The effect of epoxy/hardener composition ratio on the young’s modulus of bulk adhesive at high strain rate

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Abstract. This research investigated the effects of resin/hardener ratio to the mechanical behaviors of bulk epoxy adhesive at high strain rate loading using split Hopkinson pressure bar (SHPB) test. The variation of resin/hardener ratio are 2:1, 2:3, 1:1, 3:2, and 1:2 by weight. The results show that the bulk adhesive become more ductile as the ratio deviates from the manufacturer recommendation of ratio 1:1. The adhesive exhibits lower modulus but longer elongation as compared to the baseline. In contrast, at ratio of 3:2 where epoxy resin is slightly excessive, the modulus increases, and the elongation is shorter which indicates a brittle nature. The results facilitate the engineer to design adhesive of gradational modulus to improve joint performances.

Keywords: epoxy adhesive, composition ratio, butt adhesive joint, split hopkinson pressure bar, high strain rate

1. Introduction
The stress in the adhesive lap joint distributes nonuniformly along the adhesive layers. The stress concentrates in the edges of bondline and initiates joint failure [1–4]. Achieving high joint strength by applying high modulus adhesive worsen the stress concentration. Applying adhesive of lower modulus reduces the stress in the bondline but unable to solve the stress concentration problem. The use of bi-adhesive or gradational modulus along the adhesive layer distributes stress more evenly, reduce the stress concentration and thus, improves the joint strength [5–7].

Adding particulate filler is the most common method to modify the modulus of bulk adhesive. Addition high modulus particle such as silica increases the modulus of adhesive. Appropriate combination of silica micro and nanoparticle further improves simultaneously modulus and energy absorption of adhesive [8, 9]. However, particulate addition requires high effort, cost, and control in mixing process to produce a homogenous adhesive compound. Another simpler and cheaper method to modify the adhesive modulus is by controlling the resin/hardener ratio of two parts epoxy adhesive [10]. Modification in resin/hardener ratio controls the polymerization of the adhesive and thus, modify its mechanical properties. However, mechanical characterization of the adhesive of modified composition ratio are rare, especially at high strain loading. Such characterization is of high interest due to fact that impact loads frequently applied to the structural joints, e.g. crash of the automobile.
This present work study the effects of resin/hardener ratio on the modulus of epoxy adhesive at high strain rate using split Hopkinson pressure bar (SHPB). The experiment characterizes five different resin/hardener weight ratio of 2:1, 2:3, 1:1, 3:2, and 1:2. Each test condition runs at least five adhesive specimens. The specimen is cylindrical aluminum bar bonded by epoxy adhesive of different resin/hardener ratio. The SHPB test results are evaluated to investigate the effects of resin/hardener ratio on the stress-strain responses, Young's modulus, and elongation of the adhesive. The results of this study facilitates the design engineers to improve joint performances by appropriately designing adhesive of gradational modulus.

2. Materials and experimental setup

The tested specimen in this study is bulk epoxy adhesives of 20 millimeter in diameter and of 2 millimeter in thickness. The adhesive bonds two cylindrical bar of same diameter made of aluminum alloy AISI 6061. The adhesive consists of the epoxy resin and the hardener with recommended weight ratio of 1 epoxy : 1 hardener. There are five resin/hardener ratio in the experiments to investigate the ratio effects on the stress-strain responses of the adhesive, that are 2:1; 2:3; 1:1; 3:2; and 2:1. Each condition has at least five specimens to be tested. We control the weighting, mixing, curing process and adhesive application on the bar surfaces to avoid the effect of process variation in the measurement. Curing time is set to 16 hours in room temperature prior measurement.

![Figure 1](image.png)

**Figure 1.** Schematic arrangement (a) and the actual setup (b) of SHPB and the data acquisition used.
This study uses split Hopkinson pressure bar (SHPB) to characterize the stress-strain responses of bulk epoxy adhesive at high strain rate loading. Figure 1 shows the schematic of SHPB, its main dimensions, and the data acquisition setup. The input and transmission bar are of 20 millimeter in diameter. The striker bar is a cylindrical cone of 20 millimeter and 16 millimeter in back and front diameter, respectively. All bars are aluminum alloy AISI 6061 of 69 GPa in Young’s modulus. The striker bar impacts the input bar with a controlled speed and generates incident strain ($\varepsilon_I$) which is recorded by strain gauge 1. This incident strain propagates through the specimen to the transmission bar and recorded by strain gauge 2 as the transmitted strain ($\varepsilon_T$). The impedance mismatch between specimen and input bar reflects some part of the propagating strain. Then, strain gauge 1 records this reflected strain ($\varepsilon_R$). The dynamic strain meter records all strain signals and sends them to PC for calculation and analysis.

![Figure 2. Typical measured strain signals from SHPB test.](image)

Figure 2 shows the measured strain signals which consists of the incident ($\varepsilon_I$), reflected ($\varepsilon_R$), and transmitted ($\varepsilon_T$) pulses. The applied data processing trims and shifts those pulses to have a common starting point and loading period prior the calculation. First calculation is the compressive forces at bar-specimen interfaces, $P_1$ and $P_2$, using the Equation 1. The calculation requires the knowledge of pressure bars cross section area ($A_b$) and Young’s modulus ($E$). The equal compressive forces in the specimen during test period validates the SHPB test results as shown in Figure 3. Then, Equations 2, 3, and 4 calculates the strain rate ($\dot{\varepsilon}_n$), strain ($\varepsilon_n$), and stress ($\sigma_n$) in the specimen. These calculations require the knowledge of wave speed in the pressure bar ($C_0$), cross sectional area of bulk adhesive ($A_s$), and adhesive thickness ($L_s$). The stress and strain history in the specimen obtained from Equations 3 and 5 are plot together to obtain stress-strain curve as shown in Figure 4. We compare the stress-strain responses of five different ratio for analysis. The dashed line indicates the estimated slope that represents Young’s modulus of the adhesive. We average the Young’s modulus of five specimens for each condition and apply T-test to evaluate the significance of the resin/hardener ratio on the measured Young’s modulus.

$$
P_1 = A_b E \{\varepsilon_I(t) + \varepsilon_R(t)\} \tag{1}
$$

$$
P_2 = A_b E \varepsilon_T(t) \tag{2}
$$

$$
\sigma_n = E A_b / A_s \varepsilon_T(t) \tag{3}
$$
\[ \dot{\varepsilon}_n = \frac{2C_0}{L_s} \varepsilon_R(t) \]  \hspace{1cm} (4)

\[ \varepsilon_n = \frac{2C_0}{L_s} \int \varepsilon_R(t) \, dt \]  \hspace{1cm} (5)

\[ \begin{array}{c}
\text{Compressive force, } P \text{ (kN)} \\
\text{Time, } t \text{ (\mu s)}
\end{array} \]

Figure 3. The overlapped compressive forces in the specimen validates the measurement.

\[ \begin{array}{c}
\text{Stress, } \sigma \text{ (MPa)} \\
\text{Strain, } \varepsilon \text{ (m/m)}
\end{array} \]

Figure 4. A typical stress-strain curve of bulk adhesive of 2 epoxy : 3 hardener.

3. Results and discussion
This work has tested five bulk epoxy adhesives of different resin/hardener composition ratio using SHPB. There are five specimens at least for each test condition. Impact load applied in the SHPB deforms all specimens at a controlled high strain rate of 367±28 s\(^{-1}\) as shown in Figure 5. The strain rate history demonstrates consecutively compression and expansion of the specimen during test.
period and correlates with the loading and unloading period. The expansion period is shorter than that of compression due to separation of specimen and pressure bar. Such separation results in incomplete record of stress-strain history during unloading period. Therefore, the stress in the specimen suddenly drops to zero as shown in Figure 6.

Figure 5. Consecutive compression and expansion of the bulk adhesive induced by impact load.

Figure 6 shows the significant effect of resin/hardener ratio on the stress and strain response of the bulk adhesive. The specimens deform at different strain although the stresses induced by the impact load are comparable. The stress grows proportionally with strain then changes nonlinearly after reaches 15 MPa. The decrease of compressive stress after passing its peak value indicates the expansion of specimen to restore its original shape. However, the expansion rate is lower than the velocity of pressure bars and resulting in loose contact at the interfaces of bars and the sandwiched specimen. Therefore, the stress drops to zero immediately while the strain...
remain. The specimen falls from its sandwiched position and shows none of damage nor fracture. The used measurement technique limits the observation of yield and plastic deformation occurrence during test. However, the reference mentions that the epoxy adhesive of low-grade yields at elongation of 5.56% [www.matweb.com]. By the end of unloading phase where the stress is released completely, the remaining strain varies with composition ratio. This fact indicates different plastic deformation occurs in the specimen, except the ratio of 3 epoxy : 1 hardener.

![Figure 7](image)

**Figure 7.** The Young’s modulus varies non-linearly with the weight percentage of epoxy

The initial slope of stress-strain curve, which represents the Young’s modulus, varies significantly with the epoxy/hardener ratio. Figure 7 demonstrates the nonlinear effect of the ratio to the averaged Young’s modulus. The line indicates the standard deviation of the measurements. These deviations come from the estimation of modulus from initial slope and the variation of strain rate loading. T-test shows that the variation of the Young’s modulus is indeed caused by the composition ratio with the confident level of 95%. Here the epoxy weight fraction represents the ratio of resin/hardener to observe the sensitivity of the Young’s modulus to the modification of composition ratio. Figure 7 shows that the modulus increases linearly with the increase of epoxy resin weight percentage up to 60% (3 epoxy:2 hardener) where the adhesive becomes epoxy rich. Then, the modulus abruptly drops as the adhesive becomes richer of epoxy. This indicates that the epoxy adhesive is sensitive to the epoxy/hardener ratio and therefore must be controlled with precision in mixing process. This fact also suggests an alternative method to modify the modulus of adhesive to match the stiffness requirement by adjust the composition ratio.

The resin/hardener ratio affects the elongation and modulus of bulk adhesive simultaneously as previously mentioned. Figure 8 shows a comprehensive picture of those simultaneous effects. The 50 wt% epoxy (1 epoxy : 1 hardener) is the performance baseline as it is recommended ratio from the manufacturer. It is clearly observed that the Young’s modulus decreases but the elongation increases as the ratio deviated from the baseline ratio, except for 60 wt%. It indicates that the adhesive turns more ductile compared to the baseline. However, the excessive epoxy of 60 wt% embrittlement the bulk adhesive as indicated by higher modulus and shorter elongation. Almeida and Monteiro (1996) studied similar cases using chemical reaction measurement [10]. They demonstrated that the unbalanced weight ratio of epoxy resin and hardener reduces polymerization or crosslink density and results in the softer adhesive. They conducted compressive
tests at quasi-static loading. This present study fills the gap in the work of Almeida and Monteiro by demonstrating stress-strain responses of adhesive at high strain rate loading which are required to evaluate the adhesive joint performance under impact loading. This present work also provides knowledge of adhesive allowing the engineer to modify the behavior of the epoxy adhesive.

4. Conclusion
The present work has investigated the effect of resin/hardener ratio on the stress-strain responses of bulk epoxy adhesive at high strain rate loading using split Hopkinson pressure bar tests. Any deviation in the mixing ratio of resin and hardener leads to softer adhesive performance as compared to ratio recommended by the manufacturer. Only slightly excessive epoxy results in the stiffer yet brittle adhesive. This study provides design options of epoxy adhesive to have a desired stiffness and elongation in applications. The effect of mixing ratio on the adhesive strength should be investigated further.

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