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Study of Recyclable Thermoset Polymers Using Dynamic Covalent Bonds

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Abstract

Thermoset polymers are widely used in advanced engineering applications due to their excellent mechanical strength, thermal resistance, and chemical stability. However, their permanent three-dimensional cross-linked networks make them traditionally non-recyclable, contributing significantly to polymer waste and environmental challenges. In recent years, dynamic covalent chemistry (DCC) has emerged as a transformative approach for designing recyclable thermoset materials by incorporating reversible and exchangeable bonds within their structure. This project focuses on the study of recyclable thermoset polymers that utilize dynamic covalent bonds such as transesterification, imine exchange, disulfide exchange, and reversible Diels–Alder reactions. These dynamic bonds allow the polymer network to undergo reshaping, self-healing, and reprocessing under controlled conditions, giving rise to a new class of materials known as vitrimers. The research explores the mechanisms, properties, advantages, and limitations of such systems, along with their potential applications in aerospace, automotive, electronics, adhesives, and sustainable material design. By reviewing recent literature and analyzing the scientific principles behind dynamic covalent thermosets, this project highlights their importance in moving toward a circular materials economy. The study underlines how recyclable thermosets can play a crucial role in reducing waste and promoting environmentally responsible polymer technology.

Keywords: Thermoset, Polymers, environmental challenges, Chemical routes

INTRODUCTION

Polymers have become an integral part of modern society due to their versatility, durability, and wide range of applications across sectors including packaging, construction, electronics, transportation, and healthcare. Among the broad classes of polymeric materials, thermoset polymers occupy a special position because of their excellent mechanical strength, dimensional stability, thermal resistance, and chemical durability. These materials possess a highly cross-linked three-dimensional network structure formed during the curing process, which gives them superior performance characteristics compared to many thermoplastics. Common thermosetting materials such as epoxy resins, unsaturated polyesters, phenol–formaldehyde resins, and polyurethane networks are extensively used in high-performance applications like aerospace composites, automotive structural parts, electronic components, and adhesives.

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However, the greatest advantage of thermosets – namely their chemically permanent, cross-linked structure – also represents their most significant limitation. Once cured, thermoset polymers cannot be melted, reshaped, or reprocessed. This irreversible nature prevents them from being recycled through conventional methods such as melting and remolding, which are commonly applied to thermoplastics. As a result, thermoset waste accumulates in landfills and the

environment, contributing to long-term pollution and resource depletion. With increasing global focus on sustainability, circular economy models, and environmentally responsible materials, there is a growing need to design polymer systems that combine high performance with recyclability.

In recent years, dynamic covalent chemistry (DCC) has emerged as a promising strategy to address this challenge. DCC involves reversible chemical bonds that can break and reform under specific conditions such as heat, light, pH change, or the presence of catalysts. Incorporating these reversible bonds into a thermoset network allows the material to behave like a traditional thermoset at room temperature – retaining its rigidity and dimensional stability – while exhibiting the ability to undergo network rearrangement when triggered. This results in a new class of materials known as recyclable thermosets or vitrimers, which bridge the gap between thermosets and thermoplastics.

Vitrimers were first introduced by Ludwik Leibler and his research group in 2011, marking a major breakthrough in polymer science. These materials possess exchangeable covalent bonds, which enable the polymer network to undergo associative exchange reactions without reducing cross-link density. Unlike thermoplastics that melt completely, vitrimers soften gradually at elevated temperatures, allowing them to be reshaped, reprocessed, repaired, or welded. Some of the most commonly used dynamic covalent mechanisms in recyclable thermosets include transesterification, imine exchange, disulfide exchange, boronic ester exchange, and reversible Diels–Alder reactions.

The importance of recyclable thermoset materials extends beyond environmental sustainability. Their dynamic nature offers additional functionalities such as self-healing, stress relaxation, enhanced toughness, and adaptability to external stimuli, which can significantly improve material lifespan and reduce maintenance costs. These features are highly valuable in advanced applications such as aerospace structures, electronic encapsulation, smart coatings, and next-generation flexible devices.

Despite these advantages, several challenges remain. The design of dynamic networks must ensure that the material maintains sufficient thermal and mechanical strength in its operational state, while still allowing bond exchange at practical temperatures. Balancing performance, stability, processing temperature, and cost requires careful molecular engineering. Furthermore, industrial adoption demands scalability, compatibility with existing curing systems, and predictable long-term behavior.

This research project aims to explore the chemistry, mechanisms, and performance of recyclable thermoset polymers based on dynamic covalent bonds. It seeks to highlight how dynamic bonds are incorporated into polymer networks, the types of bond-exchange reactions that enable recyclability, and the critical factors affecting material behavior. The project also reviews recent developments in vitrimer technology, examines their potential applications across industries, and discusses future directions for sustainable thermoset design.

By presenting a detailed study of recyclable thermoset polymers, this work emphasizes the transformative role of dynamic covalent chemistry in developing environmentally responsible, high-performance materials. This exploration is aligned with global efforts to minimize polymer waste, promote circular material design, and create smarter, more sustainable alternatives to traditional polymers.

LITERATURE REVIEW

Introduction to Thermoset Polymers

Thermoset polymers are a class of materials that form irreversible cross-linked networks when cured. Because of this permanent three-dimensional structure, they exhibit excellent thermal resistance, chemical stability, dimensional rigidity, and mechanical strength. Traditional thermosets cannot be reshaped or reused after curing because the covalent bonds are permanent. This has led to growing environmental concerns, as discarded thermoset composites accumulate in landfills due to their non-recyclable nature. Recent studies emphasize the need for sustainable alternatives that enable recycling without compromising performance [1, 2].

Need for Recyclable Thermosets

Global thermoset consumption has increased drastically in coatings, aerospace, electronics, automotive, and construction industries. However, their end-of-life management remains a major challenge. Mechanical grinding leads to loss of properties, while chemical recycling is costly and energy-intensive. Thus, researchers have shifted towards designing new thermosets that possess recyclability and repairability through reversible or dynamic linkages. Dynamic covalent chemistry (DCC) has become a widely explored strategy for sustainable polymer circularity [3].

Dynamic Covalent Chemistry (DCC)

Dynamic covalent bonds can break and reform under external stimuli such as heat, pH, catalyst, or light. This allows the material to behave like a traditional thermoset during use, but like a thermoplastic during recycling. DCC introduces adaptability into polymer networks without compromising strength.

Common Dynamic Bonds Studied Include

- Diels–Alder adducts
- Disulfide linkages
- Transesterification reactions
- Imine (Schiff-base) bonds
- Boronic ester exchanges
- Vinylogous urethane exchange

The incorporation of these reversible bonds enables self-healing, reshaping, and closed-loop recycling of thermoset matrices [4, 5].

Recyclable Thermosets Based on Diels–Alder Chemistry

The Diels–Alder (DA) reaction between a diene and a dienophile is thermally reversible. Many studies show that DA-based thermosets can be depolymerized at elevated temperatures (120–150°C) and re-crosslinked upon cooling. This process allows multiple recycling cycles with minimal property loss. DA-based epoxy and polyurethane networks have demonstrated excellent recyclability, making this approach one of the most practical strategies in industry [6].

Thermosets with Disulfide Exchange Networks

Disulfide bonds undergo rapid exchange reactions under heat or UV light. Incorporation of disulfide linkages into epoxy or polyurethane matrices produces materials capable of self-repair, stress relaxation, and reprocessing. These materials retain high mechanical strength but can be ground and remolded, offering a promising route for sustainable thermoset recycling [7].

Transesterification-Based Vitrimers

Vitrimers are a modern class of recyclable thermosets that maintain network connectivity while allowing bond exchange through associative mechanisms. Transesterification is the most widely used chemistry for vitrimer design. When heated above a topology-freezing temperature (Tv), the covalent bonds rearrange, enabling flow and reprocessing. Vitrimers show excellent solvent resistance, high strength, and industrial scalability. They are considered one of the most promising future alternatives to conventional thermosets [8, 9].

Imine-Based Recyclable Thermosets

Imine (C=N) bonds are dynamic and can undergo reversible hydrolysis and exchange reactions. Thermosets containing imine linkages can be chemically recycled in mild acidic solutions and reformed into new products. These polymers enable both closed-loop recycling (same product) and open-loop recycling (different product). Imine chemistry has gained popularity for developing self-healing coatings and recyclable adhesives [10].

Recent Advances in Bio-Based Dynamic Thermosets

Sustainability research has progressed toward using renewable feedstocks such as

- Lignin
- Tannins
- Vegetable oils
- Citric acid
- Soybean-based epoxies

When these bio-based polymers are combined with dynamic covalent bonds, they offer eco-friendly alternatives with reduced carbon footprint. Several studies highlight bio-vitrimers that combine recyclability, biodegradability, and performance suitable for packaging,

SUMMARY OF RESEARCH TRENDS

The literature strongly indicates that dynamic covalent chemistry has revolutionized the recyclability of thermosets. Key trends include.

- Transition from permanent crosslinks to reconfigurable networks
- Expansion of vitrimer technology in commercial sectors
- Self-healing thermosets enabling longer product lifetimes
- Increased emphasis on bio-based and renewable monomers
- Development of closed-loop recycling systems

Overall, recyclable thermosets using dynamic covalent bonds represent a breakthrough toward a circular polymer economy.

OBJECTIVES

The objectives of this research project are as follows

- To understand the fundamental chemistry of thermoset polymers
  - To study the structural characteristics, crosslinking behaviour, and limitations of conventional thermoset materials, particularly their inability to be reshaped or recycled after curing.

- To explore the concept and mechanisms of Dynamic Covalent Chemistry (DCC)
  - To analyse how reversible covalent bonds such as Diels–Alder, imine, disulfide, boronic ester, and transesterification reactions contribute to recyclability and self-healing properties in thermoset networks.

- To examine different classes of recyclable thermosets
  - To review vitrimer systems, DA-based networks, disulfide-exchange polymers, and imine-based thermosets, and compare their recyclability potential, thermal stability, mechanical performance, and reprocessing efficiency.

- To study the role of dynamic covalent bonds in polymer reprocessing
  - To evaluate how bond exchange mechanisms allow reshaping, repair, stress relaxation, and closed-loop recycling in thermosets without degrading material properties.

- To analyse major research advancements and existing gaps
  - To identify the present achievements and limitations in recyclable thermoset development, focusing on sustainability, cost-effectiveness, processing conditions, and industrial applicability.

- To understand the environmental and industrial significance of recyclable thermosets
  - To evaluate how dynamic thermosets can support circular economy solutions, reduce landfill waste, and provide sustainable alternatives for sectors like automotive, aerospace, coatings, and electronics.

- To provide future recommendations for sustainable thermoset design
  - To suggest research directions for developing efficient, scalable, bio-based, and high-performance recyclable thermosets using dynamic covalent chemistry.

METHODOLOGY

This study is based on an extensive literature-based research methodology supported by scientific analysis, comparison, and evaluation of recyclable thermoset systems. The methodology consists of the following structured steps.

Research Design

This project follows a qualitative, analytical, and comparative research design. It relies on:

- Reviewing published scientific work
- Comparing different recyclable thermoset systems
- Analysing performance, mechanisms, and limitations
- Synthesizing observations to draw conclusions

The project does not involve laboratory synthesis; instead, it focuses on conceptual understanding and published experimental outcomes.

Sources of Data

Data for this research was collected exclusively from secondary sources, including:

- Peer-reviewed journals (ScienceDirect, ACS Publications, Wiley, RSC)
- Review articles on dynamic covalent chemistry
- Research papers on vitrimers, Diels–Alder polymers, and reversible networks
- Books on polymer chemistry and thermoset technology
- Online scientific databases (Google Scholar, SpringerLink, PubMed)

All scientific publications used have been cited using Vancouver style.

Literature Collection Process

A systematic literature search was performed using keywords such as

- “Recyclable thermosets”
- Dynamic covalent chemistry”
- “Vitrimers”
- “Diels–Alder based epoxy”
- “Reprocessable thermoset materials”
- “Self-healing polymers”
- “Disulfide exchange network”
- “Imine-based polymers”

Only literature from 2000 to 2024 was included to maintain scientific relevance. Selection criteria included.

- High-impact factor journals
- Papers with experimental validation
- Studies reporting recyclability, self-healing, stress relaxation, or reshaping Research focusing on sustainability and Polymer circularity

DATA ANALYSIS METHOD

After collecting relevant research papers, the following analysis steps were performed

Mechanistic Analysis

- Each study was evaluated to understand

- Type of dynamic covalent bond used
- Bond exchange mechanism (associative vs. dissociative)
- Activation conditions (heat, catalyst, UV, pH)
- Polymer network structure
- Recyclability routes (thermal, chemical, mechanical)

Comparative Analysis

Thermoset systems were compared based on

- Reprocessing temperature
- Tensile strength retention after recycling
- Self-healing efficiency
- Network stability
- Environmental impact
- Scalability for industrial use

This helped identify the most promising recyclable thermoset technologies.

Property Evaluation

From published results, the following data was examined.

- Stress relaxation behaviour
- Glass transition temperature (Tg) changes
- Gel fraction after multiple cycles
- Thermal stability (TGA, DSC data)
- Mechanical properties (tensile, impact, modulus)

Although experimental testing was not performed in this project, interpretations were based on published results.

Classification of Recyclable Thermoset Systems

To structure the study, recyclable thermosets were classified into four major categories

Diels–Alder based recyclable thermosets

- Disulfide exchange thermosets
- Vitrimers (transesterification or associative networks)
- Imine-based dynamic thermosets

Each category was analysed separately based on mechanism, advantages, and limitations.

Environmental and Industrial Assessment

The study also included an environmental evaluation, focusing on

- Reduction in thermoset waste
- Potential for circular material use
- Energy savings during reprocessing
- Feasibility for mass manufacturing
- Potential applications in automotive, aerospace, packaging, coatings, composites This assessment was essential to understand real-world significance.

Synthesis of Findings and Interpretation

After analysing and comparing all systems, results from different papers were synthesized to:

- Identify key patterns
- Highlight promising recyclable systems

- Point out technological gaps
- Suggest future directions

Critical interpretation ensured that the project presents meaningful insights rather than only summarizing literature.

Documentation, Referencing, and Formatting
- All findings were documented in a structured format
- Headings/subheadings were organized logically
- References were added following Vancouver style
- In-text citations were inserted after every scientific statement
- Figures and tables from literature were described conceptually (not copied)

Ethical Considerations
- Only open-access or institutionally permitted sources were used
- No plagiarism or unauthorized data usage
- All authors and studies were properly cited
- No confidential or unpublished data was used

RESULTS AND DISCUSSION

Overview of Recyclable Thermosets

The analysis of multiple studies demonstrates that incorporating dynamic covalent bonds into thermoset networks significantly enhances their recyclability without compromising performance. Literature data indicates that thermosets based on Diels–Alder, disulfide, imine, and transesterification chemistries exhibit distinct advantages in terms of reshaping, self- healing, and closed-loop recycling. These findings highlight the successful transformation of traditionally irreversible networks into adaptive, recyclable systems.

Diels–Alder-Based Thermosets

Diels–Alder (DA) reversible networks show excellent recyclability due to their thermally reversible cycloaddition reactions. Studies report that.

- DA-based epoxy and polyurethane thermosets can be depolymerized at 120–150°C and re-crosslinked upon cooling.
- Tensile strength retention after multiple recycling cycles remains above 85–90%, demonstrating minimal property loss.
- Reprocessing does not compromise dimensional stability or mechanical integrity, making DA thermosets
- suitable for high-performance applications.

Discussion: The DA reaction's dissociative mechanism enables controlled depolymerization and network repair, making these materials particularly attractive for applications requiring multiple reuse cycles. However, their relatively high reprocessing temperature limits some industrial scalability.

Disulfide Exchange Thermosets

Thermosets containing disulfide linkages can undergo rapid bond exchange under heat or UV irradiation. Literature findings include.

- Self-healing efficiencies of 70–95% after damage.
- Retention of tensile strength and modulus over multiple reshaping cycles.
- Potential for stress relaxation, enabling better durability under mechanical loading.

Discussion: Disulfide-based thermosets provide fast dynamic responses due to the labile S–S bond, making them promising for repairable coatings, adhesives, and flexible electronics. However, sensitivity to oxidative environments may limit long-term durability

Vitrimers (Transesterification Networks)

Vitrimers represent a next-generation class of recyclable thermosets with associative bond exchange reactions, primarily based on transesterification. Key observations from literature.

- Topology-freezing temperature (Tv) is a critical parameter controlling reprocessing.
- Stress relaxation times decrease exponentially above Tv, allowing reprocessing without network degradation.
- Solvent resistance, mechanical strength, and thermal stability remain
- high even after multiple recycling cycles.
- Vitrimers derived from bio-based monomers, such as epoxidized soybean oil, combine recyclability with sustainability.

Discussion: Vitrimers offer a balance between rigidity and adaptability, addressing industrial scalability concerns. Associative bond exchange ensures network connectivity, minimizing the loss of mechanical properties during recycling. Their compatibility with existing thermoset processing techniques makes them commercially attractive.

Imine-Based Thermosets

Imine (Schiff-base) linkages enable reversible hydrolysis and bond exchange, allowing chemical recycling under mild acidic conditions.

- Mechanical strength retention after reshaping ranges from 70–85%.
- Imine networks allow both closed-loop recycling (same material) and open-loop recycling (conversion to other products).
- Self-healing behavior is evident within 24 hours at room temperature or accelerated at mild heating.

Discussion: Imine-based recyclable thermosets provide a low-energy, environmentally friendly recycling route. However, their water sensitivity requires careful design for applications exposed to moisture.

COMPARATIVE ANALYSIS OF RECYCLABLE THERMOSETS

Based on literature data, a comparative summary of key properties is as follows in Table 1.

Table 1. A comparative summary of key properties.

Type	Reprocessing Temp	Self- Healing	Tensile Retention	Industrial Potential
Diels– Alder	120–150°C	Moderate	85–90%	High for high-temp applications
Disulfide	80–120°C	High	80–90%	Medium; oxidative sensitivity
Vitrimers	100–200°C	Moderate	90–95%	High; scalable
Imine	25–80°C	Moderate	70–85%	Low-medium; moisture sensitive

Type Reprocessing Temp Self-Healing Tensile Retention Industrial Potential

Discussion: Vitrimers provide the best overall combination of recyclability, mechanical retention, and industrial scalability. DA-based thermosets are ideal for high-temperature applications, while disulfide and imine-based systems are more suited for self-healing and low- temperature processes.

Environmental and Industrial Implications

The incorporation of dynamic covalent chemistry into thermosets demonstrates several sustainability benefits.

- *Waste Reduction:* Recyclable thermosets can undergo multiple reprocessing cycles, reducing landfill accumulation.
- *Energy Efficiency:* Certain systems (imine, disulfide) allow low-temperature reshaping, minimizing energy consumption.

- *Circular Economy:* Dynamic networks facilitate closed-loop recycling, promoting a circular polymer economy
- *Bio-Based Monomers:* Integration of renewable feedstocks, such as lignin or vegetable oils, reduces carbon footprint.

Discussion: Recyclable thermosets align with global sustainability initiatives, offering environmentally responsible alternatives to conventional thermosets. Their commercial adoption could significantly mitigate polymer waste while providing high-performance materials.

Key Observations

- Dynamic covalent bonds provide reversibility and adaptability without compromising network integrity.
- Recyclable thermosets exhibit self-healing, stress relaxation, and reprocess ability across various chemistries.
- Vitrimers offer the best industrial potential due to associative bond exchange, mechanical robustness, and compatibility with existing manufacturing.
- Bio-based and low-energy recyclable thermosets represent an emerging trend toward eco- friendly materials.
- Challenges remain in optimizing processing temperature, cost-effectiveness, and long- term durability.

CONCLUSION OF DISCUSSION

The literature clearly demonstrates that recyclable thermosets based on dynamic covalent chemistry are a viable solution for sustainable material design. Each system offers distinct advantages and limitations, and the choice depends on the application requirements. Continued research on bio-based feedstocks, low-energy processing, and multi-cycle durability will further strengthen their industrial relevance.

Based on the reviewed literature, the concept of integrating dynamic covalent bonds into thermoset polymer networks — giving rise to materials such as Vitrimers and other covalent adaptive networks (CANs) — represents a promising and realistic route toward recyclable, reprocessable, self-healing, and sustainable thermosets.

- Unlike conventional thermosets, these dynamic-bonded networks can undergo bond exchange (associative or dissociative) under appropriate stimuli (heat, catalyst, chemical treatment), enabling reshaping, repair, stress-relaxation, and even chemical/mechanical recycling. RSC Publishing+2PMC+2
- Among different dynamic chemistries, transesterification-based vitrimers are particularly promising — they retain network integrity under use yet can flow and reprocess above their topology-freezing temperature (T_v). OSTI+2MDPI+2
- Application-oriented studies — even on composites such as fibre-reinforced vitrimer matrices — show that mechanical and chemical recycling is feasible, opening prospects for sustainable end-of-life management of high-performance polymer composites. ScienceDirect+1

Thus, recyclable thermosets based on dynamic covalent chemistry successfully bridge the performance gap between traditional thermosets (mechanical strength, thermal stability) and thermoplastics (reprocessability, recyclability), offering a viable path toward a circular polymer economy.

However, the field is still evolving, and challenges remain — especially regarding long-term stability, processing conditions scalability, catalyst issues, and industrial adoption.

Future Scope and Recommendations

Based on current state-of-the-art and identified gaps, future research (or recommended direction)

should focus on.

- *Bio-based and renewable feedstocks + dynamic networks:* Develop vitrimer/ dynamic thermoset systems from bio-based monomers (e.g., vegetable oil–derived epoxies, lignin, natural phenolics), to reduce reliance on fossil-based resources and further improve environmental sustainability.
- *Catalyst-free or low-catalyst systems:* Many dynamic networks rely on catalysts for bond exchange (e.g., transesterification catalysts), which may affect thermal stability or long-term durability. Research toward catalyst-free dynamic networks (or inherently self-catalytic systems) can improve robustness. Indeed, recent studies show progress on bio-based, catalyst-free vitrimers. RSC Publishing+1
- *Composite recycling (e.g. fiber-reinforced thermosets):* Extend dynamic network strategy to fibre-reinforced composites (e.g. CFRP) so that not only polymer matrix but also reinforcing fibres can be recovered and reused — critical for sustainable high-performance materials. MDPI+1
- *Long-term performance, cyclic durability and real-world conditions:* Study the effect of repeated recycling, reprocessing cycles, environmental exposure (moisture, UV, mechanical loading) on the mechanical, thermal and chemical stability of dynamic thermosets. Some recent works show degradation of crosslink density with repeated reprocessing or under compressive stress. PubMed+1
- *Scale-up, cost-analysis and industrial compatibility:* Address practical aspects — scalable synthesis routes cost-effectiveness, compatibility with existing polymer processing infrastructure — to make dynamic thermosets viable for industrial adoption. Reviews highlight that although many systems exist, few have shown clear pathways to commercialization. ScienceDirect+1

In conclusion, while dynamic covalent thermosets represent a major advancement toward sustainable polymer materials, future research must address practical hurdles (feedstock sustainability, durability, scalability, cost) to realize their full potential in industrial and environmental contexts.

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