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Preface

When thermoplastics containing short fibres were first introduced onto the market it was with the intention of producing a range of new materials possessing properties that were intermediate between the high tonnage commodity plastics and the sophisticated continuous fibre reinforced composites, well established in the aerospace industry. The increase in stiffness and strength of the short fibre composite compared to the parent thermoplastic was modest but nevertheless sufficient to enable this class of material to penetrate into lightly stressed engineering applications. However during the last few years there have been some significant advances. We have seen the emergence of the very high melting point thermoplastics together with a gradual reduction in the costs of the specialist fibres such as carbon. Material manufacturers are now combining engineering thermoplastics with these more expensive fibres to produce a new range of products having properties that are approaching those of the traditional long fibre composites. There is still a long way to go, however, and significant improvements in materials design and fabrication technology are needed in order to optimize these developing reinforced thermoplastics.

This book has been written primarily for the plastics industry and describes some of the concepts on which short fibre reinforcement are based and which can be used to develop products having specified properties. Since it is the intention that this series of books concerned with Polymer Engineering should be fairly concise, it is impossible to give both depth and breath to the subject, in the space available. As far as the present book is concerned, it was decided to restrict the discussion to mechanical properties and not to give details of commercially available materials and processing equipment. The latter information, if given, would have been rapidly out of date and is best sought directly from the appropriate manufacturers. The result is an attempt to concentrate on the principles of short fibre reinforced thermoplastics. To this end, only a minimum of mathematics has been used - enough to develop the subject quantitatively but not sufficient, I hope, to deter the reader from exploring the book beyond this preface.

M. J. Folkes

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Contents

CHAPTER 1

Introduction

While Leo Baekeland recognized that wood flour was an essential additive in phenolic moulding powder formulations, this and other fillers were not considered then as essential ingredients for thermoplastics. As long as thermoplastics were inexpensive and plentiful, there was little enthusiasm for the idea of producing filled grades, at least before the OPEC price increases. However, such a need became apparent in the mid 1970's and so it is not surprising to read that about 1 million tonnes of filler were used by the American plastics industry in 1980 with more than 10% of this being used in thermoplastics. The range of fillers available is very large, extending from the mineral and inorganic fillers to the more expensive fibres such as glass, carbon and Kevlar. In principle there is really no limitation on the number of possible permutations of matrix and filler that can be combined to produce a composite. However, only a limited number have remained after the initial and extensive research and development efforts by many companies. Commercially available filled thermoplastics appear to fall into two broad categories. There are those that are based on comparatively cheap and abundant particulate fillers and ideally whose function is to reduce the overall costs of the composite, and yet at the same time to slightly improve the load bearing capabilities of the material or to impart special properties to the composite. Then there are those thermoplastics containing short fibres whose function is much more than just a simple filler. As Kelly (1973) has pointed out, fibre reinforced materials are developed in order to exploit the properties of the stiff and strong fibres and the plastic is used because it is a suitable "binder" and can be easily moulded. This is a particularly important point in the context of this book, since the mechanical properties of many short fibre reinforced thermoplastics fall well short of their maximum realisable values. While this has not been a serious drawback in the past, it is certain that major improvements in the fabrication and utilization of fibre reinforced thermoplastics will be needed. Many more reinforced thermoplastics are being used in critical load bearing applications, some of which also demand minimum component weight. The need for high specific stiffness and strength coupled with the ability to operate continuously at elevated temperatures requires that the reinforcing effect of the fibres is used to maximum advantage. These requirements have to be met by an appropriate understanding of the effects of processing conditions on the microstructure and properties of the final component. The need for improved control of fibre length and orientation during the compounding and moulding stages is particularly vital when expensive combinations of fibre and matrix are being considered. A 13% annual growth for speciality thermoplastics e.g. polyethersulphones, polyetherketones, polyimides etc is forecast for the period until 1985. At the same time, speciality fibres such as carbon and Kevlar are expected to have an annual growth of about 9%. Carbon fibre reinforced polyetheretherketone has been moulded successfully into jet engine parts, a good example of how the development of an engineering thermoplastic is extending the range of use of the more expensive fibres. These newer and more elaborate thermoplastic composites provide some exciting prospects for the production of high temperature engineering components. This book has been written with these types of material in mind.

The plan of this book is as follows. Chapter 2 discusses the relevant theory underlining short fibre reinforcement, with a view to providing a basis upon which the observed mechanical properties of short fibre composites can be compared. This comparison is made in Chapter 3 and includes some debate about the rôle of the fibre-matrix interface in influencing composite properties.

Chapter 4 is concerned with the very practical and important problem of the production of the composite feedstock and the methods of assessing the effectiveness of the compounding operation. In Chapter 5, methods for examining the microstructure of short fibre reinforced thermoplastics are discussed, together with the effects of moulding conditions on the fibre length and orientation in components. The rheological properties of reinforced thermoplastics are discussed in Chapter 6 together with the way in which this information can be used to assist in interpreting the complex pattern of fibre orientation observed in moulded components. Finally, in Chapter 7, the problems of characterizing the mechanical anisotropy in components are discussed, including a critique of the current assessment methods and how alternative testing strategies are emerging.

Reference

Kelly, A. (1973). Strong Solids. Oxford University Press, London.

CHAPTER 2
Theoretical Background

It is not the intention of this chapter to give a comprehensive coverage of the mechanical properties of fibre reinforced composites. This topic is already well covered in a number of now familiar texts - see e.g. Kelly (1973), Piggott (1980). What this particular chapter attempts to do is to provide some relevant theoretical background that highlights the rather special problems associated with short, as distinct from the traditional long fibre composites. The prediction of the mechanical properties e.g. the stiffness of a fully aligned long fibre reinforced composite is a difficult mathematical problem, especially when the load is not applied along the fibre axis. To perform the same exercise in a short fibre reinforced composite is even more difficult. The first problem involved stems from the fact that the fibres cannot be regarded as infinitely long - normally a convenient mathematical assumption. In addition, a real moulded component exhibits a very complex distribution of fibre orientations, which can itself vary from one point in the moulding to another - see e.g. Fig. 2.1. Any realistic predictive work has to include the effect of fibre length/length distribution and a distribution of fibre orientations with respect to the applied load. If such predictive work is to be of any practical use, some mathematical rigour must therefore be sacrificed to enable comparatively rapid calculations to be made. It is the stiffness of short fibre reinforced thermoplastics that has received most attention and with reason can be predicted fairly accurately.

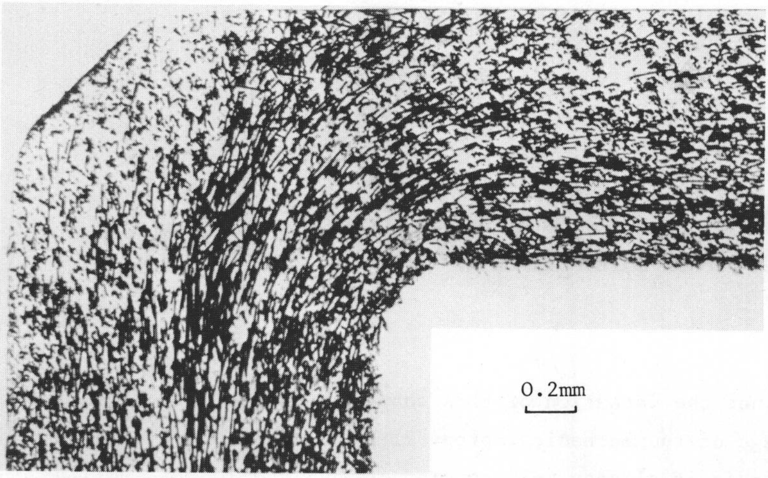

FIG. 2.1. Optical micrograph of a thin section taken from a carbon fibre reinforced nylon 66 moulding.

On the other hand, strength and toughness are difficult quantities to predict even for a long fibre composite, while the corresponding problem with short fibre reinforced thermoplastics has not yet received very much attention. Further complications that can exist in a reinforced thermoplastic may arise from the presence of a significant interfacial layer between the fibre and matrix together with molecular orientation in the matrix phase.

2.1. ANISOTROPY OF MECHANICAL PROPERTIES

A material is referred to as being anisotropic if the properties of that material depend on the direction in which they are measured with respect to some fixed axis. For example, a fully aligned fibre reinforced composite will exhibit a much larger Young's modulus when measured along the fibre direction compared to that obtained when the

the load is applied at 90^{o} to the fibre axis. In most fibre reinforced composites of practical interest, however, the properties do not change if measurements are made in different directions in a plane transverse to the fibre axis (or anisotropy axis). Referring to Fig. 2.2 , this implies that as the angle θ is varied, the properties of the composite will change but that measurements performed at different angles in the 1-2 plane will not reveal any such changes. In this case, the 1-2 plane is referred to as a plane of transverse isotropy.

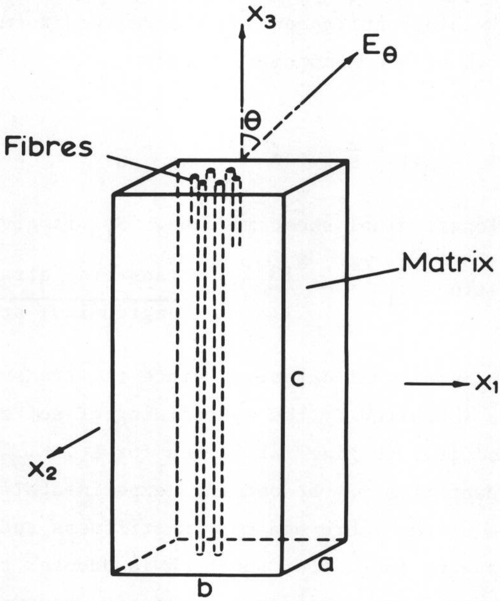

FIG. 2.2. Definition of axes used to describe mechanical anisotropy.

To characterise fully, the anisotropy of a composite would seem to require a very comprehensive set of measurements performed at a series of angles $0 < \theta < 90^{o}$. For properties such as strength or toughness this is largely true, but if elastic behaviour only is being studied then the variation of stiffness with angle θ follows a definite fundamental relationship:-

$$\frac{1}{E_{\theta}} = S_{33} \cos^{4} \theta + (2S_{13} + S_{44}) \sin^{2} \theta \cos^{2} \theta + S_{11} \sin^{4} \theta \qquad \text{................(2.1)}$$

where E_{θ} is the stiffness measured at angle θ with respect to the x_3 axis. The S's are compliances and are related to more familiar elastic properties of the composite, thus:-

$$S_{33} = \frac{1}{E_{o}} \quad ; \quad S_{11} = \frac{1}{E_{90}} \quad ; \quad S_{44} = \frac{1}{G}$$

where G is the longitudinal shear modulus. S_{13} is given by the Poisson's ratio $\nu_{13} = -\frac{S_{13}}{S_{11}} = \frac{\text{Transverse strain along } x_3}{\text{Longitudinal strain along } x_1}$

ν_{13} is likely to be a small quantity, since the transverse strain along x_3 will be inhibited by the restraining effect of the fibres. Hence E_{θ} is effectively defined if values for E_{o}, E_{90} and G are known. These quantities may be obtained experimentally or predicted from a knowledge of the fibre and matrix stiffness and fibre volume fraction. It is when the latter approach is adopted that the real complexity of short fibre reinforced composites becomes manifest. The quantity E_{o} can be easily predicted with good accuracy for a long fibre composite but the situation is much less satisfactory for short fibre composites. E_{90} and G are very difficult to predict easily and accurately for both types of composite. The next section will indicate how estimates of all these quantities may be obtained.

The above discussion has been made with reference to a fully aligned fibre reinforced composite, but the variation of E_{θ} with θ is also applicable to a partially oriented system having a single axis of anisotropy, providing the appropriate values of the compliances are used for that particular orientation distribution. Relating the elastic properties of a partially oriented system to those in the fully aligned case requires some assumptions to be made concerning the state of stress and strain throughout the composite. One approach is to assume a state of uniform strain, which gives rise to the Voigt average or to assume a state of uniform stress, which gives rise to the Reuss average. These calculations provide upper and lower bounds for the elastic properties of the partially oriented composite. The reader is referred to the work of Cox (1952), Ward (1962) and Brody and Ward (1971). Simplifications can be made to reduce the computations involved and one popular approach is to ignore the transverse stiffness E_{90} and shear modulus G i.e. to evaluate the effective stiffness of the partially oriented composite by appropriate averaging of E_o over the fibre orientation distribution - Krenchel (1964).

2.2. REINFORCEMENT USING SHORT FIBRES

The stiffness of a composite arises primarily from E_o and so it is necessary to examine the way in which this quantity depends on the properties of the fibre and matrix and the fibre length. When a load is applied along the anisotropy axis of a fully aligned fibre reinforced composite, the load is transferred to the stiff fibres via shear stresses at the interface. The calculation of the variation of shear stress and tensile stress along the fibres was reported by Cox (1952) for the case of an elastic matrix and elastic fibres. This is the now classic "shear-lag" analysis. With reference to Fig. 2.3 consider a fibre of length ℓ and radius r embedded in a matrix. We assume that the matrix as a whole is strained homogeneously by the application of a load applied parallel to the fibre axis. Since the shear stress τ at the fibre-matrix interface will vary along the fibre, so also will the tensile stress in the fibre. Considering a small element of the fibre length δx, the net tensile load δF across this element must be balanced by the shear force