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HIGH-PRESSURE MECHANICAL PROPERTIES OF
POLYMETHYLMETHACRYLATE

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HIGH-PRESSURE MECHANICAL PROPERTIES OF POLYMETHYLMETHACRYLATE

ABSTRACT

Equation of state data, including pressure-volume, static and dynamic elastic constants, failure envelope (uniaxial compression) and uniaxial strain in loading and unloading tests are presented for polymethylmethacrylate (PMMA), ($\rho_0 = 1.185 \text{ g/cc}$).

Compressibility to 40 kb is found to be relatively rate independent over the range 10^{-5} to 10^{-3} sec. In contrast, moduli that are dependent upon differential stresses and strains, and the shear strength beyond the elastic limit, appear to be very strain-rate sensitive.

INTRODUCTION

The mechanical properties of polymethylmethacrylate (PMMA) have received considerable study due to its wide use of many applications. The mechanical properties of PMMA at high pressure have been investigated here because of its use in modeling studies using high explosives (HE). Several shock wave studies have been carried out on PMMA, the most recent being that of Barker and Hollenbach¹ and Schuler.² Properties of PMMA may vary somewhat from different sources. The HE studies,³ the work of Barker and Hollenbach,¹ and our measurements were all performed using Rohm and Haas PMMA (initial density 1.185 g/cm^3). The results should therefore be directly comparable.

The equation of state (EOS) measurements described here include: hydrostatic pressure-volume (P-V) data to 6 kb, quasi-hydrostatic P-V measurements to 40 kb, the failure surface to 7-kb confining pressure, acoustic compressional and shear velocities to 10 kb, and three-dimensional, stress-strain measurements along various loading and unloading paths to 10 kb.

EXPERIMENTAL

Details of the experimental technique used for these measurements are discussed in Refs. 3 through 8. Briefly, strengths are determined from differential stress-axial strain measurements on jacketed cylindrical samples compressed to failure at various confining pressures. Strengths from these curves are then used to define the shear-stress, mean-pressure, failure envelope. Acoustic velocities are obtained by coupling the sample to pulsed electro-mechanical transducers with a resonant frequency of 1 MHz

and measuring the time-of-flight in the specimen. Precise three-dimensional stress-strain measurements are obtained with foil-strain gauges cemented to jacketed samples. These tests yield elastic and inelastic deformation moduli (coefficients relating stress and strain) over a range of stress states below and up to the failure envelope. These moduli include the effective shear moduli and Poisson's ratio and the compressibility in both hydrostatic and non-hydrostatic states. Pressure-volume data are also obtained using a piston-cylinder device to compress the sample in a quasi-hydrostatic environment to 40 kb using lead as the pressure transmitting medium.

PRESSURE-VOLUME MEASUREMENTS

Volume compressibility data for PMMA are shown in Figs. 1 and 2. The hydrostatic data are shown as filled points in Fig. 1 and the 40-kb quasi-hydrostatic data, which are less sensitive at low pressures, are shown as open points. The initial compressibility was found to be 22.7 Mb^{-1} . It may be seen that the 40-kb data are in good agreement with the hydrostatic results. Higher pressure compressibilities are shown in Fig. 2 with the earlier data of Stromberg⁹ also included. Figure 2 presents the results of a limited study of the effect of strain rate of compressibility. In this figure, no change in the β_V is apparent (within the experimental error, $\pm 5\%$) by varying $\dot{\epsilon}_V$ almost two orders of magnitude: 3×10^{-3} to $5 \times 10^{-5}/\text{sec}$. These measurements were made at a constant stress rate with the indicated strain rates being the average values for the first 10 kb.

The compressibilities were integrated to yield the P-V curve shown in Fig. 3 and compared with the results of Stromberg.⁹ The compressibilities from this work as well as all P-V data are also summarized in Table 1.

Table 1. P-V data^a for PMMA with initial density (ρ_0) = 1.185 g/cm^3 .

P kb	v g/cm^3	v/v_0	β_V Mb^{-1}	P kb	v g/cm^3	v/v_0	β_V Mb^{-1}
0	0.8441	1.000	22.7	17	0.7100	0.8411	5.27
1	0.8266	0.9793	19.64	19	0.7030	0.8328	4.83
2	0.8116	0.9615	17.04	21	0.6935	0.8216	4.48
3	0.7984	0.9459	14.88	23	0.6905	0.8180	4.15
4	0.7871	0.9325	13.01	25	0.6849	0.8114	3.88
5	0.7778	0.9215	11.62	27	0.6798	0.8054	3.62
6	0.7693	0.9114	10.53	29	0.6752	0.7999	3.42
7	0.7616	0.9023	9.72	31	0.6705	0.7943	3.23
9	0.7484	0.8866	8.20	33	0.6663	0.7894	3.07
11	0.7369	0.8730	7.11	35	0.6621	0.7844	1.92
13	0.7268	0.8610	6.37	37	0.6583	0.7799	1.78
15	0.7180	0.8506	5.77				

^aLoading and unloading are identical.

Fig. 1. Compressibility vs pressure to 6.5 kb (hydrostatic).

Fig. 2. Compressibility vs pressure to 38 kb (quasi-hydrostatic).

Fig. 3. P-V data to 40 kb.

FAILURE PROPERTIES (COMPRESSION)

Most materials tested under triaxial loading conditions (i.e., compression, extension, torsion under pressure) show an increase in strength that is usually accompanied by an increase in ductility (the amount of strain before fracture) with increasing confining pressure.¹⁰ The PMMA appears to have a normal strength behavior but an abnormal ductility dependence with pressure. In Fig. 4, the maximum principal stress (σ_1) dependence is compared to the confining pressure (σ_3) as determined in the uniaxial compression test.* Each data point (cross) is based upon both the differential stress and the confining pressure and is taken from each of the stress-strain curves compressed at a strain rate of 10^{-4} /sec. If the individual curve achieved at least 5% permanent strain beyond the yield point, the behavior was judged ductile and σ_1 was calculated at this strain; if the test was terminated by fracture at a lower strain, the response was judged as brittle and σ_1 calculated as above. All data are summarized in Table 2. Inspection of Fig. 4 shows a very nearly linear increase of σ_1 with σ_3 according to

$$\sigma_1 = 1.35 + 1.35\sigma_3,$$

σ in kb.

Table 2. Uniaxial compression data for PMMA.

σ_1 (kb)	σ_2 (kb)	σ_3 (kb)	Behavior	τ (kb)	P_m (kb)	σ_1 (kb)	σ_2 (kb)	σ_3 (kb)	Behavior	τ (kb)	P_m (kb)
1.34	0	0	Ductile	0.67	0.45	6.86	4.25	4.25	Ductile	1.31	5.12
1.35	0	0	Ductile	0.68	0.45	7.02	4.25	4.25	Ductile	1.39	5.17
1.62	0.25	0.25	Ductile	0.69	0.71	7.34	4.50	4.50	Brittle	1.42	5.45
1.95	0.50	0.50	Ductile	0.73	0.98	6.43	4.50	4.50	Brittle	0.97	5.14
2.35	0.77	0.77	Ductile	0.79	1.43	7.92	5.00	5.00	Brittle	1.46	5.97
3.33	1.51	1.51	Ductile	0.91	2.12	6.40	5.00	5.00	Brittle	0.70	5.47
4.04 ^a	2.00 ^a	2.00 ^a	Ductile ^a	1.02 ^a	2.68 ^a	9.28	6.00	6.00	Brittle	1.64	7.09
4.14	2.00	2.00	Ductile	1.07	2.71	8.65	6.00	6.00	Brittle	1.33	6.88
5.33 ^b	3.00 ^b	3.00 ^b	Ductile ^b	1.17 ^b	3.78 ^b	9.28 ^b	6.00 ^b	6.00 ^b	Brittle ^b	1.64 ^b	7.09 ^b
5.41	3.05	3.05	Ductile	1.18	3.84	6.89 ^b	6.00 ^b	6.00 ^b	Brittle ^b	0.30 ^b	6.30 ^b
6.02	3.50	3.50	Ductile	1.26	4.34	10.76	7.00	7.00	Brittle	1.88	8.25
6.53	4.00	4.00	Ductile	1.27	4.84	8.91	7.00	7.00	Brittle	0.96	7.64
6.75	4.00	4.00	Ductile	1.38	4.92						

^aCycled to 7 kb before test.

^bUnjacketed.

*In this case, $\sigma_1 > \sigma_2 = \sigma_3$ and $d\sigma_3 = 0$ with σ_1 parallel to the test cylinder axis and compression taken as positive.

Fig. 4. Principal stresses σ_1 and σ_3 at failure.

It is important to note that at a pressure of about 4.3 kb, the initial ductile behavior of PMMA disappears and the material becomes brittle. The open circles in this region show the level of σ_1 immediately after fracture. An identical response has also been reported for PMMA tested in torsional loading under confining pressure.¹¹ In torsion, the ductile-brittle transition occurs at a pressure somewhat greater than 5 kb (based on similar criteria). The relative increase in strength and transition in failure behavior with increasing σ_2 as measured in compression and torsion tests has also been observed¹² in other materials: e.g., calcite [CaCO_3] and dolomite [$\text{CaMg}(\text{CO}_3)_2$]. The abrupt change from ductile to brittle behavior near 4.3 kb reflects the pronounced (but not discontinuous) change in physical properties observed in many polymers at the "glass transition."^{13,14} Rubber also behaves similarly, showing a ductile-brittle transition at the glass transition pressure.¹⁵ This effect does not seem to be a surface phenomena nor is it apparently dependent upon the pressure history as neither pressure cycling or jacketing of the test samples changed the overall response.

Figure 5 illustrates the failure envelope for PMMA as plotted in shear-stress, mean-pressure space based on the test results used for Fig. 4. No inflection is evident at the ductile-brittle transition.

No tests were undertaken specifically to evaluate the effects of temperature or the rate of deformation on the mechanical behavior of PMMA at strains well beyond the elastic limit. However, others¹⁶ have measured these effects at moderate temperature and atmospheric pressure. PMMA deformed to large strains at high pressure can be expected to be very sensitive to both strain rate and temperature.

STRESS-STRAIN PROPERTIES AT STRESS STATES BELOW THE FAILURE ENVELOPE

Nonlinearity in the stress-strain data for PMMA makes it necessary to calculate moduli using stress and strain increments representative of a general state of stress. Although the material may be inelastic, and thus the moduli cannot have the conventional assumption of elasticity associated with them, they are still useful as coefficients relating stress to strain. Typical results for PMMA are shown in Fig. 6 where axial stress is plotted against both circumferential and axial strain for a sample loaded at 1-kb confining pressure (σ_3) in uniaxial compression.

The sample was first loaded to 1.5-kb axial stress and then unloaded, followed by loading to 2.0-kb σ_1 and unloading followed in turn by reloading to 2.6 kb. There is a pronounced nonlinearity of the loading path followed by hysteresis and permanent offset upon unloading. When the PMMA is reloaded, the initial stiffness to shear stress is recovered and the new loading curve parallels the original. Thus, the processes responsible for the nonlinearity of the curve do not affect the stiffness to shear on subsequent loading cycles. The initial μ calculated from the data in Fig. 6 is 0.013 Mb, somewhat higher than the value (~ 0.008 Mb) measured in torsion by Rabinowitz *et al.*¹¹ The value for μ from Fig. 6 is calculated at a differential stress of 200 bar while that of

Fig. 6. Axial stress vs axial and circumferential strain at 1 kb confining pressure (σ_3).

Rabinowitz may be at slightly higher stress differences where pronounced inelastic effects have begun to take place.

The shear modulus derived from the data in Fig. 6 decreases with increasing shear stress from 0.013 Mb to 0.006 Mb at 1500-bars $\sigma_1 - \sigma_3$. Poisson's ratio increases from 0.35 to 0.44 over the same interval. We measure an initial value for μ of 0.012 Mb unconfined, and 0.014 Mb at 2-kb confining pressure.

If the volume strain $(2\epsilon_r + \epsilon_\ell)^*$ observed during the last loading cycle shown in Fig. 6 is plotted against mean pressure ($3P_m = \sigma_1 + 2\sigma_3$), then it appears the effective compressibility is dependent on the amount of shear stress present. As the stress difference increases, volume is observed to decrease beyond that measured in hydrostatic ($\sigma_1 = \sigma_3$) compression (Fig. 7). Furthermore, this is observed to be a time-dependent process, with volume strain increasing with time at constant shear stress.

Examination of the moduli derived from acoustic velocity measurements (Table 3) would seem to indicate that the shear stress-strain and mean pressure-volume relations are (assuming a perfect defect-free solid) strain-rate dependent. The different initial derived μ (0.023 Mb) and β_V (17 Mb^{-1})[†] might be due to the low levels of stress ($< 10^6$ bar) involved during the passage of the 1 MHz acoustic wave. However, it has been demonstrated that while the hydrostatic compressibility is not noticeably strain-rate dependent over a range of laboratory strain rates, the shear-strain response to shear stress is

Table 3. Acoustic velocities and derived moduli PMMA.

P	ρ (g/cc)	V_P (km/sec)	V_S (km/sec)	K (Mb)	μ (Mb)	ν	E (Mb)
0	1.185	2.76	1.40	0.0589	0.0231	0.327	0.0613
0.50	1.195	2.87	1.44	0.0651	0.0245	0.333	0.0653
1.00	1.204	2.97	1.47	0.0710	0.0257	0.338	0.0689
2.00	1.219	3.15	1.53	0.0829	0.0263	0.347	0.0763
3.00	1.234	3.33	1.59	0.0945	0.0309	0.353	0.0837
4.00	1.246	3.47	1.64	0.1045	0.0334	0.355	0.0906
5.00	1.257	3.60	1.69	0.1147	0.0357	0.359	0.0970
6.00	1.258	3.72	1.73	0.1245	0.0378	0.362	0.1030
7.00	1.278	3.84	1.77	0.1342	0.0399	0.365	0.1088
8.00	1.287	3.94	1.81	0.1429	0.0419	0.366	0.1146
9.00	1.296	4.04	1.84	0.1521	0.0439	0.368	0.1202
10.00	1.304	4.13	1.88	0.1604	0.0450	0.369	0.1260

* ϵ_r is the radial strain, ϵ_ℓ is the axial strain.

† Compared to a β value of 23 Mb^{-1} from the hydrostatic compressibility data (Figs. 1, 2).

Fig. 7. Mean pressure vs volume strain in hydrostatic loading and uniaxial compression from 1-kb σ_3 .

very much dependent. That is, when an increase in the mean pressure is primarily due to an increase in shear stress, the moduli are markedly affected and appear time dependent. This observation suggests that PMMA is inherently more inelastic and strain-rate dependent in response to shear stress than to linear stress. This is supported (Table 4) by the observed decrease in Lamé's constant ($\lambda = \sigma_1/\epsilon_r$) of 25% between the acoustic and the static stress-strain results while the shear modulus

$2\mu = (\sigma_1 - \sigma_3)/(\epsilon_\ell - \epsilon_r)$ decreases by almost 50%. The former is a measure of the response of the substance to linear stress only while the latter is a function solely of shear stress. This may explain why no discernible strain-rate dependence was observed in the hydrostat, where shear stresses were not present.

Table 4. Comparison of moduli derived from dynamic and determined by static techniques.

	λ (Mb)	μ (Mb)
Static (initial loading)	0.033	0.012
Dynamic (1 MHz)	0.044	0.023

When PMMA is loaded in compression under conditions of uniaxial strain ($\sigma_1 > \sigma_3$; $d\epsilon_r = 0$) so as to simulate the conditions thought to exist upon shock loading, the path shown in Fig. 8 is followed. As observed in the sample loaded in uniaxial compression (Fig. 6), an increasingly inelastic behavior occurs with an increase in shear stress and the loading path begins to deviate away from the failure envelope. This is reflected by an increased compressibility, as shown in Fig. 7, when shear stresses are present (Fig. 9). The shear modulus decreases from 0.013 Mb upon initial loading to 0.008 Mb at a stress difference of about 1 kb. Under conditions of actual shock loading, the pronounced inelasticity that is time and therefore strain-rate dependent should be much diminished. If loading were perfectly elastic with the moduli dictated by the acoustic measurements, then Poisson's ratio ($\nu = -\epsilon_r/\epsilon_\ell$) would define the loading slope. The acoustic velocity data should reflect the actual initial shock-loading slope, while the material is still deforming elastically. The agreement between the acoustic compressional velocity (2.76 kb/sec, Table 3) and the initial shock velocity (2.76 km/sec) as determined by Schuler² suggests that this is indeed the case. As is shown in Fig. 8, the loading slope ($\nu = 0.32$) intersects the failure envelope (5% axial strain at a strain rate of 10^{-4} /sec) at about 1.7 kb, while the material is still ductile. If this were the true shock loading path, loading would continue beyond this point because failure would not be expected until much higher mean pressures at the higher rate of strain.

Ultimate failure should take place when the actual loading path either intersects the failure surface at the ductile-brittle transition or at some point in the brittle range, depending on which has the stronger strain rate dependence. An increased strain rate can be expected to lower the ductile $\rightarrow$ brittle transition pressure and at the same time, raise the entire failure envelope to higher shear stresses. Duckett *et al.*¹⁶ have shown that the yield strength at 1 atm is doubled by increasing the strain rate from 10^{-4} to 1/sec. Thus the failure envelope data presented here is not applicable to shock-wave

Fig. 8. Comparison of uniaxial strain loading and failure envelope.

Fig. 9. Mean pressure vs volume strain in hydrostatic loading and uniaxial strain loading.

processes in PMMA, and one would expect to observe shock loading to significantly higher stresses above the hydrostat than would be indicated by the data in Fig. 5. Shock-wave data of Barker and Hollenbach, when compared with our hydrostatic data (Fig. 10), indicate a shear strength (τ) of approximately 3 kb (where $\sigma_1 = 4/3\tau + P_m$). This suggests that the yield strengths (at the strain rates encountered) are more than twice those determined in this study.

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Fig. 10. Axial stress determined in shock loading compared with the hydrostat.

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