#### Research Article

Adnan Abbass Al-Azzawi\*

# Viscoelastic behavior of glass fiber reinforced rubber-modified epoxy

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**Abstract:** Epoxies as a thermoset polymer have got a great attention in different applications. To elaborate their employing and surmount their brittleness, many polymers were blended with them. The results confirm that the good mechanical properties are obtained when 6wt% of Polysulfide Rubber (PSR) is blended with epoxy and reinforced with fiber glass. The effect of rubber and glass fiber on the viscoelastic properties of epoxy were investigated using creep-recovery data under different stress levels (5, 10, 15 and 20 MPa) and temperatures (30, 50, 70 and 90°C). Polysulfide addition caused larger creep and creep recovery. In addition, the creep resistance of glass fiber reinforced blend was significantly enhanced.

**Keywords:** epoxy resin, polysulfide rubber, viscoelastic, mechanical properties

#### 1 Introduction

Epoxy resins are spaciously used as structural adhesives, matrices in fiber-reinforced composites and coatings. The crosslinking merit of treated epoxies awards a highly unfavorable property: they are proportionally brittle, having indigent resistance to crack initiation and growth [1–3]. This deficiency of toughness strictly effects the implementation of these thermoset nearly all applications [4–7]. To handle this imperfection, resin formulators have incubated technology that allows some thermosets to be toughened by the addition of a second elastomeric phase, through pressing an agreeable victimization of desirable properties [8, 9]. Rising the concentration of elastomer phase, an augmentation in impact strength of the epoxy has been accomplished [10, 11]. There are several reactive liquid elastomers which are employed to modulate or toughen epoxy resins.

Liquid polysulfide elastomer is one of the more significant reactive modifiers for epoxy resin. Polysulfide amendment epoxy adhesive systems are exceedingly used in the construction, electrical and transportation industries owing to the following utilities. One of the principal utilities is enhancement adhesion, extremely to substrates which are oily, wet, and rusted. The premium adhesion of polysulfide epoxy conducts to the utilization of these materials in surface-tolerant adhesives and coatings. Moreover, to contributing enhanced initial adhesion, polysulfide rubber provides long-term adhesion and toughness to epoxy resins. Since polysulfide is a very flexible elastomer, addition of liquid polysulfide rubber to epoxy resin awards good flexibility and influence the strength of epoxy resin [12–14]. There are some restrictions to the employ of polysulfide modified epoxy. Polysulfide rubber deactivates the most eligible properties of epoxy, they decline the glass transition temperature (Tg), tensile strength and tensile modulus of epoxy resin because of the alleviation effect of the polysulfide rubber in the epoxy matrix. This restricts the application regions in which they can be utilized. Ameliorating the fracture toughness of thermosetting resins has been the matter of essential research [15]. These approaches have consisted the addition of micron-sized soft (elastomeric or thermoplastic) or rigid (glass or ceramic) particles into the polymer matrix. These filler particles are then foreseeable to provide extrinsic toughening mechanisms. However, highly cross-linked thermosetting polymers are unable of extensive shear yielding. Thus, the effectiveness of rubber additions reduces speedily with increasing cross-link density [16–18]. Brostow et al. [19] determined the stress relaxation of polypropylene and their blends consisting of 10 and 20% of the longitudinal Polymer Liquid Crystal at constant strain of 0.5%. The consequences of nine temperatures in the range between 20 and 100°C were employed. The anticipated values corresponding with the experimental ones for all materials investigated. Paradas *et al.* [20] studied creep and recovery tests for continuous random fiber glass mat of polypropylene composite at stress ranging between 20 and 60 MPa and temperature from room temperature to 90°C. It was demonstrated that materials behaved non-linearly for all stresses and temperature. Farsani

<sup>\*</sup>Corresponding Author: Adnan Abbass Al-Azzawi: Al-Farabi College University, Baghdad, Iraq; Email: adnan.alazawi@alfaraiuc.edu.iq

et al. [21] pointed out the creep behavior of Basalt Fiber Reinforced Epoxy (BFRE) and Glass Fiber Reinforced Epoxy (GFRE) composites at high temperatures. No creep rupture failures were identified in short term (less than 10000 seconds) and high temperatures (150 and 200°C) at loads up to 15% of the tensile strength. It was also detected that the creep resistance of BFRE was more than that of GFRE and the materials are mainly attitude as non-linear for all stresses and temperatures. Badagliacco and Valenza [22] described the viscoelastic attitude of an epoxy resin amended by waste glass and rubber particles using linear fractional spring-pot model. The results specified that using waste materials as additives for polymer compounds is a practical and prospective probability when it derives to modifying their viscoelastic properties. The target of this study is to inspect the influence of polysulfide rubber and fibers concentration on the epoxy system. Mechanical properties and viscoelastic behavior will be discussed and tried to correlate between them.

### 2 Experimental work

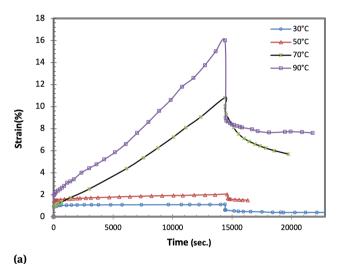
Polymer blends was attained by mixing epoxy resin with liquid polysulfide utilizing mechanical mixer for 15 minutes. The blend then heated to 70°C for 2 hours employing magnetic stirrer with speed of 500 r.p.m. The blend was thereafter degassed to attain mixture free of bubbles. A stoichiometric amount of anhydride therapy agent was then added and stirred rigorously for 15 minutes at 40°C. Test samples were acquired after curing for 7 days. The same procedure was pursued in fabrication of E-glass fiber reinforced composites. Short-term creep tests consisting of 240-minute creep followed by 120-minute recovery were attitude. Tests were implemented at four stress levels between 5 and 20 MPa with augmentation of 5 MPa and four temperature levels between 30°C and 90°C. A sample with gage length of 25 mm and width of 5 mm was employed (ASTM 2990).

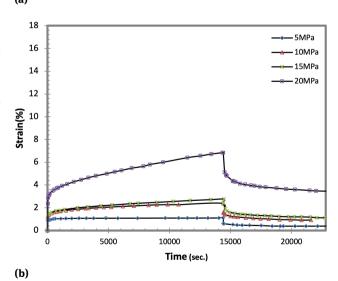
#### 3 Results and discussion

Blend of Epoxy/6wt% polysulfide rubber (PSR) which gives slightly reduction in mechanical properties was selected. Different volume fractions of E-glass fiber were added to epoxy and blend composites. The results show an optimum condition of 30% glass-reinforced blend which gives good performance and properties (Table 1).

**Table 1:** Compressive strength and impact resistance of epoxy, blend and their composites

Materials	Compression	Impact
	strength	resistance
	(MPa)	$(Kj/m^2)$
Ероху	73	0.9
Blend (Epoxy-6%PSR)	63	1.4
Epoxy + 10% glass fiber	66	4.0
Epoxy + 20% glass fiber	90	5.5
Epoxy + 30% glass fiber	108	6.5
Blend + 10% glass fiber	87	4.0
Blend + 20% glass fiber	116	4.5
Blend + 30% glass fiber	129	9

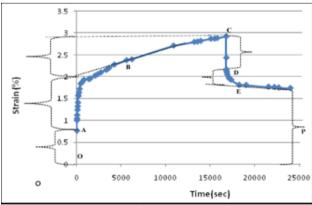




**Figure 1:** Creep and creep recovery behavior of epoxy (a) 5 MPa at different temperatures (b) Different applied stresses at 30°C

#### 3.1 Viscoelastic behavior of epoxy

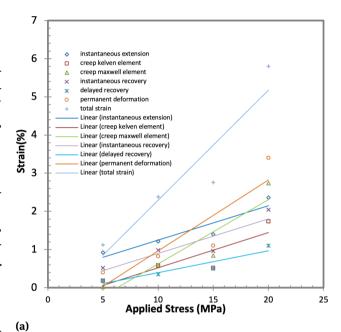
The creep stages are obviously shown in Figure 1 as instantaneous deformation, primary creep, and unfinished secondary creep processes. Tertiary stage is, however not recognized because creep rupture or failure is not regard under present condition, while a recovery stage was attitude to assess the retained strain (Figure 2). Rising the used stress at room temperature reasons condensation in instantaneous extension (Maxwell element), creep Kelvin element, creep Maxwell element, as well as, elevated in total strain (Figure 3). The resulting deformation orients to increase with time as crisscross chain segments sustain conformational alteration in reaction to external stress. Polvmers continue as a solid material even when these parts of their chains are rearranging with a view to escort the stress, and as this takes place, it generates a back stress in the material. When back stress has the same value as applied stress, the material no longer creeps. When original stress is removed, cumulated back stresses will lead the polymer regression to its original form. Material creeps, who provides prefix- viscous, and the material fully recovers, which awards suffix-elasticity [23, 24]. Instantaneous recovery (Maxwell element) grows with increasing applied stress. Total strains at 14400 second and permanent strains are more declared and raising with increasing applied stress, in contrast with instantaneous extension and instantaneous recovery. The results recorded that heightening in temperature has greater effect corresponding to the augmentation in stress. Total strain at 14400 second step-up to 16% at 90°C compared to 1.1% at 30°C, while instantaneous extension conserved on a limited increases with rise the temper-



OA: Instantaneous extension (Maxwell element), AB: Creep Kelvin element, BC: Creep Maxwell element, CD: Instantaneous recovery (Maxwell element), DE: Delayed recovery (Kelvin element), P: Permanent deformation

Figure 2: Creep and creep recovery behavior of epoxy

atures. When increasing temperature from room temperature to 90°C, more extend effect is permanent deformation, which increased from 0.4% to 7.5% by more than seventeen times. Secondary bonds (van der Waals bonds) of a polymer permanently shatter and reform due to thermal motion. Using of a stress improves some modulates over others, so molecules of polymer will progressively "flow" into preferred conformations over time [25, 26].



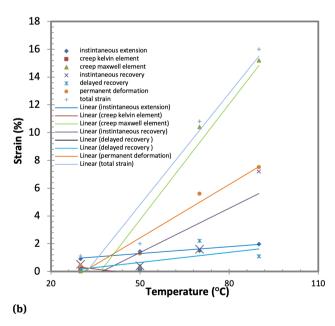


Figure 3: Variation of creep parameter of epoxy with (a) Applied stress, (b) Temperature

## 3.2 Effect of rubber on creep behavior of epoxy

Creep behavior of blend and epoxy is the same to some extent. Increasing of applied stress has more effect on instantaneous extension, permanent strain, and creep Maxwell of the blend. The instantaneous creep of blend is more, while creep Kelvin is less (Figure 4). It can be seen from Figure 5, creep Kelvin element and permanent deformation increase with increasing applied stress, whereas de-

lay recovery and creep Maxwell element have the same response as with epoxy. This phenomenon of blend at room temperature differs compared with epoxy. Instantaneous and delay recovery of the blend were higher than that of epoxy because of PSR presence. Strain percentage of 4.7 and 6.5 were recorded for instantaneous and delay recovery of blend, which is 325% and 218% higher than that of epoxy. Increasing temperature has more influence on creep behavior. Permanent deformation records an increment of 13% at 70°C compared with that of room temperature.

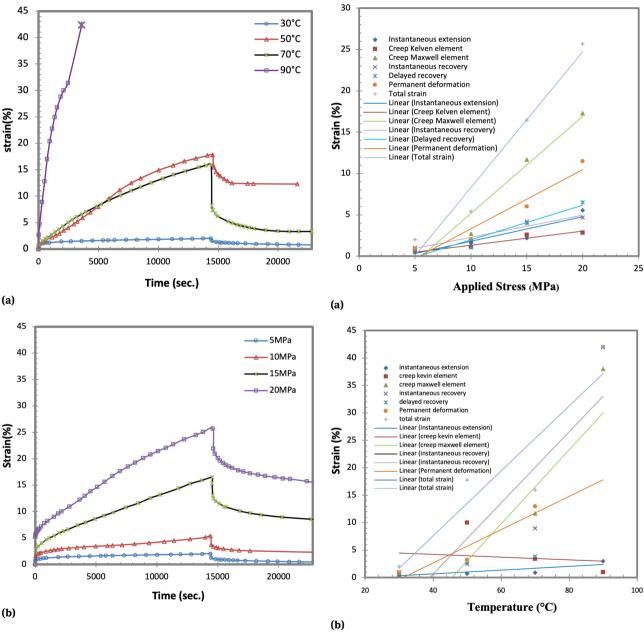


Figure 4: Creep and creep recovery behavior of Epoxy/6% PSR blend (a) 5 MPa at different temperatures (b) Different applied stresses at  $30^{\circ}$ C

Figure 5: Creep behavior of Epoxy/6% PSR blend (a) Different applied stresses, (b) Different temperatures

#### 3.3 Effect of fiber glass on creep behavior

Because of effect of polysulfide elastomer in epoxy matrix, this restricts the using reigns in which they can be employed. proceed toward to avoid this problem is integrating the benefits of adding polysulfide elastomer and glass fiber to epoxy to evolve improved matrix material with the goal to be have good ductility without decline other coveted mechanical properties of epoxy resin. Figure 6 shows glass

fiber reinforced blend not only had minimal instantaneous deformation, owing to high stiffness of glass fiber, but also elucidates a smaller creep rate than blend and epoxy creep. Strain increased with temperature, as the same time similar effect was displayed by increasing stress. With rising of temperature from  $30^{\circ}$ C to  $90^{\circ}$ C, the creep strain can increase by 135% approximately, at 14400 second (Figure 7).

This implies that creep behavior is greatly improved by presence of glass fiber in which creep strains at 90°C

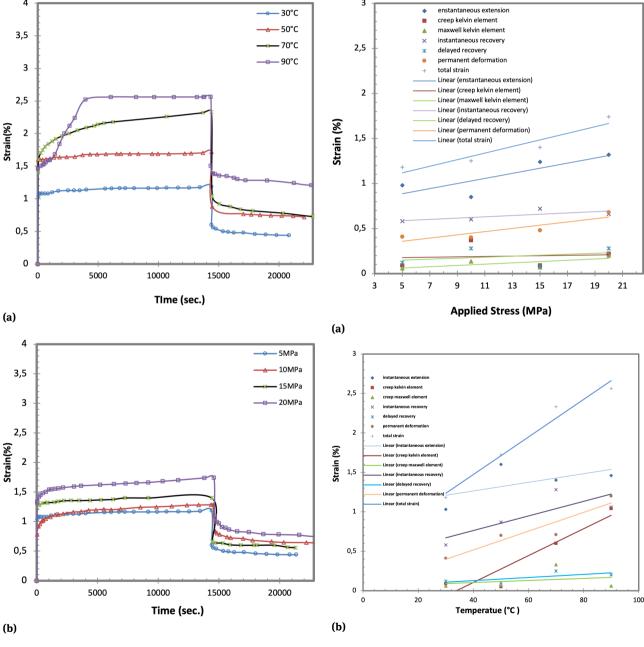


Figure 6: Creep and creep recovery behavior of Epoxy/6% PSR blend reinforced with 30% glass fiber (a) 5 MPa at different temperatures (b) Different applied stresses at  $30^{\circ}$ C

Figure 7: Variation of creep parameter of Epoxy/6% PSR blender reinforced with 30% E-glass fiber with (a) Applied stress, (b) Temperature

lowered by five, twelve times compared to epoxy and blend respectively. Results indicate that epoxy has low sensitivity to stress rising from 5 MPa to 20 MPa than to temperature step-up. The permanent deformation at 20 MPa is 0.68 strain %, which its time is lower than that of epoxy and blend.

#### 4 Conclusions

- 1. Additive of 6wt% PSR to epoxy increases creep rate, permanent deformation and recovery.
- 2. Epoxy/6% PSR blend is more affected under creep with increasing applied stress and temperature.
- 3. Reinforcing Epoxy/6wt.% PSR blend with 30vol.% E-glass fiber improves creep resistance.
- Temperature has more impact on creep parameter (Maxwell and Kelvin elements) corresponding to applied stress.

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#### References

- Sultan JN, McGarry FJ. Effect of rubber particle size on deformation mechanisms in glassy epoxy. Polym Eng Sci. 1973;13(1):29– 34
- [2] Bascom WD, Timmons CO, Jones RL, Peyser P. The fracture of epoxy-and elastomer modified epoxy polymers in bulk and as adhesives. J Appl Polym Sci. 1975;19(9):2545-62.
- [3] Ramaswamy R, Achary PS. Adhesion development in a polysulphide sealant. J Adhes. 1981;11(4):305–13.
- [4] Wang TT, Zupko HM. Phase separation behavior of rubbermodified epoxies. J Appl Polym Sci. 1981;26(7):2391–401.
- [5] Kinloch AJ, Shaw SJ, Tod AD, Hunston DL. Deformation and fracture behavior of a rubber toughened epoxy. Microstructure and fracture studies. Polymer. 1983;24(10):1341–54.
- [6] Crosbie GA, Phillips MG. Toughening of polyester resins by rubber modification. J Mater Sci. 1985;20(1):182–92.
- [7] Yee AF, Pearson RA. Toughening mechanisms in elastomermodified epoxies. J Mater Sci. 1986;21(7):2462-74.
- [8] Pearson RA, Yee AF. Influence of particle size and particle size distribution of toughening mechanisms in rubber-modified epoxies. J Mater Sci. 1991;26(14):3828-44.

- [9] Wilford A, Lee TC, Kemp TJ. Phase separation of polysulfide polymers in epoxy adhesives. Int J Adhes Adhes. 1992;12(3):171–7.
- [10] Ullett JS, Chartoff RP. Toughening of unsaturated polyester and vinyl ester resins with liquid rubbers. Polym Eng Sci. 1995;35(13):1086-97.
- [11] Singh P, Zhang M, Chan D. Toughening of a brittle thermosetting polymer: effects of reinforcement particle size and volume fraction. J Mater Sci. 2002;37(4):781–8.
- [12] Matykiewicz D. Hybrid Epoxy Composites with Both Powder and Fiber Filler: A Review of Mechanical and Thermomechanical Properties. Materials (Basel). 2020 Apr;13(8):2–22.
- [13] Ergün YA. Mechanical Properties of Epoxy Composite Materials Produced with Different Ceramic Powders. J Mater Sci Chem Eng. 2019;7(12):1–8.
- [14] Dipen Kumar Rajak. Durgesh D. Pagar, Pradeep L. Menezes, Emanoil Linul. Review Fiber-Reinforced Polymer Composites: Manufacturing, Properties, and Applications. Polymers (Basel). 2019;11:2–37.
- [15] Sim J, Kang Y, Kim BJ, Park YH, Lee YC. Preparation of Fly Ash/Epoxy Composites and Its Effects on Mechanical Properties. Polymers (Basel). 2020 Jan;12(1):2–12.
- [16] Krieg AS, King JA, Jaszczak DC, Miskoglu I, Mills OP, Odegard GM. Tensile and conductivity properties of epoxy composites containing carbon black and graphene Nano platelets. J Compos Mater. 2018;52(28):3909–18.
- [17] Marcovich NE, Villar MA. Thermal and mechanical characterization of linear low density polyethylene/wood flour composites. J Appl Polym Sci. 2003;90(10):2775–84.
- [18] Abood AN, Farhan MM, Fadhil AT. Mechanical properties of carbon/polypropylene fiber reinforced rubber-modified epoxy system. Chem Mater Res. 2015;7(5):97–108.
- [19] Brostow W, Akinay AE, Castano VM, Masimov R, Olszynski P. Time-Temperature Correspondence Prediction of Stress Relaxation of Polymeric Materials From a Minimum of Data. Polymer. 2002;43(13):3593-600.
- [20] Dasappa P, Lee-Sullivan P, Xiao X. Temperature effects on creep behavior of continuous fiber GMT composites. J Compos A Appl Sci Manuf. 2009;40(8):1071–1081.
- [21] Farasni RE, Reza Khalili SM, Daghighi V, Fazaeli R. Creep behavior of basalt and glass fiber reinforced epoxy composites. J Mech Res Appl. 2011;3(1):29–36.
- [22] Badagliacco D, Valenza A. Viscoelastic Behavior of an Epoxy Resin Modified with Recycled Waste Particles Analyzed through a Fractional Model. Processes (Basel). 2021;9(10):2–12.
- [23] Acha BA, Reboredo MM, Marcovich NE. Creep and dynamic mechanical behavior of, pp-jute composites: effect of the interfacial adhesion. Compos Part A Appl Sci Manuf. 2007;38(6):1507–16.
- [24] Ornaghi HL Jr, Bolner AS, Fiorio R, Zattera AJ, Amico SC. Mechanical and dynamic mechanical analysis of hybrid composite molded by resin transfer molding. J Appl Polym Sci. 2010;118:887–96.
- [25] Khaldi M, Vivet A, Bourmaund A, Serreir Z, Kada B. Damage analysis of composite reinforced with alfa fibers: viscoelastic behavior and debonding at the fiber/matrix interface. J Appl Polym Sci. 2016:133(31):43760-9.
- [26] Masoumi S, Slehi M, Akhaghi M. Nonlinear viscoelastic analysis of laminated composite plates-a multi scale approach. Int J Recent Adv Mech Eng. 2013;2(2):11–19.