

Enchasing Toughness And UV Aging Of Benzoxazine/Epoxy Co-Polymer Using Polyvinyl Butyral For Coating Application

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Abstract—Two types of resin composed of epoxy with benzoxazine based on styreal amine or aniline were prepared in weight ratio 50/50. These resins namely EBZ1 & EBZ2 respectively. Polyvinyl butyral was added as plasticizer to the previous resins with different wt. ratio (10, 20, and 30%). Curing behavior of prepared materials were investigated by FTIR spectroscopy. The blends were used as coating on steel to protect from corrosion. The prepared coating samples with thickness approximately 20-30 μm , using film applicator according to ASTM D4138-94, were tested as metallic coatings by carrying out visual check to get the optimum ratio of curing. Mechanical properties of the resin was measured such as (adhesion, hardness, impact and binding and tensile). Thermal stability was determined to confirm the EBZ1/20PVB have high thermal stability than EBZ1/30PVB. Chemical resistance was resulted and enhanced with compare in the neat resin. The addition of PVB was increase the toughness of materials and introduced copolymers more flexibility, processability, and easy to handle in most industrial application.

Keywords—Co-polymer, Coating, Toughness, UV ageing

I. INTRODUCTION

The improvement of a simple and proficient way to deal with plan high-toughness epoxy resin is fundamental however has stayed a gigantic test. The improvement of a high-performance environment-friendly strong epoxy sap altered with epoxidized hydroxyl-terminated polybutadiene (EHTPB) through one-step dissolve mixing was considered. The hardening impact of EHTPB on the strong epoxy sap was superior to that of a portion of the recently detailed fluid epoxy gums [1]. Be that as it may, the Polyetheramine (PEA)-modified epoxies with different sorts of PEAs were readied and separate consequences for qualities of epoxy networks were considered. The pre-owned PEAs were polyethylene glycol diamine (PEG-amine) and polypropylene glycol diamine (PPG-amine) with two diverse sub-atomic loads (i.e., 200 and 400 g mol^{-1}) [2].

Likewise, the functionalized lignin can be utilized in epoxy as a cohardener or comonomer; anyway either inadmissible mechanical properties or low lignin content remaining parts a test in using the practical biomass to supplant the petrochemical items. The great scattering of lignin-COOH in epoxy, the unbending sweet-smelling structure of lignin, and the decreased crosslink thickness in the composite [3]. Impact of both Graphene platelets (electrically conductive 2D filler) and elastic nanoparticles (0D delicate filler) can cooperate to grow electrically conductive and hardened epoxy composite adhesives [4]. Epoxy-based nanocomposites fortified with nonfunctionalized permeable graphene (NPG), carboxylated permeable graphene (CNPNG), and amine-functionalized permeable graphene (ANPG) were explored [5]. Polyethylene glycol (PEG) was effectively joined onto the outside of graphene oxide (GO) by the "uniting to" strategy [6].

By and large, the alluring qualities of every segment are consistently evident when joined as composites, and adjusted polybenzoxazine coatings by presenting other useful materials can likewise prompt a huge improvement in processability, warm steadiness, mechanical properties, thus forth.[7,8] For consumption assurance, further upgrade and some usefulness ideal for anticorrosion can likewise be gotten when polybenzoxazines consolidate with different constituents. [9,10] More critically, this is an entrancing way

which can elevate polybenzoxazine to industry soon. Among different elite materials to composite, epoxy is known to be the most traditional and great covering material in numerous viewpoints because of their incredible synthetic obstruction, great attachment properties, etc.[11,12] specifically, a steady substance bond can shape among benzoxazine and epoxy atoms in light of the fact that the epoxide gathering can respond with the phenolic hydroxyl gatherings of polybenzoxazine.[13,14] It is accepted that the fuse of an epoxy gathering could expand the crosslinking thickness of the composite with benzoxazine, which would assume a significant part in the coatings' boundary capacity against consumption medium during erosion insurance administration life.[15] Hence, understanding the impact of epoxy on the consumption opposition of the polybenzoxazine/epoxy covering will be helpful for the future improvement of business polybenzoxazine-based coatings in tending to the worldwide metallic erosion challenge.

In the ongoing years, thermoplastic interlayers are utilized to improve the crack sturdiness of overlaid composites. PolyvinylButyral (PVB) was used as plasticizer with different polymers such as epoxy and phenolic resin [16,17] an adaptable thermoplastic interlayer in types of electrospun nanoweb, arrangement, film, and powder to assess the impact of an interlayer shape on the break durability of glass-phenolic overlays [18]. Polyaniline-carboxylic multiwalled carbon nanotubes (PANI-CMWCNTs) nanocomposites were set up by a simple in-situ polymerization [19]. An epic climate amicable composite covering (P/P'- Ch-P/P') was set up by layer-by-layer gathering of polyvinyl butyral (PVB), polymethylhydrosiloxane (PMHS), and chitosan (Ch) on AA 2024 substrate to improve the consumption opposition of AA 2024 example [20]. Poly N-(vinyl) pyrrole (PNVPY) nanotubes (NTs) and nanoparticles (NPs) were readied through a UV-catalyzed green synthetic oxidative technique [21]. A polyvinyl butyral (PVB)/graphene oxide (GO) nanocomposite covering was readied through turn covering strategy to improve the anticorrosion capacity of aluminum composite [22]. An epic unattached polyvinyl butyral-polyacrylonitrile/ZnAl-layered twofold hydroxide nanocomposite film (PVB-PAN/ZnAl-LDH NCM) was set up through a stage reversal technique and utilized for salts and substantial metals expulsion from their watery solutions [23].

Herein, our work examines the influence of polyvinyl butyral incorporation on the anticorrosion behaviour of polybenzoxazine/epoxy coatings, which were studied through mechanical properties such as (elongation, hardness, adhesion and impact). The reaction between polybenzoxazine/epoxide and polyvinyl butyral was examined by Fourier transform infrared (FTIR) spectroscopy. A thermal study was done. Polybenzoxazine good absorber to UV so the addition of it to epoxy and butral was increased the UV stability and deceased low of degradation by UV. QUV weathering accelerator was used for this study this work provide guidance for the design of high-performance polybenzoxazine/epoxy with polyvinyl butyral coatings used for corrosion protection of steel.

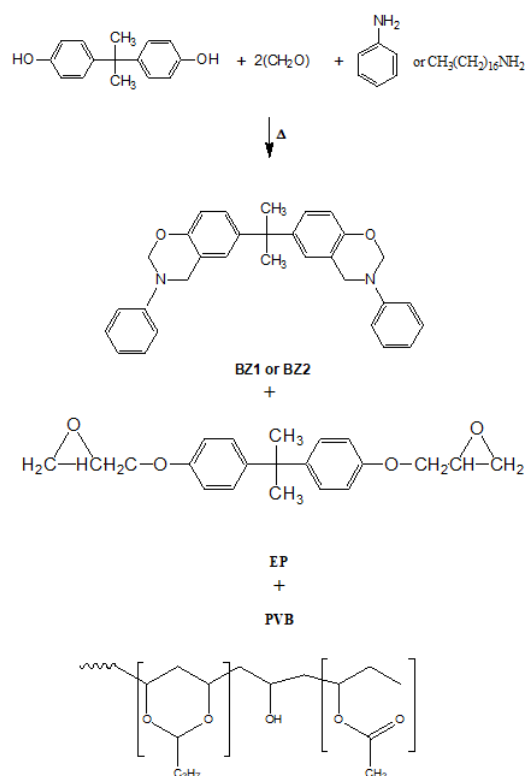
II. EXPERIMENTAL AND PREPARATION

A. Materials

PVA with Mw 85000 - 124000 g/mole 99+% hydrolysis and level of polymerization 1600 and softening point 230 °C and Butane were bought from Aldrich Company, Bisphenol A, Stearylamine, and Aniline were acquired from Kishida Co. Japan. Paraformaldehyde was gotten from Merck Co. Epoxy tar prepolymer D.E.R.331 J (epoxy identical weight rises to 186-190) in view of diglycidyl ether of bisphenol A (DGEBA) was bought from Dow Chemical Japan LTD. All synthetics were utilized minus any additional sanitization.

B. Preparation of benzoxazine/epoxy copolymers

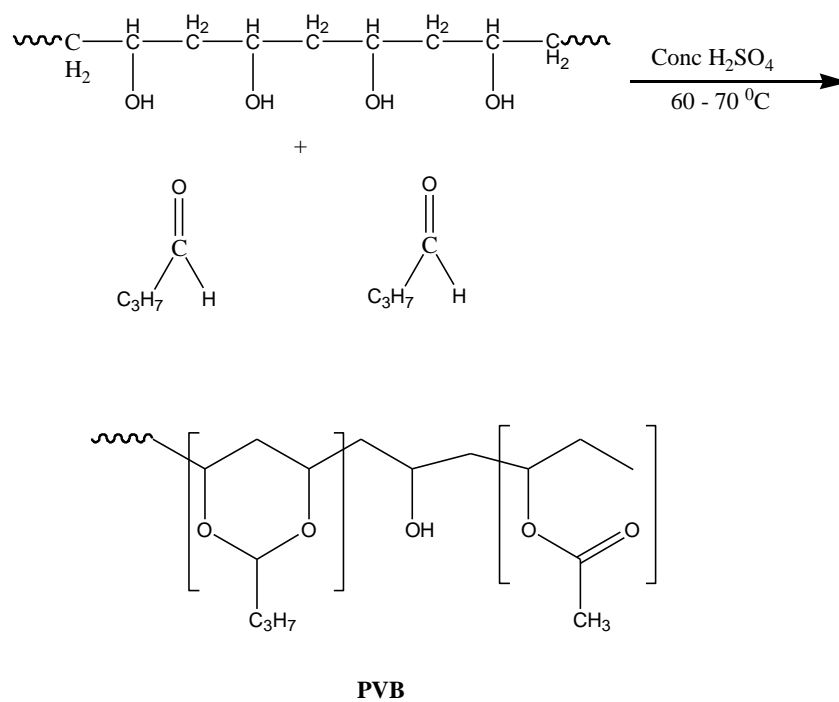
BZ was set up from stearylamine (100 mmol, 26.95 g), bisphenol A (50 mmol, 11.49 g), and paraformaldehyde (200 mmol, 6 g). Recrystallization of the item from chloroform gave white precious stones of BZ1 (36.45 g, 82.20 %, m.p. 53oC) [24]. Into 100 ml receptacle, epoxy (12.25 mmol, 4.2 g) and BZ (24.5 mmol, 20 g) were blended at 80oC Scheme 1. Benzoxaine based aniline was prepared as the past strategy test.



Scheme 1. Preparation of polybenzoxazine BZ1 & BZ2 and epoxy/ PVB

C. Preparation of Some Poly (Vinyl Butyral) [25]:

Poly (vinyl butyral) was set up by dissolving a suspension of polyvinyl liquor (PVA) in acidic corrosive and responding it with butyraldehyde within the sight of a synergist measure of a mineral corrosive (sulfuric, hydrochloric, or phosphoric); c.f. Scheme 2



Scheme 2. Preparation of PVB

D. Preparation of the Metal Coating Samples:

1 g of the readied BZ1 or BZ2/epoxy prepolymers (EBZ1 or EBZ2), was broken up in 50 ml CH₃)₂CO at room temperature, at that point PVB was added as hardening specialist with various proportions (10, 20, and 30 % wt.). The blend was mixed with sonicated for 30 min. till a homogenous arrangement was gotten. The arrangement was applied on metallic boards by stream covering strategy and left in air for 6 h to dissipate the dissolvable. The covering tests were thermally relieved in a broiler for 3 hr. at 140, 180, and 220°C to have a uniform covering thickness of around 25-40 μm estimated as per ASTM D4138-94 all the definition of the readied tests show in table 1.

TABLE (1) Samples formulations

Sample	DFT μm	Curing Temperature °C	Formulation (Ingredients Ratios %)			
			DGEBA	BZ1	BZ2	PVB
BZ1	75-85	160		100		
EBZ1/50/10	70-85	160	50	50		10
EBZ1/50/20	75-85	160	50	50		20
EBZ1/50/30	75-80	160	50	50		30
BZ2	70-85	160			100	
EBZ2/50/10	75-85	160	50		50	10
EBZ2/50/20	70-80	160	50		50	20
EBZ2/50/30	75-85	160	50		50	30

E. Testing and Co-polymer of Metal Coating Samples

Adhesion by tape test was done by ASTM D 3359-95 (B). Effect test was done by ASTM D2794-93. Scratch hardness was done by ASTM D3363-92a. Extension test was completed by ASTM D 522-93a, warm security dependent on the weight reduction for the examples in the wake of warming at 200°C for various time stretches, and substance obstruction was done by ANSI/ASTM D543-67.

III. RESULT AND DISCUSSION

A. Characterizations of Structures by FTIR

Scheme 1. Two resins of polybenzoxazine/epoxy copolymers were set up by mixing polybenzoxazine and epoxy prepolymers utilizing the distinctive amine types. The thermally initiated ring opening polymerization of benzoxazine monomer gives progressive phenolic moieties with Mannich base as rehashing joins which can respond with the oxiran gatherings of epoxy sap. Polymerization was trailed by FTIR spectra to screen the vanishing of the trademark ingestion top. Figure 1 shows the FTIR spectra of prepolymer EBZ1/20PVB and copolymer EBZ1/20PVB. The utilization of epoxy can be checked by the accompanying the trademark ingestion at 867 cm⁻¹ that vanish as the ring opening happens. The epoxide top seems to have totally vanished before the finish of the last fix at 220oC. The benzene rings that structure the foundation of a monomer were trisubstituted and showed up at 1491 cm⁻¹. During polymerization, the oxazine rings opened and bringing about the spine benzene rings become tetrasubstituted at having ingestion at 1473 cm⁻¹. The ring-opening polymerization of the benzoxazine seems to happen without obstruction by the presence of epoxy. It tends to be seen that the polymerization of the benzoxazine proceeds up to the last 220°C phase of the last fix. The epoxy seems, by all accounts, to be responding with the phenolic hydroxyl bunches when the ring-opening polymerization makes them accessible [26].

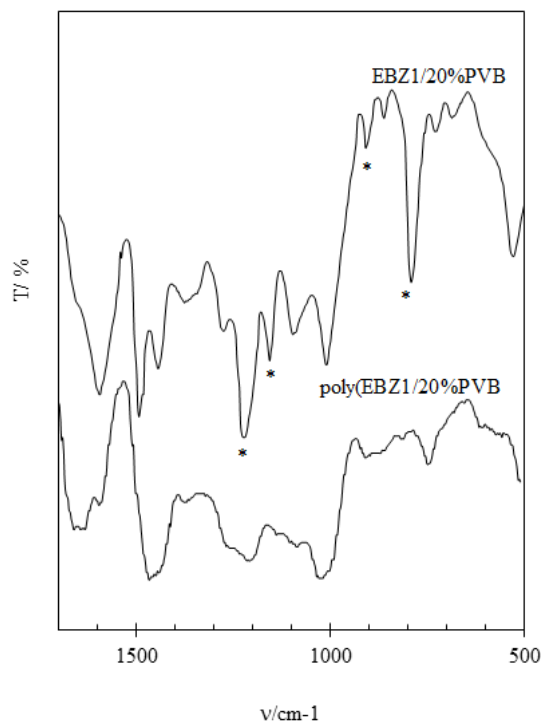


Fig. 1. FTIR of prepolymer EBZ1/20%PVB and copolymer

B. Curing of the Copolymer of Benzoxazine/Epoxy/20%Polyvinyl Butyral

EBZ1/20PVB curing attributes were gotten by DSC investigation. Figure 2 shows the DSC follows. EBZ1/20PVB the endothermic top at 49°C were credited to the softening of copolymer EBZ1/20PVB. For EBZ1/20PVB exothermic was noticed compared to the ring-opening polymerization with beginning at 160°C which was moved to lower than the benzoxazine-based stearyl amine 196°C [24] because of the impact of (epoxy and vinyl butyral) and greatest at 240°C. The ring-opening additionally creates a tertiary amine structure in the foundation of the phenolic resin. The vanished of endo. also, Exo. thermic peak to finish polymerization of the copolymer (EBZ1/20PVB)

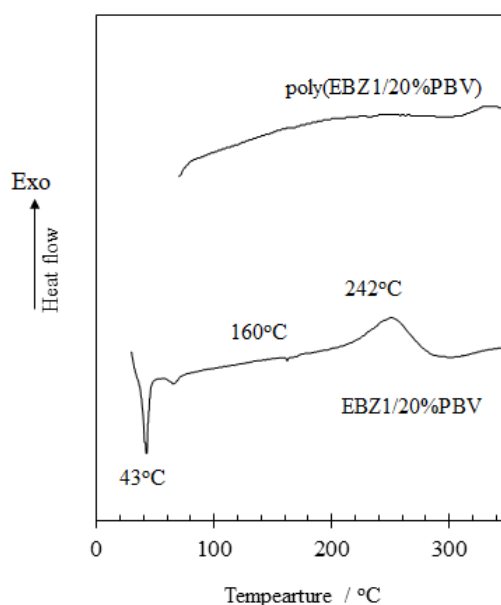


Fig. 2. DSC of curing process of EBZ1/20%PVB

C. Thermogravimetric analysis

Thermogravimetric examination of the examples was led in a nitrogen climate. The TGA follows the estimations of different disintegrations and roast yields recorded in Table 2. As we were normal the more the measure of epoxy resin the most noteworthy the deterioration temperature due to the stretch out of the organization structure and the higher the single yield by the correlation with the perfect resin of PolyBZ1 [24]. The decreased warm strength because of cutting in the organization structure, yet for this situation, the copolymerization of epoxy resin with benzoxazine may permit the organization structure to accomplish a higher crosslink thickness. Since the ring-opening polymerization of benzoxazine produces phenolic gatherings, which can respond with epoxy tars at raised temperatures, extra crosslink focuses were brought into the grid [26]. The impacts of epoxy fixation on the benzoxazine-epoxy copolymer properties have been widely concentrated from a commonsense point of view [27]. Polyvinyl butyral decreased the char yield and furthermore the security of copolymer (EBZ1 & EBZ2). Pair to their hardening impact.

TABLE 2. Thermal Properties of Copolymers

Formulation	5%DT ^a (°C)	10%DT (°C)	Char yield ^b (%)
EBZ1/10PVB	250.4	290	14.2%
EBZ1/20PVB	271.2	301.2	13.1%
EBZ1/30PVB	312.5	335.4	10.3%
EBZ2/10PVB	276.2	283	16.7
EBZ2/20PVB	262.5	278.1	14.3
EBZ/30PVB	252.8	267.3	12.4
EBZ1 ^c	284.7	312.2	9.3%
EBZ2 ^c	301.4	332.6	12.8

- a) Decomposition Temperature
- b) Char yield at 800°C
- c) Reference no 27, 26

D. Mechanical Properties of Modified BZ/Epoxy with Polyvinyl Butyral Coatings:

Mechanical properties polybenzoxazine/epoxy adjusted with polyvinyl butyral coatings for metals were completed and organized in Table (3). Results demonstrated utilizing every one of the two sorts of benzoxazine with add polyvinyl butyral increment the estimations of attachment and hardness joined with a decline in lengthening esteems. Estimations of attachment and hardness were expanded with expanding the proportion of polyvinyl butyral added to 20%. No expansion in attachment or hardness esteems past the 20% proportion of polyvinyl butyral. All altered examples with both PVF and PVB give pass sway covering tests. Estimations of prolongation expanded persistently with expanding the proportion percent of polyvinyl butyral. This conduct was coordinated with bond and hardness properties of epoxy covering tests improved by altering with 20% PVB which could be credited to acceptable similarity of polyvinyl butyral with benzoxazine and epoxy saps. This conduct might be because of the presence of the butyl bunch hanging chain in PVB, which goes about as a plasticizer [25]. The sort of benzoxazine utilized was the impact on the mechanical properties of the readied tests. Tests arranged utilizing BZ2 give lower estimations of attachment and higher estimations of hardness joined with lower estimations of lengthening than those examples arranged utilizing BZ1. This is may because of the higher aromaticity presence in the BZ2 than the aliphatic structure of BZ1.

TABLE (3): Mechanical Properties of BZ/Epoxy Modified with PVB Metallic Coatings

Sample	Curing Time (min)		Mechanical Properties			
	Touch Dry	Full Curing	Hardness	Adhesion	Impact	Elongation
BZ1	70	120	3H	5B	Pass	9
EBZ1/50/10	75	120	2H	5B	Pass	14
EBZ1/50/20	80	120	2H	5B	Pass	19
EBZ1/50/30	90	120	2H	5B	Pass	22
BZ2	60	120	3H	5B	Pass	4
EBZ2/50/10	60	120	4H	5B	Pass	7
EBZ2/50/20	70	120	4H	5B	Pass	10
EBZ2/50/30	85	120	3H	5B	Pass	14

E. Chemical Resistance:

This strategy can be applied as an adjustment in weight of the readied covering film tests after inundation for 14 days in an assortment of standard reagents and basic modern items, with indicated fixations, at room temperature (25°C).

E: Excellent, G: Good, F: Fair, P: Poor; these images were as per ANSI/ASTM D543-67.: The information got in Table 4 showed that copolymer pf polybenzoxazine/epoxy without stacking of polyvinyl butyral has magnificent protection from an alternate reagent for 14 days at room temperature 25°C as coatings for metals with film thickness 20-30 μm the various reagents specifically; water, solvents, oils, and indicated centralizations of acids, antacids, and salts. This could be ascribed to the way that all the bonds, shaped during the response of the crosslinking specialist with epoxide and hydroxyl bunches were moderately steady securities, bringing about high synthetic and dissolvable safe movies [28]. Notwithstanding, on account of EBZ1/30%PVB and EBZ2/30%PVB, The compound obstruction of these readied tests have F degree at similar conditions consequently, the high impact of polyvinyl butyral content can't avoid the reagents in this time of times. can be affirmed with the revealed information for the movies of epoxy pitch that relieved with polybenzoxazines were unaffected following 3 months drenching at 25°C by most acids, bases, ether, alcohols, water, and a few ketones. Following multi month of presentation at room temperature, the movies mollified with the accompanying materials: MEK, ethylene dichloride, hydrochloric corrosive (> 25%), sulphuric corrosive (> 25%), and hydrogen peroxide (> 15%) [29].

TABLE (4): Chemical Resistance of BZ/Epoxy Modified with PVB Metallic Coatings

Sample	Chemical Resistance												
	Water	Acids			Alkalis		Salts		Solvents			Oil	
BZ1	E	G	G	G	G	G	E	G	G	G	G	E	G
EBZ1/50/10	E	E	G	G	G	G	E	E	G	E	E	E	G
EBZ1/50/20	E	E	G	E	G	E	E	E	G	E	E	E	E
EBZ1/50/30	E	G	E	G	G	E	E	G	G	G	E	E	F
BZ2	E	G	G	G	G	G	E	G	G	G	G	E	G
EBZ2/50/10	E	E	G	G	G	G	E	E	G	E	E	E	G
EBZ2/50/20	E	E	G	E	E	E	E	E	E	E	E	E	E
EBZ2/50/30	E	E	E	G	E	E	E	G	G	G	E	E	F

F. The UV Strength of Co-polymers Coating

On the off chance that epoxy utilized outside where it is presented to daylight, all epoxies will at last yellow. This key compound response happens when epoxy is presented to bright (UV) light. Polybenzoxazine has low water take-up and high protection from UV corruption. Upon submersion in water, the relieved benzoxazines carry on in a close Fickian way, with low paces of assimilation (D) and water content at immersion [26]. The synergism between the two copolymers EBZ1 & EBZ2 presented materials can use as UV warm maturing. Table 5 finished up the copolymer of both EBZ1/PVB and EBZ2/PVB, with the distinctive heap of polyvinyl butyral way of weight when UV light introduction.

TABLE 5. Samples Weight Before and After Exposure to UV Irradiation.

Formulation	Weight before exposure to UV	Weight after exposure to UV
EBZ1	4.45	4.62
EBZ1/10PVB	4.83	5.10
EBZ1/20PVB	4.23	4.42
EBZ1/30PVB	4.91	4.52
EBZ2	3.53	3.61
EBZ2/10PVB	3.61	3.83
EBZ2/20PVB	3.91	4.21
EBZ2/30PVB	3.74	4.12

CONCLUSION

Preparation of copolymer of polybenzoxazine and epoxy as coating materials. The polyvinyl butyral was added to the prepared copolymer to enhance their toughening. Synergism between the copolymer and polyvinyl butyral introduced good materials in coating applications. The prepared copolymer EBZ1/20PVB is the optimum ratio in the mechanical and UV resistance in coating applications. Thermal stability decreased by the increasing of polyvinyl butyral content.

REFERENCES

- [1] J. Yang *et al.*, "High-toughness, environment-friendly solid epoxy resins: Preparation, mechanical performance, curing behavior, and thermal properties," *J. Appl. Polym. Sci.*, vol. 137, no. 17, p. 48596, 2020.
- [2] H. Abdollahi, A. Salimi, M. Barikani, A. Samadi, S. Hosseini Rad, and A. R. Zanjanijam, "Systematic investigation of mechanical properties and fracture toughness of epoxy networks: Role of the polyetheramine structural parameters," *J. Appl. Polym. Sci.*, vol. 136, no. 9, p. 47121, 2019.
- [3] J. Sun, C. Wang, L. P. Stubbs, and C. He, "Carboxylated lignin as an effective cohardener for enhancing strength and toughness of epoxy," *Macromol. Mater. Eng.*, vol. 302, no. 12, p. 1700341, 2017.
- [4] S. Wang *et al.*, "Graphene/nanorubber reinforced electrically conductive epoxy composites with enhanced toughness," *J. Appl. Polym. Sci.*, p. 50163, 2020.
- [5] M. Naderi, F. Ebrahimi, M. Najafi, and H. Naderi, "Reinforcing effect of amine-functionalized and carboxylated porous graphene on toughness, thermal stability, and electrical conductivity of epoxy-based nanocomposites," *J. Appl. Polym. Sci.*, vol. 136, no. 19, p. 47475, 2019.

- [6] J. S. Jayan, A. Saritha, B. D. S. Deeraj, and K. Joseph, "Graphene Oxide as a Prospective Graft in Polyethylene Glycol for Enhancing the Toughness of Epoxy Nanocomposites," *Polym. Eng. Sci.*, vol. 60, no. 4, pp. 773–781, 2020.
- [7] B. Kiskan, N. N. Ghosh, and Y. Yagci, "Polybenzoxazine-based composites as high-performance materials," *Polym. Int.*, vol. 60, no. 2, pp. 167–177, 2011.
- [8] S. M. El-Mesallamy, "Preparation, characterization and evaluation of polybenzoxazine thermoset/clay nanocomposites for metal coating," *Egypt. J. Pet.*, vol. 28, no. 3, pp. 253–260, 2019.
- [9] S.-C. Lin, C.-S. Wu, J.-M. Yeh, and Y.-L. Liu, "Reaction mechanism and synergistic anticorrosion property of reactive blends of maleimide-containing benzoxazine and amine-capped aniline trimer," *Polym. Chem.*, vol. 5, no. 14, pp. 4235–4244, 2014.
- [10] C. Zhou, X. Lu, Z. Xin, J. Liu, and Y. Zhang, "Polybenzoxazine/SiO₂ nanocomposite coatings for corrosion protection of mild steel," *Corros. Sci.*, vol. 80, pp. 269–275, 2014.
- [11] X. Shi, T. A. Nguyen, Z. Suo, Y. Liu, and R. Avci, "Effect of nanoparticles on the anticorrosion and mechanical properties of epoxy coating," *Surf. Coatings Technol.*, vol. 204, no. 3, pp. 237–245, 2009.
- [12] B. Ramezanzadeh and M. M. Attar, "Studying the effects of micro and nano sized ZnO particles on the corrosion resistance and deterioration behavior of an epoxy-polyamide coating on hot-dip galvanized steel," *Prog. Org. Coatings*, vol. 71, no. 3, pp. 314–328, 2011.
- [13] S. Grishchuk, S. Schmitt, O. C. Vorster, and J. Karger-Kocsis, "Structure and properties of amine-hardened epoxy/benzoxazine hybrids: Effect of epoxy resin functionality," *J. Appl. Polym. Sci.*, vol. 124, no. 4, pp. 2824–2837, 2012.
- [14] S. Rimdusit, P. Kunopast, and I. Dueramae, "Thermomechanical properties of arylamine-based benzoxazine resins alloyed with epoxy resin," *Polym. Eng. Sci.*, vol. 51, no. 9, pp. 1797–1807, 2011.
- [15] H. Tian, W. Li, and B. Hou, "Novel application of a hormone biosynthetic inhibitor for the corrosion resistance enhancement of copper in synthetic seawater," *Corros. Sci.*, vol. 53, no. 10, pp. 3435–3445, 2011.
- [16] A. M. Motawie and E. M. Sadek, "Adhesives and coatings based on poly (vinyl acetal) s," *J. Appl. Polym. Sci.*, vol. 70, no. 9, pp. 1769–1777, 1998.
- [17] A. M. Motawie, M. M. Badr, M. S. Amer, H. Y. Moustafa, and I. M. Ali, "Some coating studies on phenolic epoxy/poly (vinyl acetal) resins," *J. Appl. Sci. Res.*, vol. 4, no. 9, pp. 1043–1051, 2008.
- [18] H. Ipakchi, A. M. Rezadoust, M. Esfandeh, and M. Rezaei, "An investigation on the effect of polyvinyl butyral interlayer forms on the fracture toughness of glass reinforced phenolic laminates," *Thin-Walled Struct.*, vol. 151, p. 106724, 2020.
- [19] M. Sun, Z. Ma, S. Chen, G. Zhu, Y. Zhang, and W. Wang, "Anticorrosion performance of polyvinyl butyral composite coatings improved by polyaniline-multiwalled carbon nanotubes/poly (methylhydrosiloxane)," *Thin Solid Films*, vol. 712, p. 138347, 2020.
- [20] Z. Ma, M. Sun, G. Zhu, and Y. Zhang, "Anticorrosion behavior of polyvinyl butyral (PVB)/polymethylhydrosiloxane (PMHS)/chitosan (Ch) environment-friendly assembled coatings," *Prog. Org. Coatings*, vol. 144, p. 105662, 2020.

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- [21] L. Hao, K. Zhu, G. Lv, and D. Yu, "A comparative study of nanoscale poly N-(vinyl pyrrole) in polyvinyl butyral coatings for the anti-corrosion property of zinc: nanotubes vs nanoparticles," *Prog. Org. Coatings*, vol. 136, p. 105251, 2019.
- [22] G. Zhu *et al.*, "Poly (vinyl butyral)/graphene oxide/poly (methylhydrosiloxane) nanocomposite coating for improved aluminum alloy anticorrosion," *Polymer (Guildf.)*, vol. 172, pp. 415–422, 2019.
- [23] H. Azad and M. Mohsennia, "A novel free-standing polyvinyl butyral-polyacrylonitrile/ZnAl-layered double hydroxide nanocomposite membrane for enhanced heavy metal removal from wastewater," *J. Memb. Sci.*, vol. 615, p. 118487, 2020.
- [24] T. Agag, A. Akelah, A. Rehab, and S. Mostafa, "Flexible polybenzoxazine thermosets containing pendent aliphatic chains," *Polym. Int.*, vol. 61, no. 1, pp. 124–128, 2012.
- [25] M. Badr *et al.*, "Studies on coating properties of diglycidyl ether of bisphenol-a modified with poly(vinyl acetal)," *Aust. J. Basic Appl. Sci.*, vol. 6, pp. 666–674, Mar. 2012.
- [26] H. Ishida and D. J. Allen, "Physical and mechanical characterization of near-zero shrinkage polybenzoxazines," *J. Polym. Sci. Part B Polym. Phys.*, vol. 34, no. 6, pp. 1019–1030, 1996.
- [27] B. S. Rao, K. Rajavardhana Reddy, S. K. Pathak, and A. R. Pasala, "Benzoxazine–epoxy copolymers: effect of molecular weight and crosslinking on thermal and viscoelastic properties," *Polym. Int.*, vol. 54, no. 10, pp. 1371–1376, 2005.
- [28] F. Dorel and B. Alla, "Synthetic polymers technology," *Prop. Appl.*, pp. 21–22, 1996.
- [29] A. Salimi, H. Omidian, and M. J. Zohuriaan-Mehr, "Mechanical and thermal behavior of modified epoxy–novolak film adhesives," *J. Adhes. Sci. Technol.*, vol. 17, no. 13, pp. 1847–1861, 2003.