LLNL-JRNL-737373

LAWRENCE LIVERMORE NATIONAL LABORATORY

Shock Compression Response of Model Polymer/Metal Composites

D. B. Bober, Y. Toyoda, B. Maddox, E. Herbold, Y. Gupta, M. Kumar

August 23, 2017

Dynamic Behavior of Materials, Conference Proceedings of the Society for Experimental Mechanics Series

Disclaimer

This document was prepared as an account of work sponsored by an agency of the United States government. Neither the United States government nor Lawrence Livermore National Security, LLC, nor any of their employees makes any warranty, expressed or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States government or Lawrence Livermore National Security, LLC. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States government or Lawrence Livermore National Security, LLC, and shall not be used for advertising or product endorsement purposes.

Shock Compression Response of Model Polymer/Metal Composites

David Bober[1,a)], Yoshi Toyoda[2], Brian Maddox[1], Eric Herbold[1], Yogendra Gupta[2] and Mukul Kumar[1]

[1] *Lawrence Livermore National Laboratory, 7000 East Avenue Livermore CA 94550.*
[2] *Washington State University, 1455 NE College Ave, Pullman, WA 99163.*

[a)] Corresponding author: Bober1@llnl.gov

Abstract. Heterogeneous materials do not respond mechanically to an impulse in the manner of homogeneous metals and alloys. Wave propagation in a microstructure with chemically distinct identities, that are only in incidental contact with each other, is a complex process and not well understood. Here we report on a series of plate-impact experiments on a polymer-metal composite, where the volume fraction of the metallic phase was systematically varied from 0 to 40%, while other parameters like the sample thickness were kept constant. The velocity histories at the sample/window interfaces were measured to examine the continuum response corresponding to the internal materials processes. The unfilled polymer demonstrated a steady shock wave response; whereas the wave profiles obtained from mixture samples showed structured waves that depended on the volume fraction of the fill. The shock wave profiles were quantified using parameters strongly correlated to the material composition. The likely physical basis of these correlations is discussed.

Keywords: Shock response; Heterogeneous material; Particulate composite; Plate impact; Stress wave

INTRODUCTION

Shock waves propagating in matrix-particle composites exhibit complex structures that are distinct from their homogenous counterparts. This class of materials has relevance to many shock wave related applications, such as polymer bonded explosives, electronics packaging, and saturated soils. Thus, a good understanding of shock wave structures in heterogeneous materials is important for developing improved material models, which are needed for engineering hydrocode calculations. In the simple case of a matrix filled lightly with perfectly elastic spheres, shock loading results in a multiphase flow characterized by unsteady stress/velocity fields around each particle. In principle, point mass models with appropriate equations for momentum and heat transfer between the phases could be used to provide a macroscale understanding of these flows [1]. However, particle deformation, fracture, and particle-particle interaction constitute significant complications. In the most densely filled composites, like wet sand, the response is further complicated because large stresses are transmitted over long distances through particle-particle contact [2].

To amplify heterogeneity effects, we chose to study the case of stiff/dense tungsten particles embedded in a soft/light polymer matrix. The acoustic impedance mismatch between these materials represents an extreme difference achievable between two fully dense solids. Our objective was to use this pronounced heterogeneity to search for trends in the shock wave profiles and identify potentially relevant scaling parameters. To that end, a series of composites having different matrix-fill ratios were prepared and subjected to shock loading via gas-gun driven flyer plates. Analysis of the shock wave profiles is presented that quantifies their dependence on the composite's phase fraction. We close with a discussion of the potential physical mechanisms.

MATERIALS AND METHODS

A proprietary thermosetting polymer resin/catalyst mixture was loaded with varying volume fractions of tungsten particles. The tungsten particles were spheroidal, with a mean diameter of 16.5 µm. 1 mm sheets of the composite were made by pressing the uncured mixture between two flat steel plates, which were then heated to cure the polymer. The target assembly consisted of a polymer-tungsten sample (12 mm diameter, 1 mm thick) sandwiched between an aluminum buffer (25.4 mm diameter, 1-2mm thick) and a lithium fluoride (LiF) optical window (8mm diameter, 4mm thick). These were bonded using thin layers of Epo-Tek 301 epoxy (Epoxy Technology, Billerica MA). The LiF face adjoining the polymer-tungsten sample was prepared with a diffuse finish and coated with a thin layer of aluminum using physical vapor deposition. The velocity of this interface was monitored with a velocity interferometer system for any reflector (VISAR) [3]. Three additional VISAR probes directed at the release side of the polished aluminum buffer were used to determine shock arrive times and tilt. The VISAR velocity resolution was estimated at ±1%. Each probe had a diameter of 200 µm. The impactor was an aluminum disc 5.5 mm thick, which was driven by a powder gun or two-stage light gas gun. The measured tilt in every experiment was below 3.6 mrad.

RESULTS

The velocity history at the polymer-LiF interface for the neat polymer sample showed a typical sharp shock profile, as shown in Figure 1. The material reached greater than 95% of its final velocity in less than 0.8 ns. The rounded top or 'lazy S' profiles often observed in polymers were not evident. The wave profile was dramatically altered by the addition of only 5% tungsten, as shown Figure 1. In this material, the rapid initial acceleration continued only briefly, followed by a protracted period of an ever-decreasing acceleration. In contrast to the neat polymer, the 5% tungsten material did not reach 95% of its final velocity for more than 50 ns after the initial rise; a difference of approximately three orders of magnitude by the addition of only 5% filler material. Higher tungsten loading produced qualitatively similar wave profiles, and in every case the shock rise times in the W-polymer composites were far longer than in the pure polymer. These profiles match the rounded top response observed for some neat polymers and most particle-polymer composites [4-8]. In addition, steady wave behavior was confirmed for the 10% tungsten composite by the self-similar behavior observed for 1 mm and 2 mm samples. Additional experiments were performed at 2300, 3500 and 5000 m/s impact velocities, with results qualitatively similar to those in Figure 1. The principal difference observed at higher impact velocities was the tendency for the shock front to become somewhat less extended in time. These higher velocity results will be presented in a subsequent publication. For this report, the 1800 m/s experiments (Figure 1) were selected because they showed the most pronounced compositional effects.

We decided that a numerical fit for the wave profiles might be helpful in organizing the subtler differences in the wave profile and in revealing possible trends. A piecewise linear and exponential forms were chosen because they appeared likely to fit the data using a small number of parameters. The slope of the initial linear acceleration was fit via a least squares algorithm. The asymptotic velocity was taken to be steady particle velocity. The transition velocity at which the piecewise function switches from linear to exponential was found by using a non-linear optimization routine in Matlab (Mathworks, Natick MA), which simultaneously fit the exponential time constant. The transition velocity was then nondimensionalized by dividing by the peak particle velocity. The result is a quantity that represents the fraction of the total acceleration that occurred in the fast linear portion of the profile, a parameter that we term the fast rise fraction. In contrast, the exponential time constant provides a measure of time required to reach the steady state velocity after the fast rise fraction had ended. This parameter was left in units of μs, although possible normalizing time scales will be discussed later.

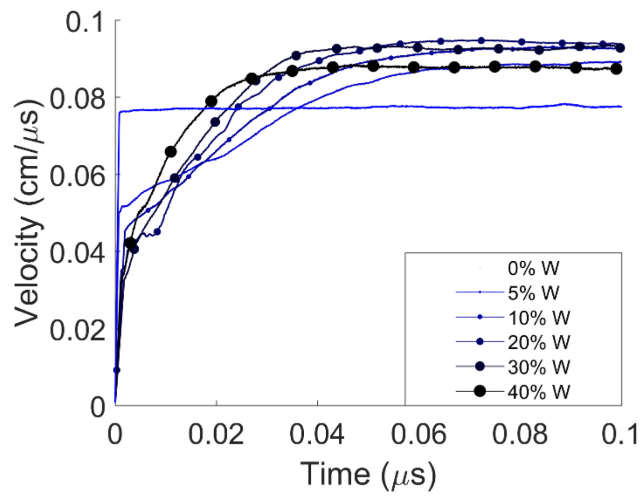
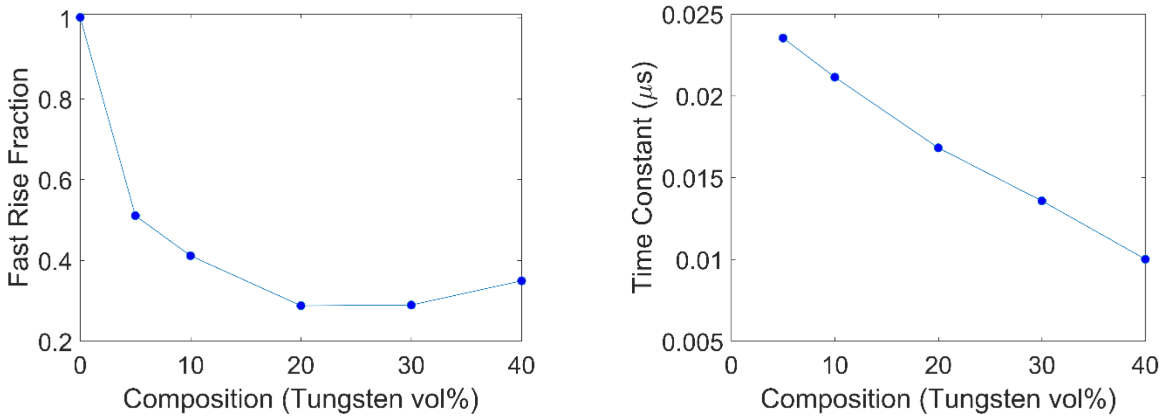


FIGURE 1. The shock profiles for the five composites are much more extended in time than that of the neat polymer (0%W). There are also noticeable differences between the composites, including a curvature which sharpens with tungsten content and the velocities at which they deviate from the neat polymer's

As expected from the introductory observations above, the value of the fast rise fraction for the neat polymer was approximately 1, and dropped to 0.51 for the 5% tungsten sample. Increasing the tungsten content caused this value to drop further until a minimum of 0.29 was reached at 20-30% tungsten. At the highest tungsten loading examined (40%), the fast rise fraction rebounded to 0.35. This trend is shown in Figure 2 (a). Tungsten content also had a significant effect on the time constant of the shock profile. As Figure 2 (b) shows, the time constant dropped monotonically as the amount of tungsten fill was increased. No time constant was determined for the neat polymer because the dominant fast linear rise left an insignificant exponential portion.



(a)

(b)

FIGURE 2. The fast rise fraction shown in part (a) quantifies how much of the acceleration follows the rapid rise seen in the neat polymer versus how much occurs in the slow exponential portion. The exponential time constants plotted in part (b) measure how long it takes the slower second phase of acceleration to bring the material to its steady state velocity. Both are plotted as functions of the tungsten volume fraction.

Organizing the polymer-tungsten materials in terms of tungsten volume fraction is intuitive from a material fabrication standpoint, but may not be the best approach to understanding the shock response. For this, some notion of the microstructural length scale may be helpful. One approach is to calculate the mean interparticle spacing (IPS), which is a measure of the distance between particles surfaces [9]. To better explain the use of IPS, consider a set of monodisperse spheres of radius r' packed together at an arbitrary 'maximum' density, for example the tapped density. The IPS of this packing is defined to be zero because most particles surfaces would be in contact. If a smaller set of spheres of radius r is inscribed concentrically within them, their IPS is given by $2(r'-r)$. Of course, the relative packing density of the r size spheres is easily calculated. More formally, the IPS is given by,

$$IPS = 2r\left(\left(\frac{\Phi_m}{\Phi}\right)^{1/3} - 1\right) \qquad (1)$$

where r is the particle radius, ϕ_m will be taken as the tapped relative density (0.60), and ϕ is the actual packing density [9]. Dividing the IPS by the particle radius produces a non-dimensional microstructural length scale. The fast linear rise fraction and the time constant are plotted in terms of this normalized IPS in Figures 3 (a) and (b). It is striking that the fast rise portion shows a nearly linear relationship with IPS in cases where the particles are widely spaces, i.e. values above about $0.9r$. As the IPS drops to about 0.9-0.5r, an abrupt transition occurs in the fast rise fraction. Figure 3 (b) also shows a distinct downward curvature in the time constant that was not evident from the volume fraction plot.

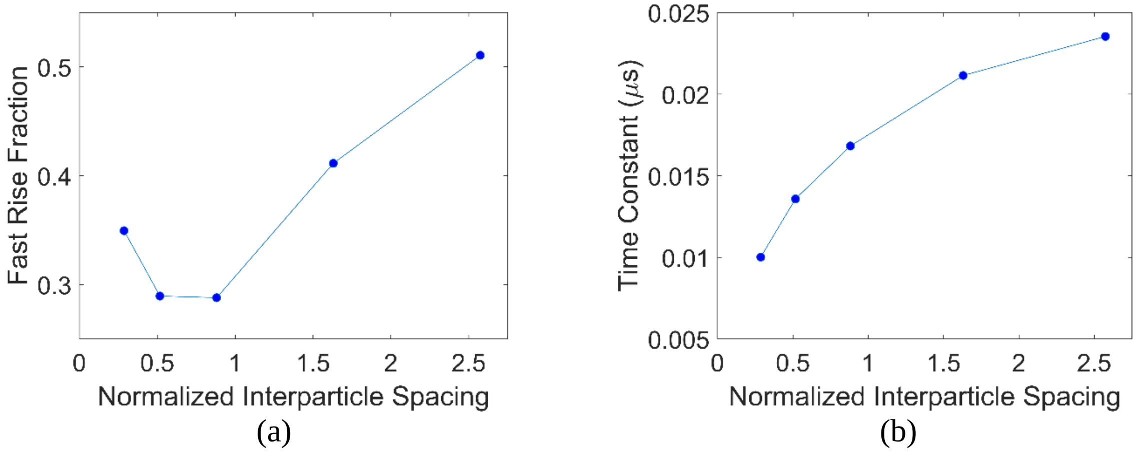


FIGURE 3. The fast rise fraction (a) and time constant (b) shown in Figure 2 are replotted here in terms of the normalized interparticle spacing, which gives a more direct measure of the microstructural length scale.

DISCUSSION

We first consider the time constant for the case of large IPS and use a working hypothesis that the particles do not interact with one another. This is to say that we will assume for the present that their local flow fields decay over short distances and no particle-particle contact occurs. Sufficiently far from any particle, the shock front will resemble a planar shock in the neat material. In those places, the sharp discontinuity separates regions of high stress/high velocity from the ambient state. Closer to a particle, the flow field becomes more complex. As the shock front passes over an isolated particle, there will be a pressure differential between the leading and trailing sides of the particle. This is the so-called inviscid unsteady force [10]. During this same period, the viscous unsteady force will also be accelerating the particle [10]. Together, these forces will act on a time scale (τ_s) set by the particle diameter (d) and the shock speed (u_s), which is given by,

$$\tau_s = \frac{d}{u_s} \text{ [1].} \tag{2}$$

The time scale over which these forces act is shorter than our measured time constants, meaning that a longer lasting force must be sought. For a particle moving slower than a surrounding fluid, this longer lasting acceleration can be supplied by the quasi-steady drag, which will persist so long as the velocity gradient lasts [10]. The time scale is given by,

$$\tau_d = \frac{\rho d^2}{18\mu} \tag{3}$$

where ρ is the particle density and μ is the polymer's dynamic shear viscosity [1]. Given that drag scales linearly with the particle radius and inertia with the radius cubed, larger particles should produce longer time constants. This agrees with the earlier observation by Herbold et al. [11] that larger tungsten particles embedded in a polymer matrix led to a slower shock response than did small particles. Setchell, Anderson and Montgomery [4] likewise found that small spherical alumina particles led to less extended shock fronts than coarse angular ones. The importance of viscosity in an epoxy-alumina composite was illustrated by Setchell et al. [12], who tested the effect of initial temperature. It was found that colder initial temperatures (-55 °C) lead to a faster shock response, while warming to 75°C lead to more extended wave profiles [12]. The inference is that more a more viscous matrix leads to faster momentum transfer and a more compact shock front [12]. The time constants measured here would imply a viscosity of ~10 Pa·S, which is line with our estimate from ongoing numerical simulations. This drag mechanism would partially justify the chosen exponential fit because it implies an accelerating force that decays as the particles gain velocity. Shock waves reflected from other particle-polymer interfaces may also contribute to a second long-time accelerating force. The relative momentum transferred via drag versus shock reverberation is difficult to estimate, though the discussion above and ongoing numerical simulations both suggest that the former dominates.

While the fast-moving polymer slowly accelerates the dense tungsten particles, the particles would in turn act to slow the mean velocity of the nearby fluid. With only a few particles in the system, the total volume of polymer that experiences a drag is minimal, and most polymer will accelerate just as in the neat case. When more particles are added, their collective volume of influence grows and so more of the polymer is subjected to the slow acceleration. Measuring a spatially averaged velocity in such a system would produce a piecewise linear-exponential behavior, with a greater portion for lower particle loadings. This matches the observed trend for the fast rise fraction, which drops for increasing particle loading, at least up to 20%. This also matches Setchell's [12] observation that increased alumina content in an epoxy composite led to longer rise times. Of course, the preceding analysis assumes non-interacting particles and is expected to break down as the IPS becomes small.

The critical value of IPS at which particle interactions become significant is unknown a priori, but the wave profiles provide some valuable clues. As a start, the conclusions of the preceding paragraph break down rapidly as the IPS drops below about $0.9r$. Beyond this point, increasing the particle loading leads to the opposite trend from that predicted for non-interacting particles. The local minimum in Figure 3 (a) can therefore be interpreted as a sign of the transition from isolated to interacting tungsten particles. At this transition, it is expected that the flow fields around neighboring particles will interact. At the small IPS values, the apparent viscosity will probably begin to depend on particle volume fraction. As is well-known in colloidal systems, the addition of rigid particles concentrates the shear flow through smaller interparticle fluid volumes [13]. An analogous concentration of shear in a shocked particle-polymer matrix has been seen in simulations, including interparticle jetting [11]. The expected effect is to amplify the drag force, even though the intrinsic matrix properties are essentially unchanged. This provides a plausible explanation for the downward slope and curvature of the time constant when plotted with respect to IPS. Increased drag would shorten the shock width by causing faster momentum transfer between the phases. This is analogous to previous observations in epoxy particle composites in which colder initial temperatures led to more extended shock profiles. While Setchell [12] observed a change due to intrinsic viscosity, here the hypothesis is that the apparent viscosity is being modified by the changing characteristic microstructural length scale.

As the IPS is decreased, the particles themselves will eventually encounter one another. At very high packing densities, this could lead to stress transmission over long distances via force chains, as is well-known to occur in granular materials [2]. Indeed, at a sufficient packing density the distinction between a matrix-particle composite and a saturated granular material disappears. For randomly packed equal sized spheres, the critical volume fraction for the emergence of continuous percolating chains is 18.3% [14]. Of course, that does not necessarily imply that these chains have sufficient lateral confinement to transmit high stresses over long distances [15]. Indeed, there is evidence in the literature that the properties of the particles remain relatively unimportant up to a density of at least 45% [16]. It has been observed that epoxy composites with alumina, aluminum, tungsten carbide, and a mixture of aluminum and tungsten particles all had similar shock responses [16]. The main difference was a shift due to the overall density, which is contrary to what would be expected if particle force chains had a dominant role [16, 17]. This conclusion agrees with direct numerical simulations which did not show evidence of force chains at 45% particle fraction [8]. Even in the case where particles are certainly close enough to interact, thin intervening layers of polymer can have a dramatic effect on wave propagation [18]. This effect can also be seen in the dependence of storage and loss moduli on composition [19]. Additional evidence for the dominance of the matrix material at packing a fraction of 43% comes from the viscous/temperature dependence [12]. White et al. [20] found that increasing the metal fill of an epoxy composite from 30% to 40% led to a percolation-like jump in the dynamic strength. In the context of these previous studies, the maximum particle fraction studied here (40%) is approaching, but may not reach, the regime in which granular force chains dominate the response.

CONCLUSION

It was found that the shock profiles of a set of polymer-metal composites depend strongly on composition. These trends can be quantified in terms of a single time constant and a transition velocity, both of which can be related to physical phenomena. The governing mechanism is believed to be a particle-matrix drag effect, in which the small dense tungsten spheres retard the acceleration of the surrounding polymer. For dilute composites, increasing the tungsten content led to more extended shock profiles, probably because the drag effect scales with the polymer volume that interacts with the filler. While we see evidence that neighboring particles interact at small interparticle spacings, long range particle to particle stress transfer is unlikely. Even at 40% tungsten, neighboring particle interaction are most likely still strongly mediated by the intervening polymer. The current hypothesis is that this leads to a rise in the apparent viscosity, which in turn causes a drop in the measured time constant. The relative role of shock reverberation requires more study, but appears to be modest. Three dimensional direct numerical simulations are planned to further refine these conclusions and better quantify the particle-matrix interaction. It is desirable to collect shock profiles for different size tungsten particles to provide valuable scaling information. The relationship between the time constant and the steady wave development time is also an area of active pursuit.

ACKNOWLEDGMENTS

This work was partly performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344. The work at Washington State University was supported in part by DOE/NNSA (DE-NA0002007).

REFERENCES

[1] Ling Y, Haselbacher A, Balachandar S, Najjar FM, Stewart DS. Shock interaction with a deformable particle: Direct numerical simulation and point-particle modeling. J Appl Phys 2013;113:013504.
[2] Nesterenko V. Dynamics of Heterogeneous Materials: Springer New York; 2013.

[3] Barker LM, Hollenbach RE. Laser Interferometer for Measuring High Velocities of Any Reflecting Surface. J Appl Phys 1972;43:4669-75.
[4] Setchell RE, Anderson MU, Montgomery ST. Compositional effects on the shock-compression response of alumina-filled epoxy. J Appl Phys 2007;101:083527.
[5] Rauls MB, Ravichandran G. Shock Wave Structure in Particulate Composites. Procedia Engineer 2015;103:515-21.
[6] Munson DE, Boade RR, Schuler KW. Stress-Wave Propagation in Al2O3-Epoxy Mixtures. J Appl Phys 1978;49:4797-807.
[7] Setchell RE, Anderson MU. Shock-compression response of an alumina-filled epoxy. J Appl Phys 2005;97:083518.
[8] Vogler TJ, Alexander CS, Wise JL, Montgomery ST. Dynamic behavior of tungsten carbide and alumina filled epoxy composites. J Appl Phys 2010;107:043520.
[9] Hao T, Riman RE. Calculation of interparticle spacing in colloidal systems. J Colloid Interf Sci 2006;297:374-7.
[10] Parmar M, Haselbacher A, Balachandar S. Modeling of the unsteady force for shock-particle interaction. Shock Waves 2009;19:317-29.
[11] Herbold EB, Nesterenko VF, Benson DJ, Cai J, Vecchio KS, Jiang F, et al. Particle size effect on strength, failure, and shock behavior in polytetrafluoroethylene-Al-W granular composite materials. J Appl Phys 2008;104:103903.
[12] Setchell RE, Montgomery ST, Cox DE, Anderson MU. Initial temperature effects on the dielectric properties of PZT 95/5 during shock compression. In: Elert M, Furnish MD, Chau R, Holmes NC, Nguyen J, editors. Shock Compression of Condensed Matter - 2007, Pts 1 and 22007. p. 193-6.
[13] Mooney M. The viscosity of a concentrated suspension of spherical particles. J Colloid Sci 1951;6:162-70.
[14] Powell MJ. Site Percolation in Randomly Packed Spheres. Phys Rev B 1979;20:4194-8.
[15] Tordesillas A, Muthuswamy M. On the modeling of confined buckling of force chains. J Mech Phys Solids 2009;57:706-27.
[16] Jordan JL, Herbold EB, Sutherland G, Fraser A, Borg J, Richards DW. Shock equation of state of multi-constituent epoxy-metal particulate composites. J Appl Phys 2011;109:013531.
[17] Millett JCF, Bourne NK, Deas D. The equation of state of two alumina-filled epoxy resins. J Phys D-Appl Phys 2005;38:930-4.
[18] Daraio C, Nesterenko VF. Strongly nonlinear wave dynamics in a chain of polymer coated beads. Phys Rev E 2006;73:026612.
[19] Veazie D, Jordan JL, Spowart JE, White BW, Thadhani NN. Model for Elastic Modulus of Multi-Constituent Particulate Composites. Exp Mech 2013;53:1213-22.
[20] White BW, Thadhani NN, Jordan JL, Spowart JE. The Effect of Particle Reinforcement On The Dynamic Deformation of Epoxy-Matrix Composites. In: Elert ML, Buttler WT, Furnish MD, Anderson WW, Proud WG, editors. Shock Compression of Condensed Matter - 2009, Pts 1 and 2. Melville: Amer Inst Physics; 2009. p. 1245-8.