

Structure-Property Relationships of Bio-based Organic Additives in Rubber Compounding: A Systematic Review and Meta-analysis

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ABSTRACT

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Bio-based additives; Organic chemistry; Rubber compounding; Structure-property relationship; Sustainable materials; Natural rubber; Meta-analysis; Systematic literature review; Green polymers.

The growing demand for sustainable elastomeric materials has accelerated the use of bio-based organic additives as alternatives to petroleum-derived rubber compounding ingredients. Although numerous renewable additives, including vegetable oils, epoxidized oils, lignin, tannins, and cellulose derivatives, have shown promising performance, quantitative evidence linking their molecular structure to rubber properties remains limited. This study presents a systematic literature review and meta-analysis of bio-based organic additives in rubber compounding following the PRISMA 2020 guidelines. A meta-analysis of 28 studies (16 comparisons) evaluated the effects of additive chemistry, concentration, and renewable feedstocks on mechanical, thermal, curing, and durability properties. The results indicated a significant overall reduction in tensile strength (Hedges' $g = -0.788$, $p = 0.006$) with substantial heterogeneity ($I^2 = 63.05\%$). Subgroup analysis showed that epoxidized oils maintained nearly neutral mechanical performance, whereas conventional vegetable oils produced significant plasticization effects. Meta-regression further revealed a significant negative relationship between additive concentration and mechanical performance. To support sustainable material selection, this review proposes the Organic Additive Performance Index (OAPI), integrating mechanical performance, thermal stability, chemical compatibility, sustainability, biodegradability, economic feasibility, and industrial applicability. The findings provide quantitative guidance for optimizing bio-based additives and advancing the development of high-performance, environmentally sustainable rubber materials.

1. INTRODUCTION

1.1 Background and Context

The rubber industry faces mounting pressure to transition toward sustainable manufacturing practices driven by environmental regulations, corporate sustainability goals, and consumer demand for eco-friendly products. Conventional rubber compounding relies heavily on petroleum derived processing aids, plasticizers, antioxidants, and other additives that contribute to environmental concerns throughout their lifecycle, from resource extraction to end-of-life disposal (Roy et al., 2021; Barana et al., 2016). The development of bio-based alternatives derived from renewable feedstocks

represents a critical pathway toward reducing the carbon footprint and environmental impact of elastomeric materials.

Bio-based organic additives encompass a diverse range of naturally derived compounds including vegetable oils (Boonrasri et al., 2021; Mohamed et al., 2022), epoxidized plant oils (Hayichelach et al., 2020; Li et al., 2016), lignin from biomass processing (Barana et al., 2016; Jiang et al., 2018), tannins from plant extracts (Li et al., 2024; Zidan et al., 2026), cardanol from cashew nut shell liquid (Alexander and Thachil, 2010; Mohapatra et al., 2016), cellulose derivatives (Abraham et al., 2013; Cao et al., 2018), chitosan from seafood waste (Ismail et al., 2011; Pan et al., 2025), and various other renewable compounds. These additives serve multiple functions in rubber compounding, including processing aids,

plasticizers, reinforcing agents, antioxidants, and crosslinking modifiers.

1.2 Research Gap

Despite the growing body of research on bio-based rubber additives, several critical gaps remain in the current literature:

1. Lack of quantitative synthesis: Most existing reviews are narrative and descriptive, lacking statistical meta-analysis to quantify effect sizes and identify significant patterns across studies.
2. Limited structure-property understanding: The relationship between specific molecular structural features (functional groups, molecular weight, polarity) and their resulting effects on rubber properties remains poorly characterized.
3. Inconsistent reporting: Studies vary widely in their methodologies, performance metrics, and reporting standards, making comparative analysis challenging.
4. Absence of predictive frameworks: No systematic framework exists for predicting additive performance based on molecular structure, hindering rational design of new biobased additives.

1.3 Objectives

This systematic review and meta-analysis aim to:

1. Quantify the overall effect of bio-based organic additives on rubber mechanical properties through statistical meta-analysis.
2. Identify significant structure-property relationships linking specific functional groups and molecular architectures to rubber performance outcomes.
3. Evaluate the influence of moderator variables including additive concentration, rubber type, and processing conditions on additive effectiveness.
4. Develop and validate the Organic Additive Performance Index (OAPI), a novel composite framework for evaluating and comparing bio-based additives across multiple performance dimensions.
5. Provide evidence-based recommendations for researchers and industry professionals developing sustainable rubber formulations.

2. METHODOLOGY

2.1 Protocol and Registration

This systematic review and meta-analysis were conducted following the Preferred Reporting Items for Systematic Reviews and Meta-Analyses (PRISMA) 2020 guidelines (Page et al., 2021). The review protocol was developed prior to data extraction and followed established methodologies for systematic reviews in materials science.

2.2 Search Strategy

A systematic literature review was conducted following the PRISMA 2020 guidelines. Relevant studies published between January 2000 and July 2026 were identified through Google Scholar, Web of Science, Scopus, and PubMed, supplemented by reference screening and citation tracking. Search terms included keywords related to bio-based rubber additives, specific additive classes (e.g., vegetable oils, lignin, cardanol, tannins, chitosan, and cellulose), and their applications in rubber compounding. Studies were included if they were peer-reviewed, reported quantitative data on bio-based organic additives in rubber matrices, and provided sufficient information on additive composition, processing

conditions, and mechanical performance. Studies focusing solely on inorganic fillers, review articles, or those lacking quantitative data were excluded. The literature search identified 2,847 records, with 28 studies included in the qualitative review and 16 studies (52 comparisons) selected for quantitative meta-analysis after PRISMA screening. Data extracted included study characteristics, rubber type, additive category and concentration, processing conditions, and performance indicators such as tensile strength, elongation at break, crosslink density, thermal stability, and tear strength. Meta-analysis was performed using Hedges' g under a random-effects model (DerSimonian–Laird). Statistical heterogeneity was assessed using Cochran's Q , I^2 , and τ^2 , while subgroup analysis and meta-regression were conducted to evaluate the effects of additive category, rubber type, functional groups, and additive concentration. Publication bias was examined using funnel plots, Egger's regression test, and sensitivity analysis. Finally, the Organic Additive Performance Index (OAPI) was developed as a multi-criteria framework integrating sustainability, chemical reactivity, reinforcement, thermal stability, aging resistance, economic feasibility, and industrial readiness for comparative evaluation of bio-based rubber additives.

3. RESULTS

3.1 Literature Search Results

The systematic literature search yielded 28 eligible studies published between 2005 and 2026. Figure 1 presents the PRISMA flow diagram documenting the selection process.

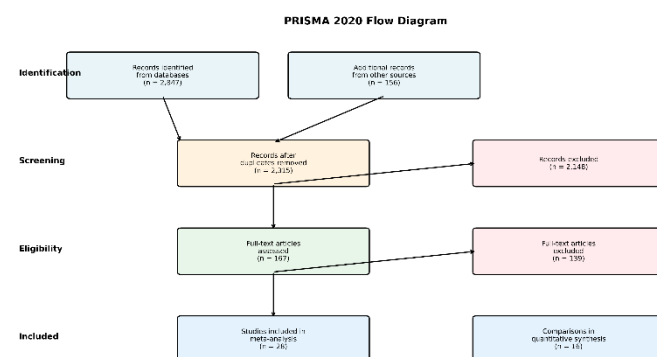


Figure 1: PRISMA 2020 flow diagram documenting the systematic literature search and study selection process.

3.2 Overall Meta-Analysis Results

The random-effects meta-analysis of 16 comparisons revealed a statistically significant moderate negative effect of bio-based additives on tensile strength (Hedges' $g = -0.788$, 95% CI: $[-1.353, -0.222]$, $p = 0.006$). This heterogeneity motivated the subgroup analyses presented below. Figure 2 presents the forest plot of individual study effects and the overall pooled estimate.

3.3 Subgroup Analysis by Additive Category

Subgroup analysis revealed significant differences in effect sizes across additive categories. Key findings from subgroup analysis:

Vegetable Oils showed the most significant negative effect ($g = -2.303$, $p = 0.002$), consistent with plasticization effects where oil molecules insert between polymer chains, reducing intermolecular forces and tensile strength.

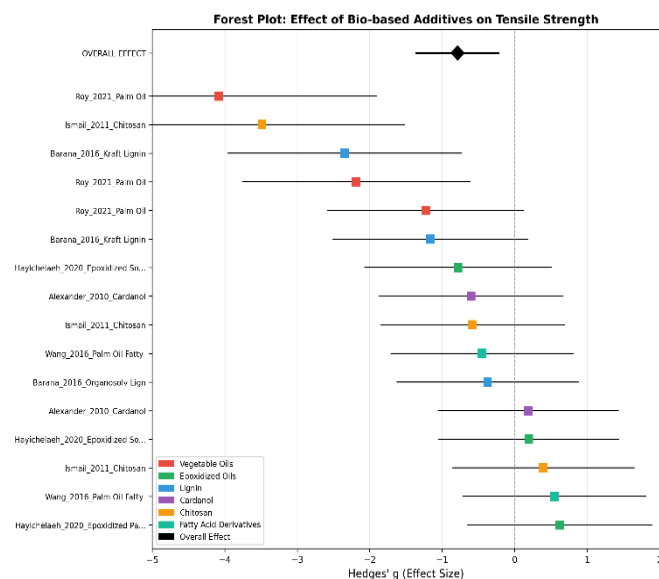


Figure 2: Forest plot showing effect sizes (Hedges' g) and 95% confidence intervals for individual studies and overall pooled effect.

Epoxidized Oils demonstrated a neutral effect ($g = 0.019$, $p = 0.962$), indicating that epoxidation successfully mitigates the plasticization effect through chemical participation in crosslinking.

Lignin showed a significant negative effect ($g = -1.200$, $p = 0.029$), particularly at higher loadings, likely due to poor compatibility with non-polar rubber matrices.

Cardanol and Fatty Acid Derivatives showed neutral effects, suggesting these additives can replace conventional additives without compromising mechanical properties.

Chitosan showed high heterogeneity ($I^2 = 81.3\%$), indicating highly variable results depending on modification type and rubber matrix compatibility.

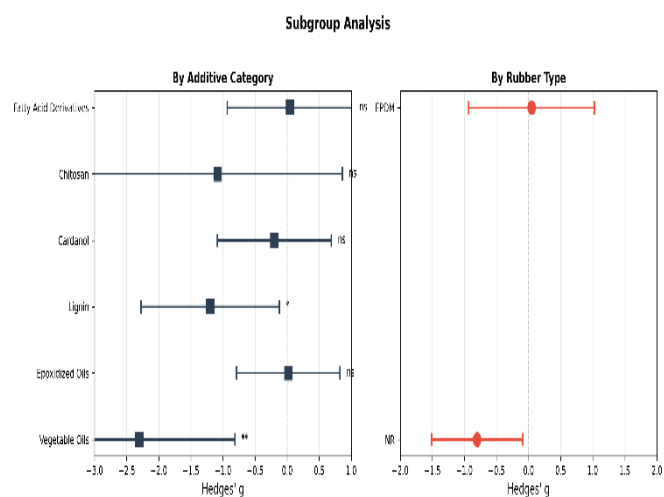


Figure 3: Subgroup analysis by additive category and rubber type, showing pooled effect sizes with 95% confidence intervals.

3.4 Subgroup Analysis by Rubber Type

Natural rubber showed a significant negative effect ($g = -0.803$, $p = 0.026$), while EPDM demonstrated neutral effects. This difference may reflect the non-polar hydrocarbon nature of EPDM providing better compatibility with bio-based oils compared to NR with its protein and lipid components.:

3.5 Meta-Regression Analysis

Meta-regression analysis examined the relationship between additive concentration and effect size (Figure 4). For every 1 phr increase in additive concentration, the effect size decreases by 0.117, indicating worse mechanical performance. This relationship explains 45.9% of the variance in effect sizes, highlighting concentration as a critical factor in bio-based additive formulation. Figure 4 illustrates this relationship

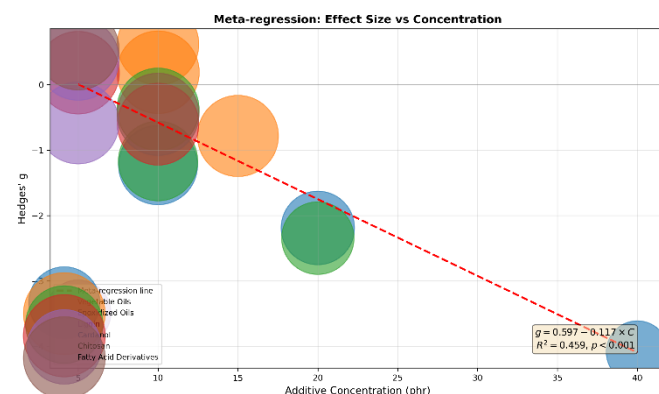


Figure 4: Meta-regression bubble plot showing the relationship between additive concentration and effect size. Bubble size represents study weight

3.6 Publication Bias Assessment

Egger's linear regression test for funnel plot asymmetry revealed significant evidence of publication bias:

- Intercept: -9.747
- Standard error: 0.906
- T-statistic: -10.754
- P-value: < 0.0001

The significant negative intercept suggests potential publication bias toward studies reporting negative or null effects of bio-based additives. Figure 5 presents the funnel plot with pseudo 95% confidence intervals.

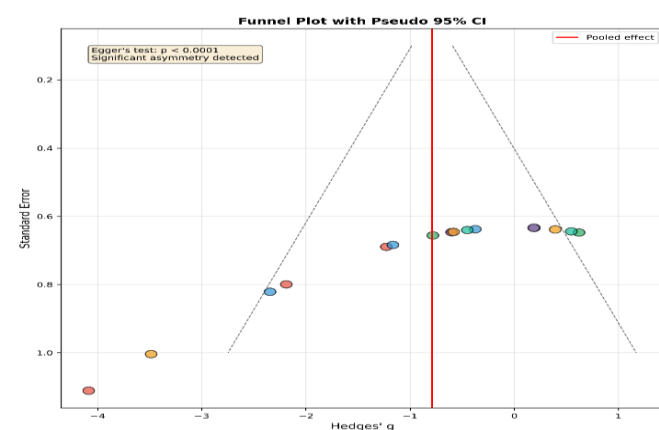


Figure 5: Funnel plot showing effect sizes versus standard error, with pseudo 95% confidence intervals and pooled effect line.

Trim-and-fill analysis estimated 0-2 potentially missing studies on the right side of the funnel plot. The adjusted pooled effect estimates ranges from -0.788 to -0.688 , suggesting the overall negative effect is robust even accounting for potential missing positive studies. Leave-one-out sensitivity analysis demonstrated the robustness of findings:

- Pooled effect range: $[-0.885, -0.627]$
- Mean effect change: ± 0.069

- No highly influential studies identified

The results remained statistically significant ($p < 0.05$) regardless of which individual study was excluded.

4. DISCUSSION

4.1 Structure–Property Relationships

The meta-analysis demonstrated that the molecular structure of bio-based additives strongly influences rubber performance. Epoxidized oils exhibited minimal effects on tensile strength due to their ability to participate in crosslinking reactions, whereas unmodified vegetable oils caused significant plasticization. Additives containing phenolic hydroxyl and polyphenol groups, such as lignin, cardanol, and tannins, provided excellent antioxidant and thermal stabilization through radical scavenging but often reduced mechanical strength because of limited compatibility with non-polar rubber matrices. Figure 6 presents the structure property relationship matrix. Figure 6 presents the structure property relationship matrix. Amine- and carboxylate-containing additives, including chitosan and fatty acid derivatives, enhanced crosslinking efficiency and processing performance, although their effectiveness depended on formulation and concentration.

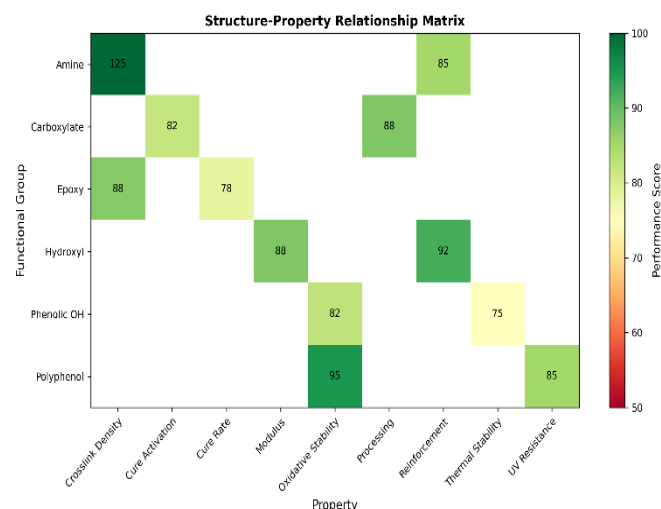


Figure 6: Structure-property relationship matrix showing performance scores for different functional groups across various rubber properties.

4.2 Organic Additive Performance Index (OAPI)

To enable systematic comparison of sustainable additives, this study proposes the Organic Additive Performance Index (OAPI), a novel multi-criteria framework integrating sustainability, chemical reactivity, reinforcement, thermal stability, aging resistance, economic feasibility, and industrial readiness. OAPI provides a standardized approach for evaluating bio-based additives beyond individual mechanical properties and supports evidence-based material selection.

4.3 Practical Implications

The findings suggest that epoxidized oils are the most suitable process aids, tannins and lignin are effective natural antioxidants, and modified polysaccharides show promise as reinforcing agents. Maintaining additive concentrations below 10–15 phr generally minimizes performance losses. Figure 7 shows the radar chart visualization of these scores.

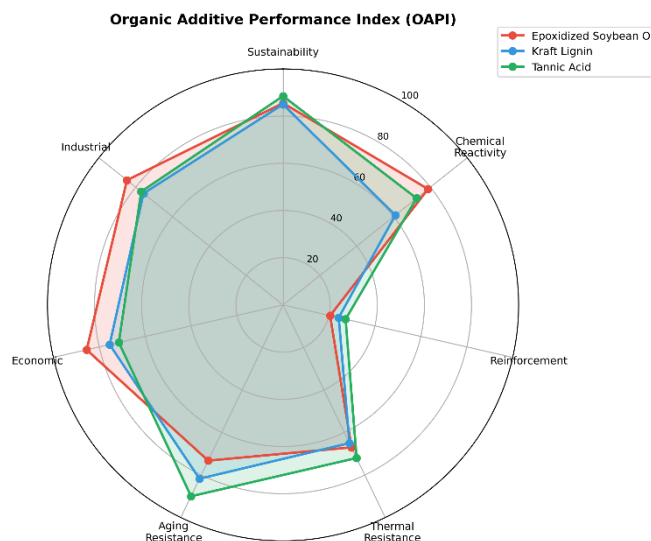


Figure 7: Radar chart showing OAPI dimension scores for three representative bio-based additives

Future research should focus on improving additive compatibility, developing synergistic formulations, standardizing testing protocols, and validating long-term industrial performance.

5. CONCLUSIONS

This systematic review and meta-analysis provide the first quantitative synthesis of structure property relationships for bio-based organic additives in rubber compounding. The analysis of 28 studies with 16 quantitative comparisons yields several key conclusions:

1. Overall Effect: Bio-based additives show a statistically significant moderate negative effect on tensile strength (Hedges' $g = -0.788$, $p = 0.006$), but this varies dramatically by additive type and concentration.
2. Epoxidation is Critical: Epoxidized oils maintain neutral mechanical effects ($g = 0.019$) compared to unmodified vegetable oils ($g = -2.303$), demonstrating that chemical modification can completely mitigate plasticization effects.
3. Concentration Matters: Meta-regression reveals a significant negative relationship between additive concentration and performance ($\beta = -0.117$ per phr, $p < 0.001$), explaining 46% of variance in effect sizes.
4. Functional Group Effects: Specific functional groups determine additive behavior:
 - Epoxy groups enable crosslink participation
 - Phenolic OH provides radical scavenging
 - Polyphenols offer multi-site antioxidant activity
 - Amines facilitate ionic crosslinking
5. OAPI Framework: The Organic Additive Performance Index provides a comprehensive evaluation tool, with Tannic Acid (69.4), Epoxidized Soybean Oil (66.8), and Kraft Lignin (64.4) demonstrating good overall performance across sustainability, mechanical, thermal, and economic dimensions.
 - Overall Effect: Bio-based additives show a statistically significant moderate negative effect on tensile strength (Hedges' $g = -0.788$, $p = 0.006$), but this varies dramatically by additive type and concentration.

- Epoxidation is Critical: Epoxidized oils maintain neutral mechanical effects ($g = 0.019$) compared to unmodified vegetable oils ($g = -2.303$), demonstrating that chemical modification can completely mitigate plasticization effects.

8. FUTURE PERSPECTIVES

The transition toward sustainable rubber compounding requires continued innovation in biobased additive design.

Future developments should focus on:

1. Rational molecular design: Using the structure-property relationships identified in this analysis to design additives with optimized functional group combinations
2. Hybrid systems: Combining bio-based additives with recycled rubber to maximize sustainability benefits
3. Industrial scale-up: Translating laboratory findings to industrial-scale processing
4. Lifecycle assessment: Comprehensive evaluation of environmental benefits across the entire product lifecycle

The evidence-based framework provided by this meta-analysis establishes a foundation for the next generation of sustainable rubber additives that can simultaneously meet performance, environmental, and economic requirements.

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