Local Shock Properties Measurement Using Time-Resolved Raman Spectroscopy

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Abstract. In this article, the dynamic response of a heterogeneous microstructure of polymer bonded composite was analyzed to a short duration shock pulse. The composite microstructure studied is a polymer-bonded sugar (PBS) with single-crystal sucrose embedded inside the polydimethylsiloxane binder. The shock pulse was created by the impact of the aluminum disk at high speeds using a laser-based projectile launch system. The mechanical response on the microscale domain was measured using ultrafast time-resolved Raman spectroscopy. The *in-situ* analysis of the change in Raman spectra from PBS during shock compression was captured in the time domain using a streak camera. The results show a steeply rising shock front after the impact where the shock pressure rise time was estimated from the time-resolved Raman spectra. The viscoplastic behavior in the local microscale domain was characterized by quantifying effective shock viscosity measured in the vicinity of the crystal-binder interface.

1 Introduction

The thermo-mechanical behavior of composite material on mesoscale and microscale is dependent on the microstructure and local morphological features. Experimentally, the overall behavior of such a complex microstructure is characterized by measuring behavior on a macroscale. However, the high strain rate material response in microscale domains such as interfaces and different components in the composite material can be significantly different from the overall behavior of microstructure. Such behavior at different scales requires significant attention as the overall behavior is governed by the local mechanical, thermal, and chemical properties of the components and can significantly change the overall response to impact loading. Measurement of experimental shock behavior on microscale posses extreme challenge due to required resolution on length and time scale.

In this study, we extended the capability of our previous work [1, 2] to measure shock behavior in microscale with nanosecond time resolution. This method provides shock measurements in a non-contact and non-invasive manner without altering the chemistry and mechanics of the microstructure. The nanosecond behavior in a local microscale domain of a polymer-bonded sugar (PBS) sample is measured using time-resolved Raman spectroscopy. The local dynamic stress rise in the vicinity of sugar and polydimethylsiloxane (PDMS)

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interface was measured to estimate local shock viscosity at high strain rates. The shock pressure is created by the impact of aluminum flyers accelerated by a laser-based projectile launch system shown in **Fig. 1**. The velocity profile during impact is measured using photon Doppler velocimetry (PDV). The temporal change in Raman spectra under shock loading provides an estimate of the shock pressure rise time and effective shock viscosity. The measurement of local shock viscosity is useful to estimate the viscous behavior of the material and correlation to the energy dissipation during the fracture and shock compression of the microscale domain.

The shock pressure can be estimated using the Hugoniot equation of state (EOS) and impedance matching between AL1100 impactor, PDMS binder, and sugar crystal. The EOS parameters used in pressure estimation for different materials are listed in **Table 1**.

Table 1. EOS paramètres for different materials

Material	ρ_0 **[g/cm^3]**	C_0 **[Km/s]**	**S**
AL1100 [3]	2.712	5.38	1.34
PDMS – Sylgard [3]	1.037	1.63	1.66
Sugar [4]	1.58	3.04	2.05

Under the assumptions of planar shock conditions, the local strain rate and shock viscosity can be estimated based on equations (1) and (2) respectively [5].

$$\dot{\varepsilon} = \frac{\sigma}{\rho C_0^2 \tau} \quad (1)$$

$$\eta = \frac{1}{4} S \sigma \tau \quad (2)$$

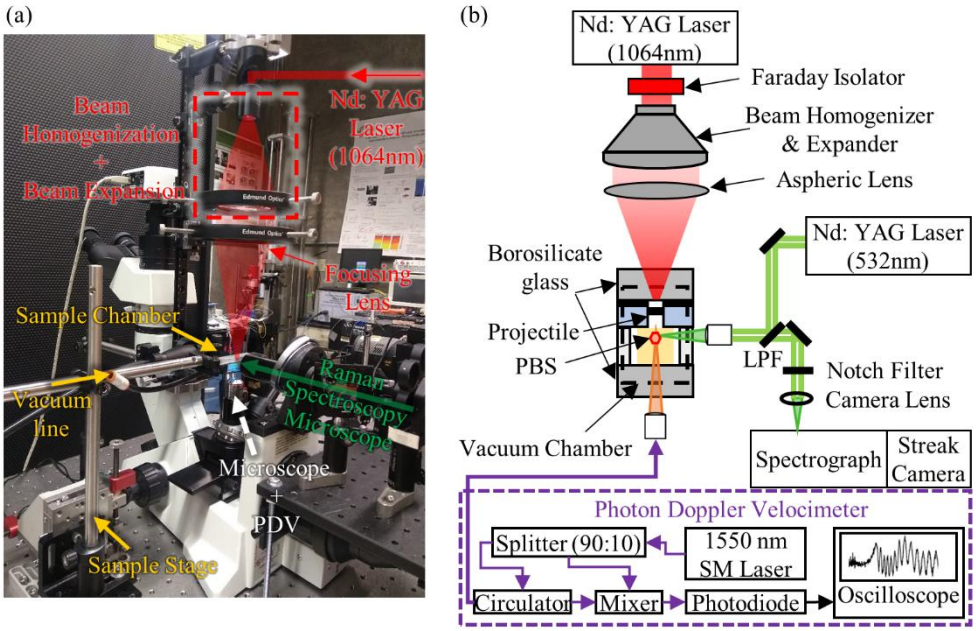

Fig. 1. (a) *In-situ* Raman spectroscopy and Impact System (b) Schematic of high-speed impact and *in-situ* Raman spectroscopy system

2 Experiment Method

Polymer-bonded sugar (PBS), also used as a mechanical simulant of cyclotetramethylene-tetranitramine (HMX) based polymer-bonded explosive (PBX), is used for this study. Due to the monoclinic crystal structure and morphological characteristics similar to that of HMX, the sugar crystal acts as a suitable mechanical surrogate for HMX crystals. In the research community, PBS has been commonly used as a mock and inert surrogate to investigate the thermo-mechanical response of PBX under inert conditions and different loading conditions. The microstructure of composite material is very complex with a high level of inhomogeneity. To reduce the complexity and obtain repeatability on sample preparation, we used a simplified microstructure with a single crystal of sucrose embedded inside the PDMS binder with a controlled orientation. The deformation of the sucrose granules is crystallographically dependent on its monoclinic crystal structure. Therefore, as shown in **Fig. 2a,** sugar crystal was oriented such that shock pressure is induced in crystal through the face (100). The sugar crystals with a dominant (100) face [6] and approximate size of 250μm were selected to be embedded inside the PDMS binder which enhances the repeatability for sample preparation.

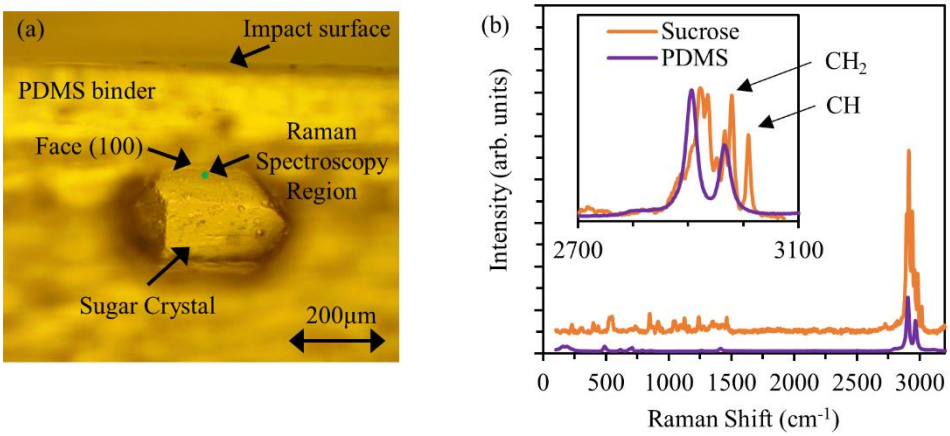

Fig. 2. (a) Simplified microstructure of polymer bonded sugar (b) Raman spectra from different phases in the microstructure

The Raman spectrum of sucrose consists of several vibrational modes as shown in **Fig. 2b**. In this work, we focused on the C-H vibrational features observed between 2900 to 3200 cm^{-1} due to relatively higher intensities compared to other groups. The CH_2 mode at 2980 cm^{-1} and CH mode at 3011 cm^{-1} [7, 8] were selected for this work, these are observed to be easily distinguishable from vibrational spectra of CH stretching in PDMS binder. The Raman spectra were collected on a spectrograph combined with a streak camera (Hamamatsu C4334-01). The vertical entrance slit on the spectrograph was 75 μm and the horizontal photocathode slit on streak scope was 75 μm. An Nd: YAG laser of 532 nm wavelength was used to excite the Raman spectra from the sample with a pulse energy of 1.5 mJ. The spectrum between 620.2 nm to 635.96 nm was collected with a resolution of 0.03 nm and 0.78 cm^{-1} resolution on Raman shift. The Raman spectra were collected from a 22 μm region [2] close to the leading interface on sugar crystal as shown in **Fig. 2a**. The time synchronization between the 532 nm Raman laser, 1064 nm flyer launch laser, and streak scope was obtained based on the time delay between flyer launch and flyer impact observed on the PDV spectrograph.

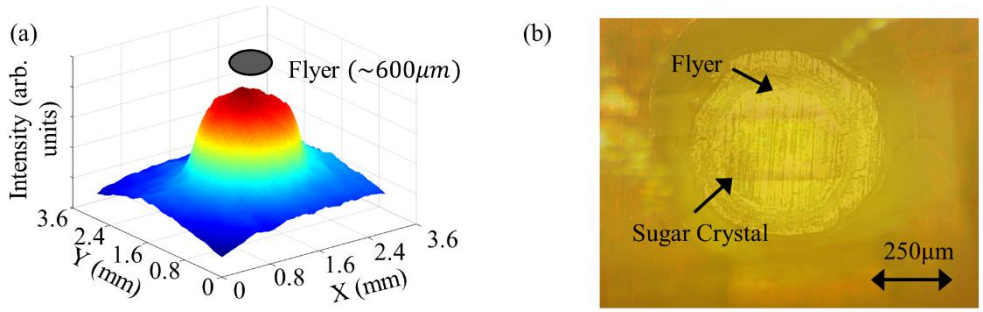

Fig. 3. (a) Profile of flyer launch laser at the launch pad (b) Assembly of flyer launch pad with PBS sample

A schematic of the experimental apparatus is similar to used before [2] with some modification and is shown in **Fig. 1b**. An Nd: YAG laser at 1064nm (Continuum laser), was used with a pulse energy of 0.75 J and a pulse duration of 7 ns. In a laser-based flyer impact system, a thin aluminum foil (AL1100, Alufoil Products Co., Inc.) of diameter ~600 µm and thickness of 50µm is launched by pulsed laser driven at the interface of foil and borosilicate glass. A spatially homogeneous beam was generated using a 25 mm diameter diffractive optical element from HOLO/OR Ltd. (RD-204-I-Y-A). After the beam homogenization optics, an aspheric lens with a 150 mm focal length (AL75150-C, Thorlabs Inc.) was used to produce 800 µm fullwidth half maximum (fwhm) focused spot. The spatial profile of the laser pulse at the launch pad is shown in **Fig. 3a**. The PDV interferogram was acquired at 20GHz and was analyzed using a short-time Fourier transform providing a resolution of 0.05ns. The launchpad was assembled with embedded sugar samples as shown in **Fig. 3b** and assembly was placed inside a vacuum chamber, reducing air drag in front of the flyer.

3 Results and Discussion

The impact experiments were performed at an impact speed of 642.6 ± 38.8 m/s measured over 400 impacts and a velocity profile measured from the aluminum flyer during the impact of PBS sample is shown in **Fig. 4a**. The particle velocity of 521m/s and a shock duration of ~ 8ns were measured from the interface of the aluminum flyer and PDMS binder, at an impact speed of 640m/s. The Raman spectra acquired on the streak camera are binned over 10 ns and time-resolved spectra from CH_2 mode and CH mode are shown in **Fig. 4b**.

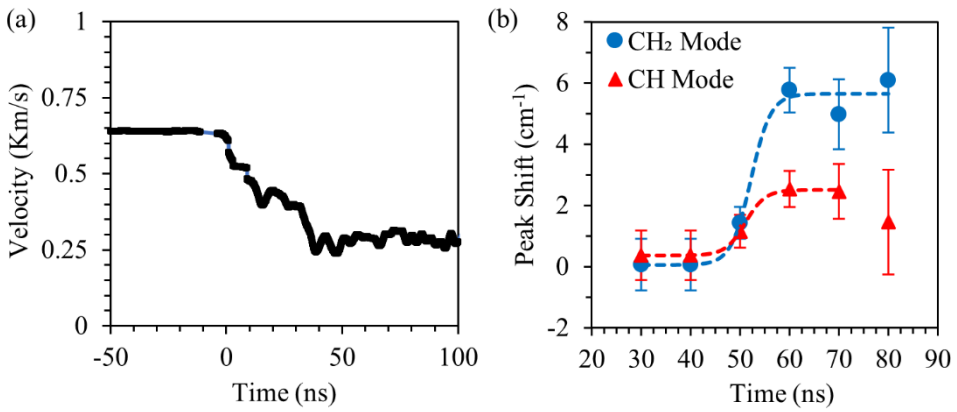

Fig. 4. (a) Velocity profile measured from the flyer through PDV (b) Time-resolved Raman spectra measured during impact

Using impedance matching, shock pressure of 1.44 ± 0.11 GPa in PDMS binder and 2.16 ± 0.17 GPa in sugar crystal is calculated at the Hugoniot state. From time-resolved Raman spectra, the rise time of 14.78ns is estimated. Using equation (2), the effective shock viscosity of 16.43 ± 1.26 Pa-s is estimated at a strain rate of 10^7/s. The measured effective shock viscosity provides an estimation of viscous stress in the vicinity of the sugar – PDMS interface where total dynamic stress is defined by equation (3),

$$\sigma(\varepsilon) = P(\varepsilon) + \eta\dot{\varepsilon} \quad (3)$$

where the pressure term (P) is calculated through the equation of state and viscous stress is obtained using effective shock viscosity. The existence of shock viscosity is attributed to the fracture and deformation of microstructure under the shock compression as shown in **Fig. 5**.

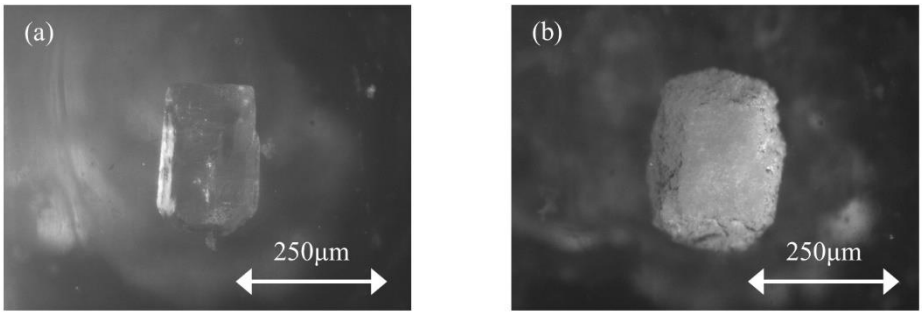

Fig. 5. (a) Sugar crystal before impact at 650m/s (b) Sugar crystal after impact at 650m/s

4 Conclusion

In this work, the shock viscosity was measured in a local microscale domain of polymer bonded sugar composite under shock compression using time-resolved Raman spectroscopy. The developed technique provides a measure of local constitutive behavior in an inhomogeneous microstructure. Future work will involve simultaneous measurement of shock pressure and temperature during shock compression to understand the local thermo-mechanical behavior.

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