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HIGH TEMPERATURE CARBON EQUATION OF STATE

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September 4, 1974

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Foreword

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HIGH TEMPERATURE CARBON EQUATION OF STATE

Abstract

The isobaric expansion technique is used to determine the equation of state of carbon at high temperatures and kilobar pressures. Glassy carbon samples are resistively heated to the melt and liquid regions, and the data

are plotted in the enthalpy-volume and resistivity-enthalpy planes. The data are also approximately fitted with a GRAY three-phase equation-of-state model suitable for use in hydrodynamics codes.

Introduction

Carbon is a useful structural material for reentry vehicle heat shields because of its very high melt temperature, its low Grüneisen gamma, and its high heat capacity per unit mass. To compute correctly the effects of thermo-mechanical loading on carbon structures, it is necessary to have an accurate three-phase equation of state (EOS). However, because of the high temperatures required, liquid carbon cannot be prepared in conventional

high-temperature experiments, and its thermodynamic properties are virtually unknown. In this report we describe a new experimental procedure for measuring the carbon equation of state in the liquid phase at kilobar pressures and high temperatures, and we present an analysis of the data collected so far. We have also attempted to fit the experimental data with a GRAY three-phase equation-of-state model suitable for use in hydrodynamic codes.

Experimental

One of the chief difficulties of high-temperature physics is the containment of a sample in equilibrium at elevated temperatures without contamination by interaction with the confining vessel. This work describes the use of a recently developed dynamic technique¹ to measure the equation of state and resistivity of carbon to temperatures much higher than

those attained in previous measurements.

A cylindrical sample is resistively heated while a constant pressure is maintained on the sample by a surrounding inert gas. The sample is constrained so that the resulting expansion is only radial. Thus the specific volume can be determined during the expansion from a streaking-camera record of the diameter,

The energy input to the sample can be determined from the current-voltage characteristic of the sample after appropriate inductive corrections have been made. From the specific volume and voltage-current data, one can construct isobars in the volume-enthalpy (energy input at constant pressure) plane and the resistivity-enthalpy plane. Previous studies of high temperature resistivities have ignored the corrections made necessary by changes in sample dimensions.² In making these constructions for a single shot, one must assume that the sample is homogeneous and never far from equilibrium during the heating. These assumptions are qualitatively checked by observing how well the trajectory of a single shot in the enthalpy-volume plane passes through the end points of shots that clearly achieved equilibrium at the end of the heating pulse.

Pressure on the sample is applied by argon or helium gas contained in a steel pressure vessel with sapphire windows at pressures up to 0.4 GPa. The gas volume is sufficiently large that sample expansion has no effect on the pressure. In addition, the energy input from the capacitor bank is kept slow enough to allow many sonic reverberations in the gas, thus preventing large pressure gradients. The cell pressure is measured by a Heise gauge.

Two lines of sight are provided through the cell (Fig. 1). One is used for a Q-switched ruby-laser shadowgraph to provide an overall view of the sample at a selected time. This diagnostic allows one to evaluate the sample homogeneity along the rod axis and the surface texture. If large inhomogeneities are observed

(e.g., the classical necking instability in magnetohydrodynamics, the shot is discounted. The other line of sight is used for a streak-camera system that records the time history of a particular diameter of the sample.

The streak-camera system is built around a TRW 1D[®] image converter camera that allows streaks of 50, 100, or 200 μ s duration to be selected. A 1-W argon-ion laser provides back lighting for the streaked shadowgraph. Timing marks are written directly on the film by the light from a helium-neon laser modulated at 1 MHz by a Pockel cell. Both the shadowgraph and streak systems use narrow band filters to reject unwanted light from each other and blackbody radiation from the sample. In addition, fast mechanical shutters limit the time integration of blackbody radiation in the pass

*Reference to a company or product name does not imply approval or recommendation of the product by the University of California or the U.S. Atomic Energy Commission to the exclusion of others that may be suitable.

Fig. 1. A schematic diagram of the optical diagnostics for the isobaric expansion experiment.

band of the filters and unwanted ambient light.

The capacitor bank for energy deposition is critically damped, giving a rise to maximum current in about 11 μ s. After an interval of 10 to 20 μ s, a crowbar system diverts the remaining bank energy from the sample, allowing control of the energy deposition by the delay of the crowbar. The pulse is slow enough to avoid nonuniform heating in the sample caused by the skin effect.

Current in the system is measured by a wideband transformer. The voltage drop across the sample is measured

between two probes arranged to minimize mutual inductance with the bank circuit.

Carbon samples used in this work were machined to a diameter of 1 mm and a length of 25 mm from rods of POCO brand AXF-5Q grade vitreous carbon of density 1.83 Mg/m³. The choice of vitreous carbon avoided the severely anisotropic mechanical properties of graphite. It is believed that the microscopic structure consists of randomly-oriented graphite grains of 1 to 10 nm. Because of the polycrystalline nature of the sample, the density is lowered from 2.24 for single-crystal graphite.

Data Analysis

The gross features of the pulse heating of glassy carbon can be seen in Figs. 2 through 6. Figure 6 in particular shows the slight thermal expansion of the sample until 6 μ s after bank fire, when the compaction to graphite density occurs. This behavior happened at all pressures measured (0.1 to 0.4 GPa). Following compaction a period of steady expansion occurs with a cusp appearing at a volume 1.1 to 1.2 times the initial volume (especially evident in Fig. 4). The expansion ends at the conclusion of energy deposition and the volume stays constant for periods of 50 to 100 μ s in well-behaved shots. More typical for the higher energy and lower pressure shots was a slow drift toward larger volume long after energy deposition was completed.

The data in Fig. 6 have been corrected for the slightly nonlinear magnification of the streaking camera. The error bars

appearing in this figure arise from the estimated errors in reading the edges of the shadowgraph. With helium as a pressure medium, the diameter could be read to better than 1% precision. Since the slit width of the streaking camera corresponds to approximately one-twentieth of the sample diameter, the surface roughness shown in Fig. 3 indicates that more accurate diameter measurements would be meaningless. In the case of argon gas as the pressure medium, the surface of the hot carbon seems to stay smoother; but the larger refractive index results in more optical distortion as acoustic waves bounce around the sample container. This effect is especially evident in Figs. 4 and 5.

Figures 7 through 9 show typical results of the current and voltage diagnostics. In Fig. 9 enthalpy is calculated after an inductive correction has been made to the voltage signal; extra

Fig. 2. Typical 50 μ s streak record in 0.2 GPa of helium. Time marks in streak are 1.028 μ s apart. Time increases vertically. Final energy: $H = 14.6$ MJ/kg. Final volume: V/V_0 (graphite) = 1.53.

Fig. 3. Ruby laser snapshot of the carbon rod shown in Fig. 2. The snapshot was fired 15 μ s after crowbar and 16 μ s after the minimum in diameter shown in Fig. 2.

energy input from the compaction is ignored.*

* An estimate of the energy input from the mechanical work done on the sample during compaction can be made. Since the process is very nearly isobaric,

$$\Delta W = -P\Delta V,$$

which for $P = 0.4$ GPa, and $\Delta V = -10^{-4}$ m³/kg (i.e. $\Delta V/V = -0.2$), gives $W = 0.04$ MJ/kg. On the scale of energies deposited by the resistive heating, this correction is completely negligible.

Fig. 4. Typical 100 μ s streak record in 0.4 GPa of argon. Time marks are spaced 2.044 μ s apart. Time increases vertically. Final energy: $H = 23.0$ MJ/kg. Final volume: V/V_0 (graphite) = 3.8.

Fig. 5. Ruby laser snapshot of carbon rod in Fig. 4. Snapshot fired 10 μ s after crowbar, 19 μ s after minimum in diameter of Fig. 4.

A decrease in relative resistance (Fig. 10) up to 6 MJ/kg is normal for many different types of carbon.³ The rapid drop between 6 and 8.5 MJ/kg correlates with the compaction of the carbon sample from its glassy state to a state with density near that of graphite. The next sharp fall in resistance at about 9.6 MJ/kg was a somewhat more variable feature. This "pre-melt" signal appeared

Fig. 6. Typical volume-time records for well stagnated carbon shots. Confining gas was 0.4 GPa of argon. Volume has been corrected for camera nonlinearity. The error bars shown are the estimated errors in reading the streaking camera record.

Fig. 7. A typical current-time record.

Fig. 8. The voltage-time record for the shot of Fig. 7. Notice sharp drops in voltage beginning at 6.5 μ s and at 9 μ s (arrows). These correspond to the compaction of the sample, and the "pre-melt" signal, respectively.

Fig. 9. The enthalpy-time record for the shot of Fig. 7. Final enthalpy is 18,004 MJ/kg.

Fig. 10. The resistance ratio vs enthalpy record for the shot of Fig. 7. Note the same features as in Fig. 8 (arrows).

on almost all of the 0.2 GPa shots, but at enthalpies ranging from 9.6 to 12.0 MJ/kg. At 0.3 and 0.4 GPa, this resistance drop appeared on only half of the shots, but always at about 13.2 MJ/kg. In most cases the magnitude of the resistivity change was 5 to 10% and it occurred in a time interval corresponding to an enthalpy change of less than 0.5 MJ/kg. As the energy is increased beyond the anomaly, the resistance shows an almost exponential rise as larger specific volumes are achieved. It becomes essentially singular at 26 MJ/kg for 0.4 GPa, 23 MJ/kg at 0.3 GPa, and 21 MJ/kg at 0.2 GPa.

Figure 11 shows the resistivity calculated from the resistance in Fig. 10 and the actual diameter vs time streak record for the same shot. The qualitative features are the same as for the resistance, but the resistivity rises faster above 10 MJ/kg. Included in Fig. 11 is a summary of static resistivity data on a variety of dense carbons, including sin-

Fig. 11. The absolute resistivity vs enthalpy for the shot of Fig. 7. Notice the same features as in Fig. 8 (arrows).

gle crystal graphite.³ From this figure it is clear that the sample resistivity after compaction joins smoothly with the resistivity of fully dense graphite.

In Fig. 12, we have collected specific volume vs enthalpy data for four shots at 0.2 GPa confining pressure. Three of these shots stagnated well and the end points of these shots are shown as the solid points. Generally speaking, the enthalpy-volume data obtained from a single shot lies 5 to 10% on the low volume side of the curve defined by the end points of the well-stagnated shots. This suggests that during the pulse heating the sample follows a slightly nonequilibrium trajectory. Therefore, one must put most credence in the points representing the final volumes of well-stagnated shots.

The degree of scatter in the volume data shown in Fig. 12 is greater than the

Fig. 12. Typical data for the expansion of carbon at 0.2 GPa (4 shots). The solid points are the end points of well stagnated shots. Static thermal expansion data for graphite is indicated by the curve beginning at $V/V_o = 1$.

estimated error in measuring the volume. This scatter may be the result of the slight deviation from equilibrium during heating or to some as yet undiscovered source of error in the measurement. The extreme variability of the occurrence of the "premelting" signal suggests that non-equilibrium at least plays a role in the scatter of the equation of state data.

One can see from Fig. 12 also that if the relative timing of the volume-time record and the enthalpy-time record is adjusted, the data can be forced to have less scatter. The amount of adjustment of the data in Fig. 12 amounts to $\pm 0.5 \mu s$, well outside the estimated error ($\pm 0.2 \mu s$) in this correlation. Also, this adjustment procedure would not reduce the scatter for many of the shots.

We have also noticed from the snapshots taken after the end of the current pulse that the rod may have diameter variations of $\pm 2.5\%$ even for well-stagnated shots. Thus, one is probably not justified in applying an ad hoc procedure to try to force the scatter out of the data.

The results of shots ending in well-stagnated volumes are used in Fig. 13 to define isobars at several pressures. This figure represents a summary of the carbon equation of state measurements made so far.

During the initial portion of the pulse heating of the glassy carbon, the resistivity fell as the temperature increased. In this situation, if the heating is by constant-current source, the sample is unstable with respect to the formation of hot filaments or cylindrical sheaths. If a filament starts getting hotter due to some perturbation, its resistivity drops and the hot filament carries more current at the expense of the rest of the sample. This situation must be kept in mind as a possible mechanism for the generation of an inhomogeneous radial temperature distribution in the pulse-heated sample. This mechanism could explain much of the scatter in our data.

One of the most basic questions to be answered in any equation-of-state measurement is the location of the phase boundaries. In the case of carbon this includes melting and vaporization. Various estimates of the melting properties of graphite suggest a melting temperature of 4000 to 5000 K and a latent heat of fusion of 5 to 10 MJ/kg.⁴ These estimates are unreliable because of the high temperatures involved and a triple point at

Fig. 13. Summary of the data for carbon along three isobars. The points represent the end points of well-stagnated shots.

roughly 0.01 GPa pressure. In view of the relatively high estimated heat of fusion, we must look for melting to occur over a considerable fraction of the heating pulse. In particular, the premelt signal (the second resistance anomaly in Fig. 10) probably cannot represent melting because the change in enthalpy is only a few tenths of a MJ/kg for the complete change. If this change corresponded to melting of only a small fraction of the carbon, then the resistivity should continue to drop as more material melted. The most likely explanation of this anomaly seems to be a new phase of carbon between graphite and melting, per-

haps chaoite.⁵ At 0.2 GPa, where the resistance anomaly appeared at variable enthalpy, it is possible that the low-temperature phase was in some cases superheated beyond the phase transition. At higher pressures the resistivity change is smaller and the enthalpy at which the transition occurs is much more stable (12.9 MJ/kg at 0.3 GPa and 13.4 MJ/kg at 0.4 GPa). In Fig. 14, a typical 0.4-GPa shot, one can clearly see the cusp in the volume-enthalpy curve slightly offset from the premelt signal in the resistivity. From 14.5 MJ/kg to around 20 MJ/kg there is a clear linear portion of the enthalpy-volume curve.

Fig. 14. Volume and resistivity vs enthalpy for a typical 0.4 GPa shot. Notice the clear energy offset between the "pre-melt" anomaly in the resistivity and the cusp in the volume data.

This seems most likely to be the region where the energy feeds the latent heat of melting; and the specific volume changes linearly with energy from the solid at the beginning of melting to the liquid at the end of melting. If this interpretation is correct, then the heat of fusion at 0.4 GPa is 5.5 ± 1 MJ/kg (16 ± 3 kcal/mole) and the volume change on melting is $\Delta V/V$ (solid) ≈ 0.5 . From the slope dH/dV in this region one can obtain the slope of the melting curve from the Clausius-Clapeyron relation:

$$\frac{dT_m}{dp} = T_m \frac{dV}{dH}.$$

This relation gives $dT_m/dp \approx 230$ K/GPa, if a value of 5000 K is assumed for T_m . Values reported in the literature vary from 70 K/GPa⁶ to 830 K/GPa,⁷ so our value at least falls in this range.

Such a low value for dT_m/dp presents problems for the consistent interpretation of the data. If we take 230 K/GPa, then the change in melting point from 0.1 to 0.4 GPa is only 70 K, an undetectable change on the enthalpy scale we use. One then cannot explain the fact that the 0.2-GPa isobar seems to break out of the linear region at about 15.5 MJ/kg, while the 0.4-GPa isobar does not break out until about 20 MJ/kg (see Fig. 13). Also, if the cusp in Fig. 14, a very stable feature at 0.3 and 0.4 GPa, represents the onset of melting, either the melting point is very high or the heat capacity is very high. The static heat capacity data for carbon⁸ shows a presublimation rise between 3500 and 4000 K. If the value of C_p at 3500 K is used and extrapolated to an enthalpy of 14.5 MJ/kg, the melting temperature would be over 7000 K, while all previous estimates range between 4000 and 5000 K.⁴ If C_p at 4000 K including the presublimation rise is used above 4000 K, the temperature at 14.5 MJ/kg is still 5700 K, much higher than previous estimates. To bring the temperature at 14.5 MJ/kg down to the 5000-K range, one must assume that the heat capacity does something extreme. For example, if the presublimation rise in the heat capacity is extrapolated for another 1000 K, the enthalpy at 5000 K is 14 MJ/kg, but C_p is now 8 kJ/kg-K (24 cal/mole-K).

One can conclude at this time that several questions remain open in the interpretation of the present carbon data.

We have definitively located neither the solid-liquid phase boundary nor the vaporization curve. The premelt signal in the resistivity is still unexplained, as is the cusp in the volume record. Optical pyrometric records could give much information on these questions.

On the other hand, we present here the first near-equilibrium equation-of-state and resistivity data for carbon at volumes over four times that of graphite and energies over twice that necessary to bring carbon to its normal sublimation point.

Equation-of-State Model

We wish now to incorporate the carbon equation-of-state data into the GRAY⁹ three-phase equation-of-state model for use in hydrodynamic calculations. This is done by varying GRAY liquid equation-of-state parameters until a satisfactory overall fit to the experimental data is achieved. The parameters to be varied are T_{mo} , the melt temperature at normal solid volume; V_J , the volume at which the van der Waals equation of state is joined to GRAY; V_b , the hard-sphere volume used in the van der Waals equation of state; and a_V , the van der Waals cohesive energy parameter.

The solid portion of the GRAY equation of state is based on Hugoniot measurements on carbon¹⁰ and the measured Grüneisen gamma. This solid equation of state is joined via a Lindemann-law melting region to the scaling-law liquid equation of state.

The final set of GRAY parameters was obtained by taking into consideration in order of decreasing importance:

- 1) smoothness of pressure-volume isotherms,
- 2) smoothness of enthalpy-volume isobars,
- 3) quantitative agreement with experimental enthalpy-volume isobars,
- 4) agreement with estimated critical point,⁴ and
- 5) agreement with estimated melting point.⁴

The optimum model equation of state was obtained by varying the liquid parameters and comparing the results with the conditions listed. No attempt was made to obtain precise quantitative agreement between theory and experiment, because the carbon melt and liquid regions are anomalous and cannot readily be described by a model designed for metals.

Table 1 contains the final GRAY parameters. The parameter V_o is the single crystal graphite volume, to which all other volumes are referred. Hugoniot parameters C and S are based on shock-wave experiments on carbon.¹⁰ The Grüneisen parameter γ_o is obtained from room temperature measurements. The electronic specific heat coefficient g_e is set to a negligible value because carbon is nonmetallic. Parameters T_{mo} , V_J/V_o , V_b/V_o , and a_V have been varied freely in order to meet required conditions of smoothness and consistency with experimental data.

Critical point and liquid-vapor tie-line parameters are given in Table 2. These data are to be used with the liquid-vapor modification of GRAY.¹¹ A previous estimate⁴ of the carbon critical point predicted that $V_c = 1.56 \text{ m}^3/\text{Mg}$, $T_c = 6810 \text{ K}$, and $P_c = 0.22 \text{ GPa}$. The

Fig. 15. Pressure-density isotherms for the GRAY carbon EOS. Here the highest isotherm is at 20 000 K, the lowest is at 2000 K, and the spacing between isotherms is 1000 K.

Fig. 16. Comparison of experimental and GRAY enthalpy isobars in the liquid-vapor region. The upper and lower isobars in each set are at 0.4 and 0.2 GPa, respectively.

Table 1. Equation of state parameters for the GRAY model of carbon.

V_0	$4.44 \cdot 10^{-4} \text{ m}^3/\text{kg}$
C	$3.96 \cdot 10^3 \text{ m/s}$
S	1.232
γ_0	0.24
a	0.24
g_e	$1.0 \cdot 10^{-7} \text{ J/(mole-deg}^2)$
T_{mo}	1000 K
AW	0.012 kg/mole
V_J/V_0	1.40
V_b/V_0	0.20
a_Y	$1.00 \text{ J-m}^3/\text{mole}^2$

unsatisfactory agreement between these estimated values and those in Table 2 is due to the constraint of smoothness imposed on the join between the van der Waals and scaling law equations of state.

Pressure-density isotherms, with emphasis on the liquid-vapor region are shown in Fig. 15. A direct comparison between experimental and GRAY enthalpy isobars is shown in Fig. 16. Once again, quantitative agreement is poor because of other constraints imposed on the equation of state and the lack of flexibility in the GRAY model when dealing with nonmetallic materials. Nevertheless, this model should be generally satisfactory for thermo-mechanical response calculations on carbon.

Table 2. Liquid-vapor tie line parameters for the GRAY carbon equation of state. The critical point parameters are: $T_c(K) = 8.2040E + 03$, $V_c(m^3/kg) = 9.9069E - 04$, $P_c(Pa) = 2.0424E + 09$, $E_c(J/kg) = 1.6806E + 07$. VMX and EVMX are the volume and energy at the vapor end of the two-phase line, and VMN and EVMN are the volume and energy at the liquid end of the two-phase line.

TEMP (K)	VMN (m^3/kg)	VMN (m^3/kg)	EVMX (J/kg)	EVMN (J/kg)	PRES (Pa)
7.4836E + 03	1.8519E - 03	6.4840E - 04	1.8519E + 07	1.4293E + 07	1.5013E - 09
6.7632E + 03	2.1773E - 03	6.1728E - 04	1.8839E + 07	1.2598E + 07	1.0580E + 09
6.0428E + 03	4.1667E - 03	5.8824E - 04	1.8839E + 07	1.1382E + 07	7.0802E + 08
5.3224E + 03	6.2500E - 03	5.6180E - 04	1.8600E + 07	1.0145E + 07	4.4000E + 08
4.6020E + 03	1.0000E - 02	5.3476E - 04	1.8237E + 07	8.8683E + 06	2.4582E + 08
3.8816E + 03	2.0000E - 02	5.1232E - 04	1.7812E + 07	7.5845E + 06	1.1685E + 08
3.1612E + 03	3.3333E - 02	4.9505E - 04	1.7184E + 07	6.2815E + 06	4.2107E + 07
2.4408E + 03	1.2861E - 01	4.7619E - 04	1.6578E + 07	4.8363E + 06	1.1184E + 07
1.7204E + 03	1.0000E + 00	4.6286E - 04	1.5854E + 07	3.5674E + 06	1.1748E + 06
1.0000E + 03	5.0000E + 01	4.5249E - 04	1.5096E + 07	1.4606E + 06	1.1126E + 04

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