

The Mechanical Performance of Polymers and Polymer Composites at High Strain-Rates

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Abstract

Polymer-based systems in general have non-linear mechanical properties which are highly-temperature dependent and strain-rate sensitive. They are widely used in applications where factors such as weight, strength and energy-absorbing properties need to be balanced. In this paper a number of low to high-rate characterisation techniques are described. A description of the output obtained is described with reference to a range of polymers, single polymer composites, polymer bonded explosives (PBX) and polymer bonded sugars (PBS): the last category are widely used as inert mechanical and density simulants for PBX systems

The importance of predictive modelling, using a limited empirically measurable parameter is of increasing importance. Advances, especially those models which start from molecular considerations, provide methods for prediction of polymer and polymer composite behaviour which allows a more focussed series of validation experiments to be conducted. This approach will be illustrated by comparison of the high-rate studies with predicted constitutive equations.

1. Introduction

The behaviour of polymers and polymer-based systems over a range of stress and strain-rate regimes is of interest for any number of reasons. Amongst these are practical advantages of formability, weight reduction, reduced sensitivity of explosive systems these can be linked to more fundamental understanding of polymer systems behaviour which is widely known to

non-linear and highly temperature dependent in comparison to metals and other highly crystalline systems.

The field of high strain-rate experimentation has expanded greatly in recent years, with the increase in high-speed diagnostics allowing sub-microsecond and in some cases femto-second time resolution to become commonplace. Increased use of predictive modelling with the implied increase in the number of materials and materials models has provided a significant drive for well controlled experiments. This forms a development circle involving experiment, theory and numerical methods. The use of multiple diagnostic systems on a single experiment, e.g. stress gauges, high-speed photography and flash X-ray systems, has provided much improved understanding of the qualitative and quantitative processes occurring at short time scales and made a large contribution to more physically based models.

Table 1 lists some of the more widely used techniques in the strain rate characterization of materials. A review on these techniques applied to a wide range of materials has been published by the authors elsewhere [1]. In this review the techniques will be discussed specifically in relation to polymeric materials. The main effect of increasing the strain rate is that the transient stress levels increase and the sudden delivery of energy allows processes with high activation energies to be accessed. Processes, which operate on long time scales, e.g. thermal diffusion, which are significant under quasi-static loading, do not have time to occur.

It is generally found that yield and fracture stresses increase with increasing strain rate [2]. The increase in failure stress is very marked at strain rates above 10^3 s^{-1} and the effect of inertia becomes significant. Ultimately the response changes from one where the sample can be assumed to be in stress equilibrium to one of a wave with associated 1-D strain moves so quickly that the material does not have time to move laterally, as seen in shock waves.

Table 1: High Strain Rate Regimes and the Associated Equipment.

Strain rate	Equipment	Stimulus duration	Comment
$10^{-6} - 10^{-2}$	Instron	100s of seconds	Quasi-static loading
$10^{+2} - 10^{+3}$	Drop weight	10s milliseconds	Generally used to determine impact ignition thresholds
$10^{+2} - 10^{+3}$	Hopkinson bars	100s microseconds	Compression, tension and torsion loading. Extensively used for PBX formulations. Constitutive models.
$10^{+4} - 10^{+5}$	Miniature Hopkinson bar	10s microseconds	For fine grain materials or single crystals. Generally metals
$10^{+3} - 10^{+6}$	Taylor impact	10s microseconds	Sometimes used for metal jacketed energetic samples
$10^{+5} - 10^{+8}$	Plate impact	microseconds	Pressures and durations similar to that of gap tests. Laser driven flier plates have sub-microsecond duration high-intensity shocks

While these represent the main techniques encountered, a range of more specialised loading techniques have been used including high rate tensile studies using the so-called “flying

wedge” system, where a notched sample is forced apart by a wedge-shaped impactor fired into the notch. The expanding ring test uses an exploding conductor or small exploding charge along the central axis of a cylinder of a material, produces a violent lateral motion of the outer walls of the cylinder inducing a rapid expansion of an annulus of the test material, closely fitted to the cylinder.

In addition, standard thermal and mechanical techniques, such as differential thermo-mechanical analysis (DMTA) are widely used in the polymer community and provide insight into the molecular-level processes occurring in the polymer system.

Polymers in many respects provide ideal study materials in that their mechanical and thermal properties can be closely controlled and varied over a range of temperatures easily accessible in the laboratory. One slightly unfortunate negative is that samples of a given material can vary significantly from batch to batch or more strongly from manufacturer to manufacturer; this can sometimes lead to contradictory results for nominally the “same” material.

A brief survey of the literature will reveal a number of models available, all with greater or lesser degree of empirical parameters. Historic spring-dashpot models are used with some degree of success in some areas, there are numerous semi-empirical models, however, there is a great drive to use models which are based on the fundamental structure of the polymer as in the group interaction model [3] is proving a fruitful area

Taking the level of complexity further it is important to understand the processes involved in single polymer composites e.g. DYNEEMA [4 and references therein] which is used to mitigate fragment impact. Here the use of the anisotropy of sound speed, fundamentally linked to stress propagation speed, within and between oriented and non-oriented forms of the same polymer is crucial to the functional use of the material. The field of single polymer composites has been recently reviewed [5] and their potential environmental benefit through increased recyclability is one of the principal driving forces.

Polymer bonded granular composites, such as PBX and PBS formulations have further complexity in the presence of interfaces between polymer and filler material. This can have the effect of increasing composite strength compared to that of the polymer, while a potential problem is that de-bonding between the filler and the binder can produce extra failure points in the system. PBX / PBS systems are also of interest from a fundamental viewpoint as the filler materials are organic crystals with elemental compositions similar to many polymer systems.

Many quasi-static mechanical methods and thermal techniques are extensively used in the polymer community and widely known to engineers and scientists, therefore the following sections will illustrate some of the material response seen across a wide range of strain-rates, concentrating on the higher-rate areas.

2. Drop-Weight

Conceptually a drop weight is a very simple device, gravity is used to accelerate a mass onto a sample and the resulting deformation and damage pattern observed. As with many experimental techniques care has to be taken with a number of experimental parameters; the geometry of the mass and sample, the contribution of friction at the interfaces, the relative movement of the upper face of the sample compared with lower face of the sample. Gauge traces from drop-weight systems are also notorious for the amount of “ringing” or oscillations seen on the signal, the result of a combination of mechanical and electrical resonance in the system. However, accurately aligned drop-weight experiments give extremely useful quantitative data e.g. stress-strain curves. Drop weight studies on polymer systems are illustrative of a number of phenomena that can be seen. Stress localisation and shear bands may also form due to the geometry of the sample. Heat sensitivity films have also been used to track the rapid, transient heated associated with the drop-weight impact [6]. Illustrative examples can be seen in figure 1 taken from [7].

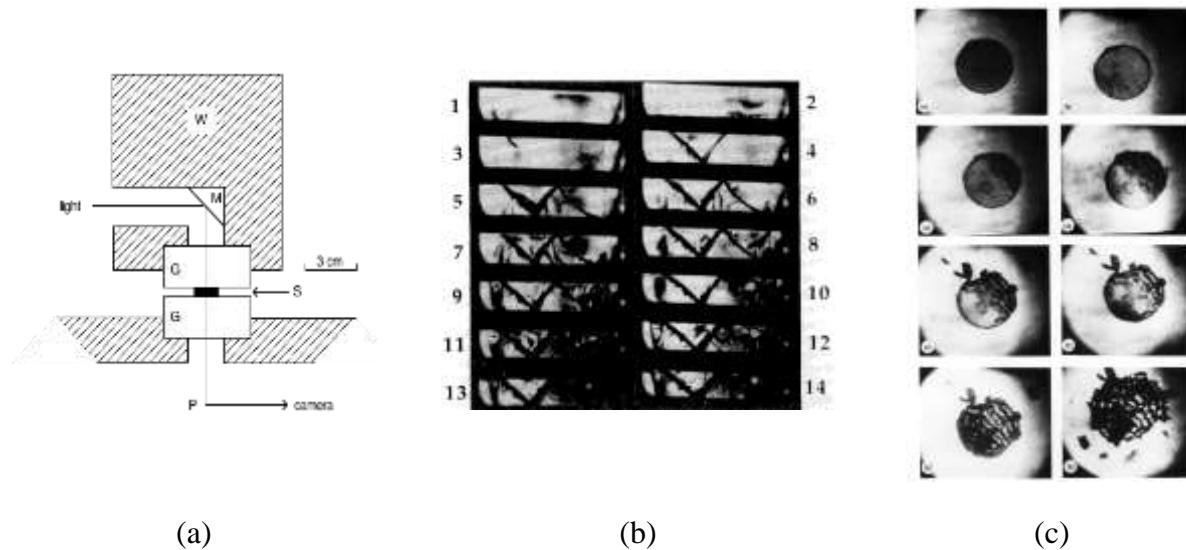


Figure 1 (a) Schematic diagram of the high-speed photography drop weight apparatus. W weight, M mirror, G glass anvil, S specimen, P prism. Mass of drop weight = 5.545 kg. (b) Selected frames, 20 microsecond interframe time shows the shearing and fracture in a polystyrene sample (22.5 x 7.3 x 3 mm side lengths) during drop weight impact. (c) Along impact view. Impact on a 5 mm diameter, 1 mm high disc of PS. Times after frame (a): (b), 35 μ sec; (c) 42 μ sec; (d) 49 μ sec; (e) 189 μ sec; (f) 196 μ sec; (g) 203 μ sec; (h) 259 μ sec.

3. Hopkinson Bar

In order to transmit stress pulses into samples rods can be used as wave guides. The rod can transmit a stress into a sample, the specimen being taken to large strains. The basic idea of the split Hopkinson pressure bar (SHPB) is that the specimen is deformed between two bars excited above their resonant frequency and chosen so that they remain elastic (small strains). This means that strain gauges can be used repeatedly to measure the signals in the bars (strain gauges normally have small failure strains). Dynamic loading is produced either by striking one end of one of the bars (the input bar) or by statically loading a section of the input bar held at some point by a clamp and then releasing the clamp so that the load propagates to the

specimen. Compression bars are nearly all of the dynamically loaded type and represent the most common and conceptually easiest system – a schematic is shown in figure 2.

Torsion SHPBs are nearly always statically loaded [8]. Tension and torsion systems have the advantage that friction between the bars and the specimen is not a problem. They have the disadvantage that the specimens are of more complex geometry and hence harder to fabricate. Also tension specimens usually have to a large length to diameter ratio so that issues of stress equilibrium and longitudinal inertia have to be carefully considered. Torsion specimens are usually thin-walled tubes. The samples are thin as the strain rate is basically dependant on the angular velocity of the sample, so small diameter sample have lower strain rates than large diameter samples when both are twisted at a given angular velocity. This raises the issue, for metals and brittle materials, as to how many grains or crystals they contain within the wall thickness and hence how representative they are of bulk material. Similarly polymer samples may buckle when loaded in such a system, while simply using thick-walled samples produces a range of strain rates through the wall thickness. One way round this is to shear simple discs of material of varying diameter [9, 10] so as to be able to subtract off the mechanical effect of the ‘dead’ material in the centre. This requires perhaps 4–5 times as many experiments to be performed per data point, but the specimens, being simple discs, are much easier to fabricate than thin-walled tubes.

In many respects the strain rate regime from $10^2 - 10^4 \text{ s}^{-1}$ represents a challenge in understanding. While most materials show an increased yield strength with strain rate there is evidence of a fall in yield strength in a number of polymers in this region, followed by a rise at the higher rates as seen in figure 3.

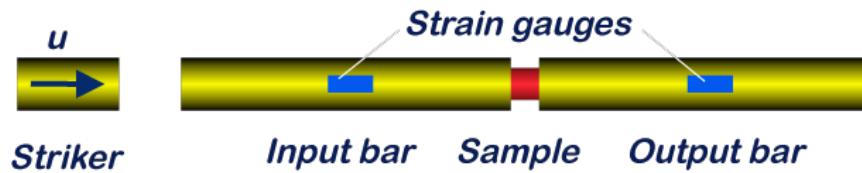


Figure 2. Schematic of a compression SHPB system

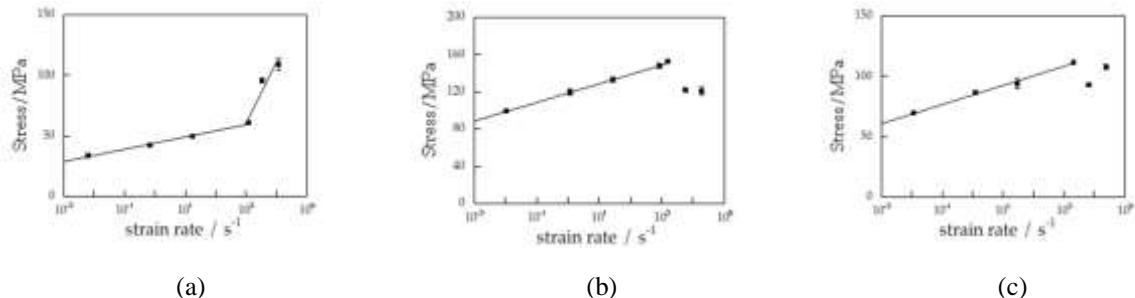


Figure 3. Yield stress versus strain rate for a number of polymers (a) A “normal” response seen in acrylonitrile-butadiene (ABS) such a response trend is seen in metals and geological materials (b) PEEK, and (c) Polycarbonate, showing a marked and reproducible drop at in yield stress at the higher rates, taken from [11]

The effect of the molecular relaxation processes in this regime has been addressed for polycarbonate and polyvinylidene difluoride [12]. In this study the use of stress rate

temperature mapping is emphasised a suitable method for understanding these polymer systems.

In granular systems composite systems this is further complicated by processes such as debonding, fracture nucleating in the binder and cracking of the filler material. Macro-properties sometime show trends in remarkable agreement with those seen in metallic systems. In the mid 2000's the effect of grain size on the high rate mechanical properties of an ammonium perchlorate (AP)/hydroxyl-terminated polybutadiene (HTPB) PBX was studied. This PBX consisted of 66% AP and 33% HTPB by mass. The AP was available in four different crystal sizes: 3, 8, 30 and 200–300 nm. The effect of grain size was most clearly seen at low temperatures (figure 4) the effect of particle size on the flow stress of the material is linear in $1/d^{1/2}$; where d is the particle size (figure 5). This has been subject to further investigation to determine if the particle size or the particle separation is dominant, this research is on-going [13].

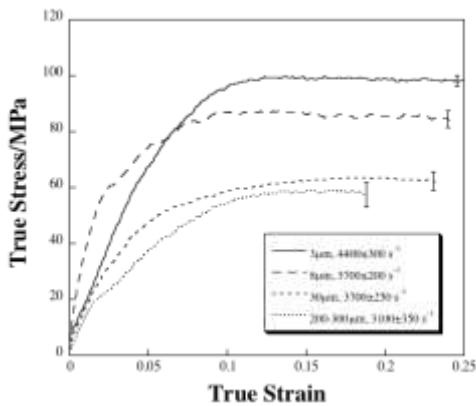


Figure 4 Effect of particle size on yield stress at -60°C

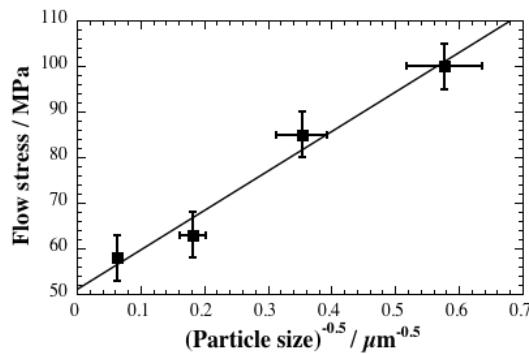


Figure 5 Plot of the flow stress versus the reciprocal of the square root of particle size for the data of figure 15b.
The linear fit has an intercept of 51 MPa and a slope of 86.5 MPa (μm)^{0.5}

More recent research has concentrated on the use of high-speed imaging to track the deformation of the sample during the loading process [14] the technique of optical cross-correlation has been used. Typical output of this technique can be seen in figure 6. This time resolved data allows the wave propagation along the specimen length to be observed. This

type of data provides a stringent test for constitutive models. This analysis technique has also been applied to granular polymer composites over a range of strain rates [15, 16].

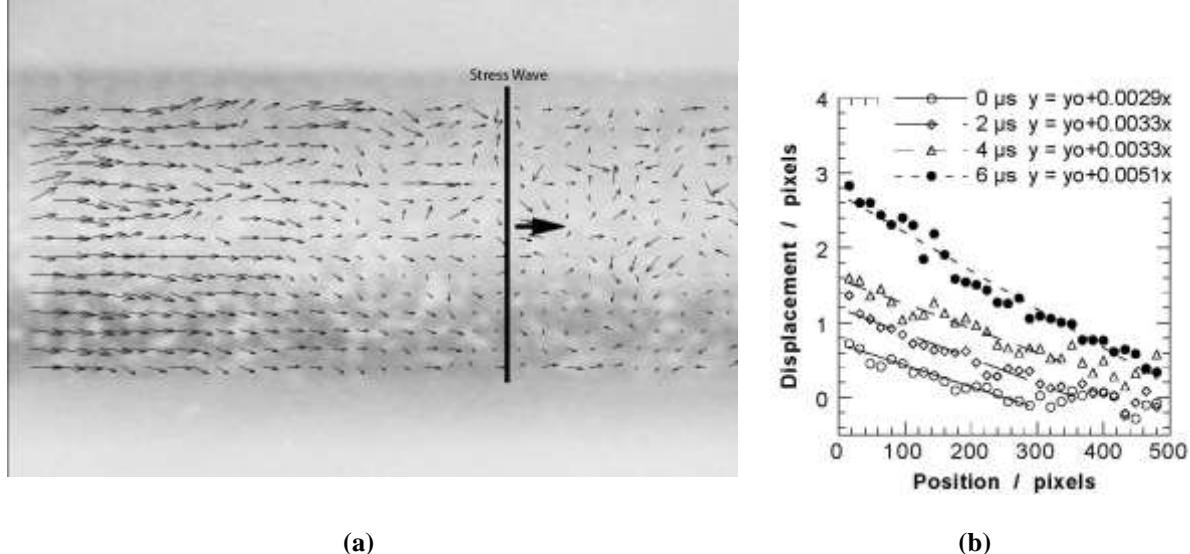


Figure 6. (a) Displacement quiver plot for a stress wave passing through a cylindrical specimen of polycarbonate in a compression Hopkinson Bar System. The stress pulse causes compression in the sample. The exposure time of this image was 1 microsecond, thus freezing the motion. Image cross correlation was used to see the wave front. Not all of the specimen is visible in this image. (b) Plot of displacement against position along a polycarbonate specimen, at four different times during loading. The horizontal scale is 53 pixels mm⁻¹, so the movement of the wave front agrees with a wave speed of 1.4 mm μs⁻¹. Taken from [14]

The use of DMTA in combination with data across a wide range of strain rates has resulted in a consistent view, in a qualitative sense of the response and damage processes in these composite systems. This is illustrated by the recent paper by Drodge *et al.* Here the effect of the binder, its thermal and mechanical properties is compared with those for a granular composite. Overall this illustrates the utility of frequency time shifts linked to strain rate effects in this area of research [13].

4. Plate Impact

A shock wave is a travelling wave front, which has a discontinuous adiabatic jump in state variables. The loading time is short compared to the inertial response, pressure pulses propagate through the body to communicate the presence of loads to interior points, and thus, the material inertia is important. A detailed review on shock wave physics was published by Davison and Graham [17]. The most common experimental method of inducing a shock wave in a target is by plate impact as illustrated in figure 7. The stress components can be measured by commercially available piezoresistive manganin gauges, which have been calibrated [18, 19] for 1-D shock loading. The particle velocity can either be measured using wire gauges, which operate by applying a constant magnetic field normal to the gauge, or by a velocity interferometric system, known as VISAR [20].

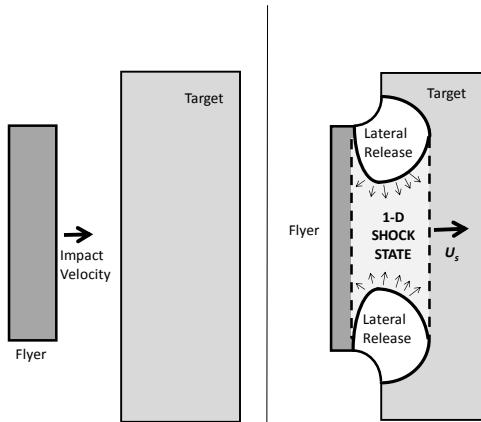


Figure 7. The plate impact experiment. A flier plate strikes a target causing a shock wave to move at a velocity (U_s) through the target and the impactor. The region of high pressure 1-D shock reduced by lateral releases as well as along-axis longitudinal wave reflections

In many solids, under shock loading there is an initial elastic region, up to the so-called Hugoniot elastic limit (HEL). Even in relatively simple cases, such as metals, below the HEL, strain and strain rate hardening as a result of dislocation accumulation is often seen. In heterogeneous materials and most polymers, a clear elastic limit is harder to determine due to the ramping caused by the differing impedances of the components or the material structure. In addition, as the passage of the wave is adiabatic, the temperature can increase dramatically. As a result, polymers may exhibit thermal softening effects as a result of this heating along with a counteracting hardening effect caused by the increased strain rate and the limited time for molecular motion to accommodate the stimulus.

At very high stress, ~20GPa, the energy deposited is sufficient to cause many polymers to chemically decompose. For some polymers a “kink” can be seen in the shock velocity – particle velocity Hugoniot. This kink occurs between 10-14 GPa in density polycarbonates for example (figure 8). A mechanism based on the removal of free volume between polymer chains has been proposed but no predictions have been made based on this hypothesis. Such a kink is not seen in high density polyethylene.

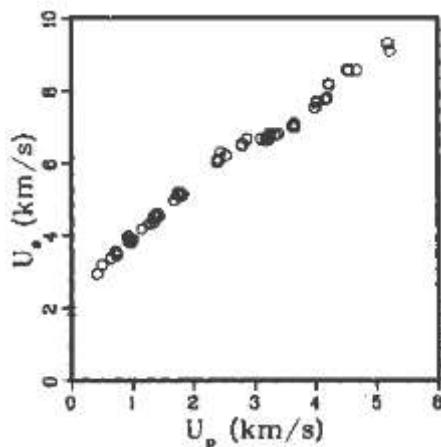


Figure 8. The shock velocity-particle velocity response of a polycarbonate. The plateau seen in the region of $U_s = 6.8 \text{ km s}^{-1}$ has been associated with the removal of free-space between the polymer chains. Taken from [21]

A similar kink is seen in poly(styrene). Porter and Gould [22] proposed that this kink is due to an activated change in structure whereby the aromatic ring transforms into a triangular prism. Molecular mechanics can predict the equation of state of polymers from their structure alone [23] and this technique has been used to predict pressure-volume for the two structures: aromatic ring and triangular prism. The prediction is compared with experimental data in Figure 9. Molecular mechanics also allows prediction of the activation energy of such a structural change, via an energy density, and this equates to a pressure of around 20 GPa [22].

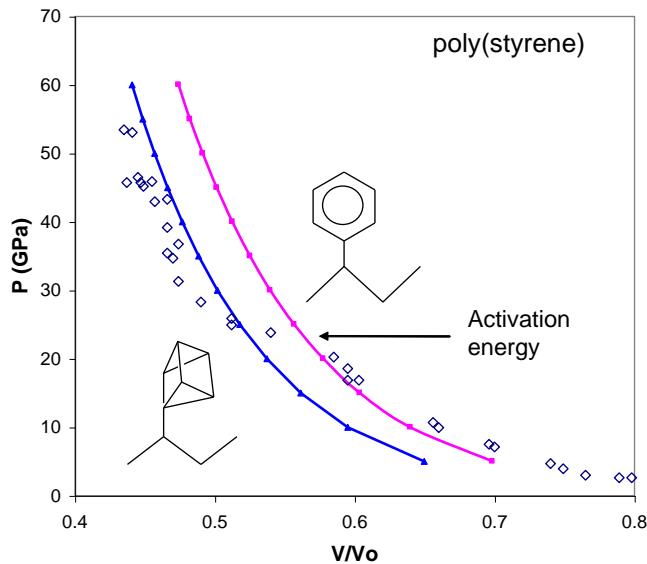


Figure 9: Comparison between predicted pressure-volume behaviour of two structures in poly(styrene) and measured data

Significant effort has been made to understand the material response both in terms of the strain-time records seen and also in the recovery of shocked samples for post impact analysis [24]. Here both strain-rate hardening and softening was seen in PTFE samples, depending on the conditions used, re-shocking of the sample material had a significant effect and there was a marked difference in the crystallinity of the recovered material.

5. Temperature Time Superposition

The previous sections have highlighted some of the techniques used and have illustrated some of the difficulties involved. Without adequate knowledge of the underlying system processes it would be difficult to encapsulate the behaviours into a coherent model. However, much effort has been made into consideration of the variation of temperature and the time and strain rate response of the material. This allows the polymer behaviour to be explained in a coherent fashion thus leaving the processes of fracture and damage more visible in the data.

This can be illustrated by considering the temperature time response of a polymer bonded energetic system. This study is one of the few that represents a complete study across a wide range of strain rates and temperatures [24].

In this study the compressive strength of the energetic composition EDC37 was measured at a temperature of $293 \pm 2\text{K}$ over a range of strain rates from 10^{-8} to 10^3 s^{-1} , and at a strain rate of 10^{-3} s^{-1} over a range of temperatures from 208 to 333 K. The results show that failure stress is a monotonic function of applied strain rate or temperature, which is dominated by the relaxation properties of the polymeric binder; this is confirmed by dynamic mechanical thermal analysis performed on both EDC37 and its binder. Similarities between the compressive strain rate/temperature data sets can be understood by temperature–time superposition; data collected at a strain rate of 10^{-3} s^{-1} over a temperature range 208 to 333K were mapped onto a plot of strain rate dependent strength at 293 K, using an empirically determined sensitivity of $-13.1 \pm 0.3\text{K}$ per decade of strain rate. Sample size had a modest effect on the stress–strain behaviour; small length to diameter ratios gave results consistent with an increased degree of confinement. Samples taken to large strains exhibited strain localization in the form of shear bands. Figure 10 shows the main experimental results used in this study: the similarity in trends is obvious.

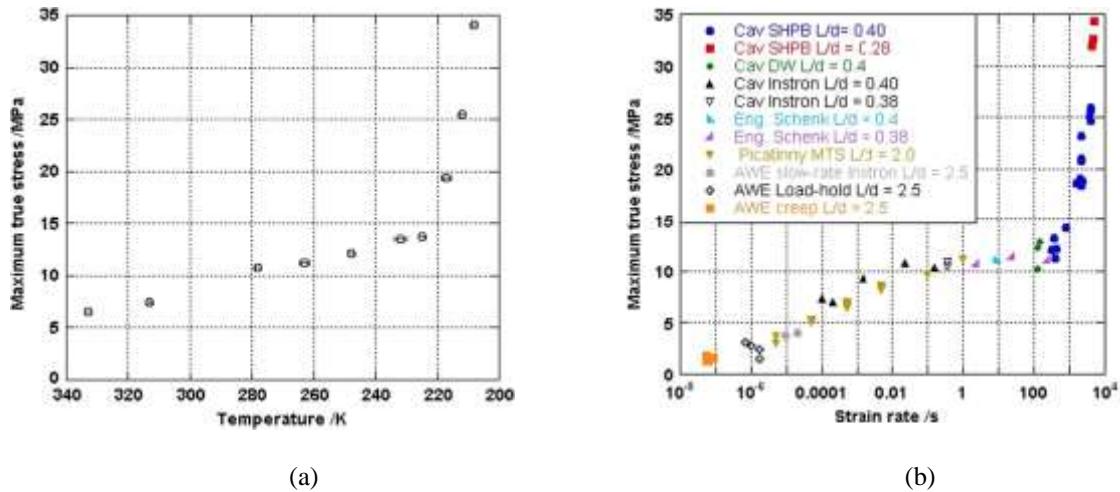


Figure 10. Yield strength of a granular composite at (a) a fixed 10^{-3} s^{-1} strain rate, varying temperature (b) failure stress as a function of strain rate tested at 293 K.

6. Constitutive Modelling

The prediction of constitutive models for polymer composites is facilitated by the plethora of good composite models available in the literature. These rely, however, on knowledge of the properties of the components and an understanding of how the presence of stiffening phases in a polymer matrix changes those properties. Conventional constitutive models for polymers cannot be used reliably for that purpose particularly at high rate and for high volume fractions of filler.

Group Interaction Modelling [3, 22] has shown that the properties of polymers at low rate, high rate and under shock can be reliably predicted from the chemistry of the polymer alone.

This also allows phase interactions to be included so as to fully inform composite models. The basis of the approach is to determine the energy-volume response of the interacting groups and to understand the degrees of freedom involved in those interactions. This allows the loss spectrum for the polymer to be determined together with its changes with temperature and rate.

A volumetric potential well is constructed for the interacting system and this predicts bulk modulus, density, thermal expansion coefficient and glass transition. The heat capacity as a function of temperature is predicted from the one dimensional Debye temperature and the degrees of freedom. Relaxation phenomena alter the transition temperatures with rate and a Vogel-Fulcher form is used to determine this dependency. The main advantage of this approach is that no fitting is required. Figure 11 shows the prediction of modulus and loss spectrum for poly(carbonate).

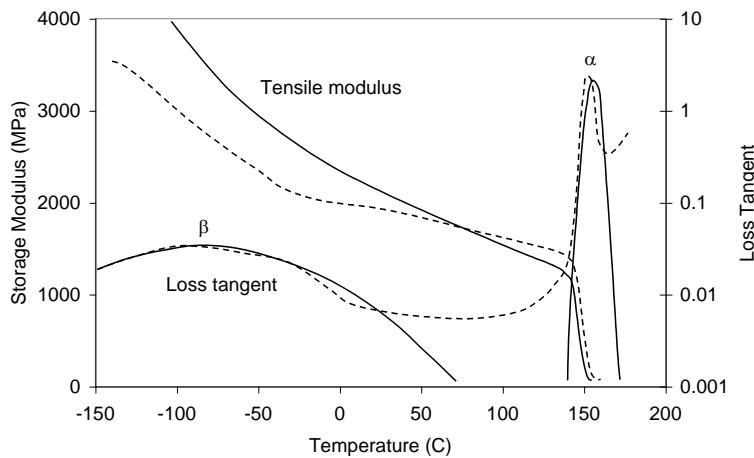


Figure 11: comparison between predicted and measured loss tangent and tensile modulus at 1Hz for poly(carbonate)

Once the loss spectrum is known then it can be combined with the predicted modulus to predict the stress/strain response in tension. Poisson's ratio gives the response in compression. This allows certain important experimental outputs such as yield stress to be predicted. Figure 11 shows the prediction of yield stress vs. rate at room temperature for two common polymers: poly(carbonate) and poly(methylmethacrylate) and compares it with available data.

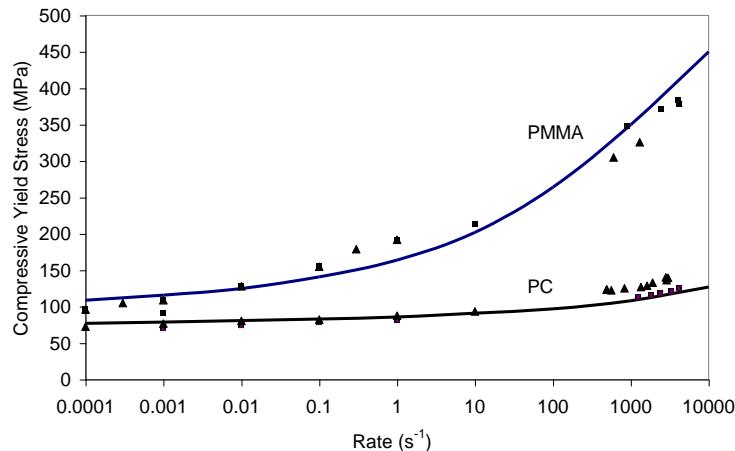


Figure 11: Prediction of variation of compressive yield stress with rate for two common polymers

7. Conclusions

In this review the response of polymer and polymer composites has been shown to be complex. A number of failure processes can occur; high rate loading heats the material but also allow less time for sample motion, resulting in a stronger response.

In a variety of polymer and polymer composites the application of temperature-time (frequency) shifts is shown to explain some of the behaviours seen. Much of the stress-strain curve can be explained through these phenomena. In a way this is unsurprising, the polymer is the softer component in most of these systems and so accommodates the majority stress and so heats more than the filler particles.

The main challenge in this area remains in quantitatively measuring the damage level included by such loadings and accommodate it into a coherent model.

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