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Shock Equation of State of Single Constituent and Multi-Constituent Epoxy- Based Particulate Composites

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SHOCK EQUATION OF STATE OF SINGLE CONSTITUENT AND MULTI-CONSTITUENT EPOXY-BASED PARTICULATE COMPOSITES

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Abstract. There are several studies in the literature regarding the equation of state of alumina-epoxy composites. Although this single component system interacts in a complex manner with shock waves, the addition of a second metal or ceramic particulate can result in even more complex interactions. This paper presents a review of shock loading studies on epoxy-based particulate composites. The relationship between equation of state parameters and particulate concentration is investigated. The measured shock properties are compared with a mixture model for two and three phases.

Keywords: epoxy particulate composites, Hugoniot, mixture model

PACS: 62.50.-p, 62.50.Ef, 64.30.Jk

INTRODUCTION

There are several studies in the literature regarding the equation of state of Al_2O_3 -epoxy [1-5] composites. Although these single component systems interact in a complex manner with shock waves, the addition of a second metal or ceramic particulate, such as in $\text{Al-Fe}_2\text{O}_3$ -epoxy [6,7] or Al-MnO_2 -epoxy, can result in even more complex interactions. The propagated wave in Al_2O_3 -epoxy has been observed to be broadened [1] at low input stress due to the time available for viscous mechanisms. As the input stress increases, the material exhibits viscoelastic behavior [3]. Additionally, the release wave is a strong function

of particle velocity and much faster than the initial shock wave [1,2].

In epoxy [8-10], Carter and Marsh [8] observed a failure of the shock velocity to extrapolate to the ultrasonic bulk sound speed, which they attributed to the distance between the chains being compressed but not the polymer backbones. Additionally, a high pressure phase transition, at ~ 23 GPa, which was attributed to interchain chemical reactions, has been observed [8].

This paper presents a review of shock loading studies on epoxy-based particulate composites. The relationship between equation of state parameters and particulate concentration is investigated. The measured shock properties are compared with a mixture model for two and three phases.

MIXTURE MODEL

Modeling the behavior of composites using the properties of the constituents offers the possibility of greatly reducing the number of experiments needed to validate the Hugoniot. Baer, *et al.* [11-13] have developed a simple model for composite materials based on the properties of the constituents. For the two phase material, the details have been published elsewhere [11,12]; however, a brief description is included here for completeness. The volume fractions (ϕ_i) of the individual constituents must sum to 1, i.e. the mixture occupies all space:

$$\sum_{k=1}^N \phi_k = 1, . \quad (1)$$

The density of the mixture is defined as

$$\phi_1 \rho_1 + \phi_2 \rho_2 = \rho_{mix}, \quad (2)$$

where ρ_i is the density of the constituent and ρ_{mix} is the density of the mixture. Equation 2 can be used to determine both the initial density and the density at pressure. From the jump conditions and characteristics theory [13], the shock velocity, U_s , is

$$U_s = c_s + u_p \quad (3)$$

where c_s is the sound speed and u_p is the particle velocity. The two-component mixture sound speed, in the limit of homogeneous flow [14], is

$$\frac{1}{c_{s,mix}^2} = \frac{1}{(u_{s,mix} - u_p)^2} = \rho_{mix} \left\{ \frac{\phi_1}{\rho_1 c_1^2} + \frac{\phi_2}{\rho_2 c_2^2} \right\} \quad (4)$$

Finally, the change in density across the shock front is

$$\frac{\rho_{mix}^0}{\rho_{mix}} = 1 - \frac{u_p}{u_{s,mix}} \quad (5)$$

where the density is defined by Equation 2 for the initial state (superscript 0) and the final state. Assuming that the particle velocity is the same in all phases, i.e. the constituents are in local mechanical equilibrium [12], the shock velocity in the mixture, along with the volume fractions at pressure, are determined by simultaneously solving Equations 1, 4, and 5.

For a three, or more, phase mixture, the same relationships are used, but are applied to create “1+2” and “1+3” constituents, which are then combined. For these “1+j” constituents, the normalized volume fraction is

$$\alpha_j = \frac{\phi_1}{\phi_1 + \phi_j}, \quad (6)$$

and the density is

$$\rho_{1j} = \alpha_j \rho_1 + (1 - \alpha_j) \rho_j. \quad (7)$$

Equations 4 and 5 can then be rewritten for the “1+j” components and solved simultaneously. Since, again, the mixture occupies all space (Equation 1), the volume fractions are

$$\phi_k = \frac{\alpha_k \phi_1}{(1 - \alpha_k)} \text{ for } k = 2, N. \quad (8)$$

Following from Equation 8,

$$\phi_1 = \frac{1}{\left(1 + \sum_{k=2}^N \frac{\alpha_k}{1 - \alpha_k}\right)} \quad (9a)$$

and

$$\phi_j = \frac{\alpha_j}{(1 - \alpha_j) \left(1 + \sum_{k=2}^N \frac{\alpha_k}{1 - \alpha_k}\right)}. \quad (9b)$$

The shock velocity of the final mixture is then determined from Equation 5, where the density is the density of the total mixture.

The properties of the constituents used in the mixture model are presented in Table 1. All of the constituents were studied under shock loading, with the exception of MnO_2 , which has been investigated under static high pressure. Using the P - V relationship from this work [16] and the Rankine-Hugoniot jump equations, a U_s - u_p relationship for this material was approximated.

TABLE 1. Density, bulk sound speed (C_0), and S for the component materials used in the Baer mixture model calculations.

Material	Density (g/cm ³)	Co (km/s)	S
Al [15]	2.712	5.332	1.3751
Al ₂ O ₃ [15]	3.969	8.14	1.28
Fe ₂ O ₃ [15]	5.00	6.24	1.39
MnO ₂ [16]	5.026	8.0	1.9
Epoxy [8]	1.185	2.69	1.51

TABLE 2. Epoxy-based composite formulations where the shaded rows have nominally the same epoxy content.

Material	Volume Fraction					Density (g/cm ³)	C ₁	C _s (km/s)	C ₀	S	Ref
	Epoxy	Al ₂ O ₃	Al	Fe ₂ O ₃	MnO ₂						
Al-MnO ₂ ¹	0.45		0.24		0.31	2.598	3.36	1.81	2.99	2.01	
A11	0.52	0.48				2.496	3.31	1.82			5
A12	0.57	0.43				2.377	3.20	1.72	2.88	1.99	5
A13	0.57	0.43				2.389	3.13	1.70			5
A14	0.57	0.43				2.391	3.03	1.59			5
fully loaded	0.57	0.43				2.28	3.16	1.68	2.93	1.63	4
300/100/20	0.58	0.42				2.37	3.40	1.57	2.93	1.94	1
F1	0.58	0.42				2.376	3.13		2.79	2.26	3
F2	0.58	0.42				2.429	3.26		2.76	2.26	3
Al-Fe ₂ O ₃	0.60		0.16	0.24		2.366	2.94	1.34	2.02	3.48	6
A15	0.62	0.38				2.233	3.03	1.59			5
A16	0.66	0.34				2.377	2.99	1.56			5
200/100/20	0.66	0.34				2.13	3.09	1.43	2.87	1.18	1
half loaded	0.75	0.25				1.87	2.78	1.4	2.63	1.66	4
Al-Fe ₂ O ₃	0.78		0.09	0.13		2.047	2.62	1.31	3.08	1.22	6
100/100/20	0.80	0.20				1.76	2.82	1.23	2.66	1.60	1
A17	0.8	0.2				2.121	2.74	1.34			5

¹ Linear fit only valid $u_p = 0.3$ -1.3

DISCUSSION

All of the experimental results presented in this paper have been published in the literature, with the exception of Al-MnO₂-epoxy, which will be published at a later date. The materials referenced in this section are presented in Table 1.

The Hugoniot for two Al₂O₃-epoxy composites are presented in Figure 1. Al₂O₃ – 57 vol.% epoxy is the most extensively studied formulation [1,3-6], and this curve is a compilation of the data from these references. The mixture model for the two formulations (57 and 80 vol.% epoxy) is also presented. It can be seen that decreasing the amount of epoxy increases the shock velocity for equivalent particle velocity. The mixture model describes the behavior of the composites within approximately 10%.

A similar graph, Figure 2, has been prepared for the multi-constituent epoxy-based composites. The mixture model has been solved for these composites and, again, describes the behavior of the composites to within approximately 10%.

In order to make the dependence on epoxy volume fraction, the pressure, at $u_p = 0.370$ km/s, for Al₂O₃-epoxy, Al-Fe₂O₃-epoxy, and Al-MnO₂-epoxy, was extracted to create Figure 3. If there was not an experimental point at this exact u_p , it

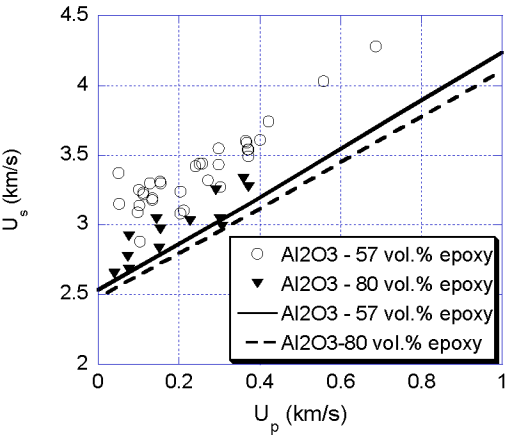


FIGURE 1. Shock velocity versus particle velocity for Al₂O₃-epoxy particulate composites, where symbols are experimental data and lines are the mixture model.

was interpolated from the next nearest points. The two-phase Baer model was used to calculate the solid line of expected pressures for Al₂O₃-epoxy. The mixture model (heavy black line) tends to under predict the pressure at this u_p . In order to determine the origin of this under prediction, an analysis of the experimental uncertainty was conducted. For both constituents, the uncertainty for the experimentally measured U_s versus that calculated using a linear fit to U_s - u_p data was determined. At $u_p = 0.370$ km/s, the uncertainty in

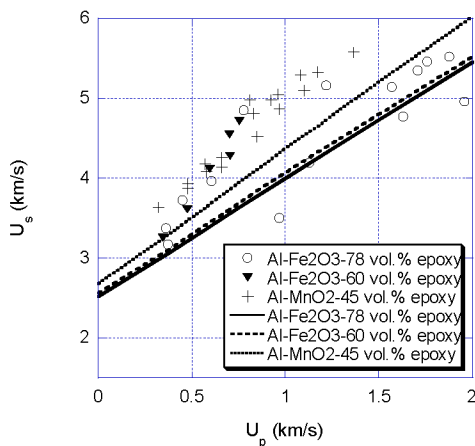


FIGURE 2. Shock versus particle velocity for multi-constituent particulate composites, where symbols are experimental data and lines are the mixture model.

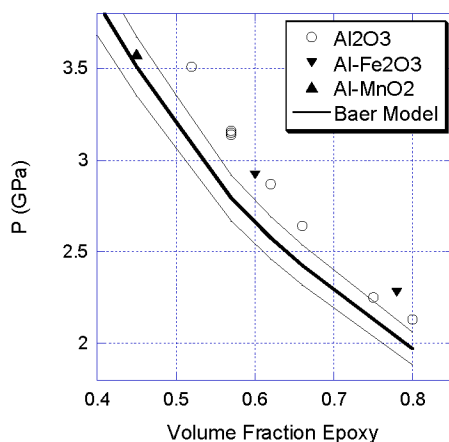


FIGURE 3. Pressure ($u_p = 0.370$ km/s) versus volume fraction epoxy, where the symbols are experimental data and the lines are the mixture model.

the epoxy is 0.03 and 0.08 in the alumina. An uncertainty analysis for the mixture model [16] yields the “error” bars shown as the light black lines in Figure 3. For higher epoxy volume fractions, the experimental points start to fall within the error bars. The uncertainty in the linear fit is not the only error in the experimentally measured properties of the constituents, which could further expand these bounds. Further refinement of the constituent properties, e.g. density, should improve the fit of the model in Figures 1 and 2.

CONCLUSIONS

A mixture equation of state for Al_2O_3 -epoxy composites and epoxy-aluminum-(iron, manganese) oxide has been generated and compared to experimental data. The mixture model predicts the behavior to within $\sim 10\%$. However, variation in the properties of the constituents, in particular density, has not been accounted for.

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