ARMY RESEARCH LABORATORY

Gelatin Equation of State Characterization Progress

by Yolin Huang

ARL-MR-0727 **September 2009**

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14. ABSTRACT
In this report the Brillouin scattering data have been reviewed and divided into two domains: (a) in the low pressure phase, for the pressure below 2 GPa, the specific heat ratio γ is assumed to be 1.5; (b) in the higher pressure phase, for pressure higher than 2 GPa, the specific heat ratio γ is assumed to be 1.0, which is valid for incompressible material, because at higher pressure, the material has transitioned into a glassy state. The calculated pressure, density data are fitted with the Vinet and the Birch-Murnaghan equations of state to find the isothermal bulk moduli and its pressure derivatives. The results compare more favorably with Hugoniot test data. More accurate measurements for the densities, the specific heats, the thermal expansion coefficient, and the sound velocities in the low pressure domain will improve the data post-processing. Lab tests in the high pressure domain above 2 GPa are not easy to perform. But the low pressure domain will cover the majority of application needs.

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1. Introduction

Ballistic gelatin has been in use as a convenient soft tissue simulant for impact studies for several decades (*1, 2*). To better understand the impact physics behind the phenomenological correlations, further details about the physical properties of the gelatin are needed. As the gelatin is extracted from the collagen in the animal hide or bone, the resultant gelatin powder is an aggregate of various molecular sizes and molecular formulae (*3*). Its composition also varies from batch to batch due to variations in the manufacturing processes. Furthermore, the procedure in mixing the gelatin powder and water into gelatin blocks for laboratory tests is also not always consistent. So the physical properties of the gelatin should be viewed as being within certain statistical variations.

The equation of state relates the pressure, the specific volume and the temperature for the material under study. One of the traditional methods of acquiring high pressure properties is by performing uni-axial strain experiments (high velocity gas gun impacts), and study the Hugoniot characteristics. This aspect of the study will be discussed in a separate report. Another method is by using the diamond anvil technique, which is to be described in this report. The laboratory tests are done at the Carnegie Institution of Washington (CIW), which is a distinguished research facility for high pressure physics. The resultant contract report (*4*) describes the Brillouin scattering test procedure, the longitudinal and transverse sound velocities acquired, and the data post-processing using these sound velocities. In the data post-processing, the densities, specific volumes and bulk moduli are calculated. The calculated bulk moduli in this contract report are higher than expected. In the following sections, the Brillouin scattering test procedure is recapped. Then, a review of the data and improved data processing is presented.

2. The Brillouin Scattering Data

In the Brillouin scattering system at the CIW, an incident laser light from a single mode Ar-ion laser (wavelength λ=514.5 nm) passing through a transparent sample, held pressurized in a diamond anvil cell, is scattered in relation to the thermally excited acoustic waves in the sample (see figure 1).

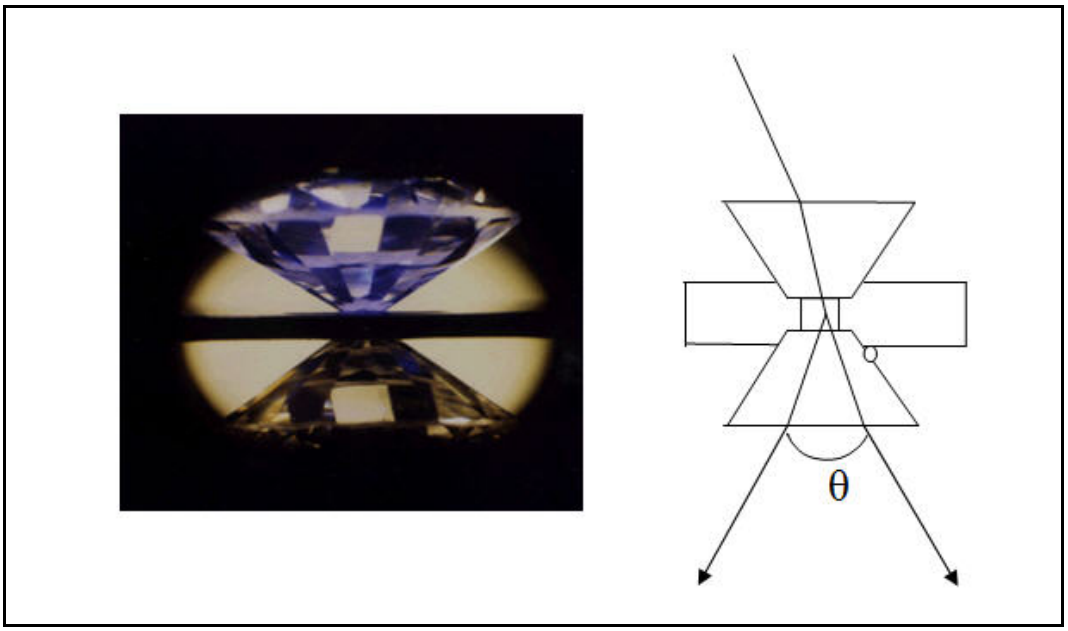

Figure 1. Diamond anvil cell for the Brillouin scattering test.

When the incident light and scattering light directions are arranged symmetrically, the frequency shift $\Delta\nu$ in the scattering light has the following relationship with the acoustic property of the sample:

$$\Delta\nu = \left(\frac{2\upsilon}{\lambda}\right)\sin\left(\frac{\theta}{2}\right) \tag{1}$$

where υ is the sound velocity, $\lambda = 514.5$ nm is the laser wavelength, and $\theta = 80°$ is the scattering angle.

The ballistic gelatin is non-linear viscoelastic; but, when the gelatin is considered as linear elastic isotropic, its density at temperature T can be related to pressure as:

$$(\rho - \rho_0)\big|_T = \int_{P_0}^{P} \frac{\gamma}{\left(\upsilon_L^2 - \frac{4}{3}\upsilon_T^2\right)} dP, \tag{2}$$

where ρ_0 and ρ are the density at pressure P_0 and P respectively, υ_L and υ_T are the longitudinal and transverse sound velocities respectively from the Brillouin scattering measurement, and $\gamma = C_p/C_v$ is the specific heat ratio. This procedure is repeated for a series of different temperatures.

The density, pressure and temperature data thus obtained are then fitted with the Vinet formulation of isothermal equation of state:

$$\left(P - P_0\right)\Big|_T = 3B_0\left[\frac{1-\left(\frac{V}{V_0}\right)^{\frac{1}{3}}}{\left(\frac{V}{V_0}\right)^{\frac{2}{3}}}\right]\exp\left[\frac{3}{2}\left(B_0^{'}-1\right)\left(1-\left(\frac{V}{V_0}\right)^{\frac{1}{3}}\right)\right], \tag{3}$$

where B_0 is the isothermal bulk modulus, B_0' is its pressure derivative at zero pressure, and V_0 is the initial specific volume at zero pressure. The calculated isothermal bulk modulus B_0 (at reference temperature 302 °K) is found to be 4.6 GPa; its pressure derivative B_0' to be 8.7 GPa and the initial specific volume V_0 to be 1.004 cm^3/g. The isothermal bulk modulus B_0 can be translated into the adiabatic bulk modulus through the relationship $B_s=\gamma x B_0$. When compared with gas gun Hugoniot test data (*5, 6, 7*), the adiabatic bulk modulus has been found to be too high.

3. Reassess the Brillouin Scattering Data

During the density calculation (equation 2), the specific heat ratio γ has been assumed to be 1. From the thermodynamic relationship

$$C_p - C_V = -T\left(\frac{\partial V}{\partial T}\right)_P^2\left(\frac{\partial P}{\partial V}\right)_T, \tag{4}$$

$C_P=C_V$ for truly incompressible material, therefore $\gamma=1$; which is valid for most geology problems. However, the specific heat ratios of some polymers at ambient atmospheric pressure are mostly greater than one, i.e., $1 < \gamma < 2$.

While there is no good data for the specific heat ratio for the ballistic gelatin, data fittings using the Vinet equation of state (equation 3) and another widely used Birch-Murnaghan isothermal equation of state,

$$\left(P - P_0\right)\Big|_T = \frac{3}{2}B_0\left[\left(\frac{V}{V_0}\right)^{-\frac{7}{3}} - \left(\frac{V}{V_0}\right)^{-\frac{5}{3}}\right]\left[1-\left(3-\frac{3}{4}B_0^{'}\right)\left\{\left(\frac{V}{V_0}\right)^{-\frac{2}{3}}-1\right\}\right], \tag{5}$$

have shown that increasing the specific heat ratio will result in lowering the calculated isothermal bulk modulus B_0. However, to lower the adiabatic bulk modulus (calculated from the isothermal bulk modulus) to the level comparable to that from the gas gun Hugoniot data will need to increase the specific heat ratio to about 6 or 7, which is most likely not physically realistic. So for the data points at higher pressures, the ballistic gelatin must have transformed into a different phase. The CIW report has indeed mentioned that a phase change has been

observed at around 2 GPa: when "the samples are compressed to around 2 GPa and stay at this pressure regime, the samples will crystallize; if they are compressed very quickly and pass through this pressure regime, the samples will not crystallize but will go into a glassy state" (*4*). Even though a comprehensive phase diagram for the ballistic gelatin is not yet available, this phase change of the ballistic gelatin at around 2 GPa is very similar to the phase change of water observed from liquid to ice VII phase at room temperature. So it is safe to say that for the ballistic gelatin at room temperature and at pressure higher than 2 GPa, it has transitioned into a different phase.

When using the Vinet equation of state (equation 3) or the Birch-Murnaghan equation of state (equation 5) to fit the isothermal density and pressure data in the higher pressure phase, the specific volume at zero pressure V_0 is a mathematical construct representing the specific volume of the material in the same molecular structure as that in the high pressure phase but hypothetically existing at zero pressure. This molecular structure cannot be reproduced in the lab for testing. This specific volume V_0 can only be calculated from the data-fitting using either the Vinet equation or the Birch-Murnaghan equation or any other models of choice. The properties thus calculated cannot be verified or compared to the data post-processed from the gas gun Hugoniot tests, because the gas gun tests at a higher pressure in that different phase cannot easily be performed.

In contrast to the CIW approach, the pressure range is now divided into two domains: (a) in the low pressure phase, for the pressure below ~2 GPa, the specific heat ratio γ is assumed to be 1.5. From the density measurement (1.058 g/cm^3) by J. Winter at 5 ~ 7 °C (*3*) and the density derivative $\partial\rho/\partial T$ (-0.000415 g/cm^3/°C) mentioned in the CIW report, the zero pressure density ρ_0 is estimated to be 1.05 g/cm^3 at room temperature (30 °C); (b) in the higher pressure phase, for pressure higher than 2 GPa, the specific heat ratio γ is assumed to be 1.0, which is valid for incompressible material, because at higher pressure, the material has transitioned into a glassy state. The zero pressure density ρ_0 is a mathematical construct, which is to be estimated from the equation of state curve-fit. The results are shown in figure 2, wherein the pink line labeled 'Vinet' is from the curve-fitting using the Vinet isothermal equation of state (equation 3), whereas the yellow line labeled 'B-M' is from the curve-fitting using the Birch-Murnaghan equation of state. The two curves don't differ much. Both show a transitional twist near 2 GPa in relation to the probable phase change.

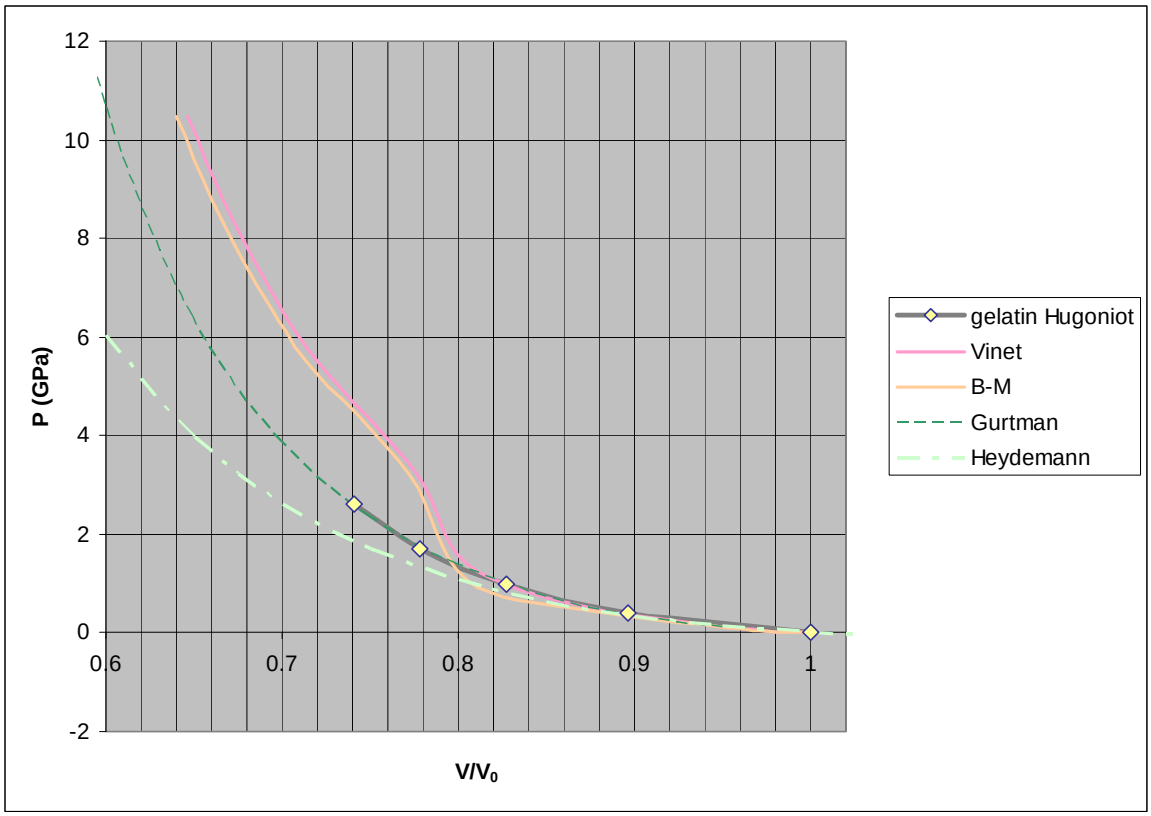

Figure 2. Estimated pressure versus specific volume ratio for gelatin, from Hugoniot data and isothermal equation of state curve-fits for both low pressure and high pressure domains. The Hugoniot and isothermal data for water are also shown for comparison.

The curve labeled 'Gurtman' came from the Hugoniot curve data for water (*8*). The curve labeled 'Heydemann' came from a Birch-Murnaghan isothermal equation of state calculation using the bulk modulus data from Heydemann's work (*9*). The phase change is not considered in these two curves related to water.

The phase change in water is seen in the phase diagram for water (see figure 3), where the curve on an isothermal plane turned at the phase change from liquid to 'Ice VII' phase.

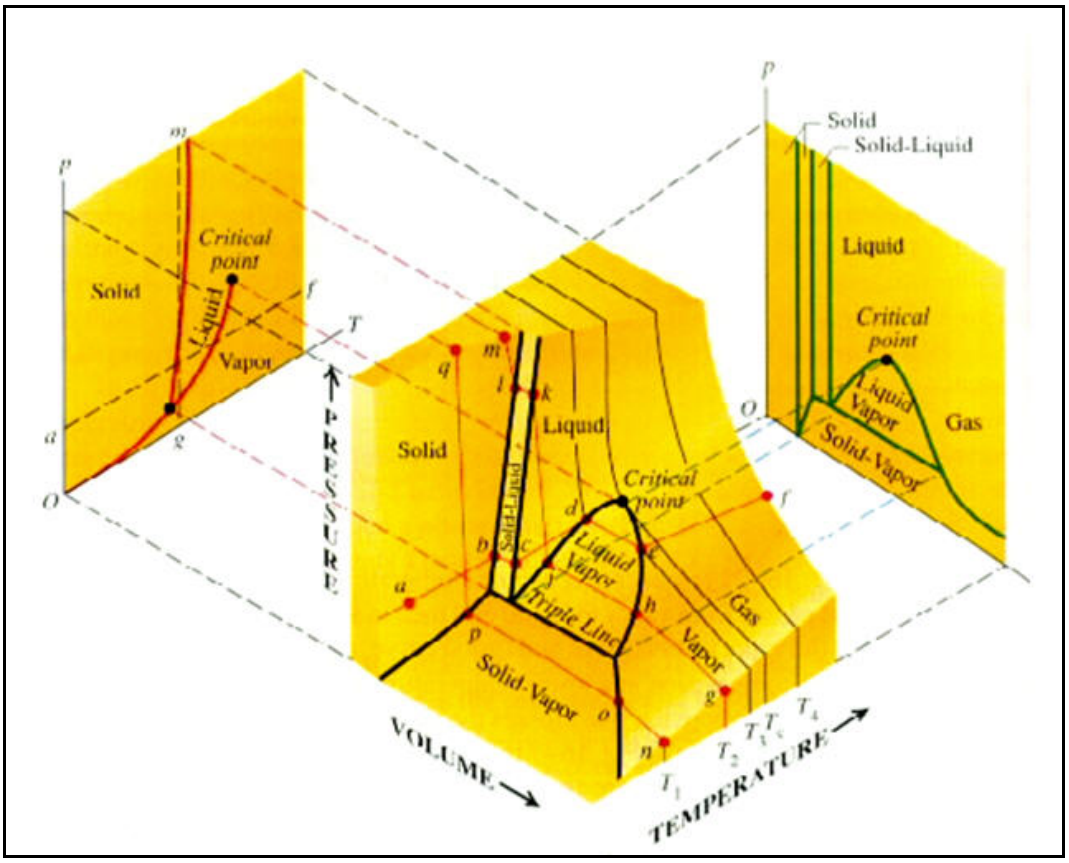

Figure 3. Three dimensional phase diagram for water.

Note: Source for figure 3 is http://www.unca.edu/~dmiller/cceqtn11_a305.ppt (last accessed August 20, 2009).

From the Brillouin scattering data, the bulk sound velocity can be calculated using the relationship:

$$\upsilon_B = \sqrt{\left(\upsilon_L^2 - \frac{4}{3}\upsilon_T^2\right)}, \tag{6}$$

where υ_B is the bulk sound velocity, υ_L and υ_T are the longitudinal and transverse sound velocities respectively from the Brillouin scattering measurement. The calculated sound velocities versus pressure is shown in figure 4.

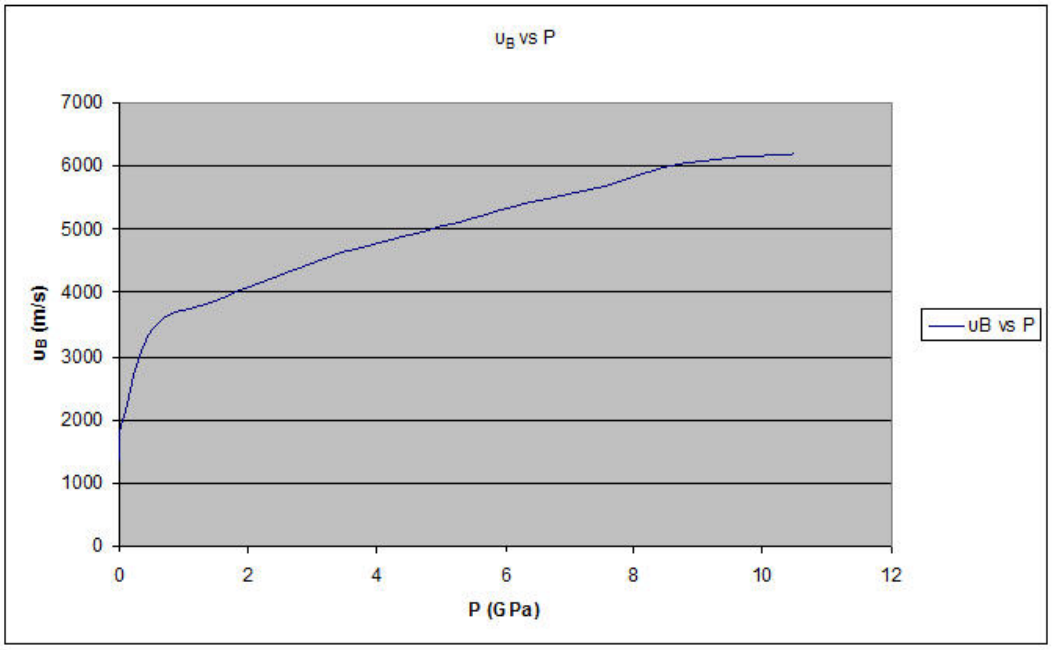

Figure 4. Bulk sound velocity versus pressure calculated from the Brillouin scattering data.

For comparison, the sound velocity versus pressure for water (*10*) is shown in figure 5.

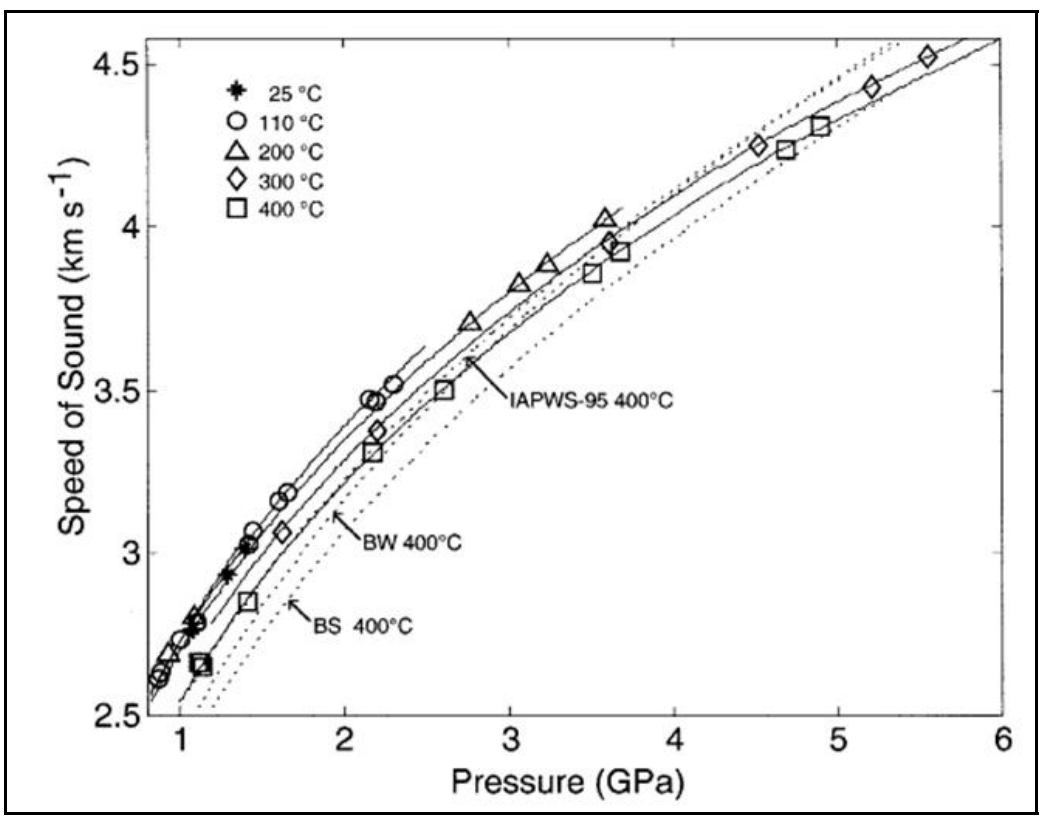

Figure 5. Sound velocity versus pressure along several isotherms for water.

From these calculated bulk sound velocities and the specific volumes from the isothermal equation of state curve-fit, the adiabatic bulk modulus can be calculated from the relationship between the sound velocity and the adiabatic bulk modulus and the density. Figure 6 shows the relation between the adiabatic bulk modulus and pressure.

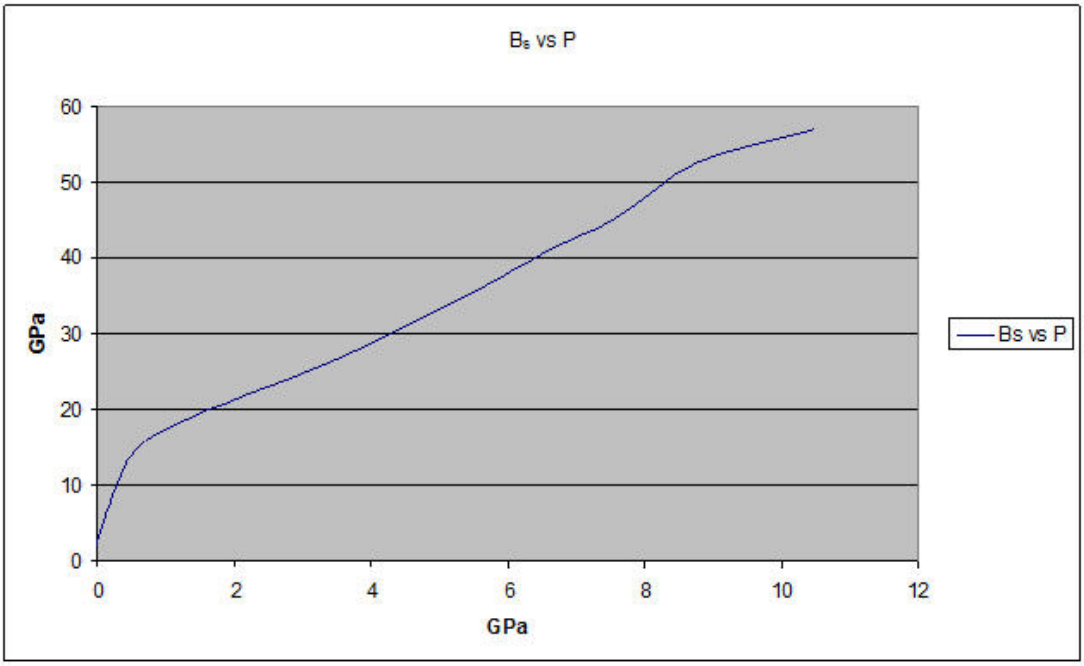

Figure 6. Adiabatic bulk modulus versus pressure calculated from the sound velocities and specific volume.

The bulk moduli at zero pressure for the low pressure domain (below 2 GPa) from the Vinet equation of state, the Birch-Murnaghan equation and from the sound velocity calculations are summarized in the following table:

Table 1. Bulk moduli (in GPa) for the low pressure domain (below 2 GPa) from Vinet equation of state, the Birch-Murnaghan equation of state and from the sound velocity calculation.

	Isothermal Bulk modulus B_0	**B_0 's Pressure Derivative B_0'**	**Adiabatic Bulk Modulus B_s**
Vinet	1.52	13.88	2.28
Birch-Murnaghan	0.94	34.04	1.40
Sound Velocity	—	—	1.95

4. Summary

The Brillouin scattering data have been reviewed and divided into two domains: (a) in the low pressure phase, for the pressure below 2 GPa, the specific heat ratio γ is assumed to be 1.5; (b) in the higher pressure phase, for pressure higher than 2 GPa, the specific heat ratio γ is assumed to be 1.0, which is valid for incompressible material, because at higher pressure, the material has transitioned into a glassy state. The calculated pressure, density data are fitted with the Vinet and the Birch-Murnaghan equations of state to find the isothermal bulk moduli and its pressure derivatives. The results compare more favorably with Hugoniot test data.

More accurate measurements for the densities, the specific heats, the thermal expansion coefficient, and the sound velocities in the low pressure domain will improve the data post-processing. Lab tests in the high pressure domain above 2 GPa are not easy to perform. But the low pressure domain will cover the majority of application needs.

5. References

1. Frasier, J. T. *The Transient Response of Gelatin Targets to Projectile Impacts*; BRL Report No. 1263; Ballistic Research Laboratory: Aberdeen Proving Ground, MD, 1964.
2. Bruchey, W. J.,Jr. *An Instrumented Range Meeting the Requirements of a Wound Ballistics Small Arms Program*; BRL Technical Note No. 1703; Ballistic Research Laboratory: Aberdeen Proving Ground, MD, 1968.
3. Winter, J.; Shifler, D. *The Material Properties of Gelatin Gels*; AD-A008 396, Marvalaud, Incorporated, 1975.
4. Aihaiti, M.; Hemley, R. J. *Equation of State of Ballistic Gelatin*; 50533-EG.1; Carnegie Institution of Washington, 2008.
5. Nagayama, K.; Mori, Y.; Motegi Y.; Nakahara, M. *Shock Hugoniot for Biological Materials*. Shock Waves **2006**.
6. Gojani, A. B.; Takayama, K. Experimental Determination of Shock Hugoniot for Water, Castor Oil, and Aqueous Solutions of Sodium Chloride, Sucrose and Gelatin. *Materials Science Forum* **2008**, Vol. 566.
7. Los Alamos National Laboratory (LANL), unfinished TCG report (2008).
8. Gurtman, G. A.; Kirsch, J. W. Hastings, C. R. Analytical Equation of State for Water Compressed to 300 Kbar. *Journal of Applied Physics* **1971**, *42* (2), 851–857.
9. Heydemann, P. L. M.; Houck, J. C. Self-Consistent Ultrasonic Method for the Determination of the Equation of State of Liquids at Very High Pressures. *Journal of Applied Physics* **1969**, *40* (4), 1609–1613.
10. Abramson, E.H.; Brown, J.M. Equation of State of Water Based on Speeds of Sound Measured in the Diamond-Anvil Cell. *Geochimica et Cosmochimica Acta* **2004**, *68* (8), 1827–1835.

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