# Properties of Densified Amorphous Polystyrene

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## Synopsis

Atactic polystyrene was subjected to an elevated pressure–temperature cycle with the resulting densification, mechanical properties, and thermal scanning behavior observed. Most densifications were carried out with the PST as a viscous liquid. In this manner, ambient residual compactions greater than 2% were produced. Pressures up to 90,000 psi and temperatures to 320°C were employed. The technique used for vitrification from the high pressure–temperature region was found to drastically affect the mechanical behavior. If the polystyrene was vitrified from the treatment region by lowering the temperature, the material exhibited enhanced yield strength, by up to 40%. If the polystyrene was quenched by raising the pressure, the samples exhibited much lower mechanical strength. While the mechanical behavior of temperature-vitrified samples is enhanced compared to the pressure-vitrified materials, their densities are comparable. The compaction achieved is primarily determined by the pressure applied as the polymer vitrifies. Thermal scanning behavior of the pressure-vitrified materials show endothermic and exothermic responses below $T_g$, while the temperature-vitrified materials do not. Annealing the compacted polystyrene at room temperature caused little change in density. However, at temperatures above 60°C, the density relaxed rapidly. Samples which had been temperature vitrified and annealed such that the compaction completely relaxed, still maintained the enhanced mechanical properties of the densified materials.

## INTRODUCTION

### Densification Behavior

By proper manipulation of temperature and pressure the densification of polystyrene, or of any polymer, may be accomplished. Until recently, most densification studies have been concerned with temperature effects only (annealing) which produce relatively small density changes. Some orientation studies have also been employed.

Brady and Yeh[1] used a general-purpose atactic polystyrene of molecular weight approximately 250,000. This material is comparable with the material used in this research. The effect of extended annealing time was to increase the density by about 0.04%. Densification by orientation of the polymer molecules using a hot stretch was also employed. In this case, densifications up to 0.19% were obtained, but the material lost its isotropy due to the aligning of the polymer molecules in the direction of stress.

Manabe[2] studied the rate of quenching on the density of PST. Specific characterization of the commercial polystyrene used was not given, but they did demonstrate that the density of polystyrene was influenced by quenching history. A very fast quench was found to produce a lower density than a slower one.

Most pressure-related densification data has been collected in conjunction with the determination of pressure effects on the glass transition. O'Reilly[3] has critically reviewed much of the literature to 1964, and Gee[4] through 1970. In 1971, Quach and Simha[5,6] studied the effect of vitrification mode on the pressure dependence of the glass transition of polystyrene.

Two types of pressure-related densification can be distinguished: those concerned with sample densities while in a hydrostatic pressure environment and those dealing with a residual compaction at ambient conditions. Typical data of the former type show densifications of 10% for polystyrene at 7 kilobars and 250°C.[7] Similar decreases in specific volume of about 10% to 12% have been found in many other polymers subjected to hydrostatic pressures of 10 kilobars or so.[8,9]

Residual densification of polystyrene of 1.3% and 0.55% have been reported by Manabe[2] and Allen.[10] Allen vitrified the polystyrene from 1.2 kilobars, while Manabe's report did not specify the pressure history. In general, increases of up to 3% are expected,[9] depending on pressure and temperature levels. Price et al.[11] vitrified polystyrene from as high as 3 kilobars and reported a density increase of about 1%. They also measured heats of solution of the densified polystyrene which they related to thermodynamic aspects of the glassy state.

One significant aspect of the pressure densification studies was that the materials being densified were above their glass transition temperature at the given pressure. This was necessary in order that maximum densification take place. If densification were attempted below $T_g$, very little compaction may be expected. Kimmel[12] has shown that compaction of the glassy state results in densification of only a few tenths of a percent.

## Densification and Mechanical Properties

Two categories of experimental results are distinguishable concerning the effect of density on polymer mechanical properties. First are mechanical property data obtained in situ at high hydrostatic pressures. Secondly, and concerning this work, are those data taken after pressure treatment has been completed.

Data of the former type have been taken on a wide variety of polymers and are summarized elsewhere.[13,14] The results vary from material to material and are also a function of the contacting pressure-transmitting medium. As one example, polystyrene becomes more ductile at high pressures if mineral oil is used as the contacting medium,[14] but remains brittle when surrounded by mercury.[15]

Little mechanical property data taken after pressure–temperature treatment may be found in the literature. Matsuoka and Maxwell[16] have

examined the dynamic mechanical behavior of pressure-densified crystalline (polyethylene) and semicrystalline (isotactic polystyrene and polypropylene) polymers. A number of relaxation studies involving the effects of temperature and time on densified materials have also been carried out.[2,11,17,18]

Regarding the effect of densification on stress–strain properties, Dale and Rogers[19] have examined the relationship between residual density changes and mechanical properties of polystyrene. The polystyrene used in their research was Dow Chemical's Styron 690, a thermally polymerized atactic polymer of viscosity-average molecular weight about 240,000 and glass transition temperature of 96°C. The molecular weight is similar to the material used in this work. Their polystyrene was molded at temperatures of 200°C and pressures up to 3000 atm, with densification of about 1% attained. Compression stress–strain specimens were then machined from the molded polymer. The observed the compressive yield stress to go through a maximum for molding pressures between 1 and 2 kilobars. A subsequent publication showed that the transport properties of the compacted films went through a minimum in this pressure range.[19] No explicit densification data are given except a relative comparison between polystyrene molded at 1 kilobar and the control. Several conclusions, not related to the yield stress maximum, were also described. The densified materials were found to exhibit lower strain to yield, faster mechanical relaxation after compressive yield, and, in a DSC experiment, an exothermic region below the glass transition.

## EXPERIMENTAL

### Materials and Premolding

The polymer material used in the experimental work was a general-purpose atactic polystyrene in bead form obtained from Sinclair-Koppers Company. Its characterization is presented in Table I. Before molding, the polystyrene beads were dried in a vacuum oven overnight at about 80°C.

The beads, after drying, were then placed into a stainless steel piston-cylinder arrangement shown schematically in Figure 1. Four such holders were machined as duplicates of one another. Preparation of these holders involved drying under vacuum at about 130°C, spray-coating with Teflon mold-release agent, wiping off excess Teflon, and screwing the separate parts together. The purpose of the premolding was to form the dried polystyrene beads into an integral specimen shape. The resultant specimen was then used either as an atmospheric sample or subjected to pressure–temperature treatment for densification.

The following procedure was employed to accomplish premolding. The dried polystyrene beads were poured into the cavity of a sample holder which was placed upright (using a stand) in a large vacuum desiccator. The vacuum desiccator was placed in an oven (nitrogen blanketed) where a
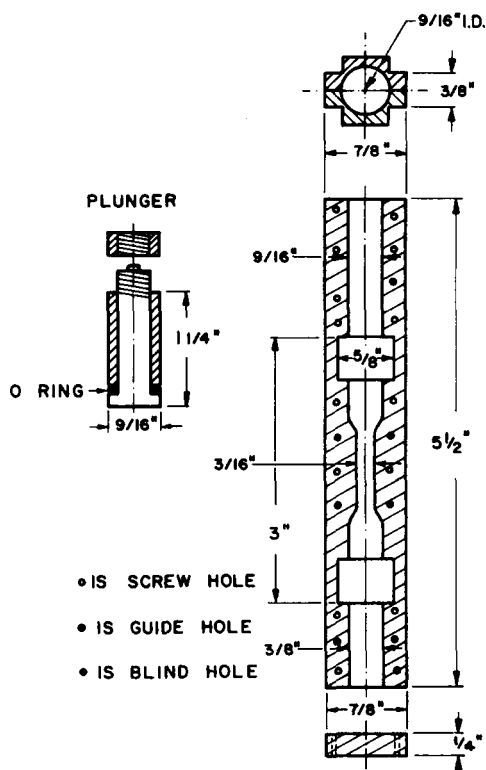
Fig. 1. Design of sample holder.

TABLE I
Polystyrene Characterization[a]

| Parameter | Value |
| --- | --- |
| Specific polymer name | Dylene polystyrene 8 KPD-1037 Crystal Beads (unmodified) |
| Molecular weight | |
| a. experimental intrinsic viscosity[b] | 0.90 dl/gr |
| b. intrinsic viscosity[c] | 0.86 dl/gr |
| c. number-average molecular weight | 110,000 |
| d. weight-average molecular weight | 274,000 |
| e. heterogeneity index | 2.5 |
| Melt flow (Condition L)[d] | 5.0 gms/10 min |
| Vicat softening point | 106°C |

[a] This characterization was conducted by Sinclair-Koppers Company and made available by Dr. E. J. Romigh of that organization.

[b] This result was obtained in toluene at 30°C.

[c] This result was calculated from gel permeation data, using toluene at 30°C.

[d] ASTM Standard Test, D 1238-70.

vacuum drawn. The oven was turned on and a temperature of 210°C reached in about $1^1/_2$ hr and held for two additional hours. The temperature in the sample holders reached approximately 170°C, as determined by a thermocouple actually in the sample holder fed through a vacuum connection. The vacuum pump was then shut off and the desiccator system allowed to reach atmospheric pressure by allowing the nitrogen to leak in slowly. The desiccator was then opened and the oven shut down. While still at 170°C and in the nitrogen environment, the plunger was placed into its receptacle. The holder was then quickly removed and a hydraulic press used to force the plunger down until a small back pressure was achieved. The sample holders were placed back into the oven and allowed to cool slowly over a period of about 5 hr.

Four holders could be premolded at one time in this manner. The final samples produced had a shape which conformed to the configuration of the sample holders in which they were molded. A typical sample is shown in Figure 2. Ultimately, section A was sawed off and broken up into density gradient specimens, discussed later; section B is used as a tensile specimen; or section C may be detached for compression testing.

## High-Pressure System

A high-pressure intensifier system and pressure vessel was purchased from Harwood Engineering Company. The system is capable of reaching pressures of 200,000 psi, or 13.8 kilobars. A 1 in. × 9 in. cylindrical cavity was provided in the pressure vessel. A water–ethylene glycol mixture was used as the pressure-transmitting medium.

The polymer sample holders, after premolding, were placed in the cylindrical cavity of the high-pressure vessel. The vessel itself is about 15 in. in height by 6 in. in diameter, with a type 316 stainless steel inner core of 4 in. diameter surrounded by a shrink-fitted carbon steel safety jacket.

Samples are placed into this cavity by removing a closure assembly located on the top surface. Sealing of the system is accomplished in two ways: by metal cone-shaped joining pieces (60° at each end) for the piping system, and by a wedge-ring seal for the closure assembly. Details of the high-pressure system and closure assembly may be found elsewhere.[13]

A high-pressure Heise gauge was used to register pressures up to 100,000 psi. The instrument features a "unitized" Bourdon tube (inlet, spring
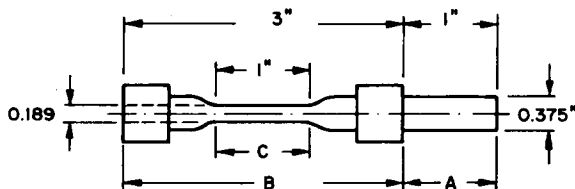


Fig. 2. Dimensions of molded specimens. Region A broken up and used for density gradient specimens. Region B used as tensile specimen, effective gauge length 1.25 in. Region C used for compression specimens.

portion, and bleeder are fabricated in one piece), so that leakage and bond-
ing effects are eliminated. A thermal compensator is provided which
maintains the gauge in calibration through ambient temperature changes
from $-32°C$ to $+52°C$. According to the manufacturer, the Heise gauge
is accurate to within $0.1\%$ of full-scale reading after application of maxi-
mum scale pressure. The Heise gauge was used for all experiments in
which the maximum pressure did not exceed 100,000 psi.

Two systems were employed to accomplish heating of the pressure bomb:
an oil system for low temperature work (less than $200°C$) and a furnace
system for high-temperature work. The resultant temperature rise was
sensed with thermocouples at various locations. The oil bath was heated
with electric band heaters placed on the outside wall of the bath vessel.

A furnace system was employed to obtain temperatures in excess of
$200°C$. Nichrome wire (#13) was wrapped around a $6^1/_{32}$-in.-inner diam-
eter, $3/_8$-in.-thick piece of grooved ceramic. This piece just fit over the
6-in.-outer diameter of the high-pressure bomb and was about 12 in. high.
The total heat output was about 7000 watts (or 24,000 BTU/hr). At the
maximum rate, a bomb temperature of $300°C$ could be reached in approxi-
mately 30 mins.

The temperature of the bomb cavity and surfaces was monitored con-
tinuously. A thermocouple, encased in a stainless steel sheath ($1/_{16}$ in. in
diameter), protruded through the closure assembly to a depth of 1 in. into
the cavity. This sheath was kept from leaking by a cone-shaped piece of
steel welded to the sheath—the apex facing away from the pressure. A
boron nitride gasket was placed over the cone-shaped piece. Subsequently,
when pressure was applied, the gasket was crushed between the cone and
the matching body of the closure, thereby producing a leak-tight seal.
Other thermocouples were placed at various locations on the surface of the
bomb and in the bath vessel.

## Densification Procedure

After premolding, the sample holder was placed into the bomb cavity for
pressure–temperature treatment. The following procedure was followed.
The closure assembly was threaded in until the wedge ring seals contacted
the cavity surface and the system was pressurized to some low value (about
2000 psi) to eliminate any large air pockets and to avoid any steam forma-
tion at the higher temperatures. The heaters were then turned on and
the system allowed to heat up until the desired temperature level was
reached. The pressure was maintained at 2000 psi. The system was then
pressurized and held at that pressure for about 30 min. As pressure is
applied, the plunger moves into the syringe receptacle (0.001 in. clearance),
thereby compressing the viscous polymer to equalize the pressure. The
length of the plunger travel is relatively small (less than $1/_8$ in.), but a
dynamic seal was necessary. This seal was accomplished with a special
nonswelling (a few per cent at most) O-ring made of carboxy-nitroso rubber

(CNR). Viton O-rings were also employed, but at the lower temperatures and pressures (less than 250°C). The temperature was then lowered to ambient while maintaining the high pressure. Finally, the pressure was reduced to atmospheric, and the holder removed by unscrewing the closure and lifting out.

After this procedure had been completed, the sample was removed from its holder by splitting the halves apart using the blind holes shown in Figure 1. The holder was then cleaned and polished before its next use.

### Density Measurement

The pressure–temperature treatment resulted in the preservation of densification for study at ambient conditions. Increases in density of from a few tenths to three per cent were observed, depending on temperature and pressure level.

Density samples were taken from the end sections of the molded specimens. After breaking apart small pieces (about $1/8$ sq in.), the density of the samples was measured in a density gradient column prepared by using aqueous sodium bromide–$n$-propyl alcohol mixtures. Densities could be measured to within $\pm 0.0002$ g/cc.

### Mechanical Testing

Tensile and compressive stress–strain tests were performed on the various samples using an Instron testing machine. The tests were conducted at room temperature only at elongation rates ranging from 0.002 in./min to 2.0 in./min. These rates represented strain rates of from 0.0016 to 1.6 in./in./min. for the tensile tests.

Yield stresses reported are based on the original dimensions of the sample. The yield stress was taken as the point at which the stress strain curve begins to deviate from linearity. This definition is comparable to various offset methods of defining an elastic limit. Since in tension the control polystyrene was brittle and the densified samples only marginally ductile, the yield stress is very close to the ultimate stress of the material. In this publication, only the tensile mechanical testing will be reported. The compression data, however, showed the same trends.

### Differential Scanning Calorimetry

A du Pont Model 900 Thermal Analysis System with a differential scanning calorimeter accessory was used at a scanning rate of 10°C/min.

## RESULTS AND DISCUSSION

### Residual Densification

This investigation was concerned with the relationship between densification and plastic yielding in high polymers. After completion of a pressure–temperature pretreatment cycle, the density was determined and the ambient tensile and compressive stress–strain properties were measured.

For this work, the conditions at the maximum temperature and pressure of densification were such that the polystyrene was in the viscous liquid region. The crosshatched section of Figure 3 depicts this operating region. Assurance of operation in the region was provided in two ways: first, by using literature values for $dT_g/dP$ in combination with the measured value of the glass transition at atmospheric pressure, and second, by conducting several runs at varying pressures for a given temperature and comparing residual densifications obtained. Both techniques agreed well. As an example of the former method, the data of Breuer[7] for a polystyrene of $T_{g0}$ equal to 96°C was plotted in Figure 3 and subsequently displaced so that the $T_{g0}$ conforms to approximately the $T_g$ for the material used in this research. Using the latter method, very small densifications (if any) were noted when the combination of temperature–pressure was such that the material was in the glassy state or in the glass-to-liquid transition zone.

For the particular pressure–temperature cycle used in this work, $T \uparrow P \uparrow$ $T \downarrow P \downarrow$, if the pressure was applied at a given temperature the residual densification increased linearly with molding pressure. Eventually at high pressures, a point was reached at which the material was transformed to a glass, and essentially no further densification occurred. Figure 4 depicts this maximum residual densification possible at a given pressure. Thus, for a given temperature, so long as the pressure is less than the glass transition pressure at that temperature, residual densification increased nearly linearly with pressure. The compactions achieved at lower pressures closely match the results of Price.[11]

If the material is still in the viscous liquid region at the maximum temperature and pressure, vitrification actually takes place during the follow-
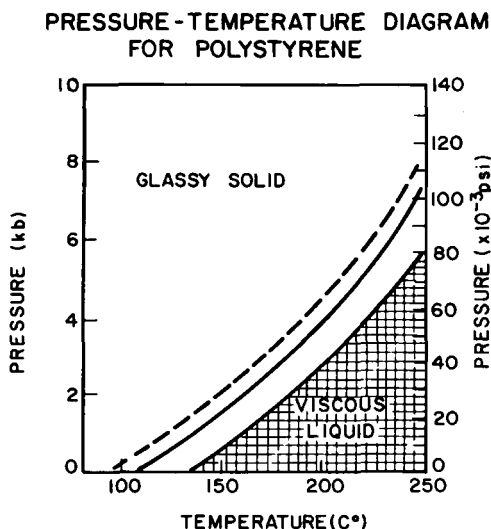


Fig. 3. Pressure–temperature diagram for polystyrene: (‒‒‒) data of Breuer[6]; (——) data of Breuer displaced 10°C.
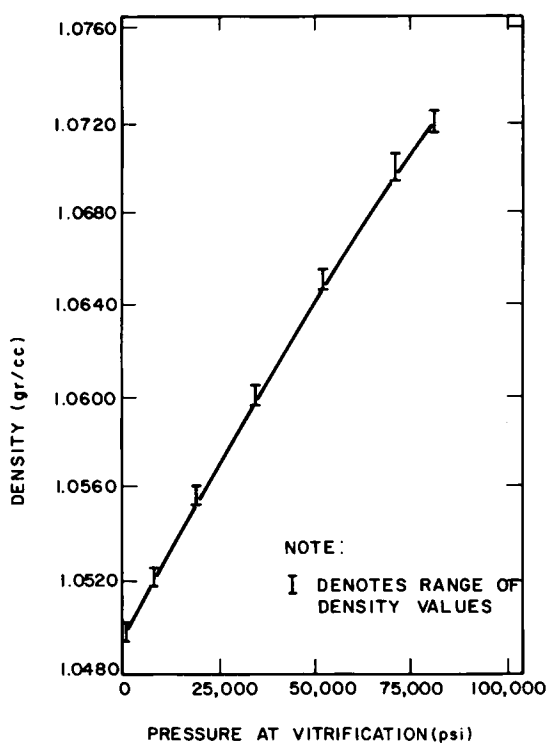
Fig. 4. Residual densification of polystyrene as a function of vitrification pressure.

ing cooling step.   Using this step, as opposed to the pressurization step to produce vitrification, was found to minimize internal stresses.   During the pressurization, there is actually a flow of material.   If vitrification were to commence as the pressure-induced flow occurred, sections of the polystyrene might become glassy while others were still flowing, resulting in frozen-in orientation.   (It is possible that if the pressure were increased *very* slowly, thereby allowing equilibration of flowing polymer, no residual stresses would be induced.)   Vitrification by cooling produces little frozen-in stresses since the material is quiescent.   The polystyrene produced by temperature vitrification was transparent and was found to be free of residual stresses using polarized light observations.

## Annealing Effects

The amount of time spent at the maximum temperature and pressure may affect the resultant density.   Kimmel[18] has found an exponential approach with time to some equilibrium density for poly(methyl methacrylate), with about 30 min necessary for constant density to be attained.

Some finite length of time is necessary for the compaction of the material to be completed.   This behavior supports the suggestion that flow of the material takes place and that by prematurely stopping this motion—as in

pressure induced vitrification—an equilibrium is not reached. Consequently, in this work, about 30 min were allowed at the molding conditions so that equilibrium could be closely approached.

The molded specimens were stored at room temperature (23°C). The densities of several specimens were measured at various time intervals, with results shown in Figure 5. While the atmospheric specimens did not relax, the densified materials recovered slightly, with the more compacted polymer recovering to a greater extent for a given time interval.

In general, little ambient relaxation occurred for the densified specimens. Moreover, a specimen density was measured within a few days of its mechanical test, thereby ensuring a proper correlation of parameters.

When the specimen temperature was raised closer to the glass transition, a relaxation of the induced densification occurred. This behavior is shown in Figure 6 for density as a function of temperature for a 20-min annealing
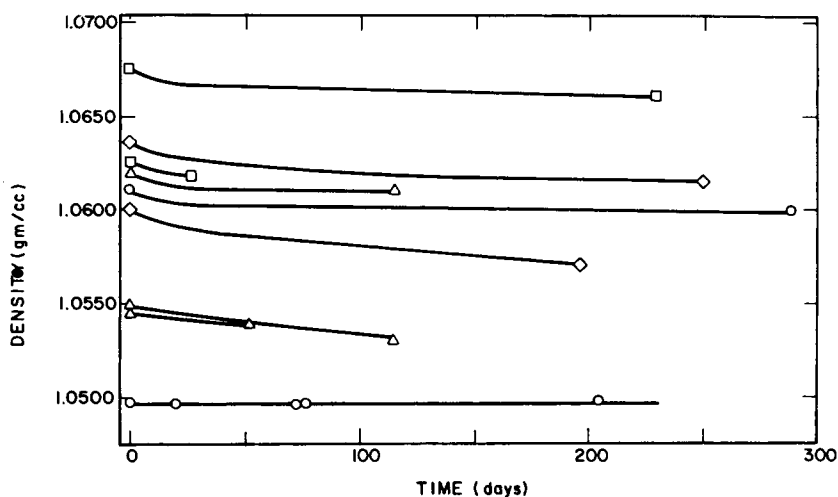


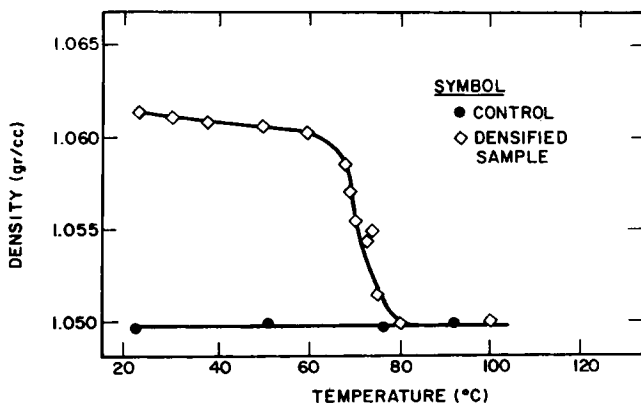Fig. 5. Relaxation of densified polystyrene at room temperature.



Fig. 6. Relaxation of density as a function of temperature: (●) control sample; (◇) densified sample.

time.   The induced densification rapidly decreases above 70°C, some 30° below the glass transition temperature.   Dale and Rogers[19] observed similar behavior for their densified polystyrene.

### Differential Scanning Calorimetry Results

Differential scanning calorimetry (DSC) results for specimens of three different densities are shown in Figure 7.   No change in the thermal behavior due to densification was noted.   Moreover, the glass transition temperature for the densified specimens was the same as the control.   This is due to the relaxation of density in the DSC cell.   By the time the polystyrene approached the $T_{g0}$, its density was essentially the same as the atomospheric material.   These samples were vitrified by cooling from the high-pressure viscous liquid region.

Samples of pressure-vitrified materials, however, exhibited significantly different behavior.   Typical differential scanning calorimetry (DSC) results are depicted in Figure 8.   The difference between the pressure-vitrified (PV) samples and the temperature-vitrified (TV) samples is directly evident and is due solely to the difference in quenching history, even though they possess equivalent densities.

Several observations may be made about Figures 7 and 8.   The greater the flaw structure, the greater the extent of the exo- and endothermic regions.   Conversely, the closer the samples approach temperature vitrification, the less these regions are noted.   One endo- and one exothermic region can be distinguished.   The endothermic region occurs in the temperature region at which compactive relaxation takes place (about 70–75°C).   Evidently, an energy absorption occurs as the internally stressed molecules relax enough to lessen the densification to the equilibrium atmospheric
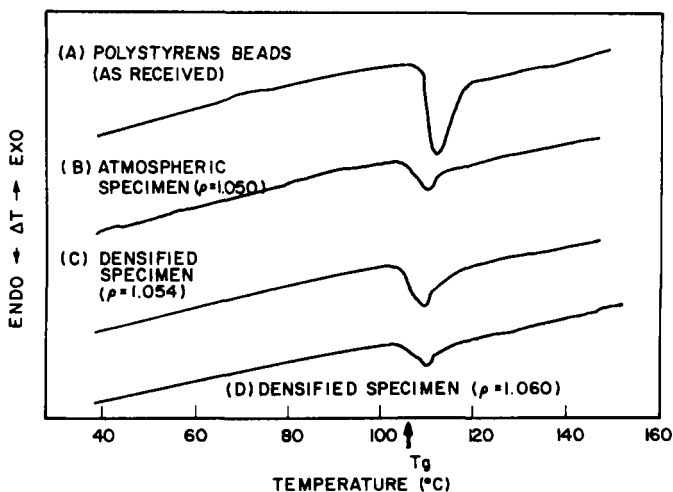


Fig. 7. DSC traces of temperature-vitrified polystyrene at a scanning rate of 10°C/min:   (A) polystyrene beads as received;   (B) atmospheric specimen, $\rho$ = 1.050 g/cc; (C) densified specimen, $\rho$ = 1.054 g/cc;   (D) densified specimen, $\rho$ = 1.060 g/cc.
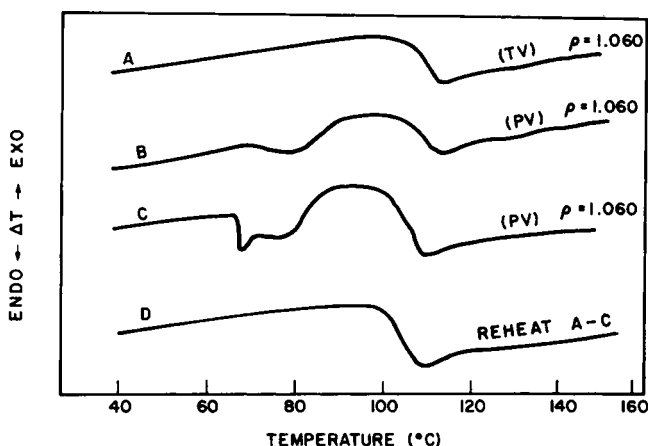
Fig. 8. DSC traces of pressure-vitrified polystyrene: (A) temperature-vitrified specimen $\rho$ = 1.060 g/cc; (B) pressure-vitrified specimen, $\rho$ = 1.060 g/cc; (C) pressure-vitrified specimen, very flawed, $\rho$ = 1.060 g/cc; (D) reheat of specimens A–C.

density. Apparently the thermal energy at this temperature is not sufficient to relax the induced internal stresses. If the temperature is raised further, the frozen-in stresses relax with the sample exhibiting an exothermic region. As with the TV samples, no variation in the glass transition is noted. Evidently, near 100°C, the segment mobility is such that all PV densification effects have been eliminated.

Similar DSC behavior to that of the pressure-vitrified samples described above has been observed by Dale and Rogers.[19] It appears that at the higher pressure ranges they investigated, their compressive specimens became pressure vitrified. A maximum in the compressive yield stress versus molding pressure was observed which may be explained by the hypothesis that the samples were passing from a region of temperature to pressure vitrification. Consequently, what they considered to be a good representation of the behavior of densified polystyrene molded under pressure was actually a combination of TV (at 1000 atm) and PV (progressively greater effect as pressure increased to 3000 atm) material effects.

### Mechanical Behavior

In tension, the atmospheric (control) specimens were brittle, while the pressurized specimens were marginally ductile, as noted from a slight bending of the curve just beyond the yield point but before fracture. All stress values were based upon original dimensions, as opposed to instantaneous ones. Such an approximation to true stress–strain behavior is good in this case since the specimens were rather brittle.

Figure 9 presents the relationship between tensile yield stress and the logarithm of the strain rate for various densities. As might be expected from an Eyring viscous flow approach,[21] the yield stress is essentially a linear function of $\log \dot{\epsilon}$. Several additional observations may also be made.

A small densification of polystyrene produces a relatively large increase in tensile strength. As noted in Figure 10, this increase in strength is more pronounced at first, gradually tapering off. Limiting values of tensile strength and density have not been reached in this work.

Secondly, in Figure 9 it may be seen that the slopes of the stress–log $\dot{\epsilon}$ curves are essentially equivalent. This implies that the activation volume of an Eyring rate model applied to the data is independent of the density of the glass.

The results for testing the densified polystyrene in compression were similar to the overall observations in tension. The yield stress increased monotonically with densification. Further details of the compression study will be published elsewhere.

In addition to the mechanical responses measured, several physical observations were also made. No crazing was observed until the approach of the yield point (at about 75% of the yield strength). The fracture surfaces of both control and densified specimens were similar. A typical surface included a circular smooth section covering about 30% of the cross-sectional area and a rough region covering the rest. The smooth section was usually located adjacent to the edge of the fracture surface. The rough region was characterized by striated concentric rings surrounding the smooth region. Craze formation initiated in the smooth area, with catastrophic failure occurring in the rough region. The physical dimensions did not change. Also no deformation bands were observed.

A comparison of samples before and after testing revealed that no change in density had occurred. It is possible that right at the fracture surface relaxation may have occurred, but the difference was not detectable using
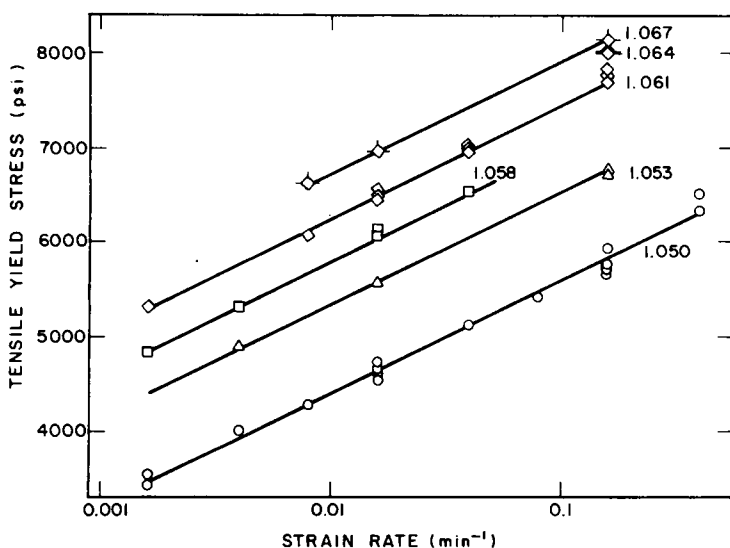


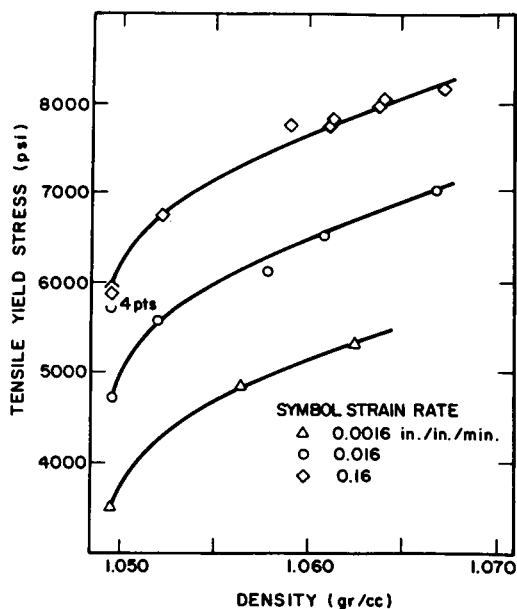Fig. 9. Tensile yield stress vs. log strain rate for densified polystyrene.

Fig. 10. Tensile yield stress of polystyrene vs. density. Strain rates: (△) 0.0016 (in./in.) min$^{-1}$; (○) 0.016 (in./in.) min$^{-1}$; (◇) 0.16 (in./in.) min$^{-1}$.

the density gradient technique. This is due to the size of the sample necessary for density measurement being relatively large compared to the fracture plane. Several specimens were also subjected to cyclic deformation in the elastic region. A very slight decrease in density was noted for about 25 cycles, with the mechanical behavior barely affected.

For the pressure-induced vitrification samples, cracks were noted to occur substantially before the yield stress was reached. The cracks expanded resulting in very low tensile strengths compared to the temperature-vitrified samples. Such behavior suggests that craze, or microflaw formation is enhanced by pressure-induced glassification. These microflaws act as sites for initiation and propagation of cracks, leading to lower mechanical strength. In contrast, temperature-induced vitrification from high pressure suppresses microflaw formation and leads to higher strength.

The role of microflaws in explaining the property changes of densified polystyrene has been explored by testing thermally annealed samples of densified polystyrene in compression.[13,22] It was found that even though the density of a compacted sample relaxed to that of the control, the material retained the higher modulus and yield strength characteristic of the densified specimen.

## CONCLUSIONS

Amorphous polystyrene was vitrified from an elevated temperature–pressure regime to produce a more closely packed, higher-density glass. If vitrification took place by lowering of temperature at approximately con-

stant pressure, the material produced was largely free from flaws and exhibited enhanced mechanical properties. Glasses produced by rapidly raising the pressure at high temperature possessed frozen-in stresses and large flaws. These samples were in some cases weaker than the control specimens.

For temperature-vitrified samples, the residual densities increased monotonically with vitrification pressure. At treatment conditions of 90,-000 psi and 320°C, upon subsequent cooling, a residual compaction of greater than 2% was observed. Reproducibility of compaction, for given conditions of pressure and temperature, was very good and relaxation of the density at ambient conditions was minimal. However, upon raising the temperature to about 30°C below the glass transition, the compaction relaxed away rapidly. The mechanical behavior of the densified materials is markedly different from that of the control specimens. In tension, the densified materials exhibited a substantial increase in yield strength, ultimate strength, Young's modulus, and toughness over the control specimens. The increase was very pronounced at first and then tapered off at the higher densities; however, no limiting value was evident.

In all tests, the atmospheric specimens exhibited brittle fracture, while the densified specimens were slightly ductile. The fracture surfaces of the densified and control specimens were essentially the same. Densified samples which were annealed at atmospheric pressure at temperatures where the compaction could completely relax maintained the higher strength and modulus characteristic of the compacted materials.

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