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ARO PROJECT No. **W911NF-05-1-0343**

REPORT TITLE: **SHOCK PROCESSING AND HIGH STRAIN RATE PROPERTIES OF
BULK METALLIC GLASSES AND THEIR COMPOSITES**

AUTHORS: **Naresh Thadhani and Morgana Martin**

The high-strain-rate mechanical properties (including deformation and fracture mechanisms) as well as high-pressure phase transformation characteristics of a Zr-based bulk metallic glass (BMG) and its composite with tungsten have been investigated through impact experiments and constitutive modeling. BMGs exhibit unique properties including high strength, deformation by shear banding, high strength-to-weight ratio, and excellent corrosion resistance; however, they undergo catastrophic failure due to localized deformation. Restricted shear band propagation and controlled fracture response can be achieved via addition of reinforcement particles or alteration of microstructure by crystallization phase transformations.

Our work has involved performing controlled impact experiments on bulk metallic glass (BMG) composites consisting of an amorphous $Zr_{57}Nb_5Cu_{15.4}Ni_{12.6}Al_{10}$ (Vitreloy106) matrix with crystalline tungsten reinforcement particles. The deformation and failure response of monolithic Vitreloy106 have also been examined to aid in the understanding of the composite. The high-strain-rate mechanical properties of both the monolithic BMG and BMG-W composite have been investigated using dynamic compression (reverse Taylor) and dynamic tension (spall) impact experiments performed using our single-stage 80-mm gas gun instrumented with velocity interferometry (VISAR) and high-speed digital photography combined with the 2-stage gun at NIMS in Japan, as well the dynamic testing facilities at Chemnitz University, in Germany. The experiments have provided information about the dynamic strength and deformation modes over a range of temperatures and strain rates, and allowed validation of constitutive models via comparison of experimental and simulated transient deformation profiles and free surface velocity traces. Equation of state (Hugoniot) measurements performed on the monolithic BMG reveal a polyamorphism transformation to a high-modulus and high-density whose formation increases the strain-rate sensitivity of strength and fracture stress.

The overall *objective* of this research has been to determine the high-strain-rate deformation and failure mechanisms and high-pressure behavior of BMG and BMG-matrix composites under dynamic compressive and tensile loads and to correlate these mechanisms to develop and validate constitutive models that describe the mechanical behavior over a range of loading conditions. The *significance* of this research has been the development of the fundamental materials-science based understanding that can enable the design of the bulk metallic glass of compositional that can undergo stress-induced polyamorphism transformation and thereby enable increased deformation and fracture resistance under high-strain-rate and shock-loading conditions. The research conducted involved collaborations with researchers at the Army Research laboratory, the National Institute of Materials Science in Japan, and the Dynamic Testing Laboratory at the Chemnitz University in Germany. The key *results obtained during the course of this program* are briefly summarized next.

High-Strain-Rate Compressive Response of W-BMG composite

Rod-shaped samples of W-Vitreloy106 ($Zr_{57}Nb_5Cu_{15.4}Ni_{12.6}Al_{10}$) composite (density – 14.59 g/cm^3) were obtained from the Army Research Laboratory (courtesy of Dr. Laszlo Kecskes). As shown in Fig. 1(a), the composite samples contain nominally 70wt% W particles embedded in Vitreloy 106 bulk metallic glass matrix. Figure 1(b) shows typical fracture surface of composite, revealing cleavage failure of W particles and the inherent brittle behavior of the glassy matrix.

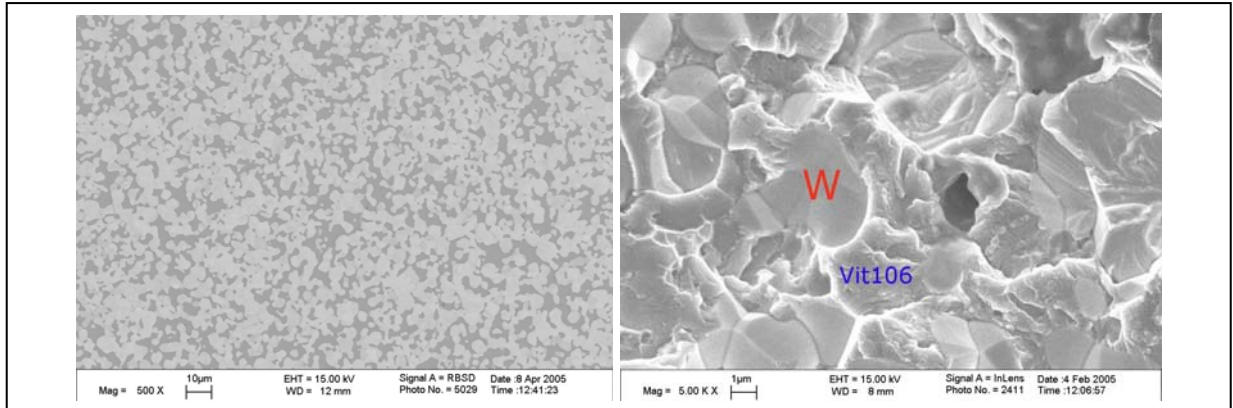


Fig. 1. SEM images showing (a) polished and (b) fracture surface of W-Vitreloy106 composite.

Figure 2 shows results from dynamic compression (anvil-on-rod impact) experiments performed on the W-BMG composite. Constitutive modeling using the Drucker-Prager strength model was performed and correlated with experimental results. The figure shows images comparing the experimental (lower half) and simulated (upper half) profiles of the deforming sample at various times during impact at (a) 134 m/s, (b) 155 m/s, and (c) 186 m/s. The simulation contours represent effective plastic strain. These comparisons show that the Drucker-Prager model provides a decent fit to the experimentally obtained final and transient deformation shapes prior to fracture initiation.

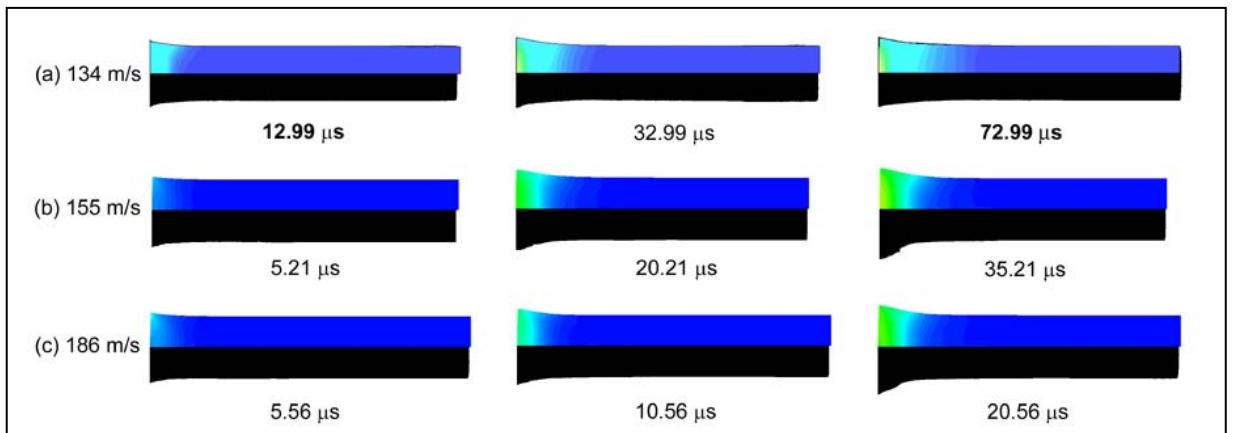


Fig. 2: Correlation between images captured during dynamic compression experiments and simulated specimen profiles of W-BMG composites.

Uniaxial/biaxial Compressive Response of W-BMG composite

The uniaxial and biaxial compressive responses of $Zr_{57}Nb_5Al_{10}Cu_{15.4}Ni_{12.6}$ -W composite were investigated over a range of strain rates ($\sim 10^{-3}$ to 10^3 s $^{-1}$) using an Instron universal testing machine ($\sim 10^{-3}$ to 10^0 s $^{-1}$), Drop Weight Tower (~ 200 s $^{-1}$), and Split Hopkinson Pressure Bar (10^3 s $^{-1}$). The temperature dependence of the mechanical behavior was also investigated at temperatures ranging from RT to 600 °C using the instrumented Drop Weight testing apparatus, mounted with an inductive heating device. The deformed and fractured specimens were examined using optical and scanning electron microscopy. Stopped experiments were used to investigate deformation and failure mechanisms at specified strain intervals in both the Drop Weight and Split Hopkinson Bar tests. These stopped specimens were also subsequently examined using optical and scanning electron microscopy to observe shear band and crack formation and development after increasingly more strain. Figure 3 shows results of stress-strain curves from drop-weight testing and micrographs of stopped specimens.

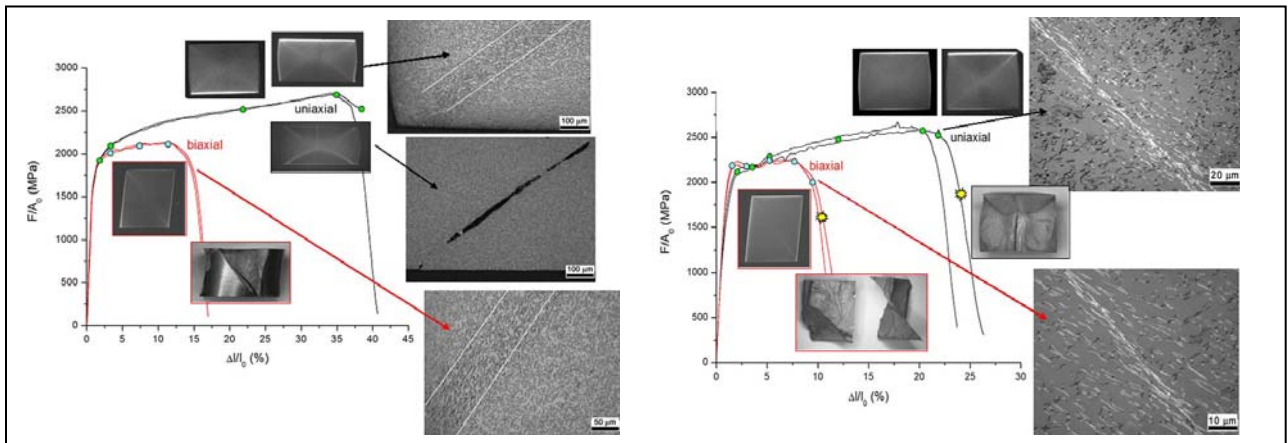


Fig. 3. Stress-strain curves obtained from (a) Drop Weight and (b) Hopkinson bar tests. Circles along curves show strain levels at which tests were stopped and specimen was examined for microstructural evidence of deformation and failure processes. Micrographs of stopped specimen sections which revealed shear bands are shown as well as photographs of failed specimens. Width of shear bands in Drop Weight tests were about ~ 50 - 100 μm , while those in Hopkinson bar tests were ~ 10 - 20 μm .

The results show an increase in yield strength with strain rate and a decrease in failure strength, plasticity, and hardening with strain rate. Comparison of uniaxial and biaxial loading showed strong susceptibility to shear failure since the additional 10% shear stress caused failure at much lower strains in all cases. Results also showed a decrease in flow stress and plasticity with increased temperature, as illustrated in Figure 4. Also notable was the anomalous behavior at 450 °C, which lies between the T_g and T_x and as such is in a temperature regime where homogeneous flow, as opposed to heterogeneous deformation via shear banding, is the dominant mechanism in the BMG. From the overall results, it can be generalized that the tungsten dominates the deformation behavior of the composite given the hardening and large degree of plasticity, which are characteristic of the BCC metal and not the BMG. However, the additional shear stress during biaxial loading causes the BMG to play a strong role. Additionally, at temperatures between T_g and T_x , the BMG deforms homogeneously and this mechanistic change is so significant that the deformation of the BMG plays a significant role in the overall deformation of the composite in spite of its minor volume content (30%).

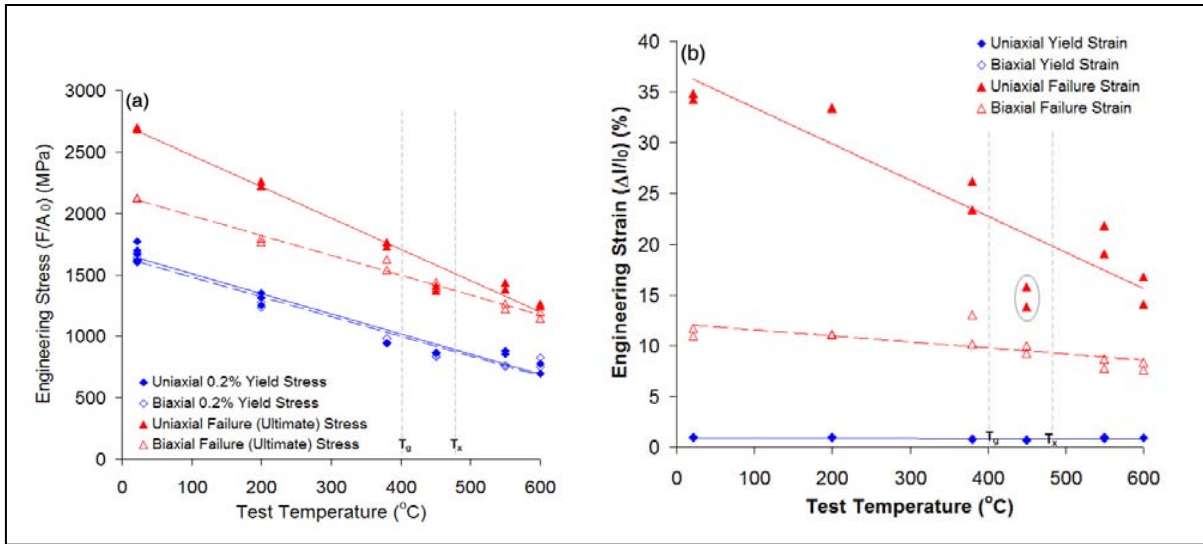


Fig. 4. Temperature dependence of mechanical behavior of uniaxial and biaxial LM106-70W specimens tested at $\sim 200 \text{ s}^{-1}$. (a) Yield and failure stresses and (b) yield and failure strains as function of temperature. In (a) and (b) the data from 450 °C tests (circled) deviate from otherwise linear trend. For both uniaxial and biaxial specimens, yield stress decreases with increasing test temperature, and this decrease occurs at the same rate, regardless of specimen configuration. Additionally, failure stress decreases with increasing test temperature, but the uniaxial failure stress, which is always above that of the corresponding biaxial specimen, decreases at a much higher rate such that at 600 °C the uniaxial and biaxial failure stresses are approximately same. Strain to failure decreases with increasing test temperature, as shown in (b). Both the failure stress and failure strain measured for the 450 °C test temperature, deviate from otherwise nearly linear trend (circled points), due to difference in deformation mechanism in the temperature range.

High-Strain-Rate Compressive Response of Monolithic Vitrealoy BMG

The dynamic compressive response of the monolithic BMG, using anvil-on-rod impact experiments, has been investigated using sleeved and un-sleeved samples to probe the effect of the imposed radial confinement stress. Figure 5 shows the schematic and photographs of the fractured BMG rods recovered after being impacted at (a) 59, (b) 98 and (c) 131 m/s. Black lines indicate starting sleeve/specimen configurations and gray lines indicate fracture and final/recovered configurations. The photographs show the obvious difference in failure mechanisms when comparing the sleeved and non-sleeved specimens (Fig. 5 (b)), both of which were impacted at 98 m/s. Because the sleeve allows for preservation of shear planes, we can also see the shear plane failure angles and the differences in orientation and locations of these planes. The photographs also reveal the fracture response as a function of impact velocity, illustrating almost single shear failure (42.5° angle) at 59 m/s, to multiple shear failure at 98 m/s, and conical symmetry failure at 131 m/s. These differences are attributed to the triggering and propagation of single, double, and multiple (symmetry) shear bands leading to failure under compression.



Fig. 5. Monolithic BMG rods (confined by stainless steel sleeves) recovered post-impact after anvil-on-rod dynamic compression tests performed at (a) 59 m/s, (b) 98 m/s, and (c) 131 m/s.

High-Pressure Phase Stability and Equation of State

The high-pressure stability of the BMG was investigated via Hugoniot equations of state measurements, based on PVDF+VISAR experiments performed at Georgia Tech and streak camera experiments performed at the National Institute of Materials Science. Fig. 6 (a) illustrates U_s-U_p data, in which bulk sound speed (C_B) and longitudinal sound speed (C_L) were calculated from elastic properties data, and U_p values obtained either using impedance matching (in high pressure regime >67 GPa), and experimentally determined, are plotted in the mixed phase and the low pressure regions. All lines are linear fits to the corresponding data. Figure 6(b) shows a plot of pressure as a function of density. The lines in the elastic and mixed phase regions are linear fits and the lines in the low and high pressure regions are Birch-Murnaghan equation fits. For low pressure phase, the Birch-Murnaghan Equation is plotted using the known bulk modulus (K_0) value and K_0' is taken as $4S-1$. For the high pressure phase, the best fits of K_0 , K_0' and ρ_0' were determined using least squares regression.

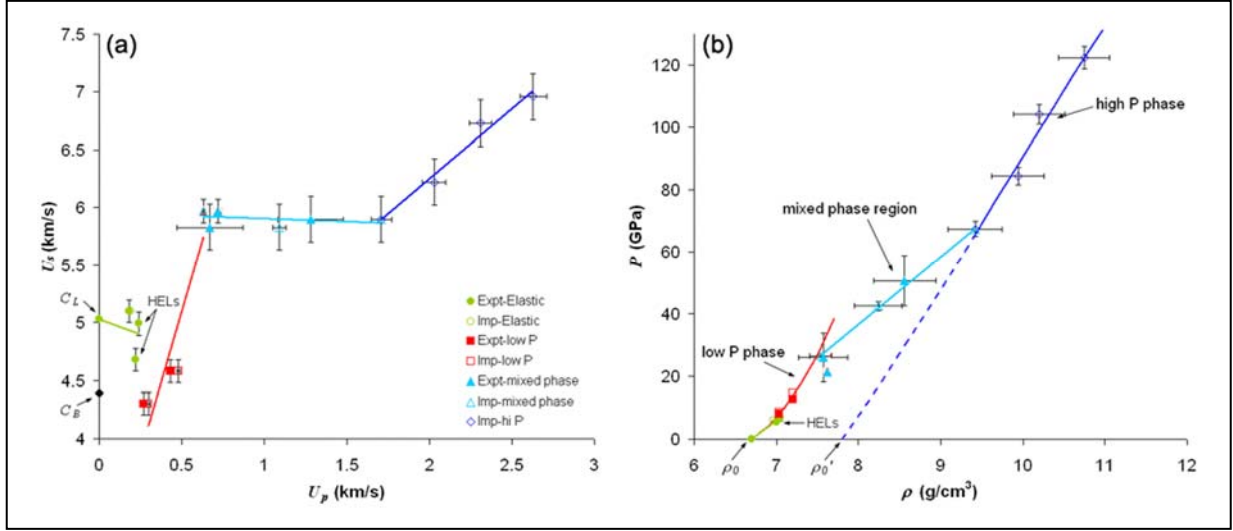


Figure 6: Equation of state data plotted as (a) U_s - U_p and P - ρ plots showing evidence of a transition to a mixed phase at 26 GPa and a transition to a second phase at 67 GPa.

The U_s - U_p plot shows four regions; (1) HEL region ($U_p \leq 0.24$ km/s), (2) the low pressure plastic region ($0.27 \leq U_p \leq 0.72$ km/s), (3) the mixed phase region ($0.72 \leq U_p \leq 1.71$ km/s), and (4) high pressure plastic region ($U_p \geq 1.71$ km/s). The onset stress for the phase transition is about 26 GPa. The U_s - U_p relation in the elastic region is given by $U_s = 5.03 - 0.54U_p$ ($U_p \leq 0.24$ km/s). The negative slope is not unexpected due to the existence of dispersed elastic shock fronts in amorphous materials, because of the negative first pressure derivative of elastic modulus. However, because of only one point causing the negative trend, it possibly could be due to experimental error and thus the slope may be closer to horizontal or slightly positive. The U_s - U_p relations in the plastic regions are given by $U_s = 2.63 + 4.95U_p$ ($0.27 \leq U_p \leq 0.72$ km/s), $U_s = 5.96 - 0.05U_p$ ($0.72 \leq U_p \leq 1.71$ km/s), and $U_s = 3.83 + 1.21U_p$ ($U_p \geq 1.71$ km/s). The pressure versus density plot also shows four distinct regions: an elastic region, a low pressure phase, a mixed phase region and a high pressure phase. The Birch-Murnaghan equation was used to calculate the pressure-density relationship for the BMG in respective low and high pressure regions. For the low pressure phase, the equation was plotted using known value of K_0 (118 GPa) and K_0' was taken to be $4S-1$, where S was determined from the U_s - U_p plot. For the high pressure phase, the values of K_0 , K_0' and ρ_0' (zero pressure density of the high pressure phase) were determined using least squares regression to determine the best fit to the experimental data, and were found to be 288 GPa, 2.3 and 7.8 g/cm³, respectively. The analysis confirms that the BMG goes through polymorphism transition to high-pressure phase having a bulk modulus of 288 GPa, which is $\sim 144\%$ higher than the ambient pressure phase, but not unrealistic.

Effect of Polyamorphism on Strain Rate Sensitivity

The polyamorphism transition has an influence on the yield and failure strength (normalized by elastic and plastic modulus, respectively) of the BMG as shown in Figure 8, which compare the strain rate effects in the BMG with pure BCC-tungsten and a BMG-tungsten composite. While the BCC-tungsten and the BMG-W composite (with tungsten dominating its response) have an obvious effect of strain rate on strength and failure response, the monolithic BMG shows a substantial increase in yield (strengthening) and fracture (toughening) stress. The effect is attributed to the influence of polyamorphism, with the high-pressure phase contributing to strengthening and the mechanical (PdV) work being dissipated in producing the phase change rather than in crack propagation. The experimental results thus illustrate that the change in mechanical behavior of the monolithic BMG at high strain rates, due to energy dissipation associated with high-pressure polyamorphism, can potentially be used to design BMGs with high yield strength and fracture toughness.

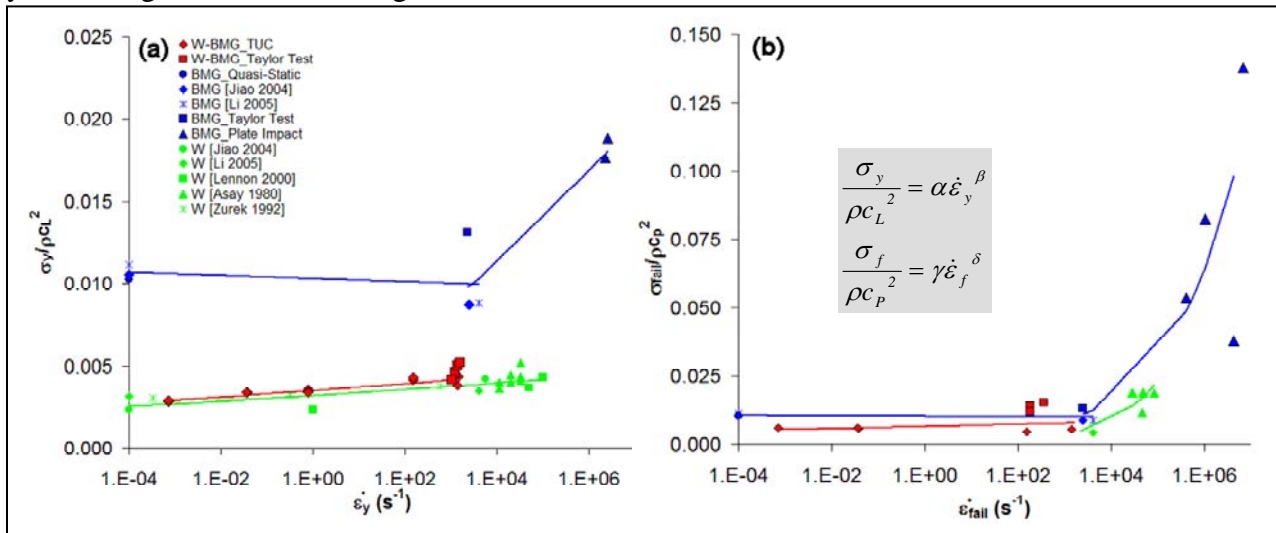


Fig. 8. Strain rate sensitivity of yield and failure strengths (normalized by elastic and plastic modulus, respectively) of monolithic BMG, with pure BCC-tungsten, and BMG-tungsten composite.

Summary and Concluding Remarks

The mechanical behavior of a Zr-based bulk metallic glass and its composite with tungsten has been investigated to determine the deformation response over a range of stress states, strain rates, and temperatures. The equation of state (EOS) of the monolithic glass has also been investigated to determine its phase stability. Anvil-on-rod impact experiments performed on the BMG (with and without steel sleeve) reveal the deformation, fracture, and elastic-plastic wave propagation response characterized by the Drucker-Prager model. The deformation response of the composite is dominated by the flow and failure characteristics of tungsten. The equation of state experiments performed over a wide range of shock pressure show a polyamorphism transition starting at 26 GPa. The bulk modulus of the high-pressure phase is ~144 times that of the ambient pressure phase. Correlation of normalized yield and fracture stress with strain rate shows that the phase transition contributes to a substantial increase in yield and fracture stress at strain rates $>10^4$ s⁻¹, which is more so than that observed for bcc-tungsten and BMG-W composite.

GRADUATE Dissertation

Dr. Morgana Martin, HIGH STRAIN RATE PROPERTIES OF BULK METALLIC GLASSES AND THEIR COMPOSITES, completed May 2008.

LIST OF PERSONNEL INVOLVED

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Grad Student: Morgana Martin (Ph.D./MSE)

UG Students: Adam Jakus, Jim Painter, Arsalan Siddiqqi, and Daniel Miller

- (a) Number of Undergraduates – 4
- (b) Number of UGs graduated – 1
- (c) Number of UGs graduated with science/math/eng/tech degrees – 1
- (d) Number of UGs graduated and will be pursuing grad degree – 1
- (e) Number of UGs graduated and will work for DoD – Nil
- (f) Number of UGs graduated with >3.5 GPa – 1
- (g) Number of UGs graduated funded on DoD CoE – Nil
- (h) Number of UGs graduated and will receive fellowship – 1

Collaborations: Dr. Laszlo Kecskes, Army Research Laboratory, MD; Dr. Toshimore Sekine, National Institute of Materials Science, Tsukuba, Japan; and Dr Lothar Meyer, Chemnitz University, Germany.

PUBLICATIONS:

- ^{1.} M. Martin and N.N. Thadhani, “Mechanical Behavior of Bulk Metallic Glasses,” A review Article invited for publication in the Progress in Materials Science, 2008.
- ^{2.} M. Martin, L. Meyer, L. Kecskes, N.N. Thadhani, “Uniaxial and biaxial compressive response of a bulk metallic glass composite over a range of strain rates and temperatures,” Journal of Materials Research, **accepted for publication, September 2008.**
- ^{3.} N.N. Thadhani, M. Martin, T. Sekine, L.W. Meyer, High-Strain-Rate Deformation And Shock-Compression Behavior Of Zirconium-Based Bulk Metallic Glass, Proc. of ESHP International Conference, Kumamoto University, Japan, Sept 10-12, 2008.
- ^{4.} M. Martin, L. Kecskes, and N.N. Thadhani, “High-Strain-Rate Dynamic Mechanical Behavior of a Bulk Metallic Glass Composite,” J. Matls. Research, Vol. 23, No. 4, April, 2008, pp. 998-1008.
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PRESENTATIONS:

- ❖ N.N. Thadhani, M. Martin, T. Sekine, L.W. Meyer, High-Strain-Rate Deformation And Shock-Compression Behavior Of Zirconium-Based Bulk Metallic Glass, ESHP International Conference, Kumamoto University, Japan, Sept 10-12, 2008 (invited).
- ❖ N.N. Thadhani and M. Martin, "High Pressure Phase Stability and High-Strain-Rate Mechanical Properties of Zr-based Bulk Metallic Glass & Composite," Institute of Non-Linear Mechanics, Dec 11, 2007 (invited)
- ❖ M. Martin, L. Kecskes, N.N. Thadhani, "Dynamic compression and tension of a Zr-based bulk metallic glass," IDEA League Summer School on Multiscale Modeling in Materials Science and Engineering, Eifel Mountains, Germany, July 23-28, 2007 (poster).
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- ❖ M. Martin, T. Sekine, T. Kobayashi, L. Kecskes, N.N. Thadhani, "High pressure equation of state of a Zr-based bulk metallic glass," American Physical Society Shock Compression of Condensed Matter, June 26, 2007 (poster).
- ❖ Mishra, M. Martin, N.N. Thadhani, B. Kad, M.A. Meyers, "High-Strain Rate Response of Ultra-Fine Grained Copper: Experiments and Analysis," American Physical Society Shock Compression of Condensed Matter, Kohala Coast, Hawaii, June 25, 2007 (poster).
- ❖ M. Martin, T. Sekine, T. Kobayashi, L. Kecskes, N.N. Thadhani, "High pressure equation of state of a Zr-based bulk metallic glass composite," Dynamic Behavior of Materials Symposium, The Minerals, Metals & Materials Society Annual Meeting, Orlando, FL, Feb 28, 2007.
- ❖ Mishra, B. Kad, M. Martin, F. Gregori, N.N. Thadhani, Edward A. Kenik, M.A. Meyers, "Microstructural evolution in copper subjected to severe plastic deformation: experiments and analysis," The Minerals, Metals & Materials Society Annual Meeting, Orlando, FL, Feb 27, 2007
- ❖ M. Martin, T. Sekine, T. Kobayashi, N.N. Thadhani, "High pressure equation of state of a zirconium-based bulk metallic glass composite," Japanese Society for the Promotion of Science Summer Program Closing Ceremony, Aug 22, 2006, invited.
- ❖ M. Martin and N.N. Thadhani, "High-Strain-Rate Mechanical Behavior and High-Pressure Stability of Bulk Metallic Glasses," Plenary talk to be presented at International Conference on Impact Engineering in Daejeon, Korea, September 17-19, 2007 (invited).