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Measurement of residual stresses in CFRP laminates by X-ray diffraction method

C BALASINGH* and VANDANA SINGH†

Materials Science Division, National Aerospace Laboratories, Bangalore 560017, India

†Present address: LEOS, ISRO, Peenya, Bangalore 560058, India

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Abstract. The fabrication of CFRP laminates from prepregs involves curing at elevated temperatures. Residual stresses are set up due to the difference in thermal expansion coefficient between the matrix and the fibre. In this investigation, the X-ray diffraction method is used to measure the curing stresses in CFRP laminates by incorporating a very fine layer of aluminium particles during the lay up of the laminate. A calibration procedure is followed to correlate the strain in the crystalline particles, as measured by X-rays, with the composite strain and stress. Curing stresses measured by this technique are quite close to the value calculated from the differential coefficient of thermal expansion.

Keywords. CFRP laminates; residual stresses; X-ray diffraction.

1. Introduction

The fabrication of composite laminates involves curing at fairly high temperatures. Residual stresses are set up due to the difference in thermal expansion coefficients of the matrix material and the fibre used for reinforcement (Hahn and Pagano 1975; Jeronimidis and Parkhyn 1988). The curing stresses in certain graphite/epoxy laminates cause premature failure upon tensile loading or can be large enough to cause ply failure (Doner and Novak 1969) even in the absence of applied stress. Theoretical models have been developed for the calculation of stresses in laminates of different lay up sequences. However, experimental measurement of residual stresses in such systems is limited.

Residual stresses in laminates are of two types: (i) microstresses or pseudo–macrostresses (p–m stresses) in each of the constituent phases within a given ply. These arise because of the differences in thermal and moisture expansion between the phases and (ii) macrostresses in each ply, considered to be homogeneous, having anisotropic thermal and moisture expansion. These stresses arise because of the constraints by the neighbouring plies. The macrostresses have been experimentally measured using strain gauges, embedded in various plies during the lay up of the laminate, and from warping deflection measurements of the laminate. Laminated plate theories have also been used to calculate the macrostresses.

The purpose of this investigation is to use the X-ray diffraction method for the experimental determination of the p–m stresses in graphite/epoxy laminates. Although X-ray diffraction method has been very widely used for measuring residual stresses in crystalline materials, the method has not been used with polymer composites because the phases present do not give strong diffraction lines. However, in principle, it is possible to measure residual stresses in these systems, if small amounts of crystalline

*Author for correspondence

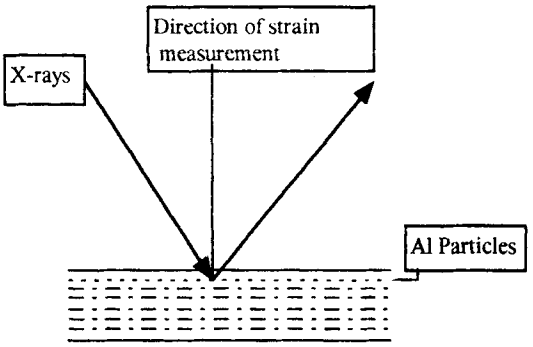

Figure 1. A seven-layered laminate showing the diffraction conditions.

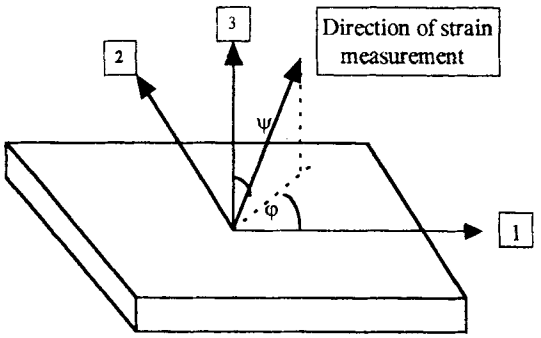

Figure 2. Direction of strain measurement with respect to the laminate axis.

particles with suitable diffracting characteristics are incorporated into the composite (Barrett and Predecki 1976; Predecki and Barrett 1979).

2. Principle of the method

A very fine layer of crystalline particles is uniformly sprinkled between two layers of prepreg plies in a unidirectional laminate during lay up. Figure 1 shows the crystalline particles in between the 6th and 7th layers in a 7 ply unidirectional laminate. The laminate is cured at the recommended temperatures and pressures. In the curing process, stresses develop in the matrix and the fibre, and some residual strain is transferred to the crystalline particles also. These strains in the crystalline particles are manifested as changes in the lattice planes of these particles. These changes in the lattice planes are measured by the X-ray diffraction method.

The interplanar spacings of high angle reflection lines are measured, and a comparison of these with the spacings in the unstressed state will yield strain values in the particles. If the unstressed spacing is indicated by d_u and that inside the laminate by d, the strain in the particles in the direction normal to the diffracting plane is

$$\frac{d - d_u}{d_u}.$$

The direction of strain measurement and the interplanar spacing $d_{\varphi\psi}$ is defined by the angles φ and ψ relative to the sample axes are shown in figure 2.

The generalized equation for the strain $e_{\varphi\psi}$ is given by

$$e_{\varphi\psi} = [\varepsilon_{11}\cos^2\varphi + \varepsilon_{12}\sin 2\varphi + \varepsilon_{22}\sin^2\varphi - \varepsilon_{33}]\sin^2\psi + \varepsilon_{33} + [\varepsilon_{13}\cos\varphi + \varepsilon_{23}\sin\varphi]\sin^2\psi, \quad (1)$$

where ε_{ij} are the strains. Direction 3 is taken as normal to the surface of the laminate. When referred to the principal axes, the above equation reduces to

$$e_{\varphi\psi} = [\varepsilon_1\cos^2\varphi + \varepsilon_2\sin^2\varphi - \varepsilon_3]\sin^2\psi + \varepsilon_3. \quad (2)$$

In unidirectional laminates the fibre direction is taken as $\varphi = 0$. When $\varphi = 0$, (2) becomes

$$e_{\varphi\psi} = \frac{d_{0,\psi} - d_u}{d_u} = \varepsilon_{1p} + \varepsilon_{3p}\cos^2\psi$$

or

$$\varepsilon_{1p} = \frac{d_{0,\psi} - d_u}{d_u}\frac{1}{\sin^2\psi} - \varepsilon_{3p}\cot^2\psi \qquad (\psi \neq 0). \quad (3)$$

Similarly when $\varphi = 90°$, we get

$$\varepsilon_{2p} = \frac{d_{90,\psi} - d_u}{d_u}\frac{1}{\sin^2\psi} - \varepsilon_{3p}\cot^2\psi, \quad (4)$$

and

$$\varepsilon_{3p} = \frac{d_{\varphi,0} - d_u}{d_u}, \quad (5)$$

ε_{1p}, ε_{2p} and ε_{3p} refer to strains in the crystalline particles in the principal directions.

The principal stresses σ_{1p}, σ_{2p} and σ_{3p} in the particles can be calculated from isotropic elasticity expressions. (The filler particles are assumed to be elastically isotropic). Since the strains and stresses in the particles are not the same as in the composite, the method is calibrated by comparing the measured particle strain ε_{1p} with the composite strain ε_1^* obtained with strain gauges.

3. Experimental

3.1 *Laminate*

The laminates were made from T300 graphite/epoxy prepregs imported from Hexcel Composites, Duxford, UK. The weight percentage of the fibre in the prepregs is between 67 and 69%. These prepregs are being used to fabricate aircraft components. The procedure followed in making the laminates for this investigation is similar to the one followed in fabricating the aircraft components. Unidirectional CFRP laminates of size 25 cm × 30 cm were made by stacking seven layers of prepregs. For each laminate, seven pieces of 25 cm × 30 cm prepregs were cut and the lay up done in a clean room. The cut prepregs were laid one above the other on an aluminium plate coated with a releasing agent. On the sixth layer of prepreg a very fine layer of aluminium powder was sprinkled. Care was taken to spread the powder as uniformly as possible. The seventh layer of prepreg was laid and another aluminium plate coated with releasing agent placed on top. Loads were applied and the curing done at 130°C for 30 min

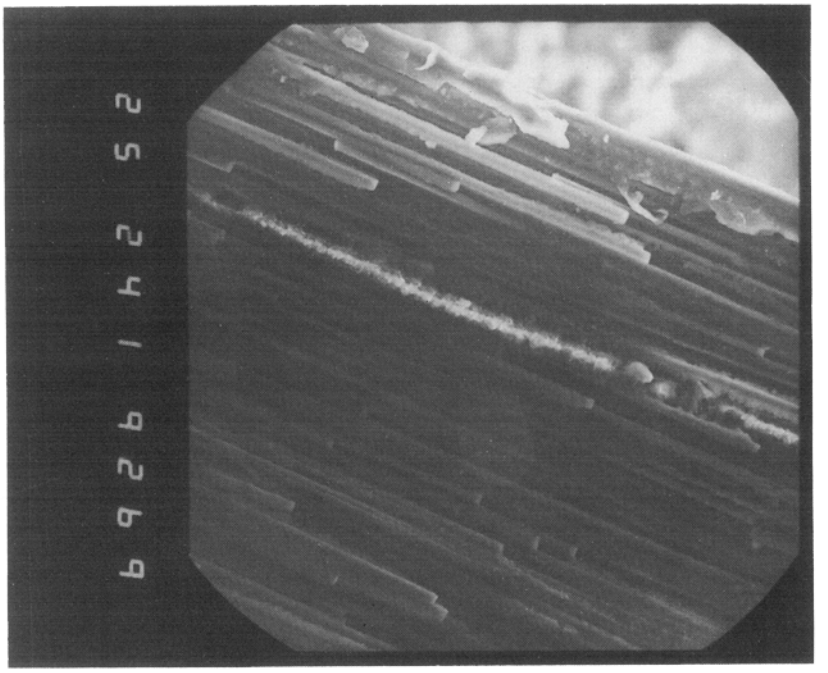

Figure 3. Cross-section of unidirectional carbon fibre/epoxy laminate embedded with Al particles.

followed by 175°C for 2h. The thickness of the laminate was 1·1 mm. One laminate was cured in an oven and the other in an autoclave. In both cases the curing sequence of temperature and pressure were the same (pressure 90 psi ~ 0·62 MPa).

During curing, some of the aluminium particles bleed out with the resin in the plane of the lamina. But, as seen from the SEM photograph in figure 3. there is a negligible migration in the transverse direction. The photograph also shows that the aluminium crystalline particles are almost uniformly spread, and it is a very thin layer.

3.2 *Tensile testing*

For tensile testing, specimens of 140 mm length and 15 mm width were cut from the laminates. Tapered end tabs (from aluminium sheet of 1·5 mm thick) of 45 mm length were bonded before testing. Two strain gauges were fixed in the axial direction on either side of the tensile specimen. The testing was done in an INSTRON machine and the strains measured using a BLH Strain indicator. From each laminate two samples were subjected to tensile testing. The average Young's Modulus of the laminate is 127 GPa.

3.3 *Loading jig*

The strains and stresses in the crystalline particles are not the same as those in the surrounding resin. Hence to get the strain in the matrix from a measurement of strain in the crystalline particles a calibration procedure is adopted. Calibration is done by

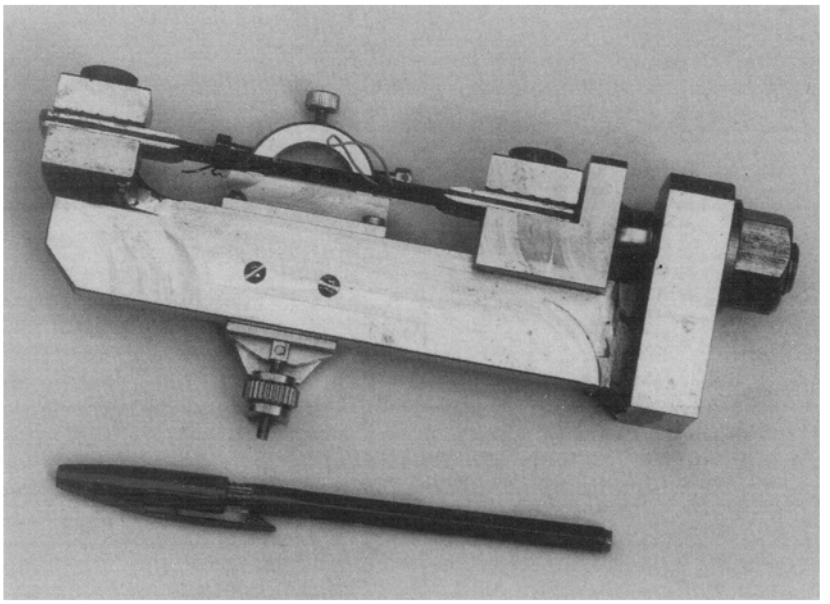

Figure 4. Loading jig.

applying known loads to the composite and measuring the strains produced in the aluminium particles by the X-ray diffraction technique. For *in situ* loading, a loading device, which can be mounted on the specimen shaft of a Philips Vertical Goniometer, was designed and fabricated. The loading device, shown in figure 4, can apply loads up to 640 kg. The jig was tested on 20 mm wide laminates up to a strain of 2500×10^{-6}. In the fixture used to mount the loading device on the shaft, there is a provision to adjust the level of the specimen, so that any desired layer can be brought on the goniometer axis. Very fine adjustment is possible. This provision is necessary to ensure that the embedded layer of crystalline particles is on the axis of the goniometer.

4. Results and discussion

4.1 *Calibration curve*

For calibration purpose, specimens of length 135 mm in the fibre direction and 20 mm width were used. The loading jig requires specimens with holes at both ends. These holes were made by the ultrasonic method. Aluminium end tabs with matching holes were fixed on both ends. A strain gauge was fixed in the fibre direction (axial) to the side opposite to that exposed to X-rays.

A Philips Vertical Goniometer with provision to set inclination angles ψ was used. The loading jig was mounted on the goniometer shaft using the fixture, which was adjusted to bring the layer of aluminium particles on the axis of the goniometer. Cobalt Kα radiation was used, which gives the Al(420) reflection at about 162° 2θ. Loads were applied to produce composite strains in steps of 100 or 200×10^{-6}, and the corresponding strains in the aluminium particles measured at $\varphi = \psi = 0$ and $\varphi = 0, \psi = 45°$.

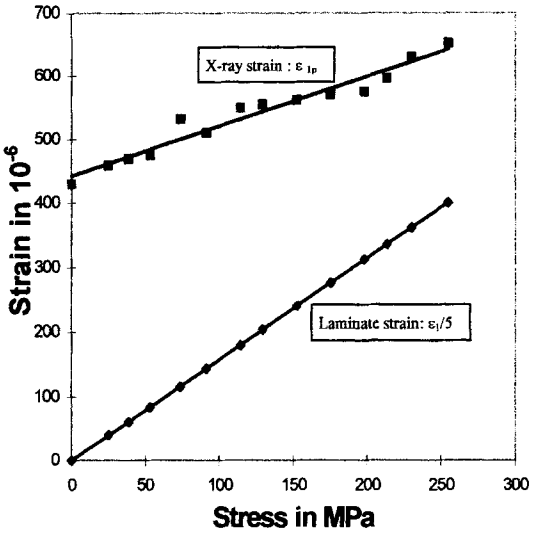

Figure 5. Variation of particle strain (as measured by X-rays) and laminate strain with applied stress.

Table 1. Residual strain in Al particles (in 10^{-6}).

Sample No.	ε^r_{1p}	ε^r_{2p}	ε^r_{3p}
1	492	−337	−610
2	516	−385	−587
3	560	−370	−618
4	520	−332	−592
5	551	−347	−620
6	542	−391	−602
Average	530	−360	−605

The X-ray strain should be proportional to the composite strain up to the yield point in the crystalline particles. The initial set of experiments showed scatter. This necessitated repeated loading and the use of a number of specimens. Counting time was also increased to decrease the statistical errors. The best set of results is shown in figure 5. It is seen that as expected, the X-ray strains in the particles increase linearly with the applied stress in the composite.

4.2 *Curing stresses*

For the determination of curing p–m stresses in the laminate, coupons of size 4 cm × 4 cm were cut from the laminate. X-ray strain measurements were done at $\varphi = \psi = 0$; $\varphi = 0$, $\psi = 45°$ and $\varphi = 90°$, $\psi = 45°$. The three principal strains in the particles ε^r_{1p}, ε^r_{2p} and ε^r_{3p} are calculated using (3), (4) and (5). The values of the particle strains obtained from six samples are listed in table 1. The average value is also given. If these average strain values are converted into stress values using $E = 71$ GPa and

$\nu = 0{\cdot}343$, we get the residual stresses in Al particles as $\sigma^r_{1p} = 8$ MPa, $\sigma^r_{2p} = -33$ MPa and $\sigma^r_{3p} = -51$ MPa.

The residual strain in the resin matrix can be found by replotting the particle strain ε_{1p} as a function of the composite strain ε^*_1 from the calibration curve. The particle strain ε^r_{1p} of 530×10^{-6} corresponds to a matrix residual strain of 5337×10^{-6}. This observed matrix strain can be compared with the value of ε^r_{1m} calculated using differential coefficient of thermal expansion.

$$\varepsilon^r_{1m} = (\alpha_m - \alpha_f)\Delta T,$$

where α_m is the coefficient of thermal expansion of the resin, equal to $45 \times 10^{-6}/°C$, α_f coefficient of thermal expansion of carbon fibre in the fibre direction, equal to $-1{\cdot}8 \times 10^{-6}/°C$ and ΔT is taken as 152°C.

Using these values we get $\varepsilon^r_{1m} = 7114 \times 10^{-6}$. Compare this value of 7114×10^{-6} with the experimentally obtained value of 5337×10^{-6}. The experimentally obtained value is quite close to the theoretically calculated value. Assuming the resin modulus to be 3·54 GPa the experimentally obtained residual strain corresponds to a stress of 18 MPa. The theoretically calculated strain of 7114×10^{-6} corresponds to a stress of 25 MPa. Calculation of residual stress in these systems by a finite element method (Predecki and Barrett 1979) gave a matrix residual stress of 26 MPa.

The slight discrepancy between the experimentally obtained value and theoretically calculated may be attributed to: (a) many particles are present in the 1, 2 plane and quite likely the development of full residual stress is inhibited and (b) also $\varepsilon^r_{2p} \neq \varepsilon^r_{3p}$, which should be the case if the matrix is unaffected by the presence of the crystalline particles. It is to be noted that the unidirectional laminates containing no filler particles are transversely isotropic.

The p–m stresses in the fibres in the fibre direction will be compressive. In a two-phase material, the p–m stresses should satisfy the equilibrium equations (Abuhasan *et al* 1990)

$$(1-f)^{\mu}\langle\sigma_{11}\rangle^{\alpha} + f^{\mu}\langle\sigma_{11}\rangle^{\beta} = 0,$$

where f is the volume fraction of the second phase. The p–m stresses in the fibres can be found from the equilibrium equations, if the volume fractions are known.

The measurements can be improved by using fewer crystalline particles. However, the use of fewer particles is beset with many practical difficulties. When the number of crystalline particles is reduced the diffraction intensity decreases. This necessitates very long counting time to get appreciable diffracted X-ray intensity. The long term stability of the X-ray diffractometer system has also to be taken into account.

5. Conclusion

The X-ray diffraction method can be used to measure residual stresses in polymer laminates by measuring the strains in the crystalline particles incorporated between prepreg layers during lay-up. The experimentally determined value is quite close to the theoretically calculated value. The slight discrepancy may be due to the perturbation caused by the presence of the particles themselves. The non-destructive XRD method could be very effectively used to monitor the changes in residual stresses in the laminates caused by factors like moisture absorption.

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