

Introduction

I. M. WARD

Department of Physics, University of Leeds, Leeds, UK

Until comparatively recently the only aspect of the mechanical properties of polymers which could be given a satisfactory treatment at a molecular level was rubber-like behaviour. Our understanding of all aspects of the mechanical properties started with the so-called amorphous polymers, and treated crystallising polymers as extensions, albeit complicated and awkward extensions. For example, in viscoelasticity there was the classic survey of the linear viscoelastic behaviour of polyisobutylene by Marvin, based on a collation of international data, the comprehensive studies of time-temperature equivalence by Ferry and his collaborators which were formulated in the WLF (Williams-Landel-Ferry) equation, and numerous dynamic mechanical measurements of viscoelastic relaxation processes. The treatment was at a phenomenological level, and although the importance of molecular structure was recognised in terms of the chain conformation and chain flexibility, the links between structure and mechanics were usually imprecise, relying on such ideas as free volume, the theory of activated processes, or the dynamics of molecular chain models.

Structural techniques such as nuclear magnetic resonance and dielectric relaxation led to semi-quantitative molecular interpretations of the relaxation processes, but on the whole the subject lacked a definitive approach at a structural level, particularly for crystalline polymers.

During the last ten years the situation has altered beyond recognition and the group of papers which form this issue of the *Journal of Materials Science* illustrate the improvement in an admirable fashion. Two factors in particular have been important in bringing about the transformation. These are:

(1) The discovery of chain folding of molecules in crystallisation, and increasing evidence for lamellar morphology in bulk polymers.

(2) A gradual recognition that detailed correspondence between mechanical properties and structure must be sought on oriented polymer

specimens of well-defined structure.

Central to our understanding of the issues involved are the papers by Keller and Pope and by Point, Dosière, Gilliot and Goffin. These papers are essentially concerned with establishing the links between the macroscopic deformation and identifiable molecular processes such as interlamellar slip and intralamellar slip. There is a difference of viewpoint even between these two closely related papers on polyethylene and nylon-11 and at this juncture it may be necessary to sound a note of caution. In establishing a subject, and such has been the aim of much of the work to date, it is useful to test the validity of general hypotheses and models. These two papers are testing hypotheses concerned with the geometrical aspects of the shear deformations, and this approach is carried one stage further in the next paper by Owen and Ward. We would expect the most general hypotheses to carry us only to the first stage of understanding and therefore should not be disturbed if one type of generalisation holds for low density polyethylene and another for nylon-11. Indeed, it is these sorts of differences between materials which we should anticipate and seek to establish, as we reach the second phase in the development of the subject.

The recognition of the complex morphology of crystalline polymers has led to detailed studies of drawing, rolling and more recently hydrostatic extrusion. These aspects are well represented in this issue with a comprehensive review by Peterlin, two papers by Gezovich and Geil on rolling, a paper by McConkey, Darlington, Saunders and Cannon on hot drawing and one on solid state extrusion by Imada, Yamamoto, Shingematsu and Takayanagi. These papers emphasise the many-sided nature of this research. First, one seeks to understand the morphological changes subsequent to yield. Although there are many observations and speculations and Peterlin's article gives an excellent summary of these, it is still very uncertain how the final structure depends on the

stress and strain history. In some cases there is evidence to suggest that the final structure depends on the final strain only and not on history, but one would hesitate very much before concluding that this applied in general.

Secondly, one may seek to relate the properties, and particularly the mechanical properties, of the final product to its final structure. The paper by Andrews and Reeves is an admirable example of this type of work and contains several surprises which serve to underline the difficulties which would occur if one were to attempt a facile explanation in semi-phenomenological terms. Yield and fracture in oriented polymers are represented here by contributions from Simpson and Hinton, and Anderton and Treloar, respectively. In both cases the oriented polymers provide much more insight at a structural level than comparable studies on isotropic polymers.

It is perhaps a little disappointing to find that there were few contributions on a molecular level, e.g. concerned with nmr spectroscopy or dielectric relaxation. Such investigations, as can be seen from the paper on molecular motion in oriented polyethylene, are often rather involved, but it is important to remember that there is a *dynamic* as well as a static aspect to our understanding of polymer properties and that this is an area which has not benefited so greatly from the recent advances in structural knowledge.

In conclusion, these papers may be commended as representing a wide cross-section of approaches to the understanding of oriented polymers, and their contents emphasise that studies of oriented polymers now form an important part of polymer science.