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Split Hopkinson Pressure Bar Measurements of Polymer Energy Dissipation and Identification of the Glass Transition Condition

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Abstract. We explore the idea of using a split Hopkinson pressure bar to measure polymer energy dissipation. For an elastic specimen the sum of the mechanical energies within the transmitted and reflected pressure waves ought to be equal to mechanical energy in the incident pressure wave. Any deficit is due to inelastic dissipation by the specimen. The amount of energy dissipated can be plotted as a function of temperature and will be greatest at the glass transition condition. We use historical split Hopkinson pressure bar data to carry out a calculation of dissipated energy at different temperatures. The ratio of dissipated to stored energy in the split Hopkinson pressure bar specimen is directly compared to Dynamic Mechanical Analysis measurements of loss tangent. The temperatures of the peaks identified are used to demonstrate the validity of our proposed method.

BACKGROUND

Understanding the dynamic properties of polymers is important for applications involving varying strain rates, such as impact conditions. Of importance is energy dissipation within the material, which is maximal at the glass transition condition and other polymer relaxation conditions (e.g. secondary transitions) [1]. We constrain relaxation conditions by a combination of strain rate and temperature, at ambient pressure.

The quantity of the loss tangent ($\tan \delta$) is related to energy dissipation [2]. One of its definitions is given as the ratio of energy lost to energy stored during one deformation cycle. It can also be identified as the ratio of loss modulus to storage modulus, and the tangent of the angle of phase difference between applied strain and resultant stress in an oscillatory experiment [1]. Since the loss tangent is related to dissipated energy, its peak is related to the glass transition temperature at the test strain rate. The temperature of glass transition, and hence the temperature where the loss tangent peaks, increases with increasing frequency (or increasing strain rate) in Dynamic Mechanical Analysis (DMA) tests, according to the time-temperature superposition principle [3].

We investigate whether we can identify the glass transition condition at higher strain rates than the DMA, which is limited by a maximum frequency of order 100 Hz. A well-known experimental technique that achieves strain rates of $10^2 - 10^4 \text{ s}^{-1}$ is the split Hopkinson pressure bar (SHPB) technique, originally developed to its split form by Kolsky [4], based on Hopkinson's pressure bar technique [5]. The SHPB technique is conventionally used to obtain dynamic stress-strain relationships for a range of materials, including soft and viscoelastic materials, such as polymers [6]. It is possible to identify whether the polymer is glassy or rubbery by the shape of the stress-strain curve, but not to precisely identify the glass transition temperature. Our proposition is to use energy calculations to work out the amount of energy dissipated by the sample, thus obtaining a measure of $\tan \delta$ from SHPB measurements at different temperatures. We investigate the merit of our proposed novel analysis route for SHPB measurements by utilising historical data, collected by Clive Siviour for his PhD thesis [7] on Polycarbonate and HTPB. Provided we obtain distinct peaks in $\tan \delta$ at the expected temperatures using our method, it is then worth optimising our own experiments for making energy loss measurements. The purpose of such experiments would be to increase the range of strain rates over which the glass transition temperature can be accurately identified and hence validate models of time-temperature superposition by considering plots of glass transition temperature against strain rate. The method is also applicable to lower temperature secondary relaxations of polymers, which would be shifted to higher and more accessible temperatures at the high strain rates of SHPB experiments.

In the following sections, we outline the experimental methods used in the collection of the SHPB data and our proposed method of analysis to calculate the loss tangent. We present the resulting curves of $\tan \delta$ against temperature from SHPB, compared with complimentary DMA data of the same materials. We use Group Interaction Modelling (GIM) [8] to identify the peaks of $\tan \delta$ with polymer relaxations and the time-temperature superposition principle to assess whether the peak temperatures we deduce from SHPB are reasonable, given the peaks from the DMA measurements. We use our conclusions from this re-evaluation of historic data to determine the merit of using this

method on our own dedicated campaign of experiments and to identify routes of optimising the setup for the purpose of isolating viscoelastic dissipation.

METHODS

Split Hopkinson pressure bar

In SHPB experiments the specimen is sandwiched between two bars, called the incident and the transmission bar. A striker bar is fired onto the incident bar by means of a gas gun and generates a compressive stress wave, which propagates through the incident bar (F_I). Depending on the impedance mismatch between the bars and the sample, a fraction of the incident wave is reflected back through the incident bar and is detected as the reflected wave (F_R). The remaining fraction of the incident wave is transmitted to the transmission bar (F_T). Strain gauges on the two bars are used to detect the waveforms as illustrated in Figure 1. Subscripts I , R and T are used to refer to quantities derived from the incident, reflected and transmitted wave respectively.

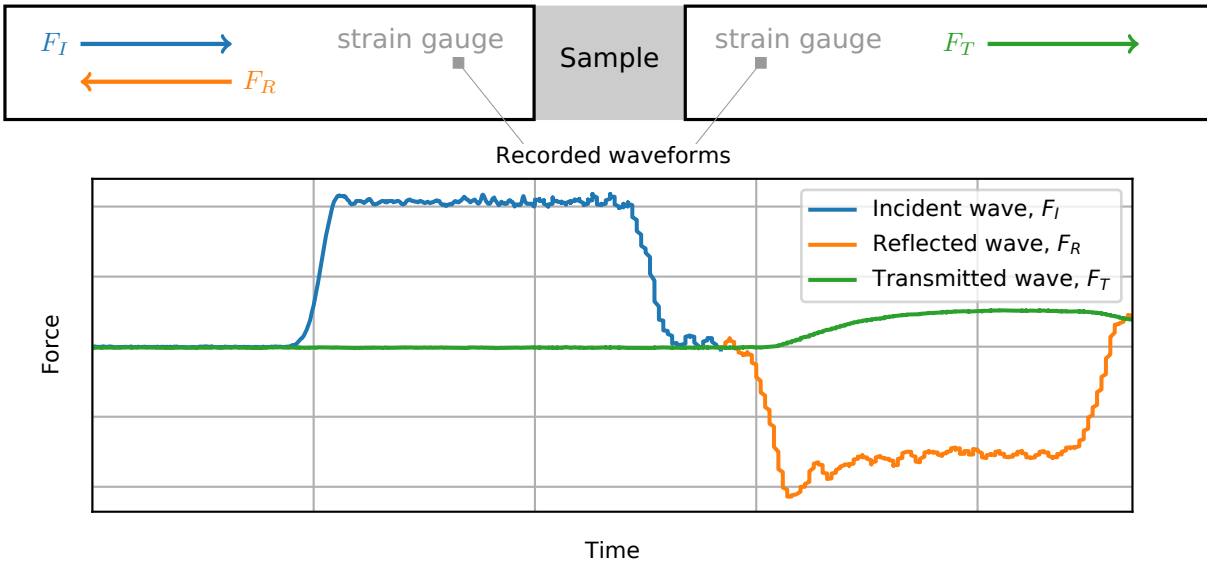


FIGURE 1. Schematic of the waves travelling through the bars in the SHPB setup and a visualisation of the waveforms recorded.

We identify the mechanical energy (W) in each pulse as follows:

$$W_I = \int_{\text{incident pulse}} \frac{F_I^2}{AZ} dt; \quad W_R = \int_{\text{reflected pulse}} \frac{F_R^2}{AZ} dt; \quad W_T = \int_{\text{transmitted pulse}} \frac{F_T^2}{AZ} dt \quad (1)$$

,where A and Z are the cross-sectional area and mechanical impedance of the bars respectively. The force-time profile (F) of each wave is obtained from the voltage detected by the strain gauges, following conventional procedures and analysis, as described by Gray [9]. In the case of an elastic experiment, it is expected that $W_I = W_R + W_T$. A polymer sample, however, dissipates some energy which we identify with an energy loss as follows:

$$W_{\text{loss}} = W_I - (W_R + W_T) \quad (2)$$

To obtain a measure of $\tan \delta$, we need the energy stored in the specimen. The area under the stress-strain curve extracted from the SHPB data, corresponds to the volumetric work done on the specimen. This is a combination of stored energy, viscous loss and plastic work and it is difficult to separate out the components. Nevertheless, provided we keep the final strain of each experiment the same, the volumetric work is proportional to the stored energy:

$$W_{\text{stored}} \propto \int \sigma \cdot \epsilon d\epsilon \quad (3)$$

A measure of the loss tangent is then obtained from the ratio of energy loss to energy stored:

$$\tan \delta \propto \frac{W_{loss}}{W_{stored}} \propto \frac{W_I - (W_R + W_T)}{\int \sigma \cdot \epsilon d\epsilon} \quad (4)$$

The data analysed here are from historical experiments, carried out by Clive Siviour. The strain rates ($\dot{\epsilon}$) achieved in the experiments were circa 3000 s^{-1} for HTPB and 5500 s^{-1} for Polycarbonate. These values were calculated in the usual manner for SHPB measurements [9]:

$$\dot{\epsilon} = \frac{-2F_R}{ZAl_s} \quad (5)$$

where l_s is the length of the sample. Both campaigns included experiments at different temperatures, which allow the construction of $\tan \delta$ curves with temperature.

Dynamic Mechanical Analysis

To assess whether the peaks we obtain from SHPB are reasonable, we compare with a standard technique that provides $\tan \delta$ data, DMA [10]. In this experiment, strain is applied sinusoidally and the resulting force (or stress) is measured. The phase difference δ between the two defines the loss tangent ($\tan \delta$) and the complex modulus ($E^* = E' + iE''$) can be calculated using the stress and strain amplitudes σ_0 and ϵ_0 :

$$|E^*| = \frac{\sigma_0}{\epsilon_0}; \quad E' = |E^*| \cos \delta; \quad E'' = |E^*| \sin \delta \quad (6)$$

We define a representative strain rate for the DMA as the maximum slope of the strain sinusoid at frequency f :

$$\dot{\epsilon} = 2\pi\epsilon_0 f \quad (7)$$

Other definitions for the strain rate of a DMA test in the literature [11, 12, 13, 14] differ to ours by factors of order one. The strain rate throughout the DMA test varies from zero to our given value, so which value ought to be chosen is debatable. According to time-temperature superposition models [15, 16, 17, 18], we expect the glass transition temperature to depend on the logarithm of strain rate, hence a choice of the right order of magnitude is sufficient.

The DMA data on HTPB were taken by Siviour, using a dual cantilever configuration at frequencies 1, 10, and 100 Hz and strain amplitude 0.085%, which correspond to strain rates of 5.34×10^{-3} , 5.34×10^{-2} , $5.34 \times 10^{-1} \text{ s}^{-1}$ [7]. For Polycarbonate we make use of original data taken in our laboratory TA Instruments Discovery 850 machine. The single cantilever geometry was used with frequencies 0.1, 1, and 10 Hz and strain amplitude 0.0226%, corresponding to strain rates of 1.42×10^{-4} , 1.42×10^{-3} , 1.42×10^{-2} . A frequency sweep on a temperature ramp was carried out, so that we could construct curves of $\tan \delta$ with temperature at different frequencies.

RESULTS

The summary of loss tangent data for the two materials studied is presented in Figure 2. The y axis is scaled in both cases, since the curves generated from SHPB are proportional to $\tan \delta$; they are not absolute measurements. The measure of loss tangent from SHPB is missing volumetric factors and is likely to contain additional losses, such as plastic work and energy dissipated by mechanisms other than viscoelastic dissipation within the sample (e.g. friction with experimental components). Any peak it shows however, can only be explained by the viscoelastic dissipation being maximum upon the glass transition or other secondary transitions. Therefore, despite any upward shifts and scaling in the SHPB data relative to DMA $\tan \delta$ curves, we can readily derive the peak temperatures.

We use Group Interaction Modelling (GIM) to fit these $\tan \delta$ curves and identify the peaks as the glass transition temperature (T_g) and the beta transition temperature (T_β) in Polycarbonate. The peaks are fitted to Gaussian distribution functions centred on the transition temperatures, with standard deviations σ_g and σ_β respectively, in accordance to previous work on GIM [2]:

$$\tan \delta = \tan \delta_b + A \exp \left[-\frac{(T - T_g)^2}{2\sigma_g^2} \right] + B \exp \left[-\frac{(T - T_\beta)^2}{2\sigma_\beta^2} \right] \quad (8)$$

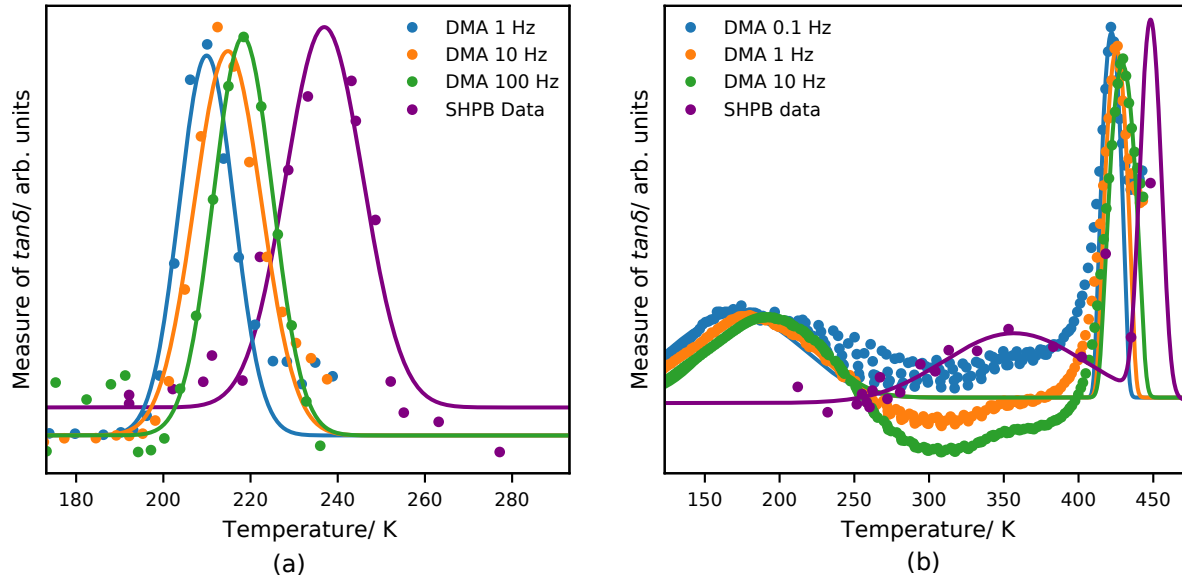


FIGURE 2. The ratio of energy lost to energy stored calculated from SHPB data and loss tangent obtained from DMA are plotted against temperature for (a) HTPB and (b) Polycarbonate. The y-axis is scaled for ease of comparison.

The solid lines in Figure 2 are fits of the data to the expression above, where we fit for peak temperatures, widths, and constants A and B . The background loss tangent $\tan \delta_b$ was fixed at a representative constant for each case. The paper by Porter and Gould [2] contains details on how to estimate such a value from the model, and how the loss tangent is described within GIM. We have identified two peaks for Polycarbonate, the glass transition and the lower temperature beta transition. Our treatment of the SHPB data reveals both peaks, although the glass transition peak is incomplete (ideally we would require data at higher temperatures as well). The data for HTPB shows a clear peak that we can identify as the glass transition peak, and no secondary peaks, so B is set to zero.

DISCUSSION

To assess whether the magnitude of the shifts of $\tan \delta$ curves of SHPB data relative to DMA data is plausible, we use time-temperature superposition. According to this principle, there is an equivalence of increasing the strain rate at fixed temperature to reducing the temperature at a fixed strain rate [3]. Hence, we expect the whole curve with the peak, to shift to higher temperatures, or to the right, with increasing strain rate, as observed in Figure 2.

In Figure 3 the assessment is shown visually by plotting the peak temperatures against strain rate (calculated using Equations 5 and 7) on a logarithmic scale. We estimate the shift factors using a linear relationship between the temperature shift and the logarithm of strain rate, an approach similar to that taken by Siviour et al. [18] to map different datasets to each other, for example yield stresses at different temperatures and strain rates, using shift factors obtained from DMA. The premise is the same here, but we are using the position of the transition peaks to indicate shifts on the temperature scale. The magnitude of the shift in temperature per decade we find is in agreement with rule-of-thumb values of 4 K/decade for the glass transition and 10 K/decade for secondary transitions of polymers. Typical values calculated using GIM are also in agreement with the shifts seen here [2]. Our shift factors for Polycarbonate agree with Siviour's [18] within error, except the beta transition peak deduced from SHPB is shifted more than the DMA peaks relative to each other. Siviour's linear fit approach works well for a reasonable range of strain rates, but when several decades are covered, we expect that the actual relationship to have some curvature. Models such as Williams-Landell-Ferry [15], Vogel-Fulcher-Tammann [16, 17] and Arrhenius type approaches [19], do actually have curvature, so the increased shift at higher strain rates is a plausible behaviour. The actual value of the shift and the curvature of the relation depends on the material, the type of transition, and the strain rate considered.

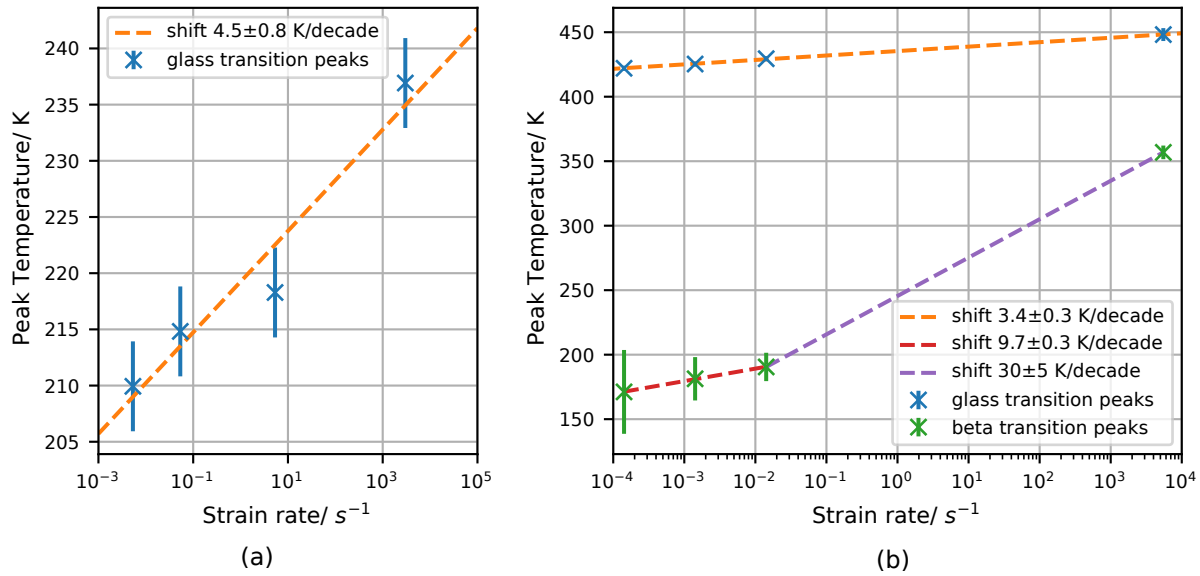


FIGURE 3. Peak temperatures of $\tan \delta$ curves of (a) HTPB and (b) Polycarbonate, plotted against strain rate for each test.

CONCLUSION

A novel methodology of SHPB data analysis has been demonstrated, that allows for identification of the amount of mechanical energy dissipated and stored by the specimens. The ratio of the two, which is proportional to the loss tangent in a DMA experiment, allows for the determination of loss peaks, and hence the glass transition and secondary transition temperatures at high strain rates. We were able to construct SHPB curves of $\tan \delta$ with temperature that look like DMA curves of the same, albeit shifted to higher temperatures, by a magnitude commensurate with what we would expect. Our analysis has demonstrated that this idea has merit to be used with our future experimental results.

OUTLOOK

From the analysis presented in this paper we have identified points to be mindful of when conducting SHPB experiments specifically for the purpose of extracting a loss peak. Firstly, we should ensure that the duration of experiments (pulses) is short compared to the useful record time of our SHPB system, to allow integration over the waves before any wave overlap occurs. If the desired focus is on viscous loss, one should minimise the plastic work, which in practice means keeping the final strain relatively small (<10%) and consistent throughout the series. Minimising additional losses should remove any vertical shifts present between the SHPB and DMA curves. Finally, to characterise Gaussian loss peaks, we should gather data at temperatures on either side of the peaks. This is on the immediate plans to carry out for a more reliable identification of the glass transition peak of polycarbonate at high rates. A more detailed comparison with time-temperature superposition models will be carried out with our own set of data and an investigation of the curvature of the relations between transition temperatures and strain rate.

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