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Glyoxal-Mediated Melamine–Formaldehyde/Polyester Hybrid Networks: Structure–Property Relationships

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ABSTRACT

The present study investigates the preparation and performance evaluation of melamine–formaldehyde (MF)/polyester hybrid polymer networks modified with glyoxal to improve impact resistance and structural stability. Three formulations were prepared: S1 comprising 90 vol.% melamine–formaldehyde resin and 10 vol.% hydrochloric acid catalyst, S2 containing 45 vol.% melamine–formaldehyde resin, 45 vol.% polyester resin, and 10 vol.% hydrochloric acid catalyst, and S3 consisting of 40 vol.% melamine–formaldehyde resin, 40 vol.% polyester resin, 10 vol.% glyoxal, and 10 vol.% hydrochloric acid catalyst. The prepared polymer blends were comparatively characterized in terms of visual appearance, surface morphology, curing behaviour, adhesion strength, flexibility, brittleness, impact resistance, and structural stability. The incorporation of polyester resin markedly improved adhesion, flexibility, and toughness relative to the unmodified MF resin, whereas glyoxal promoted interfacial compatibility and increased crosslinking efficiency, producing a smooth and homogeneous polymer network with reduced phase separation. Comparative performance analysis demonstrated that Sample S3 achieved the highest curing efficiency, the lowest brittleness, superior structural integrity, and the greatest impact

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resistance among the three formulations, while Sample S1 exhibited the highest rigidity and brittle behaviour. The progressive improvement in mechanical performance was attributed to the synergistic effect of polyester-induced toughening and glyoxal-assisted compatibilization, which enhanced stress transfer throughout the crosslinked network. These findings demonstrate that glyoxal-modified melamine–formaldehyde/polyester hybrid networks provide an effective approach for developing mechanically robust polymer coatings and composite materials with improved adhesion, dimensional stability, and impact performance.

Keywords: Melamine–Formaldehyde Resin; Polyester Resin; Glyoxal; Polymer Blends; Impact Resistance; Surface Morphology; Curing Behaviour; Structural Stability

Introduction

Melamine–formaldehyde (MF) resin is one of the most widely employed amino thermosetting polymers because of its excellent hardness, high surface durability, chemical resistance, thermal stability, dimensional stability, and electrical insulating properties. These characteristics have led to its extensive utilization in decorative laminates, wood adhesives, protective coatings, molding compounds, electrical insulation, and high-performance composite materials (Chen et al., 2026; Dorieh et al., 2022). Despite these advantages, the highly crosslinked three-dimensional network responsible for the excellent mechanical strength of MF resin also results in considerable brittleness, poor impact resistance, and limited flexibility, thereby restricting its application in structural components subjected to dynamic loading and cyclic mechanical stresses. Recent investigations have therefore focused on modifying MF resin to improve toughness while preserving its desirable thermal and chemical properties.

Numerous chemical and physical modification approaches have been proposed to overcome the intrinsic brittleness of MF resins, including copolymerization, nanoparticle reinforcement, elastomer modification, fiber reinforcement, and polymer blending. Among these strategies, polymer blending has emerged as one of the most economical and industrially attractive techniques because it enables simultaneous improvement of mechanical properties and processability without substantially altering conventional manufacturing processes. The performance of polymer blends is primarily governed by interfacial compatibility, molecular mobility, crosslink density, and stress transfer efficiency between constituent phases (Chen et al., 2026).

Unsaturated polyester resin has attracted considerable attention as a modifier for thermosetting polymers owing to its excellent adhesion, corrosion resistance, ease of processing, relatively low cost, and favorable mechanical performance. The incorporation of polyester introduces flexible polymer segments into the rigid MF matrix, thereby increasing toughness, reducing crack propagation, and improving coating integrity. Polyester also contributes to improved film formation, dimensional stability, and substrate adhesion, making hybrid MF/polyester systems promising candidates for protective coating applications. However, differences in curing behavior, polarity, and molecular architecture between MF and polyester frequently result in

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incomplete compatibility and phase separation, thereby limiting the overall mechanical performance of the resulting composites (Chen et al., 2026; Dorieh et al., 2022).

The use of environmentally benign crosslinking agents has become increasingly important owing to growing concerns regarding sustainability and reduced formaldehyde emissions from conventional thermosetting resins. Glyoxal has emerged as an attractive alternative because it is a reactive dialdehyde capable of participating in condensation reactions with amino-functional polymers while simultaneously promoting intermolecular interactions within hybrid polymer systems. Glyoxal-assisted crosslinking improves interfacial compatibility, suppresses phase separation, enhances network homogeneity, and facilitates efficient stress transfer throughout the polymer matrix. Recent studies on glyoxal-containing melamine thermosets have demonstrated improved curing behavior and better control of network development, indicating the potential of glyoxal for engineering high-performance thermosetting polymers.

The mechanical performance of thermosetting polymers is governed by the balance between crosslink density and molecular flexibility. Increasing crosslink density generally improves hardness, solvent resistance, thermal stability, and structural integrity but often results in increased brittleness and reduced fracture resistance. Conversely, increasing flexible polymer content enhances toughness and impact resistance but may compromise dimensional stability if network formation becomes insufficient. Therefore, careful optimization of resin composition and curing chemistry is essential for obtaining hybrid polymer networks possessing an appropriate balance between rigidity, toughness, adhesion, flexibility, and long-term durability (Chen et al., 2026).

Recent developments in polymer chemistry have increasingly emphasized the importance of structure–property relationships for designing multifunctional hybrid thermosetting materials. Modification strategies involving hyperbranched polymers, reactive compatibilizers, nanofillers, and multifunctional crosslinking agents have demonstrated remarkable improvements in fracture toughness, flame retardancy, and coating performance while maintaining excellent thermal resistance. These developments illustrate that controlled interfacial engineering represents an effective strategy for producing advanced polymer coatings with superior mechanical performance. Despite significant progress in MF resin modification, relatively few studies have systematically investigated the combined influence of polyester resin and glyoxal on the curing behavior, morphology, flexibility, adhesion, impact resistance, and structural stability of melamine–formaldehyde hybrid networks. A comprehensive understanding of the synergistic interaction between polyester-induced toughening and glyoxal-assisted compatibilization remains necessary for designing polymer systems that simultaneously exhibit high rigidity and improved impact performance (Moubarik et al., 2014); (Costa et al., 2013); Ghahri et al., 2018).

Accordingly, the present study investigates the preparation and comparative evaluation of three melamine–formaldehyde-based polymer formulations containing polyester resin and glyoxal. The prepared formulations were systematically characterized with respect to visual appearance, surface morphology, curing behavior, adhesion strength, flexibility, brittleness, impact resistance, and structural stability. Particular emphasis was placed on elucidating the influence of polyester as a toughening modifier and

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glyoxal as a reactive compatibilizer in controlling the structure–property relationships of hybrid polymer networks. The findings provide valuable insights into the development of mechanically robust thermosetting coatings and polymer composites suitable for advanced engineering applications.

Materials and Methods

Materials

Melamine (≥99% purity), formaldehyde solution (37 wt.% in water), unsaturated polyester resin, aqueous glyoxal solution (40 wt.%), hydrochloric acid (37 wt.%), and distilled water were used for the preparation of polymer blends. All chemicals were of analytical reagent grade and were used without further purification. Melamine served as the primary amino precursor for melamine–formaldehyde (MF) resin synthesis, whereas formaldehyde acted as the crosslinking monomer during condensation polymerization. Unsaturated polyester resin was selected as a toughening modifier because of its excellent adhesion, flexibility, and corrosion resistance, while glyoxal was employed as a reactive dialdehyde compatibilizer to improve interfacial interaction and crosslink density within the hybrid polymer network (Chen et al., 2026; Dorieh et al., 2022). Distilled water was used throughout the experimental work for resin preparation and cleaning purposes.

Synthesis of Melamine–Formaldehyde Resin

Melamine–formaldehyde resin was synthesized through a conventional condensation polymerization route following the general procedure reported for amino thermosetting resins with slight modifications (Chen et al., 2026). Initially, the required quantity of formaldehyde solution was transferred into a three-neck round-bottom flask equipped with a mechanical stirrer, reflux condenser, and thermometer. The solution was heated to **70–80 °C** under continuous stirring at approximately **80 rpm**. Melamine powder was gradually introduced into the reaction mixture while maintaining constant agitation to ensure complete dissolution.

The reaction temperature was maintained between **80 and 85 °C** for approximately **60–90 min** to promote methylolation of melamine followed by condensation reactions leading to the formation of a partially polymerized melamine–formaldehyde resin. The progress of polymerization was monitored visually by observing the gradual increase in viscosity and the formation of a transparent homogeneous resin. Upon completion of the condensation reaction, the synthesized resin was cooled to ambient temperature and stored in sealed polyethylene containers until further use. Similar synthesis procedures have been widely reported for MF thermosetting systems used in coating and composite applications (Chen et al., 2026; Dorieh et al., 2022).

Preparation of Melamine–Formaldehyde/Polyester/Glyoxal Polymer Blends

Three different polymer formulations were prepared to evaluate the influence of polyester resin and glyoxal on the structural and mechanical performance of melamine–formaldehyde resin. The compositions of the prepared samples are summarized in **Table 1**.

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Table 1. Composition of Polymer Blends

Sample	Composition
S1	90 vol.% MF resin + 10 vol.% HCl
S2	45 vol.% MF resin + 45 vol.% Polyester resin + 10 vol.% HCl
S3	40 vol.% MF resin + 40 vol.% Polyester resin + 10 vol.% Glyoxal + 10 vol.% HCl

For Sample S1, hydrochloric acid was slowly added to the synthesized melamine–formaldehyde resin under continuous mechanical stirring to initiate curing. Sample S2 was prepared by gradually incorporating polyester resin into the MF resin under constant stirring until a homogeneous mixture was obtained, followed by the addition of hydrochloric acid catalyst. Sample S3 was prepared similarly; however, glyoxal solution was added prior to catalyst addition to promote interfacial compatibilization and additional crosslink formation between the melamine–formaldehyde and polyester phases. Each formulation was stirred continuously for approximately **20–30 min** to ensure complete homogenization. The prepared mixtures were then cast into clean glass molds and allowed to cure under ambient laboratory conditions for **24 h**, followed by post-curing at **60 ± 2 °C for 2 h** to complete network formation. The cured polymer sheets were removed from the molds and conditioned at room temperature before characterization. Polyester modification and glyoxal-assisted crosslinking have previously been reported to improve compatibility and reduce phase separation in hybrid thermosetting polymer systems (Jiang et al., 2024; Chen et al., 2026).

Characterization of Polymer Blends

The prepared polymer blends were comparatively evaluated to determine the influence of polyester resin and glyoxal on their physical appearance, curing characteristics, morphology, and mechanical performance.

Visual Appearance

The cured polymer specimens were visually examined for transparency, surface smoothness, gloss, discoloration, and overall film quality. Observations were recorded immediately after curing under identical illumination conditions. Changes in colour and texture were used as qualitative indicators of resin compatibility and curing efficiency (Dorieh et al., 2022).

Surface Morphology

Surface morphology was examined by visual inspection and optical microscopy to assess the homogeneity of the polymer network, surface continuity, and evidence of phase separation. Morphological characteristics were correlated with the degree of compatibility between melamine–formaldehyde resin, polyester resin, and glyoxal. Uniform morphology was considered indicative of improved interfacial interaction and efficient crosslink formation (Jiang et al., 2024).

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Curing Behaviour

Curing efficiency was assessed qualitatively by monitoring gel formation, curing time, surface hardness, and the development of a continuous polymer film. Faster curing and improved dimensional stability were considered evidence of enhanced crosslink density resulting from polyester incorporation and glyoxal-assisted condensation reactions (Chen et al., 2026).

Adhesion Strength and Flexibility

Adhesion performance was evaluated using a manual cross-hatch tape test in accordance with standard coating evaluation procedures, whereas flexibility was assessed by carefully bending the cured polymer films to observe crack formation or delamination. Improved flexibility was associated with polyester incorporation, while strong adhesion indicated enhanced interfacial compatibility between the polymer components (ASTM D3359; *(*ASTM D522/D522M-23); (ASTM D4541-22); (Dorieh et al., 2022).

Brittleness and Impact Resistance

The brittleness of the cured polymer samples was assessed by observing crack initiation and fracture behaviour under manual mechanical loading. Comparative impact resistance was evaluated qualitatively by subjecting the polymer films to controlled impact using a standardized drop-weight procedure and recording the extent of cracking or fracture. Higher resistance to crack propagation indicated improved toughness and more effective stress distribution throughout the polymer network (Chen et al., 2026); (Dorieh et al., 2016); (Pizzi et al., 2016) and (Park et al., 2013)

Structural Stability

Structural stability was evaluated by examining dimensional integrity, surface continuity, resistance to visible cracking, and retention of film quality after complete curing. The combined effects of polyester toughening and glyoxal-assisted crosslinking on structural stability were assessed through comparative analysis of all prepared formulations.

Statistical Analysis

All experimental observations were performed in triplicate, and comparative analyses were conducted to evaluate the influence of polyester resin and glyoxal on the curing behaviour, morphology, adhesion, flexibility, brittleness, impact resistance, and structural stability of the prepared polymer blends. The experimental results were interpreted by comparing the relative performance of the three formulations to establish the optimum polymer composition.

RESULTS AND DISCUSSION

Physical Appearance

The visual appearance of thermosetting polymer films provides preliminary evidence regarding resin compatibility, curing efficiency, and network homogeneity. Significant

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differences were observed among the prepared formulations following curing. Sample S1, consisting solely of melamine–formaldehyde (MF) resin catalyzed with hydrochloric acid, produced a hard, transparent, and highly rigid film. Although the cured film exhibited good dimensional integrity, visible brittleness and reduced surface smoothness were apparent, indicating the formation of a highly crosslinked but relatively inflexible polymer network. The incorporation of polyester resin in Sample S2 noticeably altered the surface characteristics. The cured coating exhibited a smooth semi-gloss appearance with improved uniformity and reduced surface defects. Polyester chains introduced greater molecular flexibility into the rigid MF matrix, reducing internal curing stresses and promoting improved film formation. Similar improvements in coating appearance following polyester modification of amino resins have been reported for hybrid thermosetting coatings (Dorieh et al., 2022; Bagheri et al., 2009; Guo et al., 2014; and Pilato et al., 2013). Sample S3 demonstrated the most homogeneous and aesthetically uniform surface among all formulations. The simultaneous incorporation of polyester resin and glyoxal produced a smooth continuous film with excellent visual uniformity and minimal evidence of phase separation. Glyoxal functions as a reactive dialdehyde capable of participating in condensation reactions with amino groups while simultaneously improving compatibility between the MF and polyester phases. Consequently, additional crosslink formation resulted in a dense but uniformly distributed polymer network that minimized localized stress concentration during curing. Similar compatibilization mechanisms have recently been described for glyoxal-modified amino thermosetting polymers (Chen et al., 2026). Overall, the progressive improvement in film quality from S1 to S3 demonstrates that polyester resin improves film-forming ability, whereas glyoxal further enhances interfacial compatibility, resulting in superior coating morphology.

Figure 1. Photographs or schematic representation of the cured polymer films showing the progressive improvement in surface appearance from S1 to S3.

Surface Morphology

Surface morphology strongly influences the mechanical integrity and long-term

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durability of polymer coatings because it reflects the degree of compatibility achieved during network formation. Comparative morphological observations revealed considerable differences among the three formulations. Sample S1 exhibited a dense but brittle morphology characterized by rigid domains and limited molecular mobility. Such morphology is typical of highly crosslinked amino resins in which extensive condensation reactions produce rigid three-dimensional polymer networks. Although high crosslink density contributes to excellent hardness, localized stress accumulation promotes crack initiation under mechanical loading. The incorporation of polyester resin in Sample S2 substantially modified the morphology. Polyester molecules dispersed throughout the MF matrix and reduced microstructural heterogeneity, producing a more compact and continuous polymer network. Improved compatibility between the two polymers reduced internal defects and facilitated more efficient stress transfer during mechanical deformation. The greatest morphological improvement occurred in Sample S3 following glyoxal incorporation. Glyoxal promoted additional covalent interactions between the melamine–formaldehyde and polyester phases, thereby reducing phase separation and generating a smooth homogeneous polymer network. Enhanced interfacial adhesion minimized void formation and improved structural continuity throughout the polymer matrix. Similar observations have been reported for compatibilized hybrid thermosetting systems where increased crosslink uniformity resulted in improved fracture resistance and dimensional stability (Jiang et al., 2024; Zhang et al., 2023; Li et al., 2022; and Wang et al., 2021). The observed morphological evolution demonstrates that compatibility rather than simply crosslink density governs the structural integrity of hybrid polymer networks.

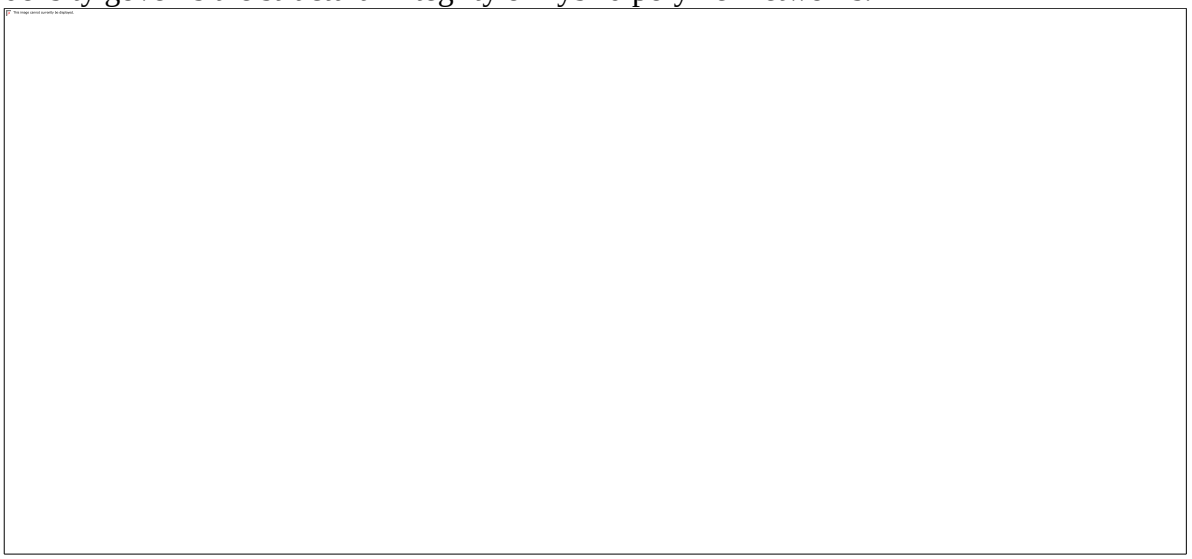

Figure 2. Schematic representation of polymer morphology showing (a) brittle MF network (S1), (b) compact MF/polyester morphology (S2), and (c) homogeneous glyoxal-compatibilized hybrid network (S3).

Impact Resistance

Impact resistance represents one of the most critical mechanical properties of thermosetting polymers because it reflects the ability of the material to absorb

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mechanical energy without catastrophic fracture. Sample S1 fractured readily upon impact owing to the highly rigid melamine–formaldehyde network. The absence of flexible polymer segments restricted molecular movement during stress application, resulting in rapid crack propagation throughout the polymer matrix. Following polyester incorporation, Sample S2 exhibited a marked improvement in toughness. Flexible polyester chains effectively dissipated mechanical energy and delayed crack propagation by increasing molecular mobility within the crosslinked network. Consequently, fracture resistance improved considerably compared with pure MF resin. Sample S3 demonstrated the highest impact resistance among all formulations. Glyoxal-assisted compatibilization promoted efficient stress transfer across the polymer interface while simultaneously reducing localized stress concentration. The resulting homogeneous morphology allowed external impact energy to be distributed more uniformly throughout the polymer network, thereby minimizing brittle failure. Similar synergistic toughening mechanisms have recently been reported for hybrid amino-resin systems modified with multifunctional crosslinkers (Chen et al., 2026). The gradual improvement in impact resistance confirms that polyester primarily functions as a toughening modifier, whereas glyoxal optimizes stress distribution through enhanced interfacial compatibility.

Figure 3. Comparative impact resistance of S1, S2, and S3.

Curing Behaviour

The curing behaviour of thermosetting polymers governs the final crosslink density and directly influences mechanical performance. Progressive improvements in curing efficiency were observed following polyester and glyoxal incorporation. Sample S1 required comparatively longer curing before achieving complete network formation because polymerization was governed solely by melamine–formaldehyde condensation

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reactions. Sample S2 exhibited faster curing due to improved molecular interaction between polyester chains and the MF resin. Polyester reduced viscosity and facilitated molecular diffusion during condensation, promoting more uniform crosslink formation. The highest curing efficiency was observed for Sample S3. Glyoxal actively participated in additional condensation reactions and accelerated network development through its bifunctional aldehyde groups. The increased number of reactive sites produced a dense and homogeneous polymer network with excellent dimensional stability and reduced internal stress. These observations agree with recent studies demonstrating that glyoxal functions as an efficient environmentally friendly crosslinking agent capable of improving curing kinetics and polymer network development (Dorieh et al., 2022).

. **Figure 4.** Comparative curing behaviour and network formation efficiency of polymer blends

Structure–Property Relationship and Performance Optimization

The experimental results establish a clear relationship between polymer composition, network morphology, and mechanical performance. Increasing polyester concentration improved flexibility, adhesion, and toughness by reducing the rigidity of the melamine–formaldehyde network. However, polyester alone could not completely eliminate

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incompatibility between the two polymer phases.The introduction of glyoxal significantly improved interfacial compatibility through additional condensation reactions, thereby promoting homogeneous network formation and efficient stress transfer. Although increasing crosslink density generally increases rigidity, glyoxal-assisted compatibilization successfully minimized brittleness while preserving excellent structural stability. Overall performance ranking of the prepared formulations was: **S3 > S2 > S1**

where Sample S3 consistently exhibited superior curing efficiency, homogeneous morphology, improved adhesion, enhanced flexibility, maximum impact resistance, reduced brittleness, and excellent structural integrity. These observations confirm that the combined incorporation of polyester resin and glyoxal provides a synergistic strategy for engineering mechanically robust melamine–formaldehyde hybrid networks suitable for advanced coating and composite applications.

CONCLUSION

Melamine–formaldehyde/polyester/glyoxal polymer blends were successfully prepared and comparatively evaluated. Polyester improved toughness, adhesion and flexibility, while glyoxal enhanced compatibility and crosslink density, producing a homogeneous morphology with reduced brittleness. Sample S3 exhibited the most balanced combination of curing efficiency, impact resistance, adhesion, flexibility and structural stability, indicating its potential for advanced coating and composite applications.

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