SESAME Equation of State Number 8010:
Boron Loaded Silicone Potting Material

J. C. Boettger

Los Alamos
NATIONAL LABORATORY

Los Alamos, New Mexico 87545

SESAME Equation of State Number 8010:
Boron Loaded Silicone Potting Material

by

J. C. Boettger

ABSTRACT

A new SESAME equation of state (EOS) for boron loaded silicone potting material has been generated using the computer program GRIZZLY. This new EOS has been added to the SESAME EOS library as material number 8010.

I. INTRODUCTION

In recent years, SESAME equation of state (EOS) library users have expressed considerable interest in the EOSs of polymer composites; i.e., materials produced by embedding some relatively hard substance inside a softer polymer matrix. This interest has been driven by a desire to develop materials whose properties can be tailored for particular applications. In response, there has been an ongoing effort to expand the SESAME library to include a number of interesting polymers and polymer composites. For example, the SESAME library already includes EOSs for carbon phenolic (carbon embedded in phenolic; material number 7542) and phenolic refrasil (fiberglass embedded in phenolic; material number 7580).[1]

In this investigation, an EOS has been constructed for boron loaded silicone pot-

ting material (hereafter B-Silicone), a polymer composite with an ambient density (ρ_0) of 1.542 gm/cm^3 that is formed by mixing boron powder (63% by weight) with a soft silicone rubber (37% by weight).[2] B-Silicone was selected to be studied at this time as a result of recently released Hugoniot data.[2] EOSs for additional materials will be produced as experimental data become available. The new EOS for B-Silicone will be added to the SESAME library as material number 8010.

In the next section, the models and parameters used to generate the EOS for material number 8010 are described in detail. The new SESAME EOS for B-Silicone is presented and discussed in Section III.

II. METHODOLOGY

Most of the EOSs in the SESAME library are partitioned into three terms for the pressure P, the internal energy E, and the Helmholtz free energy A:

$$P(\rho,T) = P_s(\rho) + P_n(\rho,T) + P_e(\rho,T) \tag{1}$$

$$E(\rho,T) = E_s(\rho) + E_n(\rho,T) + E_e(\rho,T) \tag{2}$$

$$A(\rho,T) = A_s(\rho) + A_n(\rho,T) + A_e(\rho,T) \tag{3}$$

where ρ is the density and T is the temperature. (In the SESAME library, discrete values of ρ and T are chosen to form a mesh on which P, E, and A are stored.) The subscripts s, n, and e denote the contributions from the static lattice (i.e., frozen nuclei) cold curve (zero temperature isotherm), the nuclear motion, and the thermal electronic excitations, respectively. It is thus possible to calculate (or update) each term

independently using any desired model. Here, all three pieces of the EOS have been generated with GRIZZLY,[3] the locally developed multipurpose computer program for calculating SESAME EOSs.

In GRIZZLY, the only model currently available for calculating the thermal electronic contributions is the Thomas-Fermi-Dirac (TFD) model.[4] First thermal electronic contributions were generated for each individual atom in B-Silicone. Those monatomic thermal electronic EOSs were then combined via additive volume mixing[3] to form the total electronic contribution to the EOS. This part of the calculation requires the number fraction (N_i), atomic number (Z_i), and atomic mass (A_i) for each species of atom (i). As noted above, B-Silicone is composed of 63% boron and 37% silicone, by weight. It is assumed here that the silicone has the same ideal composition as that used by Dowell[5] in constructing an EOS for Sylgard 184 (a silicone rubber), $Si(CH_3)_2O$. Table 1 gives the atomic composition used here for B-Silicone and the values of A_i and Z_i for each constituent.[6]

Table 1: Atomic Composition Of B-Silicone

Atom	N_i	A_i	Z_i
B	0.5400	10.81	5.0
H	0.2760	1.008	1.008
C	0.0920	12.01	6.0
O	0.0460	15.999	8.0
Si	0.0460	28.086	14.0

The thermal nuclear contributions for B-Silicone were obtained with the JDJNUC nuclear model[7] in GRIZZLY using a Gruneisen function of the standard CHART-D form.[8] In this model, the material is treated as a Debye solid at low temperatures and as an ideal gas at high temperatures. The JDJNUC model smoothly switches between these two limits for temperatures near the melt line, which is obtained from the Lindemann law. At the melting point on each isotherm, the JDJNUC model builds in a small two-phase region to simulate the melting transition. In addition to the data in Table 1, this part of the calculation requires ambient values for the Gruneisen parameter (γ_0) and the melt temperature (T_m). The Debye temperature (Θ_D) is computed internally from T_m. For silicone rubber, Dowell[5] calculated an intermolecular Θ_D of 107 K and an intramolecular Θ_D of 1799 K. For boron, Θ_D is about 1250 K.[6] Based on these numbers, B-Silicone's Θ_D has been estimated to be about 1000 K. This value of Θ_D corresponds to $T_m \approx 2100$ K. The input value of γ_0 used here (0.5) was chosen so that the theoretical EOS reproduced experimental thermal expansion data for B-Silicone;[9] see Section III.

The static lattice cold curve for modest compressions (≤ 2.0) was calculated by removing thermal contributions from a Hugoniot that was read in as a table of particle and shock velocities (U_p, U_s) chosen to give a reasonable fit to the available Hugoniot and sound speed data;[2,9] see Table 2. For high compressions, the cold curve was required to smoothly extrapolate to a mixed TFD cold curve. This method of calculation ensures that the compressed EOS will reproduce the experimental Hugoniot and also have the correct asymptotic behavior. In the expanded region ($\rho < \rho_0$), the cold

curve was forced to have a generalized Lennard-Jones form[3] that was constrained to smoothly connect with the compressed portion of the cold cui /·· and have the correct cohesive energy (E_c). E_c is estimated here to be about 85 kcal/av-atom-mol. In addition to the parameters already given, the expanded cold curve calculation requires a parameter $FACLJ^3$ (here 0.2) which controls the shape of the expanded cold curve.

Table 2: Input Hugoniot Points

U_p (km/s)	U_s (km/s)	U_p (km/s)	U_s (km/s)
0.00	1.1940	2.50	6.9874
0.50	2.6500	3.00	7.7439
1.00	4.1000	3.50	8.4396
1.25	4.7000	4.00	9.0743
1.50	5.2800	4.50	9.6481
1.75	5.7383	5.00	10.1611
2.00	6.1699		

III. RESULTS

Figure 1 shows a selected group of P vs. ρ isotherms from the new SESAME EOS for B-Silicone. The most outstanding feature of the EOS is its exceptionally large thermal expansion at constant pressure. As the temperature is increased from 298 K to 1160 K, the volume increases by roughly an order of magnitude. This result is due, in part, to the exceptionally small bulk modulus ($B \approx 2.2$ GPa) produced by the bulk sound speed used here ($C_0 = 1.194$ km/s).[9] To ensure the quality of the SESAME EOS, the value of γ_0 was adjusted to match the experimental thermal expansion

5

coefficient α, as noted in Section II. Two separate measurements of α have been carried out for B-Silicone over the temperature range 255 K $\leq T \leq$ 322 K,[9] yielding $\alpha = 1.1 \times 10^{-4}$ K^{-1} and 1.6×10^{-4} K^{-1}. (These values for α are roughly 2 to 5 times larger than those found for the alkali halides at room temperature.[6]) Over the experimental temperature range, the theoretical EOS for B-Silicone shown in Fig. 1 yields $\alpha = 1.6 \times 10^{-4}$ K^{-1} or 1.7×10^{-4} K^{-1}, depending on the type of interpolation function used to find the untabulated values of P, T, and ρ. These values agree quite well with the experimental data for such a sensitive EOS parameter.

Figure 2 compares the theoretical Hugoniot for B-Silicone with the experimental Hugoniot and sound speed data.[2,9] There are two experimental values for the bulk sound speed shown in Fig. 1.[2,9] The larger of the two values for C_0 (1.194 km/s) was used here in the input Hugoniot (see Table 1) because the smaller value (0.816 km/s) implies a bulk modulus of only 1.0 GPa, which is 50% smaller than the bulk modulus used by Dowell[5] for pure silicone rubber. A noteworthy feature of the Hugoniot in Fig. 2 is the rapid drop in U_s as U_p approaches 0, which is characteristic of soft polymers and reflects the very weak interchain binding relative to the intrachain binding. Thus, as the interchain spacing decreases under pressure, the bulk modulus and shock velocity both increase rapidly. It is clear from Fig. 2 that the new SESAME EOS for B-Silicone provides an excellent fit to the the experimental Hugoniot data and thus should be reliable for all applications involving shock loading. The new EOS will be added to the SESAME library as material number 8010.
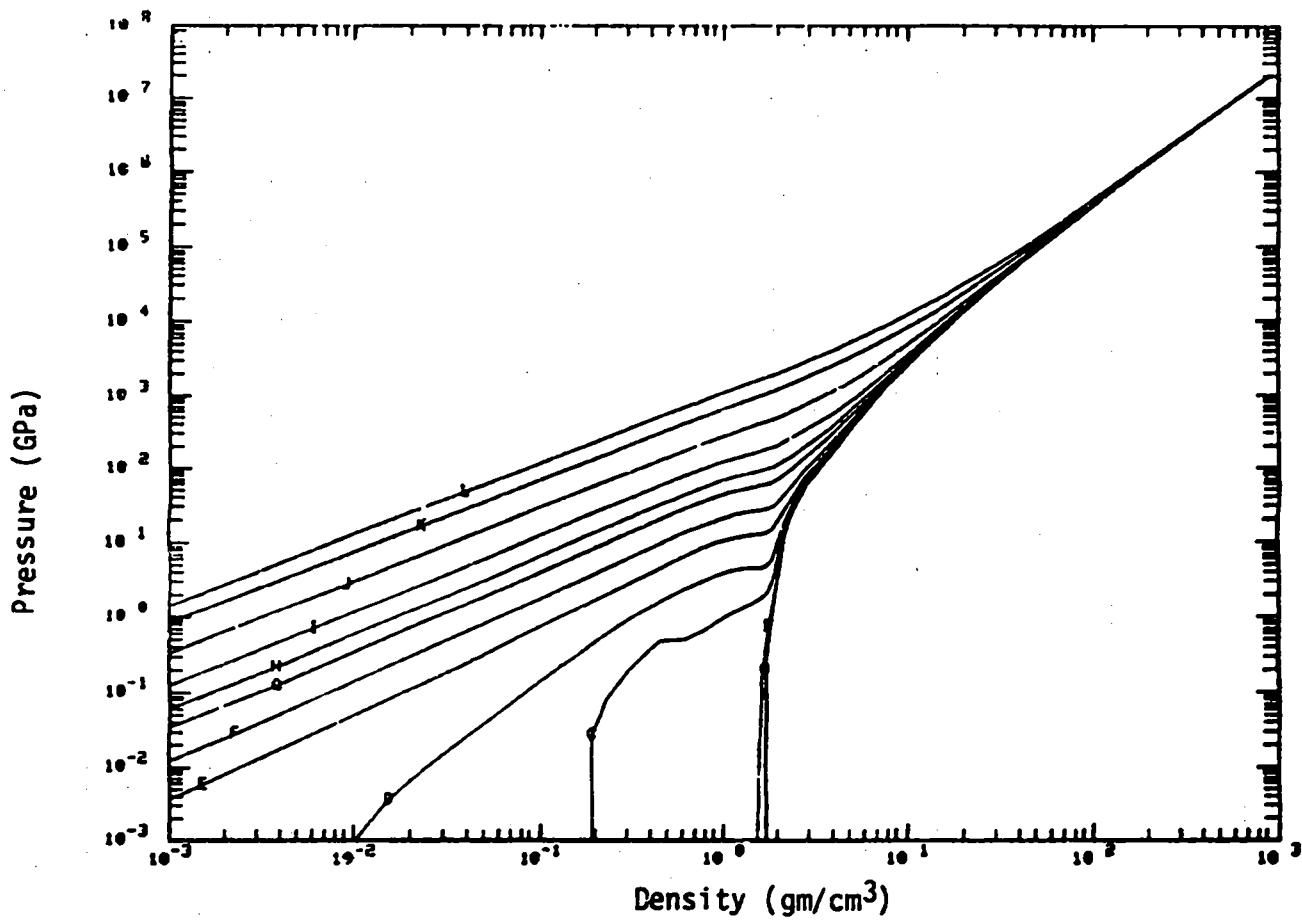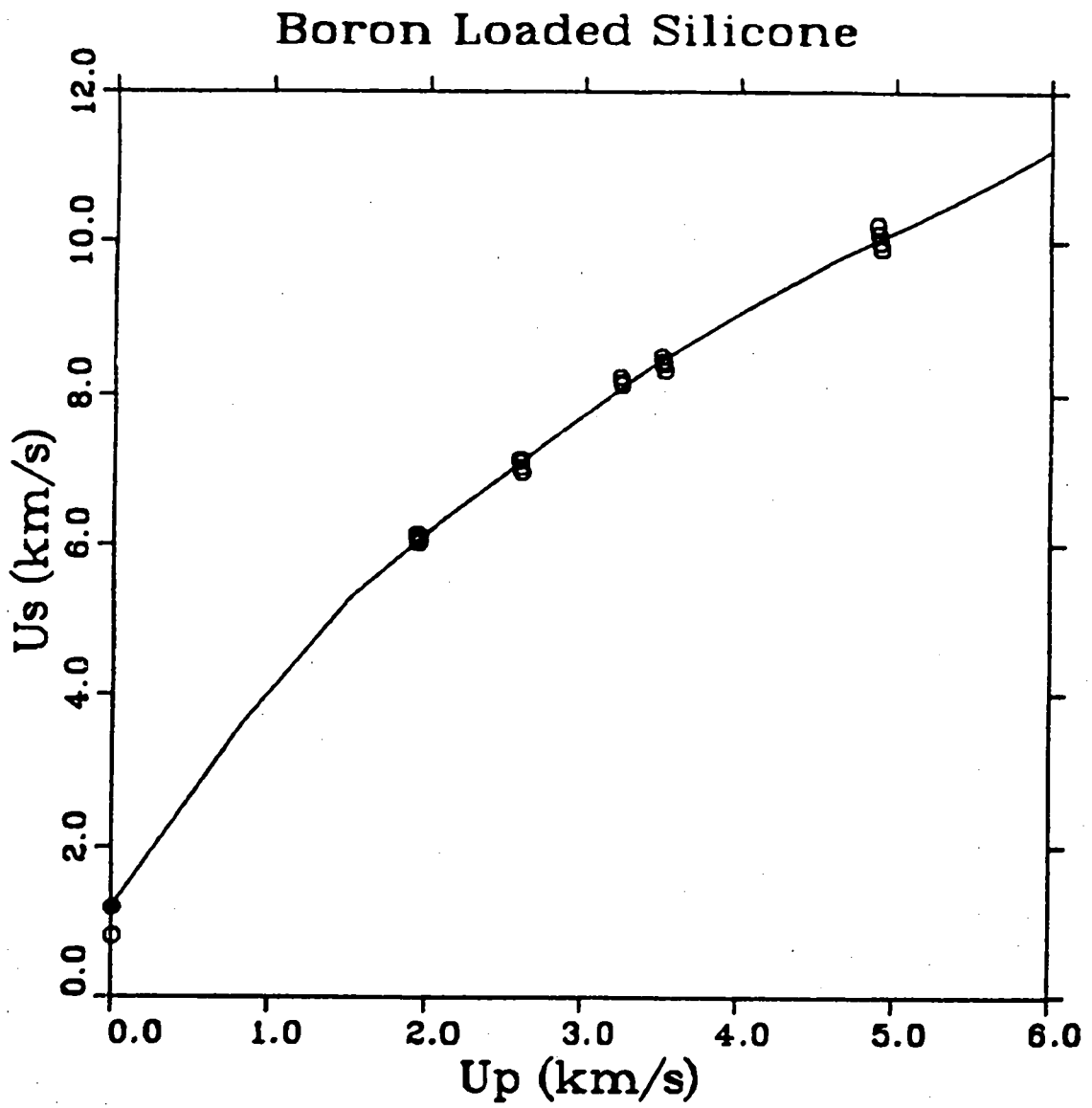
6

Figure 1.   Selected isotherms from the theoretical SESAME EOS for B-Silicone.
(A: 0 K; B: 298 K; C: 1160 K; D: 2320 K; E: 5800 K; F: 11600 K; G: 23200 K;
H: 34800 K; I: 58000 K; J: 116000 K; K: 232000 K; L: 348000 K.)

Boron Loaded Silicone

Figure 2.   The theoretical Hugoniot for B-Silicone is compared with Hugoniot data and two experimental sound speeds. (Open circles are from Ref. 2, and the solid circle is from Ref. 9.)

8

REFERENCES

1. J. C. Boettger, "SESAME Equation of State for a Polymer Composite: Phenolic Refrasil," Los Alamos National Laboratory Report LA-11512-MS (1989).

2. B. Olinger, R. McQueen, J. Fritz, and C. Morris, unpublished preliminary report, Los Alamos National Laboratory (Sept. 1992).

3. Joseph Abdallah, Jr., "User's Manual for GRIZZLY," Los Alamos National Laboratory report LA-10244-M (September 1984).

4. R. D. Cowan and J. Ashkin, *Phys. Rev.* 105, 144 (1957).

5. F. Dowell, "A simple EOS for the Silicone Rubber Sylgard 184," Los Alamos National Laboratory Report LA-10164-MS (1984).

6. N. W. Ashcroft and N. D. Mermin, *Solid State Physics* (Holt, Rinehart, and Winston, New York, 1976).

7. J. D. Johnson, "A Generic Model For The Ionic Contribution To The Equation Of State," Los Alamos National Laboratory Report LA-12003-MS (1991).

8. S. L. Thompson and H. S. Lawson, "Improvements to the CHART-D Radiation-Hydrodynamics Code III; Revised Analytic Equations of State," Sandia National Laboratories report SC-RR-710714 (1972).

9. C. Sandoval, unpublished report, Los Alamos National Laboratory (Jan. 1993).