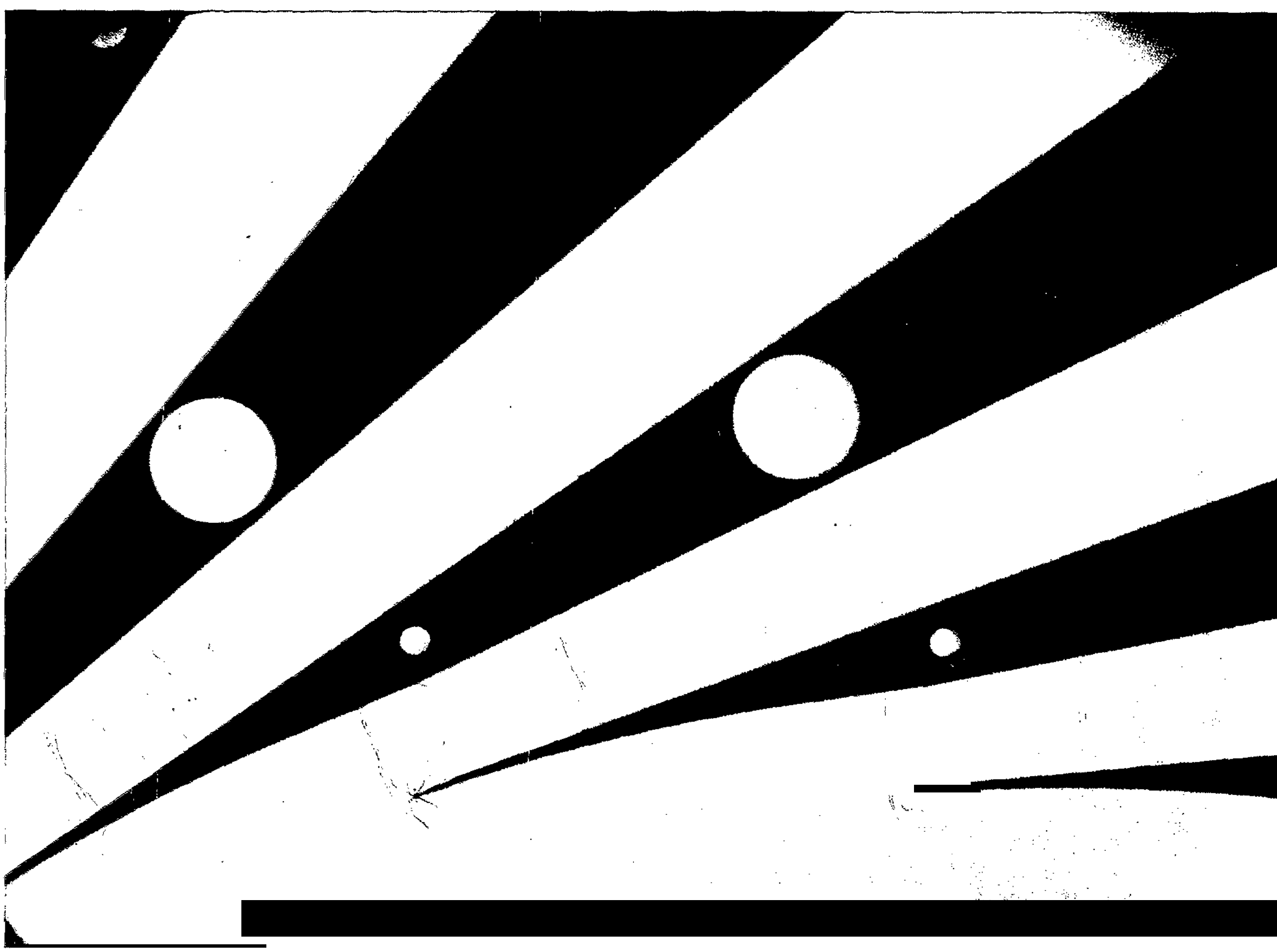

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SYNTHESIS REPORT FOR PUBLICATION

CONTRACT No: 13 RE2-CT92-0139
PROJECT N°: BE-508 1

TITLE: **Remote non-destructive monitoring of spatial fibre orientation in polymer composites**

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Approved by: Head of L3C laboratory

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TITLE : Remote non-destructive monitoring of spatial fibre orientation in polymer composites. Synthesis report

AUTHORS : J.C. Krapez, L. Mc Donnell, G. Busse, M. Diener, K. Nixdorf, W. Rippel, Ritter, J. Stanullo, R. teegmüller, D. Wu, S. Puzziello, T. Girasole, G. Gouesbet, G. Grehan, J.-N. Letoulouzan, " Mroczka, K.F. Ren, D. Wyzoczanski, G. Gardette, R. Gouyon

(individuals identities)

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- ONERA: thermal inspection by laser stimulated thermography (thermal ellipsommetry as well as by an eddy current method;
- CTRC: destructive characterization of the samples to provide reference dots;
- IKP: microwave transmission testing, thermal inspection, dielectric spectroscopy, ultrasonic backscattering, 3D speckle interferometry;
- ISRIM: preparation of autoclave, pultrusion, and injection moulding samples, as well as mechanical testing;
- LESP: fibre orientation monitoring by a light scattering approach..

KEY WORDS :

NDE - FIBRE ORIENTATION - COMPOSITE ANISOTROPY - INJECTION MOULDING

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REMOTE NON-DESTRUCTIVE MONITORING OF SPATIAL FIBRE ORIENTATION IN POLYMER COMPOSITES

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ABSTRACT

The high specific strength of modern materials is provided in most cases by suitably oriented fibres. Quality control of such materials requires methods that respond to fibre orientation in such a way that a map may reveal areas with deviations from the assumed or calculated orientation fields, In this project several non-destructive evaluation (NDE) methods were tested to explore how well they are suited to monitor and resolve in-depth fibre orientation in polymer composites, more specifically in injection moulded thermoplastics composites. Different types of composite materials were processed with either continuous fibres or short fibres to provide samples with either well controlled or unknown fibre orientation. 130th destructive fibre orientation characterization (by implementing scanning force microscopy) and mechanical testing were finally performed on a series of Round Robin test samples in order to assess the potential of the NDE methods for quality control.

It turned out that four NDE methods could be candidates for successful applications in industrial environment. First of them relies on the analysis of the thermal field induced by local heating of the composite surface and has two variants: thermal ellipsometry and lock-in thermography. As they are only sensitive to the thermal anisotropy generated by the particular fibre orientation, the fibres have to be more thermally conductive than the matrix, Thermal methods are thus efficient only in the presence of carbon fibres. An other remote method, i.e. microwaves transmission is applicable to all non-conducting fibres. Due to the same constraint regarding transmission, the light scattering method can only be successful if the matrix-filler system and the respective concentrations are such that light passes through the inspected material. The last method (ultrasonics) has the disadvantage of requiring liquid coupling to the inspected part. However, it responds to all kinds of fibres.

These methods are capable not only to display depth averaged orientation fields but also to indicate (at least to some degree) how orientation changes with depth.

INTRODUCTION

Because of weight reduction and equal or better structural properties with respect to metals, composites are increasingly used in engineering applications. The cheapest of them are probably the thermoplastics reinforced by short fibres because they can be produced by injection moulding. Most common fibres for this purpose are glass and carbon fibres. It is well-known that injection moulding process introduces a stratification as far as the fibres orientation is concerned. This stratification which is dependent upon the parameters of the process (thermal gradients, matrix viscosity, . ..) strongly influences the mechanical properties of the final product. The knowledge of the *lateral distribution and the depth profile of fibre orientation* is thus crucial, on one hand to *control the quality* of the final products, and on the other hand to consequently improve the fabrication process.

Fibres orientation characterization is often made *destructively* by slicing suitable sample sections and analyzing the micrographs obtained from them. Such a procedure is of course intrusive and furthermore *time consuming and expensive*. Quality control of such materials would require methods that respond to fibre orientation in such away that a map may reveal areas with deviations from the assumed or calculated orientation fields. The requirements imposed on such methods are not only high sensitivity but also the ease of application (untrained people) and no constraints on sample size and geometry. Therefore remote inspection is an essential feature.

The objectives of present Brite/EuRam project were to develop non-destructive evaluation (NDE) techniques, namely:

thermal ellipsometry in steady-state regime and phase sensitive modulation thermography,

microwaves transmission,

light scattering,

3D speckle analysis,

dielectric spectroscopy,

ultrasonic backscattering,

eddy-current analysis through magneto-optic imaging,

for the quantitative lateral and in-depth resolved characterization of short fibres orientation in injection moulded thermoplastics composites.

This research was to be carried out on polymer composites containing carbon or glass short fibres (somewhat less than 1 mm to 12 mm). Long, continuous fibres were nevertheless considered in a first step for in-depth sensitivity assessment of the different NDE techniques.

The work had to be oriented towards composite structures of relatively low thicknesses, typically the ones which are met in the automotive industry, i.e. from 1 to 5 mm, and of fibre content ranging approximately from 5 to 40%.

The first objective of the project was to define the most sensitive NDE method with respect to the type of filler used. The problem of lateral and in-depth resolution had also to be addressed.

In order to realize this performance analysis, comparisons were planned between, on one hand, the measurements provided by the NDE methods, and on the other hand, the real fibres orientation as a function of depth which were to be inferred from destructive tests.

The final objective was to propose generic correlations between the fibres orientation distribution as monitored by a particular NDE method or a combination of such methods and the quality of the composite as evaluated by mechanical tests.

This research project was set up with the support of the BRITE/EURAM program of the CEC. Five partners were involved:

ONERA: French national establishment for aerospace research. Leader of the project (L3C **laboratory).** Involved in NDE by thermal ellipsometry and in corresponding theoretical analysis as well as in NDE by eddy-currents with magneto-optic imaging.

CRTC Cork: Irish Technical College. Involved in the intrusive fibre orientation evaluation to serve as reference.

IKP Stuttgart: German University. Involved in NDE by phase sensitive modulation thermography (lock-in thermography), microwaves transmission, electronic speckle pattern interferometry (ESPI), dielectric spectroscopy, and ultrasonic backscattering.

ISRIM: Italian research institute. Involved in sample preparation and mechanical testing.

INSA Rouen: French University. Involved in NDE by light scattering and corresponding theoretical analysis.

This Brite/EuRam Project had four endorsers: Aérospatiale CCR Suresnes (F), Bayer (G), Fiat Research Centre (I), Manducher (F) who also supplied the partners with representative samples.

1. SAMPLE PREPARATION

To evaluate the possibilities of the NDE methods, samples with known and unknown fibre distribution have been prepared at ISRIM by applying industrial techniques such as Autoclave's lamination, Pultrusion, Compression Moulding and Injection Moulding. First three techniques were applied to prepare samples with well controlled fibre orientation such as ladder orientation (see Fig. 1.1), dual step orientation (see Fig 1.2), skin core orientation (see Fig. 1.3), quasi-isotropic orientation, and with different fibre content. After that the NDE methods have been tested on samples with unknown fibre distribution realized by Injection Moulding (see Figs 1.4 and 1.5).

1.1. Samples with controlled fibre orientation

Compression Moulding, Pultrusion, and Autoclave techniques have been applied on thermosetting matrices, such as polyesters and epoxies, reinforced with glass or carbon fibres.

Compression Moulding technique has been applied first to realize a ladder sample 1 mm thick by moulding together prepreg plies in a 70 *x* **140** mm mould and then to prepare filled (with calcium carbonate) and unfilled polyester matrices samples 3 mm thick to have a baseline correction on light scattering technique and to test its capability to see through a non transparent material. The conditions adopted for the preparation of the samples were: 180 "C and high pressure for the prepreg plies, and 120 °C and small pressure for the polyester samples.

The Pultrusion technique has been adopted to produce samples with different fibres content, in fact using an ad-hoc die. 30 mm wide and 3 mm thick specimens, made of unsaturated polyester and glass fibres, have been obtained with a fibre content varying from 35 to 60 wt %. Moreover samples with and without filler (Dolomite) but with the same fibre content have been prepared to test the NDE methods on the possibilities of discern between fibres and filler.

The Autoclave technique has been adopted to prepare different types of samples to test the possibilities of the NDE methods to reveal changes in fibre orientation. The ladder samples have been produced by laminating on a flat 200x **200** mm surface plies of carbon-epoxy or glass-epoxy prepregs to obtain a pattern as described in Fig. 1.1. The ladder step were, for example, of 30 mm on the borders and then of 20 mm in between with a ply in the 0/90 ^{0} direction on the top/bottom of the sample. The dual step samples were prepared with plies directions as shown in Fig. 1.2. The number of plies in the 0_i direction as compared to those in the 90° direction was varied from one sample to the other. The skin-core orientation samples were prepared to obtain symmetrical 0°/900/00 stackings, as shown in Fig. 1.3, by varying the number of the plies in each strata to simulate different conditions of skin-core composition. The last type of samples were those with a quasi-isotropic orientation in which plies of 150 x 150 mm were placed from 0° to 90° with rotation step of 15° or 30° and with symmetry in respect to the middle layer.

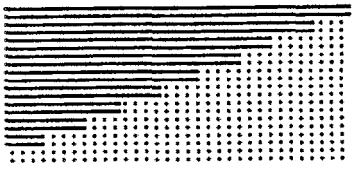


Fig. 1.1 - Continuous fibres samples realized by ISRIM with ladder-type fibre orientation (plies at either 0° or at 90°)

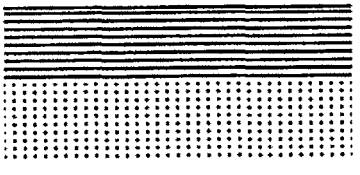


Fig. 1.2 - Continuous fibres samples realized by ISRIM with dual step orientation (plies at either 0° or 90" with different ratios for layers thicknesses)

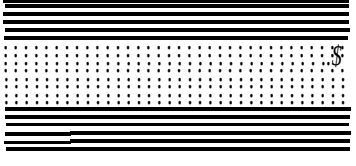


Fig. 1.3 - Continuous fibres samples realized by ISRIM with the [0/90/0] skin-core-skin structure

1.2. Samples with unknown fibre orientation

Previous samples had known fibres orientations and were thus rather academic. Samples with unknown fibre orientation were then prepared using the Injection Moulding process.

The dog-bone and rectangular plates samples, shown in Figs 1.4 and 1.5 respectively, were obtained using polycarbonate as matrix and short glass fibres as reinforcement and by injecting the melt material in corresponding moulds. Different percentage of reinforcement were chosen to assess the NDE methods and also to permit a study on the influence of the reinforcement and of its distribution on the mechanical properties of the material. The percentage of the fibre composition ranged form O to 40% in steps of 5%. The rectangular plates were injected at a single point inducing thus a radial flow. A series of such rectangular plates with different fibre concentrations were chosen as Round Robin Test samples.

Dog-bone samples made of Makrolon 8030 (polycarbonate + 3070 glass fibres) were also supplied by Bayer for the light scattering and microwaves approaches.

Aérospatiale supplied the partners with injection moulded rectangular plates made of C/PEEK. Those plates were 150 x 150 mm large and had a 14 mm hole at the centre which induced disturbance to the flow during the injection and therefore a weld line downstream from the hole (see fig. 1.6). A couple of such plates from the same batch were chosen as Round Robin Test samples which, after various NDE measurements, were submitted respectively to destructive fibre orientation evaluation and to mechanical testing.

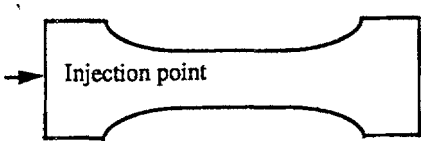


Fig. 1.4 - Short fibres samples realized by ISRIM with dog-bone *geometry (G/polycarbonate)*

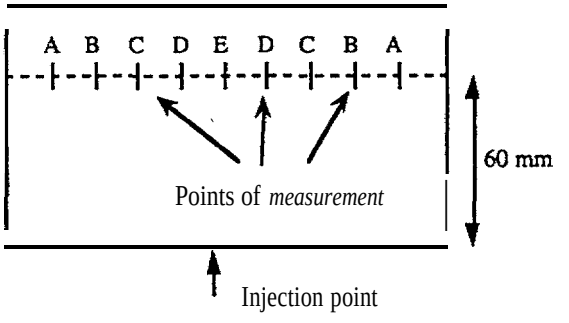


Fig. 1.5- Short fibres samples realized by ISRIM with rectangular geometry and radial injection (G/polycarbonate)

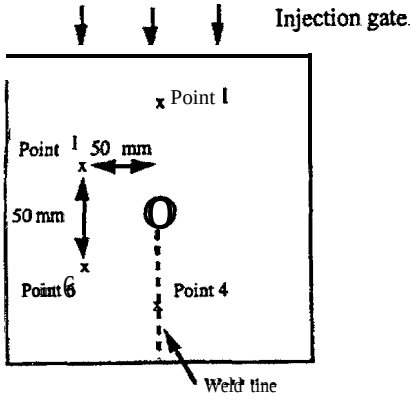


Fig. 1.6- Short fibres samples realized by Aérospatiale with square geometry, uniform injection, and a central hole inducing a weld line downstream (C/PEEK)

2. NON-DESTRUCTIVE EVALUATION OF FIBRE ORIENTATION

A series of seven NDE techniques were initially considered to solve the problem of fibre orientation characterization in composites with lateral and in-depth resolution. After an exploratory phase it rapidly turns out that some methods had a negligible potential for industrial application. Sensitivity of the measured physical variables to the fibre orientation were too low regarding the

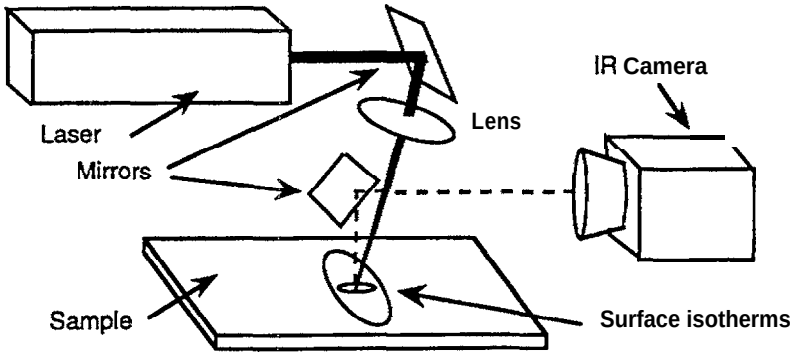
nature of commonly used fibres and the available instrumentation. For this reason, eddy -current imaging with magneto-optic sensor, dielectric spectroscopy and electronic speckle pattern interferometry were to be discarded.

It turned out that four NDE methods could be candidates for successful applications in industrial environment: thermal diffusion through its two variants thermal ellipsometry and lock-in thermography, microwaves transmission, light scattering, and ultrasonic backscattering.

Thermal ellipsometry and lock-in thermography indeed correspond to the same approach i.e. the analysis of the thermal field induced by local heating of the composite surface. The former initially assumed constant heating and referred to the steady-state regime whereas the latter is based on sinusoidal heating and provides amplitude and phase images of the temperature modulation map.

2.1. Thermal ellipsometry

The experimental set-up implemented at ONERA is depicted on figure 2.1: a 0.1-0.2 W Argon laser beam, focused down to 0.8 mm in diameter was used for heating the samples and an infrared camera (Agema 880LW or Amber 4128) was used to record the temperature distribution around the spot. The samples were hold horizontally in order to prevent the temperature field from convection-induced asymmetrical distortions.



Fig, 2.1- Experimental set-up for thermal ellipsometry.

Assuming that all fibres are parallel in the material, the isotherms are in that case elliptical: their common ellipticity corresponds to the square root of the ratio of the principal in-plane thermal conductivities. A model was then necessary to understand which effect do internal fibre orientation changes have on heat transfer in the context of a thermal ellipsometry experiment.

ONERA therefore developed a general analytical model to simulate at best the thermal behaviour of injection moulded plates. The application of thermal ellipsometry to the 3-layer morphology generally met in such composites is as follows: near the heated point the isotherms shape is mainly determined by the upper skin layer anisotropy, whereas only the widest isotherms are influenced by the core layer properties (anisotropy, thickness and depth). The analysis of the isotherm aspect ratio distribution around the heated spot thus provides an insight on the depth of the two skin/core interfaces. We recall on figure 2.2a particular theoretical result obtained with the 3-D analytical model of heat transfer in orthotropic laminates. It corresponds to the particular case of steady-state regime with symmetrical stackings in which the core has relative thickness of 1/3 or 2/3 (plies of C/epoxy and typical experimental conditions were considered). The aspect ratio of the isotherms r_x/r_y was reported vs. their relative mean radius, i.e. $(r_x r_y)^{1/2}/e$ where e is the plate thickness. The aspect ratio of the smallest isotherms is relevant of the skin layer anisotropy and of the heating spot size. Far away from the centre the aspect ratio reaches an asymptotic value which is merely representative of the core relative thickness. Only the isotherms in a transition region extending from about 0.3 to 10 times the total thickness can thus help to evaluate the core layer depth.

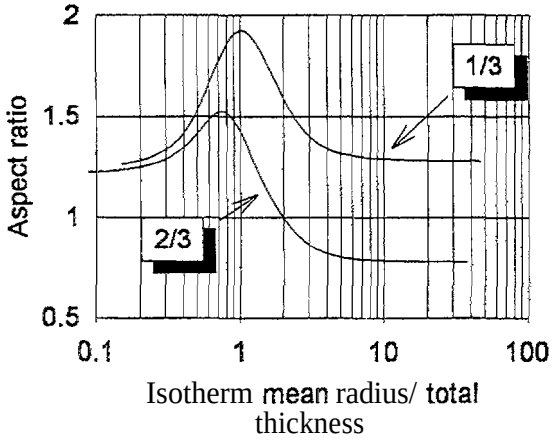


Fig. 2.2 - Isotherm aspect ratio profiles obtained in steady-state regime in the case of two symmetrical skir#core/skit stackings (core oriented at 90° to the skin). Core relative thickness of either 1/3 or 2/3.

From this observation, an inversion method was then devised to infer both the core layer depth and its thickness from the steady-state thermal field by fitting experimental and theoretical distributions of the isotherm aspect ratio. The proposed method was applied on various samples provided by ISRIM and Aérospatiale.

Fig. 2.3 indicates the IR images obtained with four symmetrical C/Epoxy samples 2.2 mm thick which were made of continuous fibres plies and where the core relative thickness was 1/6, 1/3, 1/2, and 2/3 (refer to fig. 1.3 for sample geometry).

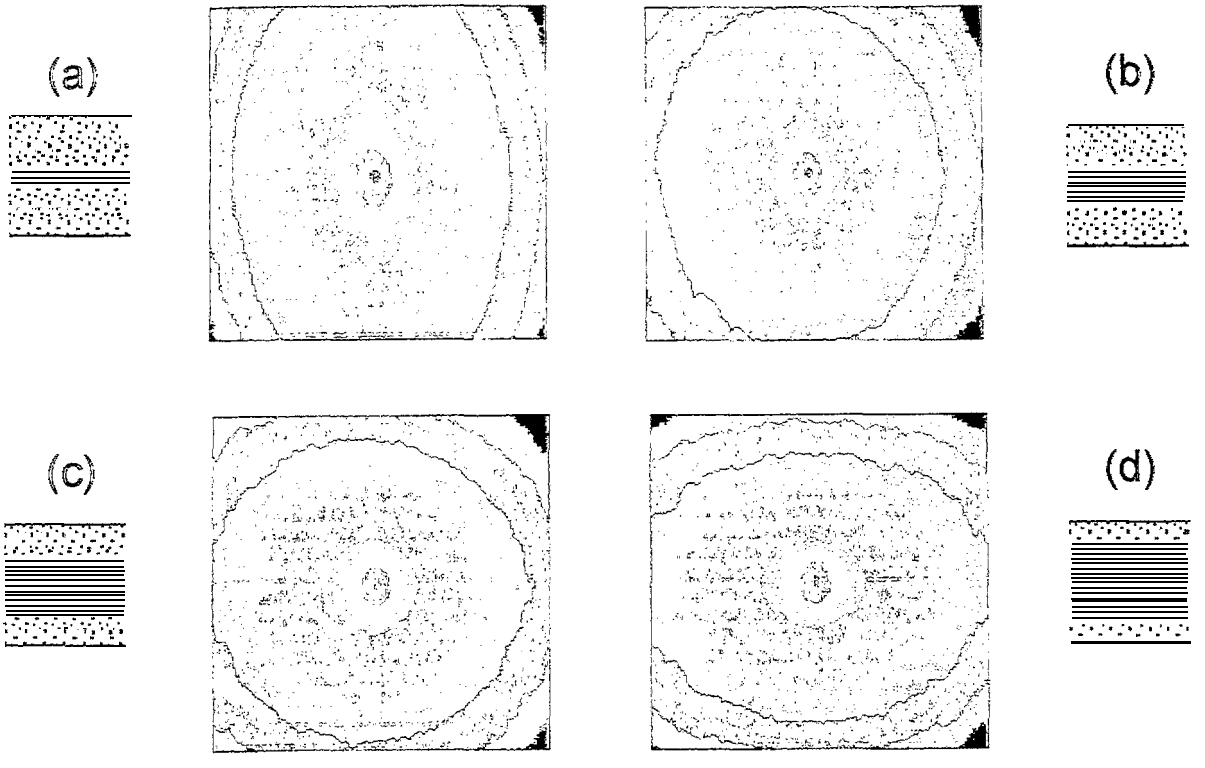


Fig. 2.3 - IR images obtained with C/epoxy samples having a [0/90/0] structure (core relative thickness: 17,33, 50,67%)

After inversion, the retrieved values of the relative thickness and depth were as reported in Table 2.1. The results about core thickness are reasonably precise (less than about 5% error). However, the precision of its mean depth gets less satisfactory as the upper interface gets deeper into the composite.

Table *2.1. Results of the inversion of IR images presented on fig.* 2.3

Core layer actual thickness	Calculated thickness	Calculated mean depth
0.167	0.146 ± 0.013	0.664 ± 0.050
0.333	0.341 ± 0.021	0.501 ± 0.024
0.500	0.553 ± 0.015	0.581 ± 0.016
0.667	0.694 ± 0.019	0.533 ± 0.015

Thermal ellipsometry was also performed at several locations of an injection moulded short fibres C/PEEK sample having an insert which induced a flow bifurcation and a weld line downstream (refer to Fig. 1.6). The isotherms were all elongated in the flow direction, thus indicating that the core layer relative thickness was everywhere less than 0.5 (see fig. 2.4). After inversion the core layer was generally found nearer from the upper surface than it was in reality (as revealed by microscopy after sectioning the sample). Anyway, the calculated core thickness variations across the sample were in close relation with the actual counterpart. Furthermore we found a good correlation between the mean isotherm aspect ratio and the part of fibres which are in the mean flow direction (see fig. 2.5).

Finally, by comparing thermal ellipsometry results and mechanical testing results (see fig. 2.6), a significant correlation was found between the mean aspect ratio at each test location and the flexural modulus measured on coupons extracted therefrom (correlation coefficient of 0.96), thus highlighting the potential of thermal ellipsometry for the quality assessment of injection moulded composites made of carbon fibres.

Other temporal regimes, in particular, the periodic regime, were tackled during the experiments. A lock-in technique recently developed at ONERA was applied together with thermal ellipsometry arid led to a new approach for in-plane principal thermal diffusivities measurement.

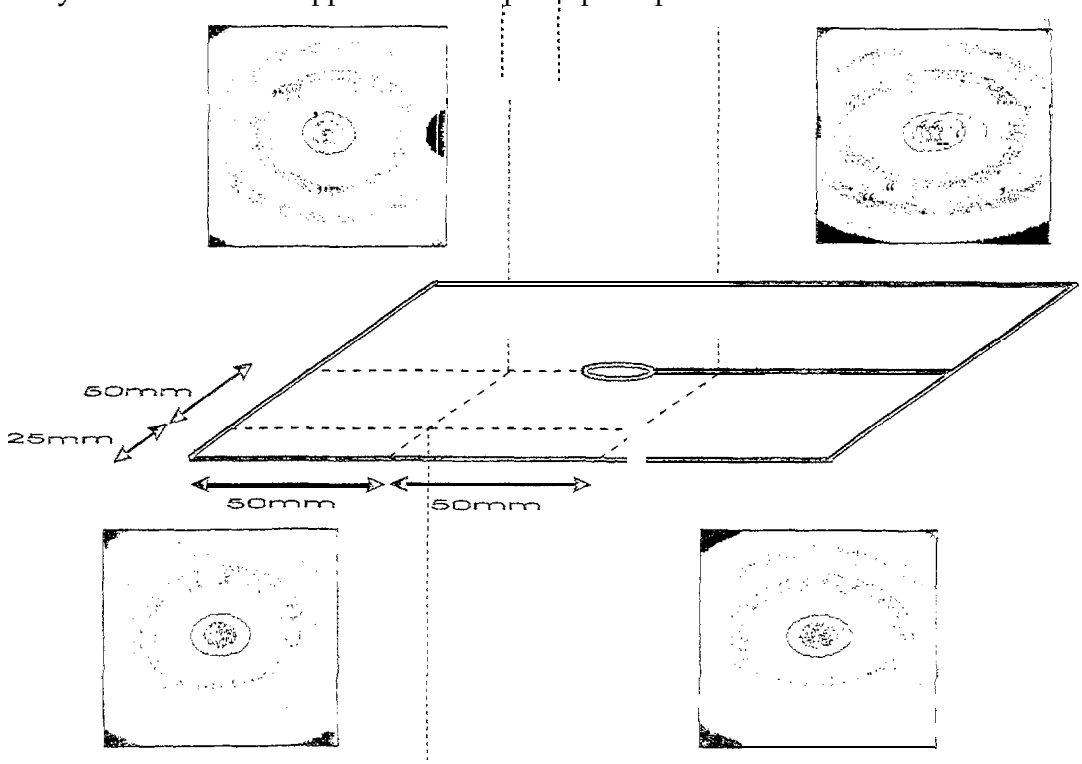


Fig. 2.4- Thermal ellipsometry of a C/PEEK short fibre injection moulded sample with a 14 mm hole and a weld line downstream.

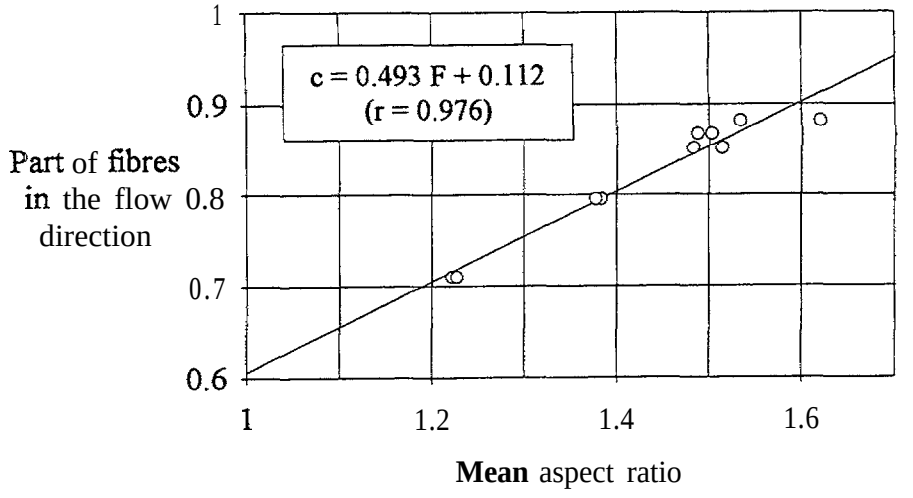


Fig. 2.5- Correlation between Thermal Ellipsometry results and actual fibre orientation distribution inside the C/PEEK sample reported in fig. 2.4.

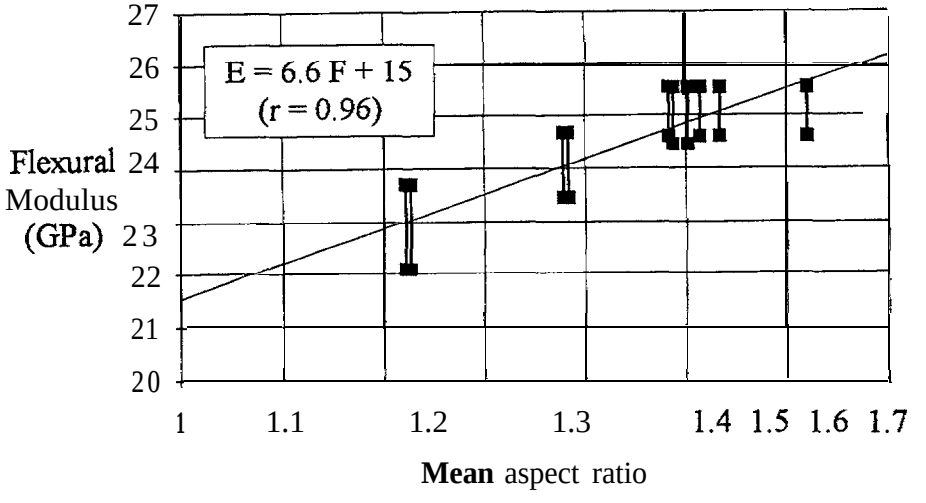


Fig. 2.6- Correlation between Thermal Ellipsometry results and local mechanical properties of the C/PEEK sample reported in fig. 2.4.

2.2. **Phase sensitive modulation thermography ("lockin-thermography")**

This method is based on thermal waves and on their detection using photothermal radiometry which is known to respond to hidden boundaries. Depth range of this remote method depends on modulation frequency in such a way that some millimeters in polymer materials require a frequency well below 1 Hz. At such low modulation frequencies the time to make a raster scan image exceeds by orders of magnitude the industrial needs.

Therefore a multiplex technique is desirable where many picture elements are monitored in parallel during each modulation cycle. In that case the time for image generation is given rather by the modulation period than by its product with the number of picture elements. This technique is now capable to provide within 3 min. a phase angle image even when the modulation period is 1 min. thereby giving a depth range exceeding 5 mm in most polymer materials.

The phase field around a modulated circular focal spot is an array of elliptical patterns whose ellipticity depends on their distance from the focal spot in a way which is determined by the depth profile of orientation. Therefore the analysis of such a spot allows to characterise the orientation. If many spots are projected and modulated simultaneously the three dimensional orientation field may be visualized within 3 min. without any physical contact to the sample. As this method is based on heat transport it is applicable for materials where the fibre has a substantially

higher thermal diffusivity than the matrix. So orientation depth profiling with lockin-thermography is feasible only for CFRP.

Single point measurements were performed with a diode laser of 30 mW at 812 nm wavelength. For the multispot arrangement we used an Ar^+-laser of 4 W split into 49 beams by a holographic grating. Using this technique we could inspect e.g. the orientation step CFRP sample within three minutes. Also the fibre orientation fields in short fibre samples can be analysed.

2.3 **Microwave anisotropy imaging**

Fibres differ from the matrix material around them not only by their them-ml properties but also by their dielectric properties which can be probed by microwaves. The orientation of the local dielectric tensor which is correlated with the fibres orientation can be monitored with polarised electromagnetic waves. At a microwave frequency of 30 GHz "local" means a spot of about 6 mm diameter.

In transmission experiments the obtained result is always an average across sample thickness. This may be a real disadvantage for injection moulded samples where orientation in the "core" differs from the. "skin". Therefore we investigated how depth-weighted information can be derived from measurements based on standing waves. This concept is applicable if sample thickness is less than half the microwave wavelength inside the material. So the set-up requires a rotatable microwave unit, a scanner unit moving the sample in a raster fashion, and a movable mirror. Samples to be inspected remotely with microwaves must be electrical insulators like GFRP or LCP materials.

This method is confined to plate-like samples where local curvature can be neglected. It was demonstrated that one can monitor the orientation of long fibres and short fibres in injection moulded samples. Also in this case three dimensional information is obtained.

2 . 4 Ultrasonic backscattering

Ultrasonic inspection is based on a pulse-echo-technique which requires a liquid coupling between the sample and the transceiver. If the transceiver is inclined with respect to the sample surface there will be echoes only from boundaries inside the sample, like from fibres that are perpendicular to the acoustic beam. By moving the transceiver on a cone one can monitor at which angles there is an echo and which depth it comes from. This allows for local orientation depth profiling of all fibre reinforced materials which may be exposed to water. The technique is not applicable to big objects and to complex surface geometries.

As the reflection of elastic waves is determined by a fourth rank tensor one obtains sharp reflexes from the fibres thereby allowing for high angular resolution. The result of such a local measurement is an angular B-scan displaying the intensity of echoes in a plane given by angle and time. This method should also have the potential to evaluate the component of fibre orientation perpendicular to the sample surface.

2.5 **Light scattering**

INSA Rouen created a Monte-Carlo code to predict the backward and the forward electromagnetic field scattered when a beam impinges a cloud of fibres. The fibre direction was assumed to be function of depth and multiple scattering phenomena were eventually taken into account. Application to light and microwave scattering was performed in the case of matrix-filler systems, reinforcement concentrations, and orientation evolutions found in industrial composites as proposed by Bayer, Isrim and Aérospatiale.

Based on Maxwell's equations, the code allows to predict the scattering of any electromagnetic wave, including propagative microwave and infrared radiation, which are of interest for matrix-filler systems. The Monte-Carlo method simulates a complex process as a succession of elementary events. The theoretical tool assumes that the elementary event is the scattering of a plane wave by an infinite cylinder. The media which contains the cylinders is assumed to be transparent and homogeneous, at least on a given scale around the particle. The

cylindrical particles have a length much larger than the wavelength of the incident beam and a ratio length/diameter much larger than 1. The regime of multiple scattering may occur or not and smoothing of fine details of the scattering patterns in the process is assumed. The scattering of a Gaussian beam is modelled as being the one of a plane wave, except for a large scale phenomenon associated with the Gaussian beam. Simulation characteristics studied in a general geometry without limitation are determined by four entries: the sources of light, the interfaces, the scattering media and the detectors.

INSA Rouen developed anew non-destructive optical light scattering diagnosis to measure fibre orientation and concentration for a depth-resolved quantification in composite polymers: this simple and direct method is based on the analysis of the scattered image of a laser beam crossing the cloud of fibres of the sample. Orientation and concentration of the fibres are independently and directly determined, respectively by the orientation and the length of the main axis of the ellipse which fits at best the scattering image of the sample.

The qualification of the "two crossed polarization camera system" prototype, devoted to laboratory developments, was achieved with industrial samples (glass fibres samples) supplied by Bayer and ISRIM.

Information	independent and direct characteristics on concentration and orientation of fibres
Samples	"transparent" polymeric matrix (transmission not lower than 0.00 1) with long or short glass fibres
Resolution volume	1 mm x 1 mm x 0.1 mm (height x width x depth) at maximum depth resolution
Working distance	remote measuring method with no limitation on working distance
Measurement time	1 min (actual prototype) to 1 s (industrial prototype)
Orientation accuracy	depending on selected intensity levels range typically 6% measured with L5 ISRIM sample (fig. 1.5 with 5% fibres) with an integrated value on depth of the sample
Orientation measurement range	0°-360° (no limitation)
Concentration measuring range	up to 30% fibre volumic fraction (limitation, linked with multiple scattering phenomena, could be overtaken with an indirect measurement method with further developments)
Concentration accuracy	typically less than 3% measured with L5 ISRIM sample with an integrated value on depth of the sample
Reproducibility	depending on intensity levels. Typically, for L5 sample (point 5, depth = 1.55 mm, focused incident laser beam V-polarized): - orientation angle :-13° ± 10 - concentration (large axis in pixel) :53 ± 10
Restrictions	in-depth resolution depending on optical thickness no edge effects as the laser beam provides a local probe (1 mm x 1 mm)
cost	50 kECU
Physical set up	0.5 m × 0.2 m × 0.2 m
Industrial transfer	* laboratory prototype including various versions for developments * light scattering codes including multiple scattering to evaluate industrial versions of prototypes and/or modelling new methods and prototypes

3. MECHANICAL TESTING

To establish a correlation between the fibre orientation derived from the NDE methods and the properties of the materials two different destructive approaches have been followed. First of all the samples have been characterized by determining their mechanical properties and then, on specimens chosen as Round Robin samples on which the different NDE methods have been applied, by microscopic analysis of the fibre distribution and orientation in different strata of the material. ”

The mechanical properties have been measured following the recommendation of the European Norm EN 63 for the determination of the flexural properties of plastic reinforced materials; the value obtained for the injection moulded samples prepared by ISRIM and for those supplied by Aérospatiale are reported in Tables 3.1 and 3.2 (see respectively figs 1.5 and 1.6 for the locus of the analyzed points).

Table 3.1 - Mechanical properties of polycarbonate-glass fibres injection moulded samples (10, 25, and 40% glass fibres)

SAMPLE	Ultimate Flexural Strength (MPa)	Ultimate Yield Strain (mm)	Flexural Modulus (MPa)
A1O	104.2 ± 0.6	8.45 ± 0.23	3289 ± 63
B1O	100.7 ± 1.8	8.87 ± 0.33	3083 ± 88
C10	99.1 ± 1.1	9.05 ± 0.25	3027 ± 44
D1O	101.1 ± 0.6	8.82 ± 0.24	2991 ± 70
E1O	96.9 ± 0.8	8.60 ± 0.33	2877 ± 28
A25	120.7 ± 2.7	4.55 ± 0.29	5022 ± 283
B25	118.4 ± 9.6	4.53 ± 0.35	4945 ± 35
C25	116.1 ± 4.3	4.52 ± 0.26	4989 ± 312
D25	118.9 ± 3.2	4.31 ± 0.28	5163 ± 330
E25	117.4 ± 3.8	4.03 ± 0.31	5271 ± 374
A40	131.3 ± 2.6	3.48 ± 0.13	6948 ± 79
B40	121.3 ± 1.3	3.62 ± 0.09	6417 ± 79
C40	121.4 ± 1.2	3.38 ± 0.10	6647 ± 92
D40	140.1 ± 1.1	3.13 ± 0.05	7953 ± 132
E40	145.4 ± 1.5	2.97 ± 0.05	8507 ± 65

The results obtained for the injection moulded polycarbonate samples revealed that, as one could expect, all the mechanical properties depend on the fibre content, but there are also variations from point to point due to the radial flow in the specimens which was induced by the chosen injection geometry.

Table 3.2- Mechanical properties of C/PEEK injection moulded sample

Point n°	Ultimate Flexural Strength (MPa)	Ultimate Yield Strain (mm)	Flexural Modulus (MPa)
1	311.5 ± 17.1	2.920 ± 0.018	22890 ± 820
4	342.3 ± 1.3	2.653 ± 0,017	25070 ± 467
5	347.4 ± 3.8	2.83 ± 0.06	24070 ± 608
6	351.2 ± 5.4	2.75 ± 0.12	24995 ± 541

For the PEEK/Carbon sample supplied by Aérospatiale the difference in the mechanical properties for the different analysed points is very clear and the maximum value is on the weld line downstream from the hole.

4. SAMPLE INTRUSIVE CHARACTERIZATION

In order to determine the effectiveness of the non-destructive evaluation (NDE) techniques it is essential to compare the results of the NDE techniques with the fibre orientation that prevails within the test samples. The determination of the actual distribution of fibres within fibre reinforced polymer composites requires destructive sectioning of the test sample at a desired location, polishing of the exposed surface and finally, observation of the polished surface using an appropriate microscopic technique. Conventionally, optical microscopy has been used for the latter observation phase. Within this project, the technique of scanning force microscopy (SFM) was used at CRTC, in addition to optical microscopy, during the observation phase. This is a novel application of the SFM technique.

4.1. Technical description

4.1.1. Sample Preparation

Samples were sectioned from the fibre-reinforced plates using standard techniques. Firstly, a region was marked out that was approximately twice the size of the final sample and for which the section to be cut was central. This piece was then cut with a diamond wheel close to the desired section. The sample was then mounted using a cold casting resin and the section surface was then polished using standard polishing techniques with the final polish using a 0.25 micron grade diamond polishing compound. The polishing process was monitored using optical microscopy and after a satisfactory surface finish was obtained, the polished surface was cleaned by washing, using an ultrasonic bath when required.

4.1.2. Strategy *for Microscopy*

The overall strategy for the microscopy of the polished surfaces was determined from an examination of the capabilities of the optical and scanning force techniques with regard to the particular characteristics of the composite samples. Particular advantages for SFM include a higher spatial resolution and the ability of the technique to see optically transparent objects and to image material properties.

While the SFM is slower than optical microscopy in terms of actual image acquisition, this is not a problem given that the actual image acquisition time is a very small part of the process to determine fibre distributions in these materials.

The strategy developed was to use optical microscopy for low to medium resolution imaging and to use scanning force microscopy for high resolution.

4.1.3. *SFM System*

A Topometrix Explorer SFM system was automated by adding an inertial drive x,y positioning stage for the sample. This prototype stage was assembled from component parts and provides 7 mm of linear movement for each of the two axes. This stage has been modified to suit the resin mounts used to hold the polished composite samples. The stage is mounted inside a large suspended mass plate which serves to reduce external vibrations. The translation stage is under computer control and it is possible to program a sequence of steps and scans in order to acquire SFM images automatically. The overall schematic for the automated SFM system is shown in Figure 4.1 and is self-explanatory.

The most important issue from the SFM imaging perspective was to obtain images that showed high contrast between the fibres and the matrix. High contrast simplifies computerised

image processing and object recognition. Three distinct SFM imaging modes investigated: topography mode, modulation mode and lateral force mode. The best contrast images for the widest range of composite types were obtained in lateral force mode. The effectiveness of this mode is illustrated in Figure 4.2 which shows a lateral force microscopy image of a polished section through a carbon fibre reinforced injection moulded polymer composite.

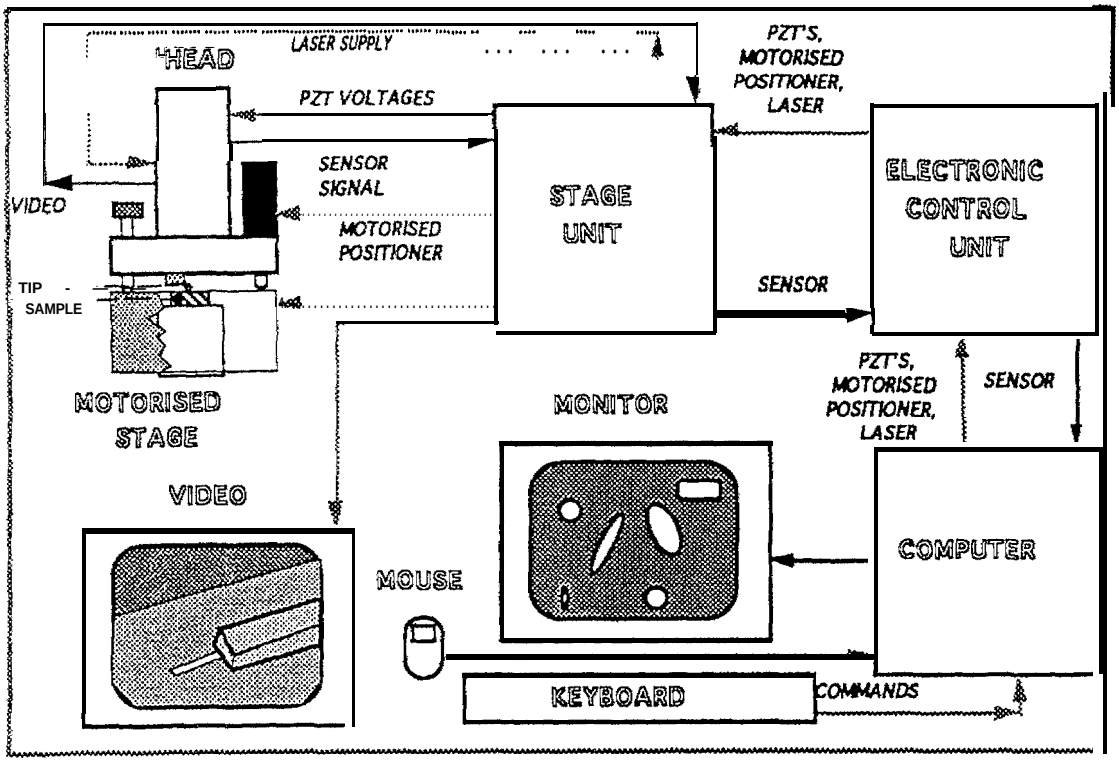


Fig. 4.1- Schematic of the SFM system

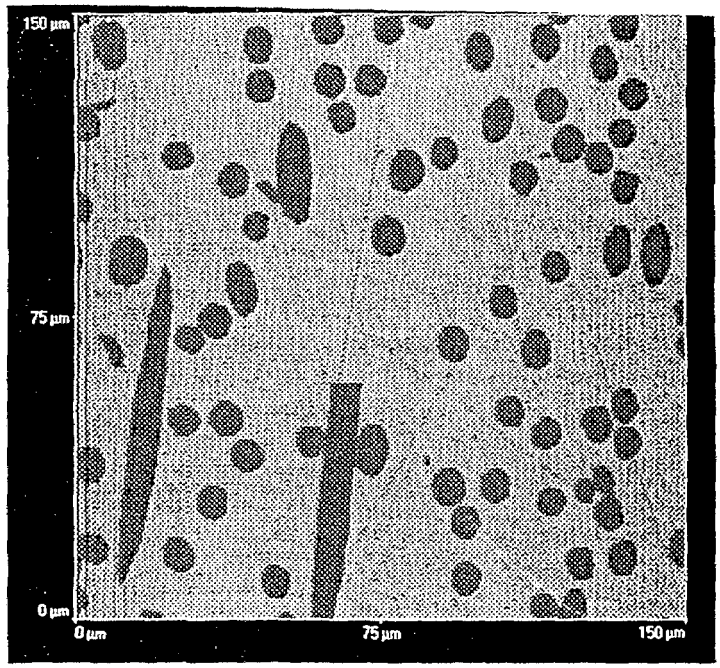


Fig. 4.2- Lateral force microscopy image of a polished section through a carbon fibre injection moulded composite

4.1.4. Image Analysis

Despite the use of powerful commercial Optilab software and contrast SFM images, considerable manual effort was required to edit the images and to analyse the fibre orientations. Also during step and scan imaging, the individual steps were kept less than the scan length of 150 μm in order to ensure that no portion of the surface was missed. Consequently, each successive image contained redundant information that required further manual editing. While a complete automation of step and scan SFM image acquisition *and* analysis is feasible, this would have required considerable effort and was deemed to be outside the scope of this particular project.

4.2. **Results**

The automated SFM system has been used to image the polished sections of a range of glass fibre and carbon fibre polymer composites. For each section surface the fibre count and the fibre orientations relative to the sample axes were determined for 150pm thick depth slices and the data was presented in spreadsheet format and also graphically. Particularly extensive studies were carried out on the injection moulded "Round Robin" samples that were used in the final phase of the project to evaluate the capabilities of the NDE techniques developed during the project.

5. CONCLUSION

A series of seven non-destructive methods were assessed and eventually improved in order to provide efficient remote techniques for the fibre orientation evaluation in polymer composites.

Different kinds of samples were prepared for the assessment: they were either academic samples containing continuous fibres with well controlled orientation (C/epoxy, G/epoxy, G/polyester) or short fibre samples representative of the composite materials used in the automotive or aeronautic industry (C/PEEK, G/polycarbonate).

Three methods (speckle analysis, eddy-current analysis by magneto-optic imaging, and dielectric spectroscopy) were soon discarded since their applicability is difficult or confined to the lab. The other methods complement each other with respect to materials and applicability.

If the material is transparent enough in the visible spectrum (transmission higher than 0.001) the light scattering approach provides valuable information both on fibre orientation and fibre concentration. In order to prevent the filler particles from scattering too much the incoming light, the fibre and other additive (i.e. colour pigments) concentration has to remain relatively low.

If the sample is flat, not sensitive to water and small enough to be immersed in a tank, then ultrasonic backscattering may be applicable with an acceptable effort, especially since conventional C-scan units can be modified.

If water coupling is not acceptable then the choice between microwaves transmission method and heat transfer method (thermal ellipsometry in steady-state or periodic regime) depends whether on the inspected component is filled with heat conductive fibres or not. Depth information is not as easily obtained as with ultrasonics, and equipment requires more effort than just a simple modification of existing commercial units. Though the thermal methods use conventional cameras, orientation measurements require either an array of focused beams (possibly time-modulated) or a scanning unit which moves one laser focus across the sample in a raster fashion.

For industrial applications the time required for inspection is an important quantity. From the present experience it seems that microwaves and ultrasonics require about half an hour for an raster image of orientation. As far as the thermal approach is concerned, a couple of minutes is necessary for the measurement of the mean fibre orientation in each 40 x 40 mm area.

Scanning force microscopy has been successfully used to provide high contrast images of polished sections of glass fibre and carbon fibre polymer composites. These images were subsequently analysed to provide the actual fibre distribution within the composites. An automated

step **and** scan SFM system was developed to obtain composite images 150 μm wide and up to 7 mm long.

Mechanical properties of a series of Round Robin samples were finally measured to provide local flexural modulus, ultimate flexural strength, and ultimate yield strain.

The data obtained by the NDE methods, by the destructive fibre orientation analysis, and by the mechanical properties measurement were finally compared to each other in order to assess the merits of the retained NDE methods. Correlations were found between actual fibre orientation distribution, mechanical properties of the materials, and NDE results.

6. **ACKNOWLEDGEMENTS**

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