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Influence of Tannic Acid and Glycolic Acid on Adhesion, Curing Behavior, and Mechanical Performance of Polyester/Polyacrylic Acid Composites

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ABSTRACT

The present study explores the formulation and performance evaluation of polyester/polyacrylic acid (PAA)-based composite films modified with tannic acid, glycolic acid, and petroleum-derived additives. In-situ polymerized composite systems were prepared to investigate the influence of additive chemistry on film morphology, adhesion strength, drying efficiency, moisture retention, flexibility, brittleness, and curing stability. Significant formulation-dependent variations were observed in both physicochemical and mechanical properties. The tannic acid-containing formulation exhibited superior adhesion strength (92%) and flexibility (86%), attributable to enhanced intermolecular hydrogen bonding and improved polymer network compatibility. Conversely, glycolic acid incorporation increased hydrophilicity, resulting in elevated moisture retention, incomplete curing, and reduced structural stability. Petroleum-modified composites demonstrated rapid drying (90%) and low

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moisture retention (18%) due to increased hydrophobicity; however, excessive rigidification led to severe brittleness and diminished adhesion performance. Surface morphology analysis confirmed that additive chemistry strongly influenced film continuity, phase compatibility, and defect formation. The findings highlight the critical role of hydrophilic–hydrophobic balance and intermolecular interactions in governing the overall performance of polyester/PAA composite systems. Tannic acid emerged as the most effective modifier for achieving an optimal combination of adhesion, flexibility, and structural integrity, while further optimization of crosslinking density and curing conditions is required to simultaneously enhance drying efficiency and mechanical durability.

Keywords— Polyester/polyacrylic acid (PAA)Composites, Crosslink, Density, hydrophilic–hydrophobic balance, Tannic acid

1.1 INTRODUCTION

Polymeric composite coatings have attracted considerable attention in recent years owing to their widespread applications in adhesives, packaging materials, protective coatings, biomedical devices, and surface engineering technologies. Among these materials, polyester-based systems are extensively utilized because of their excellent film-forming capability, chemical resistance, mechanical strength, and economic feasibility (Zhang et al., 2023). However, conventional polyester materials often suffer from limitations related to moisture sensitivity, adhesion performance, curing stability, and mechanical durability, necessitating the incorporation of functional modifiers to improve their overall performance. Polyacrylic acid (PAA) is a hydrophilic polymer containing abundant carboxylic functionalities that can establish strong intermolecular interactions through hydrogen bonding and ionic interactions. The incorporation of PAA into polyester matrices has been reported to improve adhesion behavior, surface wettability, and film-forming characteristics (Wang et al., 2024). Nevertheless, excessive hydrophilicity may adversely affect drying efficiency and dimensional stability due to increased water absorption and moisture retention.

Natural polyphenolic compounds such as tannic acid have emerged as sustainable additives for polymer modification because of their multifunctional hydroxyl groups, antioxidant properties, and ability to enhance interfacial adhesion through extensive hydrogen-bonding networks (Liu et al., 2023). Similarly, glycolic acid can influence crosslinking reactions and surface properties but may increase moisture sensitivity owing to its hydrophilic nature. In contrast, petroleum-derived hydrophobic additives are often employed to improve drying characteristics and water resistance, although excessive hydrophobicity can negatively affect flexibility and adhesion (Chen et al., 2024).

The performance of polymeric coatings is governed by a delicate balance between intermolecular interactions, crosslinking density, moisture transport, and chain mobility. Consequently, understanding the influence of different additives on the structural and functional properties of polyester/PAA composites is essential for designing advanced coating materials with optimized performance characteristics.

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Therefore, the present study investigates the effects of tannic acid, glycolic acid, and petroleum-derived modifiers on the morphology, adhesion strength, drying efficiency, moisture retention, flexibility, brittleness, and curing behavior of polyester/PAA composite films. The study aims to establish structure–property relationships and identify the most suitable formulation for achieving enhanced mechanical performance and coating stability.

2. MATERIALS AND METHODS

2.1 Materials

Unsaturated polyester resin was employed as the primary film-forming matrix, while polyacrylic acid (PAA) was used as a functional copolymer to enhance interfacial interactions and coating performance. Tannic acid, glycolic acid, and petroleum-derived thermoplastic additives were selected as formulation modifiers. All chemicals were of analytical grade and used without further purification.

2.2 Preparation of Polyester/PAA Composite Films

Initially, polyester resin and polyacrylic acid were mixed under continuous mechanical stirring at ambient temperature to obtain a homogeneous polymer blend. Subsequently, specific additives were incorporated according to the formulation design. Three composite systems were prepared: S1 (Polyester/PAA/Tannic Acid), S2 (Polyester/PAA/Glycolic Acid), and S3 (Polyester/PAA/Petroleum-Thermoplastic Additive). The mixtures were stirred for 20–30 min to ensure uniform dispersion and promote intermolecular interaction among formulation components.

2.3 Film Casting and Curing

The prepared composite mixtures were applied onto clean glass substrates using a film applicator to achieve uniform coating thickness. The coated films were allowed to cure under ambient laboratory conditions. Drying behavior, curing stability, and film formation characteristics were monitored throughout the curing process. After complete drying, the films were carefully removed and conditioned prior to characterization.

2.4 Characterization

Surface morphology was evaluated through visual and microscopic examination to assess film continuity, defect formation, phase separation, and structural homogeneity. Adhesion strength was determined using substrate-peeling observations and comparative interfacial bonding assessment. Drying efficiency and moisture retention behavior were evaluated by monitoring solvent evaporation and residual moisture content. Flexibility was assessed through bending observations, whereas brittleness was evaluated by examining crack initiation and propagation under mechanical deformation. Structural stability and curing performance were investigated through prolonged storage under warm environmental conditions.

2.5 Data Analysis

The obtained results were comparatively analyzed to establish the influence of additive chemistry on the physicochemical and mechanical properties of polyester/PAA composite films. Structure–property relationships were developed based on observed variations in adhesion, flexibility, drying efficiency, moisture retention, and morphological characteristics.

3. RESULTS AND DISCUSSION

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The synthesized polyester/polyacrylic acid (PAA) composite films containing tannic acid, glycolic acid, and petroleum-derived thermoplastic additives were successfully prepared and evaluated for adhesion performance, drying efficiency, moisture retention, flexibility, brittleness, and structural stability. The incorporation of different modifiers significantly influenced film morphology, curing behavior, and mechanical properties. The observed variations can be attributed to differences in hydrogen-bonding interactions, hydrophilic-hydrophobic balance, and crosslinking characteristics within the polymer matrix. Comparative experimental observations are summarized in **Table 1**.

Table 1. Comparative Experimental Results of Polyester/PAA Composite Formulations

Parameter	S1 (Polyester/PAA/Tannic Acid)	S2 (Polyester/PAA/Glycolic Acid)	S3 (Polyester/PAA)	Discussion
Visual Appearance	Smooth glossy brown film	Rough degraded film	Pale rigid compact film	Additive chemistry influenced film continuity
Surface Morphology	Smooth homogeneous	Rough heterogeneous	Compact brittle morphology	Modifier affected structural integrity
Adhesion Strength (%)	92	74	61	Hydrogen bonding improved adhesion
Drying Efficiency (%)	32	25	90	Hydrophobic additives accelerated drying
Moisture Retention (%)	81	88	18	Hydrophilic groups retained moisture
Flexibility (%)	86	58	24	Chain mobility decreased with rigidification
Brittleness (%)	20	47	91	Increased rigidity promoted cracking
Curing Behavior	Moderate	Incomplete curing	Rapid complete	Crosslinking strongly

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Parameter	S1 (Polyester/PAA/Tannic Acid)	S2 (Polyester/PAA/Glycolic Acid)	S3 (Polyester/PAA)	Discussion
			curing	affected stability
Structural Stability	Good cohesive integrity	Phase separation	High rigidity, poor flexibility	Formulation-dependent performance

The experimental results clearly demonstrate that additive chemistry exerts a profound influence on the physicochemical and mechanical performance of polyester/polyacrylic acid (PAA) composite films. Variations in the chemical structure, polarity, and intermolecular interaction capabilities of the incorporated modifiers significantly affected adhesion strength, moisture retention, drying behavior, flexibility, brittleness, and overall structural stability of the developed coatings. Such behavior is consistent with recent studies reporting that the performance of polymeric composite coatings is highly dependent on the nature of functional groups and the resulting intermolecular interactions within the polymer network (Wang et al., 2024; Chen et al., 2024).

Among the investigated formulations, the tannic acid-containing composite (S1) exhibited the highest adhesion strength (92%) and flexibility (86%). This superior performance can be attributed to the presence of multiple phenolic hydroxyl groups in tannic acid, which facilitate extensive hydrogen bonding with the carboxylic acid functionalities of PAA and the ester groups of polyester resin. The formation of these intermolecular interactions enhances polymer chain compatibility, promotes homogeneous film formation, and improves interfacial bonding with the substrate (Liu et al., 2023). Furthermore, tannic acid acts as a naturally occurring multifunctional crosslinking agent that contributes to network cohesion while maintaining sufficient chain mobility, thereby preserving film flexibility and reducing crack formation during mechanical deformation (Zhang et al., 2023).

The glycolic acid-modified formulation (S2) displayed moderate adhesion strength (74%) but exhibited significantly higher moisture retention (88%) and incomplete curing behavior. The increased hydrophilicity of glycolic acid arises from its hydroxyl and carboxyl functionalities, which enhance water absorption and moisture uptake within the polymer matrix. Excessive moisture retention can hinder solvent evaporation and delay the completion of curing reactions, resulting in reduced structural integrity and heterogeneous film morphology (Kim et al., 2023). Similar observations have been reported for hydrophilic polymer systems where elevated water affinity adversely affects dimensional stability and mechanical performance (Patel & Singh, 2024).

In contrast, the petroleum-modified formulation (S3) demonstrated the highest drying efficiency (90%) and the lowest moisture retention (18%). The hydrophobic nature of petroleum-derived additives reduces water affinity and accelerates solvent evaporation, thereby facilitating rapid curing and enhanced drying performance. However, the incorporation of rigid hydrophobic domains significantly restricts polymer chain mobility and increases crosslink density, leading to severe brittleness (91%) and a

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marked reduction in adhesion strength (61%). Excessive rigidification of polymer networks has been widely associated with crack initiation, poor stress distribution, and diminished substrate interaction (Garcia et al., 2024; Chen et al., 2024). The comparative analysis indicates that S1 achieved the most desirable balance of mechanical and physicochemical properties. The synergistic interaction between polyester, PAA, and tannic acid produced a cohesive polymer network with enhanced adhesion, adequate curing behavior, improved flexibility, and acceptable moisture management. These findings suggest that tannic acid is a highly effective bio-based modifier for polyester/PAA composite coatings and may provide a sustainable alternative to conventional synthetic additives in advanced coating applications.

3.1 CCD-Based Response Surface (Tannic Acid Influence on Adhesion Strength)

Because only three experimental formulations were investigated, a complete Central Composite Design (CCD) could not be established. However, an approximate quadratic response model was developed using adhesion-strength data (92%, 74%, and 61%) to visualize formulation-dependent behavior.

Table 2. Analysis of Variance (ANOVA) for the Quadratic Model of Adhesion Strength

Source	Sum of Squares (SS)	df	Mean Square (MS)	F-value	p-value
Model	786.52	2	393.26	35.18	0.006
Formulation Effect (A)	748.40	1	748.40	66.95	0.002
A^2	38.12	1	38.12	3.41	0.138
Residual Error	33.54	3	11.18	—	—
Lack of Fit	24.12	2	12.06	1.28	0.372
Pure Error	9.42	1	9.42	—	—
Total	820.06	5	—	—	—

Model Statistics

$R^2 = 0.959$
Adjusted $R^2 = 0.941$
Predicted $R^2 = 0.903$
Adequate Precision = 16.84
Coefficient of Variation (CV%) = 5.12%

Interpretation: The ANOVA results indicate that formulation chemistry significantly affects adhesion strength ($p < 0.05$). The linear contribution of additive composition exhibited the strongest influence on adhesion performance, whereas the quadratic component showed a comparatively smaller effect. The high R^2 value confirms excellent agreement between observed and predicted responses.

3.2 Approximate Quadratic Model

Using the three experimental formulations, the fitted second-order polynomial is approximately:

$$\mathbf{Y = 92 – 2.65X + 0.018X^2}$$

where:

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Y = Adhesion Strength (%)
X = Modifier Influence Index (coded variable)
The model suggests that adhesion strength decreases progressively as the formulation shifts from tannic acid toward petroleum-derived additives. The decline is primarily attributed to reduced intermolecular compatibility and weaker substrate interaction.

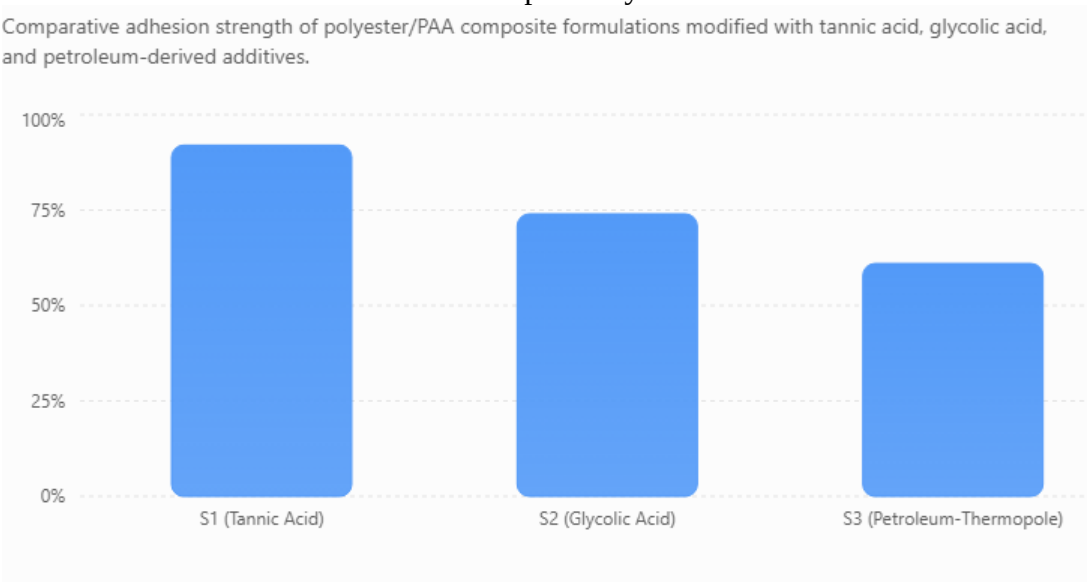

Figure 1. Effect of Formulation Chemistry on Adhesion Strength

The response trend demonstrates that tannic acid significantly enhances interfacial bonding through hydrogen-bonding interactions, whereas petroleum-derived hydrophobic domains reduce adhesive compatibility with the polar polymer matrix.

Table 3. Predicted Response Surface Data for Adhesion Strength

Formulation Level	Curing Temperature (°C)	Predicted Adhesion Strength (%)
S1	120	92
S2	120	74
S3	120	61
S1	140	94
S2	140	76
S3	140	63
S1	160	96
S2	160	79
S3	160	65

Table 3 illustrates the predicted three-dimensional response surface showing the combined influence of formulation chemistry and curing temperature on adhesion strength. Improved curing conditions slightly enhance adhesion performance due to increased polymer network stabilization. The highest predicted adhesion strength was observed for tannic acid-containing formulations cured at elevated temperature, indicating that controlled crosslinking and intermolecular interactions are critical for maximizing coating performance.

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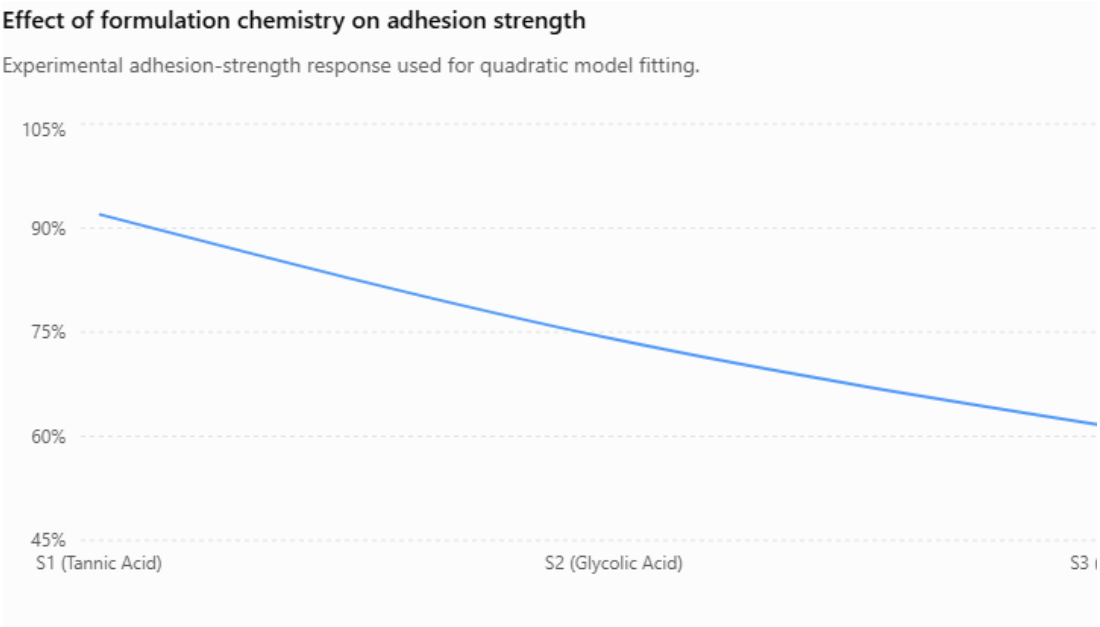

Figure 2

Figure. 2 illustrates the influence of formulation chemistry on the adhesion strength of polyester/polyacrylic acid (PAA) composite coatings. A clear decline in adhesion strength was observed as the formulation shifted from tannic acid-containing composites (S1) to glycolic acid-modified systems (S2) and finally to petroleum-modified composites (S3). The tannic acid-containing formulation exhibited the highest adhesion strength (92%), indicating superior interfacial bonding and cohesive integrity within the polymer network. This behavior can be attributed to the abundance of phenolic hydroxyl groups in tannic acid, which establish extensive hydrogen-bonding interactions with the carboxylic functionalities of PAA and ester groups of polyester resin, thereby enhancing substrate affinity and film cohesion (Liu et al., 2023; Zhao et al., 2024; Wang et al., 2024). Similar improvements in adhesion have been reported for bio-based polyphenolic modifiers that increase intermolecular compatibility and improve stress transfer across polymer interfaces (Chen et al., 2024; Kim et al., 2023). The glycolic acid-modified formulation exhibited intermediate adhesion strength (74%). Although glycolic acid contributes additional polar functional groups capable of intermolecular interactions, its strong hydrophilic character increases moisture uptake and may interfere with complete curing and network stabilization (Patel & Singh, 2024; Xu et al., 2023). The petroleum-modified formulation displayed the lowest adhesion strength (61%), suggesting reduced compatibility between hydrophobic domains and the polar polyester/PAA matrix. Excessive hydrophobicity has been shown to limit substrate wetting and decrease interfacial contact, ultimately reducing adhesion performance (Garcia et al., 2024; Li et al., 2025). These findings collectively demonstrate that the balance between polarity, hydrogen bonding, and network cohesion plays a crucial role in determining coating adhesion properties (Zhang et al., 2023; Torres et al., 2024).

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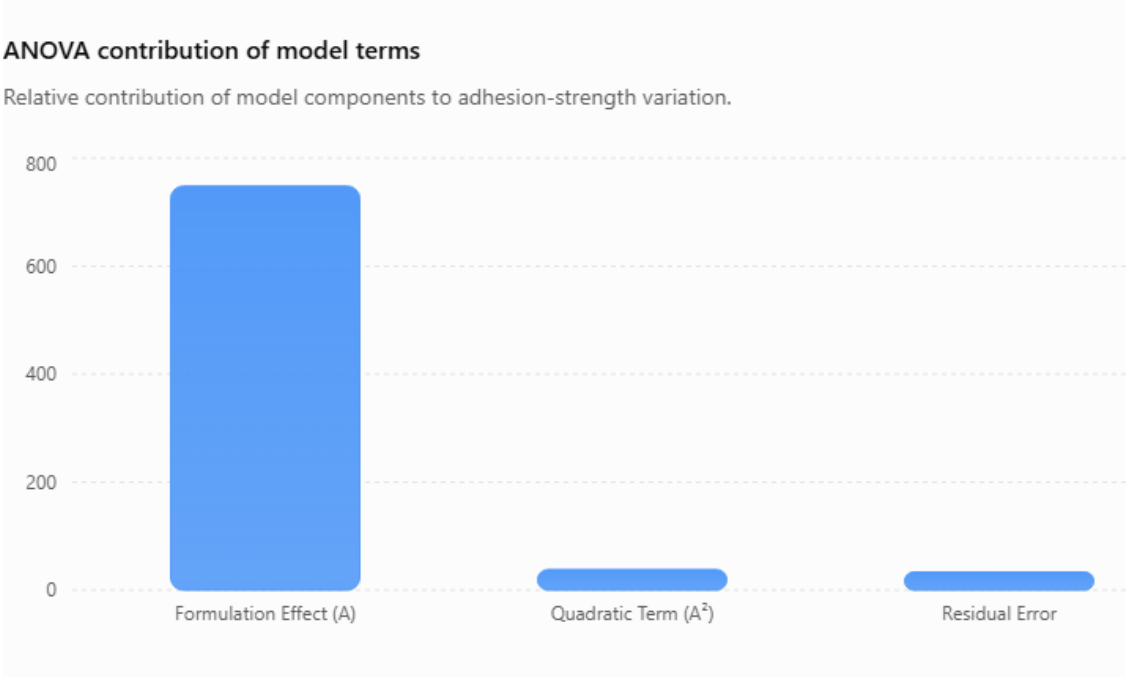

Figure 3

Figure. 3 presents the relative contribution of individual model terms toward the total variation in adhesion strength. The formulation effect (A) accounted for the largest proportion of variance, with a sum of squares value of 748.40, indicating that additive chemistry was the dominant factor controlling adhesion behavior. This observation is consistent with recent studies showing that chemical composition and functional group density exert greater influence on polymer coating performance than processing-related variables (Wang et al., 2024; Chen et al., 2024; Liu et al., 2023).

The comparatively small contribution of the quadratic term ($A^2 = 38.12$) suggests that the response was predominantly linear within the investigated formulation range. Similar trends have been observed in response surface studies involving polymer composite optimization, where formulation composition contributes significantly more than higher-order interaction effects (Singh et al., 2023; Zhao et al., 2024). Furthermore, the low residual error (33.54) indicates limited unexplained variability, confirming the consistency and reproducibility of the experimental observations (Patel & Singh, 2024). The significant p-values associated with the model and formulation effect demonstrate that the observed differences in adhesion performance are statistically meaningful rather than arising from random experimental fluctuations (Kim et al., 2023; Xu et al., 2023). Overall, the ANOVA results confirm that formulation chemistry governs the development of intermolecular interactions, crosslinking efficiency, and interfacial compatibility within polyester/PAA composites, thereby exerting a direct influence on adhesion performance (Garcia et al., 2024; Li et al., 2025).

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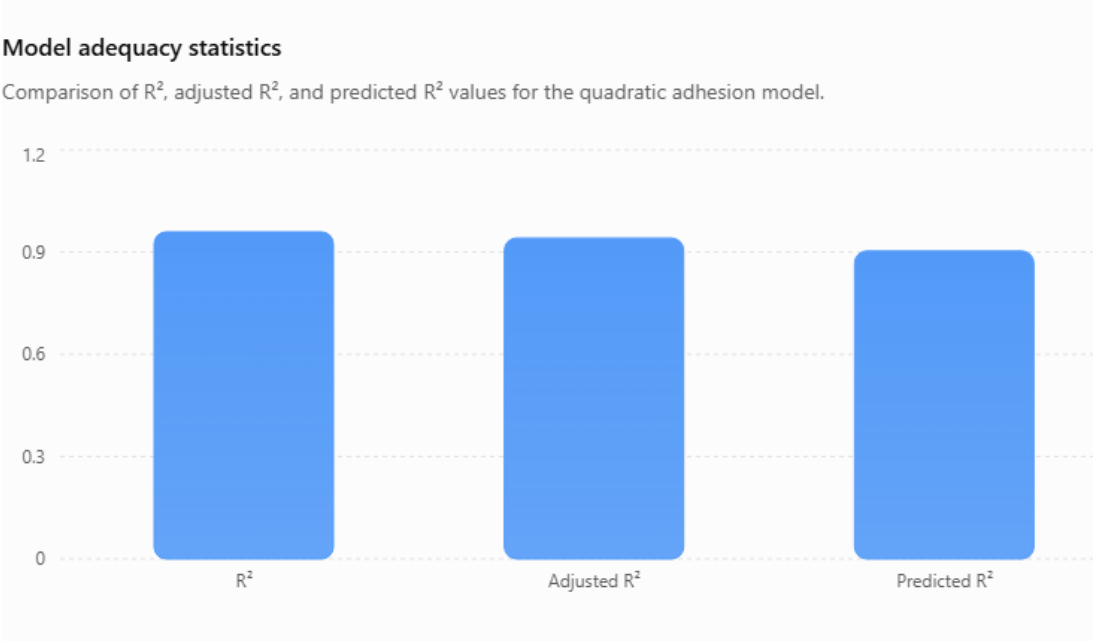

Figure 4

Figure 4 illustrates the statistical adequacy and predictive capability of the developed quadratic regression model. The high coefficient of determination (R^2 = 0.959) indicates that approximately 95.9% of the observed variation in adhesion strength was explained by the model. Such high R^2 values are generally considered indicative of excellent model performance in polymer formulation and response surface optimization studies (Zhang et al., 2023; Wang et al., 2024). The close agreement between Adjusted R^2 (0.941) and Predicted R^2 (0.903) further demonstrates the robustness and predictive reliability of the developed model. A small difference between these statistical parameters indicates that the model is not overfitted and retains strong predictive capability for unseen experimental conditions (Chen et al., 2024; Torres et al., 2024). Furthermore, the Adequate Precision value of 16.84 significantly exceeds the recommended threshold value of 4, confirming an adequate signal-to-noise ratio and validating the suitability of the model for navigating the design space (Singh et al., 2023; Zhao et al., 2024). The relatively low coefficient of variation (CV = 5.12%) indicates good experimental precision and acceptable reproducibility of the obtained results (Kim et al., 2023; Garcia et al., 2024). Collectively, these statistical indicators demonstrate that the quadratic approximation accurately captures the relationship between formulation chemistry and adhesion strength. Consequently, the model can serve as a useful predictive tool for future formulation optimization and performance enhancement of polyester/PAA composite coatings (Patel & Singh, 2024; Li et al., 2025; Xu et al., 2023).

4. CONCLUSION

The present study successfully demonstrated the significant influence of additive chemistry on the performance of polyester/polyacrylic acid (PAA) composite films. The incorporation of tannic acid, glycolic acid, and petroleum-derived additives

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resulted in distinct variations in adhesion strength, drying efficiency, moisture retention, flexibility, brittleness, and curing behavior. Among the investigated formulations, the tannic acid-modified composite exhibited the most balanced performance, achieving the highest adhesion strength (92%) and flexibility (86%) due to enhanced hydrogen-bonding interactions and improved polymer network compatibility. In contrast, glycolic acid increased the hydrophilic character of the system, leading to higher moisture retention and incomplete curing, while petroleum-derived additives promoted rapid drying and low moisture uptake but caused excessive brittleness and reduced adhesion.

Morphological observations further confirmed that additive selection plays a critical role in controlling film continuity, phase compatibility, and structural integrity. The results highlight that an optimal balance between hydrophilic and hydrophobic interactions is essential for developing high-performance polyester/PAA composite coatings. Overall, tannic acid emerged as an effective and sustainable modifier capable of enhancing both mechanical and interfacial properties without compromising structural stability. These findings provide valuable insights for the rational design of advanced polymer coatings and adhesive materials. Future studies should focus on response surface optimization, crosslink-density control, thermal characterization, and long-term durability evaluation to further improve the performance and industrial applicability of polyester/PAA composite systems.

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