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The time-dependent mechanical properties of fibre reinforced polymers

- by -

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A report of work carried out during the period 1st October, 1967 to 31st March, 1968

Contents

Nomenclature

c_{ij}	elastic stiffnesses	Matrix notation where i, j are integers taking the values 1, 2, 3, 4, 5, 6.
s_{ij}	elastic compliances	
σ_i	stress	
ϵ_i	strain	
E	Young's modulus	
ν	Poisson's ratio	
k	Bulk modulus	
v	volume fraction	
α_i, β_i	Fourier coefficients, i are integers	
$f(\theta)$	distribution of fibre orientation	
t	time	
p	transformed variable	
S_i	magnitudes of discrete retardation spectrum	
γ_i	associated retardation times	
$\underset{\sim}{C}$	stiffness matrix.	

suffixes

f	fibrous phase
m	matrix phase

superfixes

U	upper limit of compliance
L	lower limit of compliance

A tilde, ~, below a letter denotes a matrix.

A circumflex, ∧, above a letter denotes a Laplace transform.

1. General Progress

The purpose of this project is to measure the time-dependent mechanical properties of composite materials formed from a matrix, having time-dependent properties, reinforced with elastic fibres and to compare these with theoretical predictions calculated from certain basic assumptions.

The matrix material to be used is an epoxy resin of the Araldite range and the reinforcing fibres will be continuous strands of E-glass. Consequently the matrix material will be taken to be isotropic and any anisotropy of the composite materials will be due to the orientation of the fibres. The time-dependence of the mechanical properties of the resin can be described by a knowledge of the creep properties and the behaviour of the creep contraction ratio. The E-glass, presuming that the stress-strain relationships are independent of time, can have its relevant mechanical properties fully described by its Young's Modulus and Poisson's Ratio. In order to fully characterise these two materials, therefore, it is necessary to carry out a set of creep tests on the resin at several stresses and measure the variation of longitudinal extension and perpendicular contraction with time. The Young's Modulus and Poisson's Ratio of the glass are not easy to measure and it is initially proposed to take their values from the manufacturers literature.

In order to find out how the two phases interact it is necessary to orientate the fibrous phase according to different distributions and measure the resulting mechanical behaviour under stress. If we define an 'ideal' fibre-reinforced composite as one in which the fibres are straight and continuous throughout the system and which contains no voids or inclusions, then the simplest 'ideal' composite to produce is one in which the fibres are all aligned in one direction. Consequently the first samples of fibre reinforced composites will be of this uniaxially reinforced type.

During the six months from October 1967 to March 1968, apparatus for producing these simple specimens has been developed and tested as has a tensile creep testing machine on which they can be subjected to a uniaxial stress and the resulting extensions measured. Alongside this apparatus development, theoretical work to calculate the limits, within which the creep of composite materials lie, has taken place.

2. Apparatus Design

2.1 Tensile Creep-Testing Machine

The tensile moduli of the materials to be tested are likely to range between 0.3 and 4×10^6 psi and in order to produce extensions large enough to be measured accurately, stresses must be applied to the specimens that are larger than can conveniently be applied using a direct loading system. Consequently it has been decided to use a lever arm with a ratio of 10:1 to apply the stress to the specimens.

The tensile creep-testing machine that has been designed (see Fig. 1) is sufficiently strong and rigid to allow a load of 1000 lb to be applied to the specimen without any undue deflection of the machine itself. The main framework is formed from 4″ x 2″ mild steel channel, machined to ensure that the loading system can be perfectly aligned. The lever arm itself is fabricated from a piece of 'T' section mild steel tapering away from the fulcrum attached to a solid mild steel block, housing the roller bearings. This particular form was chosen as giving small lateral instability and vertical deflection without being excessively heavy.

It is important that the system is designed so that the load is applied along the geometric axis of the specimen. Several defects can cause non-axiality of the load. These are:-

i) Large friction forces in the bearings supporting the specimen assembly which may cause rotation of the links as the lever arm rotates. These have been prevented by the use of a suitable type of roller bearing in which the torque required to rotate the bearing is less than .05 lb in.

ii) The specimen might not be correctly lined up in each jaw and a bending moment could be transmitted to the specimen. This has been prevented by pushing a thin rod through the centre of each jaw and the specimen and applying a light tension to the specimen to line it up before tightening the jaws. As the holes in the specimen are on the centreline of the specimen, this procedure also ensures that the specimens are gripped on the centreline of the jaws.

iii) The possibility that the gripping faces of the jaws might not be at rightangles to the pin through the jaws could cause a bending moment in the specimen but this is prevented by the machining procedure.

It is possible that there may be some small variation in the initial length of the specimens due to shrinkage effects and it is consequently necessary to be able to raise or lower the position of the fulcrum. In order to allow for this movement, the fulcrum is supported by a yoke which can be raised or lowered by a threaded rod.

The bottom jaw assembly is mounted on a slide so that it can be moved directly beneath the upper jaw assembly and locked in position.

In order to ensure that the specimens start in a stress free condition there is a counterweight which can be adjusted until there is no load in the specimen.

2.2 Longitudinal Extensometer

The moduli of the specimens to be tested are sufficiently high for an extensometer to be attached directly to the specimen without causing any large deflections of the specimen. The sensitive element of the extensometer is a Sogenique linear displacement differential capacitance transducer having a travel of 0.1". These can read the separation of the two ends of the gauge length to within 0.00001" and consequently over a 3" gauge length, the resulting strain can be measured to within 1% if the minimum strain measured is 0.03%.

The extensometer designed to house this transducer is shown in Fig. 2 and is of the direct reading type to eliminate any possible friction between moving parts. It is clamped to the specimen at each end of the gauge length by means of two silver steel rods sliding in a slot and held together by two Allen screws. These clamps automatically sit at right angles to the specimen whereas if knife edges had been used instead of the round bars this would have been difficult to achieve. In order to ensure that the gauge length is always the same, the clamps are fastened to the specimen using a simple jig which also ensures that the clamps are at right angles to the axis of the specimen.

2.3 Filament Winding Machine

The purpose of the filament winding machine is to produce flat plates of uniaxially reinforced epoxy resin from which specimens may be cut.

The most convenient form in which continuous 'E'-glass fibres are produced commercially is that of rovings. These consist of several parallel strands each composed of 204 individual filaments, bundled together but not twisted. As the main difficulty associated with filament winding is the complete impregnation of the rovings with resin, it was decided to use a roving with as few strands as possible. The roving being used is produced by Fibreglass Ltd. and consists of 12 strands.

The principle of the machine is very simple. The glass is wound off its cheese, through a resin bath and onto a mandrel where it is left while the resin cures. In order that the fibres shall be laid down a constant distance apart on the mandrel, the resin bath is moved slowly along a screw thread at a constant rate. The processes through which the fibres pass are shown pictorially in Fig. 3. The fibres are pulled through the resin bath and off the cheese by the mandrel which is rotating at 16 rpm. They are wound off the cheese and through a guide into the traveller containing the resin bath. In order to try and separate the strands the fibres pass over a serrated roller in the resin and expose a large surface area to the resin. On leaving the resin the fibres pass through a cone leading to a $^1/_{16}$" hole to remove most of the excess resin from the surface of the fibres. In order that the glass-resin ratio can be varied simply, the fibres then pass between two rollers which are held together with a variable pressure. The screw thread on which the resin bath assembly travels is driven by a variable speed motor so that the spacing of the fibres on the mandrel can be varied. Fig. 4 shows a general view of the machine.

During preliminary tests on the performance of this machine it has been found that a considerable amount of air remains trapped between the fibres in the final laminate, and variation of the production parameters do not change the amount of air present noticeably. It has been discovered that if a high local pressure is applied to a bubble rich area that the bubbles are completely squeezed from the area. Consequently the mandrel is removed from the winding machine after winding has been completed and while the resin is still in the liquid state and it is passed several times betwen two rubber rollers, whose distance apart can be varied. When a high pressure is exerted the air foams out of the laminate in front of the rollers and the resulting laminate can be made air free. Figs. 5A and 5B show typical laminates that have been prepared. The first shows the amount of air present if the laminate is not rolled while the second shows the total absence of air in a rolled laminate.

As the winding procedure takes about an hour to place one layer of glass onto the mandrel, the resin must be of a type that is slow to gel. Also if the resin is of a low viscosity the air is more easy to remove from the system. Consequently Araldite MY753 with HY951 hardener was chosen as being the most suitable system as it has a gel time of $2\frac{1}{2}$ hours at room temperature and a viscosity of less than 20 poises.

To ensure that it is not difficult to remove the laminate from the mandrel when it has been allowed to cure for a day, the mandrel is formed from a polypropylene sheet, $^{3}/_{8}''$ thick, which is spring loaded in position. The epoxy resin does not adhere to the polypropylene and so no release agent is required.

2.4 Specimen Preparation

It is important that the specimens produced to be tested should have a parallel sided gauge length that is symmetrical about the centre line, and that the centre line should pass through the centres of holes drilled at either end of the specimen.

Using a template, two holes are first drilled through the laminated sheet of material to be tested. These define the centre line of the specimen and it must be ensured that this is at the required angle to the direction of the fibres. A template has been made that defines the shape of the specimen and is fastened to the sheet of material through the two holes that have been drilled.

Using a Tensilkut router with a diamond impregnated cylindrical cutting tool, rotating at 20,000 rpm, the specimens are cut out around the template. The harmful glass and resin dust is removed from the immediate region of the tool by the use of a vacuum head situated above the tool.

After the samples have been cut out of the sheet, they are placed in a constant temperature oven at 50°C to cure until all the attendant shrinkage has ceased.

3. Theoretical Predictions

3.1 Introduction

The stiffness of a composite material depends both on the geometry of the structure of the material and on the stiffness of the component phases. The composite materials under consideration here are fibre reinforced linear viscoelastic materials and consequently any analysis of their stress-strain characteristics must take the geometry and orientation of the fibrous phase into account as well as the time dependence of the matrix.

Cox (1951) has analysed a mat of ideal fibres, assuming that those fibres have no flexural stiffness and that in consequence they can only transmit loads in tension. He characterises the orientation of the fibres in the mat by a distribution function. This represents the number of fibres at a given angle to a specified direction in a unit width perpendicular to their axial direction. The assumptions made by Cox seem valid in the context of a mat with no means of interconnection between the fibres. Using this analysis Arridge (1963) has combined an ideal fibrous mat with an elastic matrix by assuming that the strains in the two phases are equal. These principles of Cox and Arridge have been extended to allow for the matrix material being linearly viscoelastic by Dootson (1968) who has obtained Volterra integral equations relating the creep compliance of a composite to the geometry and stiffness of the two phases. These equations have been solved using several techniques (see Mikhlin (1964)) and the calculated compliances compared with the experimentally obtained compliances of several glass fibre/ polyester resin systems. In a composite material it seems likely that, due to the connection between the fibres, the fibres affect the stiffness of the whole other than axially. Bishop (1966) has tried to overcome this by introducing two hypothetical lateral fibres to act in conjunction with each fibre. While this artifice can be used empirically to improve predictions of the mechanical properties of the composite, it is not very satisfactory from a theoretical point of view.

In this work it is intended to use a more rigourous elastic analysis, based on the variational principles used by Hashin and Rosen (1964), to calculate the five elastic stiffnesses required to characterise a simple composite element. Summing these using a distribution function in the same way as Cox has done, the elastic solution for a fibre reinforced composite can be obtained.

Using the correspondence rule, by which a viscoelastic problem is associated with an elastic one, that has been proved by Biot (1954) for the general anisotropic case this elastic solution can be used to yield the viscoelastic solution required. This technique is explained by Williams (1964) who suggests that the complicated transform inversion involved can be bypassed by an approximation method such as the collocation method proposed by Schapery (1962).

3.2 Moduli of a representative composite element

In order to analyse the elastic behaviour of a composite material it is necessary to assume a mode of combination of the component phases. Previous assumptions that have been made have assumed that the fibres have no flexural stiffness and that the two phases undergo equal strains but have no interaction with each other. Calculations made on the basis of these assumptions would be expected to be in error for values of the shear stiffness and the tensile stiffness, perpendicular to the fibre axis of the composite.

To try and eliminate these errors we will consider a fibre reinforced composite as being formed from a number of representative composite elements. Each of these is composed of many long parallel fibres embedded in a cylinder of resin with its axis parallel to the direction of the fibres. Both phases are also assumed to be isotropic and homogeneous.

Hashin and Rosen (1964) have derived the macroscopic elastic constants for elements of this kind by using variational principles. As these elements exhibit transverse isotropy only five elastic constants are needed to fully characterise their stress-strain relationship. The stress-strain relationship for an element can therefore be written as

$$\begin{bmatrix} \sigma_1 \\ \sigma_2 \\ \sigma_3 \\ \sigma_4 \\ \sigma_5 \\ \sigma_6 \end{bmatrix} = \begin{bmatrix} C_{11} & C_{12} & C_{12} & & & \\ C_{12} & C_{22} & C_{23} & & & \\ C_{12} & C_{23} & C_{22} & & & \\ & & & (C_{22}-C_{23})/2 & & \\ & & & & C_{66} & \\ & & & & & C_{66} \end{bmatrix} \cdot \begin{bmatrix} \epsilon_1 \\ \epsilon_2 \\ \epsilon_3 \\ \epsilon_4 \\ \epsilon_5 \\ \epsilon_6 \end{bmatrix} \qquad (1)$$

in the usual matrix notation taking 01 as the direction of the symmetry axis (see Hearman (1961)).

3.3 Extension to a complete composite

The elastic constants of the representative composite element form a fourth rank tensor and consequently obey the tensor transformation laws as described by Hearman (1961). If we rotate the composite element through an angle θ about the 3-axis the elastic constants become

$$
\underset{\sim}{C}(\theta) = \begin{bmatrix}
C(\theta)_{11} & C(\theta)_{12} & C(\theta)_{13} & & & C(\theta)_{16} \\
C(\theta)_{12} & C(\theta)_{22} & C(\theta)_{23} & & & C(\theta)_{26} \\
C(\theta)_{13} & C(\theta)_{23} & C(\theta)_{33} & & & \\
 & & & C(\theta)_{44} & C(\theta)_{54} & \\
 & & & C(\theta)_{45} & C(\theta)_{55} & \\
C(\theta)_{16} & C(\theta)_{26} & C(\theta)_{36} & & & C(\theta)_{66}
\end{bmatrix} \qquad (2)
$$

where the relationships between $C(\theta)_{ij}$ and C_{ij} are described in Hearman (1961). The stress-strain relationship for a cylindrical composite element oriented at an angle θ to the 1-axis in the 1,2 plane is thus

$$
\underset{\sim}{\sigma} = \underset{\sim}{C}(\theta) \cdot \underset{\sim}{\epsilon} \qquad (3)
$$

If we now consider a composite formed from these elements we have to add the effects of these elements orientated at various angles to the 1-axis Let us define an orientation distribution function $f(\theta)$ which describes the number of fibres oriented at any angle θ to the 1-axis. By assuming that these elements are subjected to a uniform strain field we obtain as the stress-strain relationship for the composite

$$
\underset{\sim}{\sigma} = \int_0^\pi \underset{\sim}{C}(\theta) \cdot f(\theta)\, d\theta \cdot \underset{\sim}{\epsilon} \qquad (4)
$$

Alternatively we may make the dual assumption that the elements are subjected to a uniform stress field which gives

$$
\underset{\sim}{\epsilon} = \int_0^\pi \underset{\sim}{C}^{-1}(\theta) \cdot f(\theta) d\theta \cdot \underset{\sim}{\sigma} \qquad (5)
$$

as the stress-strain relationship. These alternative assumptions give as the upper and lower bounds to the stiffness matrix

$$
\underset{\sim}{C}^U = \int_0^\pi \underset{\sim}{C}(\theta) \cdot f(\theta) d\theta
$$

$$
(\underset{\sim}{C}^L)^{-1} = \int_0^\pi \underset{\sim}{C}^{-1}(\theta) \cdot f(\theta) d\theta \qquad (6)
$$

As the distribution function, $f(\theta)$, is periodic it can be written as a Fourier series

$$\pi f(\theta) = 1 + \alpha_1 \cos 2\theta + \alpha_2 \cos 4\theta + \dots + \beta_1 \sin 2\theta + \beta_2 \sin 4\theta + \dots \quad (7)$$

where further terms do not affect the stiffness matrix. We can therefore write the two bounds to the stiffness matrix in terms of α_1, α_2, β_1, β_2, and the five elastic constants for the composite element. These in turn are functions of the Young's Modulus and Poisson's Ratio of the two phases as well as the volume fractions.

3.4 Solution for a fibre reinforced linear viscoelastic composite

Biot (1954) has proved for the general anisotropic case that any visco-elastic problem can be associated with the corresponding problem where all the components are elastic. Using this correspondence principle in conjunction with the elastic solution that has already been obtained we can obtain the solution for the viscoelastic case. The method depends on transforming the equilibrium, compatability and boundary conditions with respect to time and thus obtaining an associated set of equations in the transform plane in terms of the transformed variable, p. Having solved these associated equations, the final step involves the inversion of the transformed solution back to real time.

Before obtaining this solution, it is necessary to define the time-dependent behaviour of the matrix material in general terms. First let us consider a general description of a creep curve. Dootson (1968) has shown that the use of a discrete spectrum of retardation times is probably the best general solution with which a high degree of accuracy can be obtained. This gives the general creep curve as

$$\epsilon_m(t) = \left\{ S_o + \sum_{i=1}^{n} S_i (1 - e^{-t/\gamma_i}) \right\} . \sigma_m \quad (8)$$

where the γ_i are the chosen retardation times and the S_i are the associated values of the spectrum. Secondly it is necessary to define the behaviour of the Poisson's Ratio of the matrix material. Turner (1966) has suggested that the assumption that the Bulk modulus is independent of time gives an acceptable approximation to the Poisson's Ratio, and this assumption will be used here initially.

If the elastic solution was

$$\underset{\sim}{\sigma} = \underset{\sim}{C} \cdot \underset{\sim}{\epsilon} \quad (9)$$

then the associated solution becomes

$$p . \hat{\underset{\sim}{\sigma}}(p) = p . \hat{\underset{\sim}{C}}(p) \cdot p \hat{\underset{\sim}{\epsilon}}(p) \quad (10)$$

where $\hat{\underset{\sim}{C}}(p)$ is known in terms of the transformed modulus, $\hat{E}_m(p)$, and the transformed Poisson's Ratio, $\hat{\nu}_m(p)$. If we use the description of the matrix material discussed above, then these become

$$p.\hat{E}_m(p) = \left\{ S_o + \sum_{i=1}^{n} S_i \left(\frac{1}{1+p\gamma_i} \right) \right\}^{-1} \tag{11}$$

and

$$p.\hat{\nu}_m(p) = \frac{1}{2} \left\{ 1 - \frac{p}{3K_m} \cdot \hat{E}_m(p) \right\} \tag{12}$$

If we wish to calculate the strain response to a given stress input we must first invert the matrix of the transformed stiffnesses and then take the inverse transform of the resulting expression. To invert these transforms exactly requires the use of either transform tables or of a formal inversion, both of which are much too complicated for this particular case. Let us consider how this inversion can be effected for the particular case where the stress is applied as a step input and the composite creeps. If we assume that the creep curve of the composite can be described by an expression of the same form as is used in equation 8, we can transform this expression into the p plane and equate it to the transform to be inverted. Using a linear regression technique to find the optimum values of the constants to provide the best fit we thus obtain the 'viscoelastic constants', describing the composite behaviour.

This solution is not complicated mathematically but a large quantity of numerical work arises that cannot be managed manually. A computer program is consequently being produced that will be able to calculate the time-dependent behaviour of any planar fibre reinforced composite from a knowledge of the volume fractions of the two phases, the Young's Modulus and Poisson's Ratio of the fibres, and the values of S_i and γ_i for the matrix.

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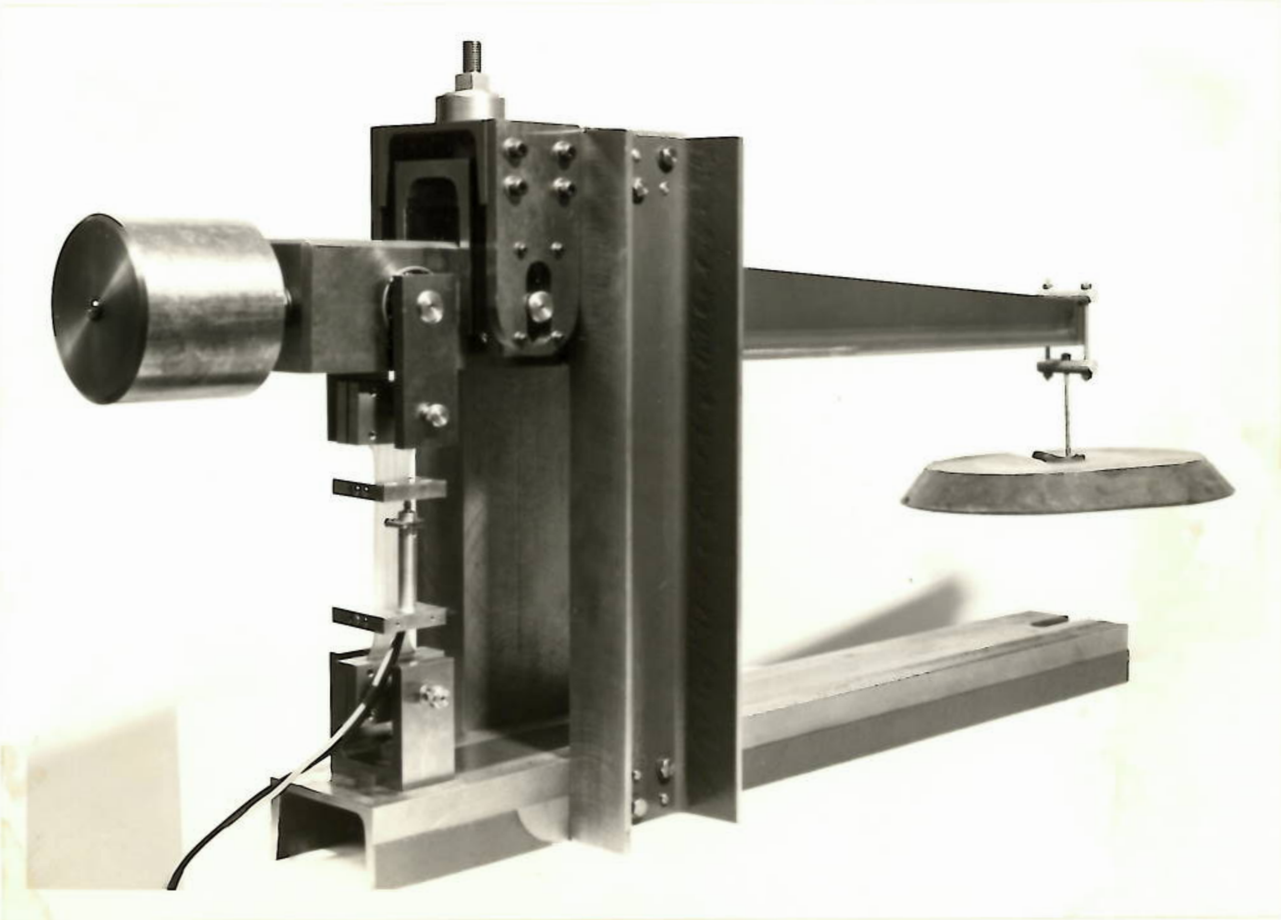

Figure 1 Tensile Creep-Testing Machine

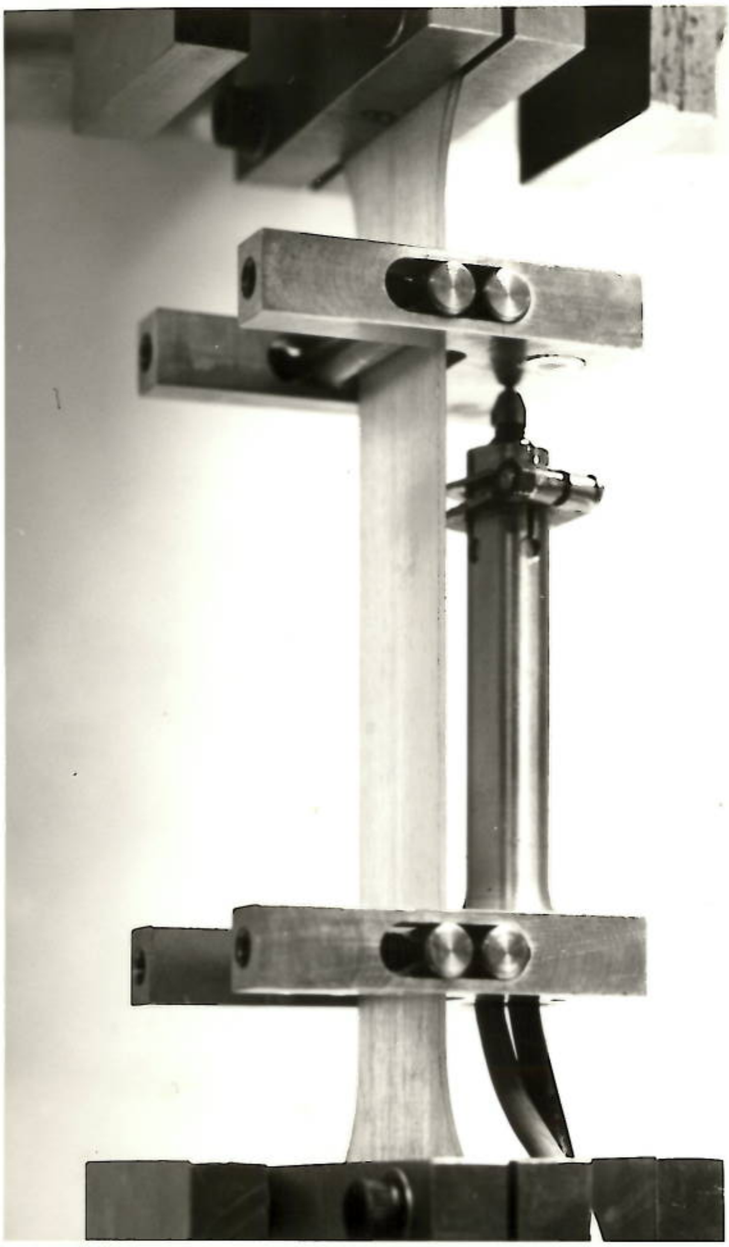

Figure 2   Longitudinal Extensometer

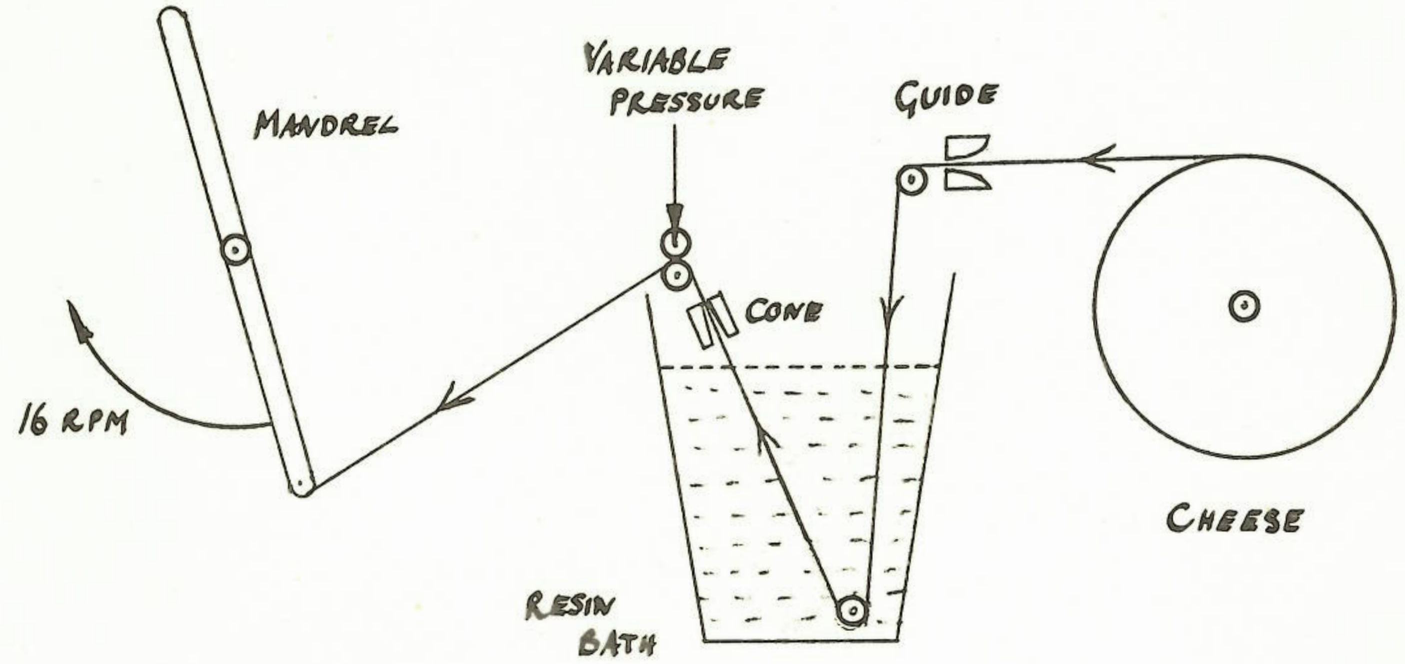

Fig. 3 PRINCIPLES OF FILAMENT WINDING MACHINE

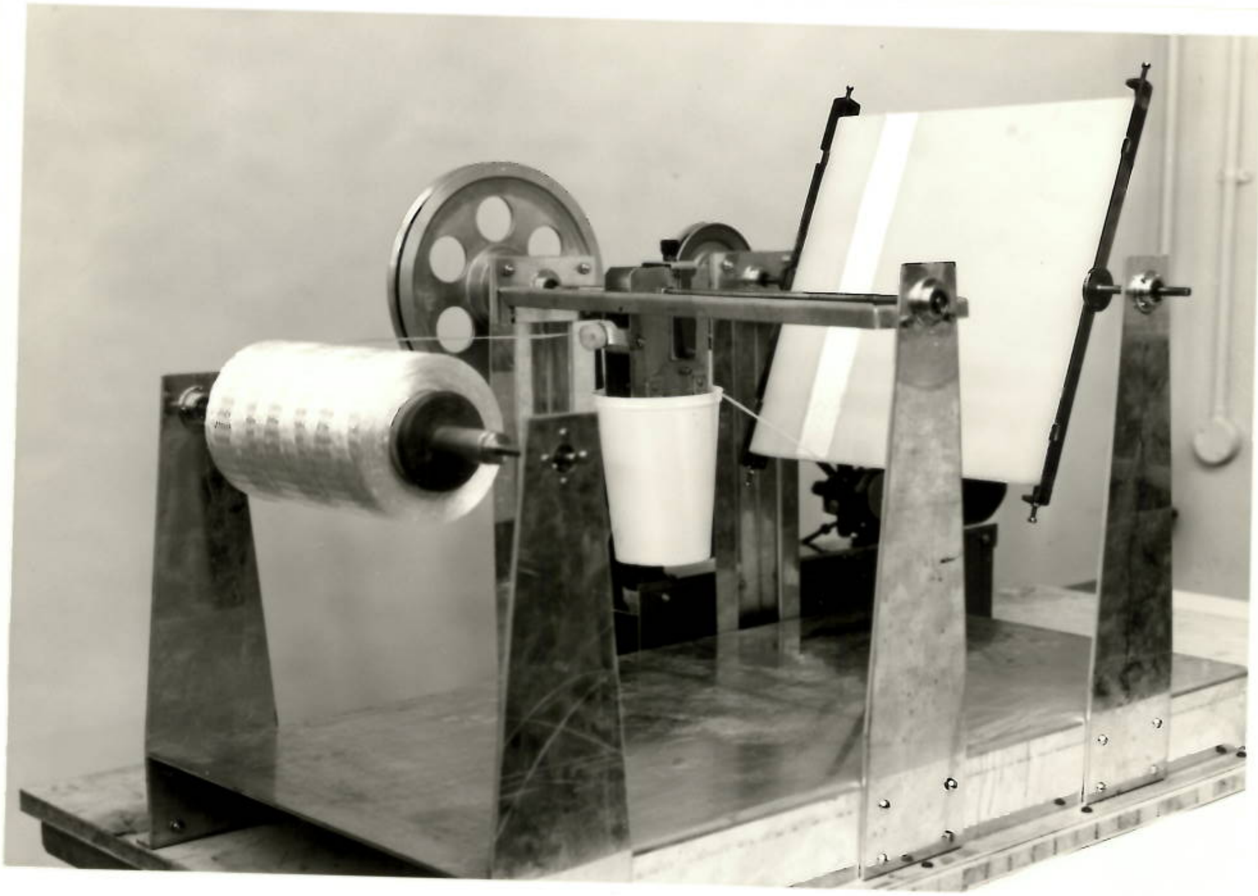

Figure 4   Filament winding machine

Figure 5A   Composite before rolling (x 120)

Figure 5B   Composite after rolling (x 120)