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Green Bonds: Advancing Eco-Friendly Hot-Melt Pressure-Sensitive Adhesives through Functional Polymer Design

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Abstract

Hot-melt pressure-sensitive adhesives (HMPSAs) represent a solvent-free adhesive technology that integrates the rapid solidification of hot-melt adhesives with the viscoelastic tack and removability of pressure-sensitive adhesives. While conventional HMPSAs offer advantages such as high-speed processability and low volatile organic compound emissions, growing regulatory and environmental pressures necessitate the development of sustainable and eco-friendly alternatives. This review critically examines polymer design strategies for HMPSAs with emphasis on bio-based polymers, renewable tackifiers, viscoelastic optimization, and recyclability-oriented formulation approaches. Quantitative structure–property relationships involving glass transition temperature, melt viscosity, peel and shear strength, and rheological moduli are discussed to highlight performance–sustainability trade-offs. Advanced characterization techniques, including probe tack testing, shear adhesion failure temperature analysis, time–temperature superposition, and atomic force microscopy, are reviewed to elucidate interfacial and bulk adhesion mechanisms. End-of-life challenges such as adhesive residue during paper and plastic recycling are addressed, alongside emerging solutions including clean-removal and debond-on-demand HMPSAs. The review provides a comprehensive framework for designing next-generation sustainable HMPSAs that balance performance, processability, and environmental responsibility.

Keywords:

Hot-melt pressure-sensitive adhesives (HMPSAs); Thermoplastic adhesives; Pressure-sensitive adhesion; Rheological characterization; Viscoelastic behavior; Polymer formulation; Tackifiers and plasticizers; Bio-based polymers

1. Hot-Melt Adhesives (HMA)

Hot-melt adhesives (HMAs) are thermoplastic adhesive systems applied in the molten state and solidified upon cooling to form cohesive bonds. Unlike solvent-borne or water-based adhesives, HMAs are 100% solids and rely solely on physical solidification rather than chemical curing or solvent evaporation.[1] This solvent-free nature enables rapid bonding, high line speeds, and reduced environmental emissions, making HMAs widely used in packaging, woodworking, hygiene products, and automotive interiors.[2]

Despite these advantages, conventional HMAs exhibit intrinsic limitations that restrict their performance in demanding applications. Their thermoplastic nature leads to thermal sensitivity, creep under load at elevated temperatures, and limited resistance to prolonged heat exposure.[3] In addition, non-polar polymer backbones commonly used in HMAs often result in poor adhesion to low-surface-energy or polar substrates without chemical modification or surface treatment. [4-6]

From a mechanistic standpoint, adhesion in HMAs is governed by melt flow, substrate wetting, and mechanical interlocking during cooling. The balance between melt viscosity and solid-state cohesion is therefore critical, as excessive viscosity limits wetting while low viscosity compromises cohesive strength. [5,8-9] These limitations have driven the evolution of HMPSAs, which incorporate pressure-

sensitive viscoelastic behavior into hot-melt systems to achieve immediate tack at ambient conditions while retaining solvent-free processing.

2. Pressure-Sensitive Adhesives (PSA)

Table 1 presents the key characteristics of common PSA base polymers

Polymer System	Representative Monomers / Composition	Key Properties & Advantages	Limitations	Typical Applications	References
Acrylic PSAs	2-Ethylhexyl acrylate (2-EHA), butyl acrylate (BA), methyl acrylate (MA), copolymerized with polar monomers such as acrylic acid (AA), methacrylic acid (MAA), or hydroxyethyl acrylate (HEA)	Excellent optical transparency and color stability. High UV and oxidation resistance. Adjustable tack and peel strength via monomer ratio Good aging resistance and weatherability	Moderate heat resistance (typically <120 °C). Relatively low cohesive strength compared to rubber PSAs	Labels, protective films, medical tapes, and optical electronics	[14]
Rubber-based PSAs	Natural rubber (cis-1,4-polyisoprene), styrene–isoprene–styrene (SIS), styrene–butadiene–styrene (SBS), butyl rubber (IIR), or ethylene–propylene rubber (EPR)	High initial tack and peel adhesion Excellent low-temperature flexibility Compatible with a wide range of tackifiers and plasticizers Economical and easy to process	Poor UV and oxidation resistance. Limited thermal stability (softening above 80–100 °C). Susceptible to aging and yellowing	Packaging tapes, masking tapes, footwear, industrial labels	[15]
Silicone PSAs	Polydimethylsiloxane (PDMS) elastomers and resins crosslinked via Si–O–Si backbone	Outstanding chemical inertness and thermal stability (service up to 250 °C). Excellent release properties and low surface energy adhesion. Stable under humidity, radiation, and oxidation. Maintains tack over a broad	High cost of raw materials. Limited cohesive strength unless reinforced. Requires specialized curing and surface treatment	Aerospace tapes, medical dressings, electronic masking, high-temperature labels	[16]

		temperature range			
Polyurethane PSAs	Linear or lightly crosslinked polyether or polyester-based urethanes, synthesized via diisocyanate reactions	Tailorable viscoelastic spectrum via soft/hard segment ratio. High elastic recovery and creep resistance. Excellent mechanical strength and transparency. Potential for bio-based synthesis	Sensitive to moisture and hydrolysis (for ester-based types). Processing requires controlled curing	Flexible electronics, biomedical adhesives, specialty labels	[17]
Polyolefin PSAs	Amorphous poly-α-olefins (APAO), ethylene–propylene copolymers, functionalized with polar groups (e.g., maleic anhydride, hydroxyl, or epoxy)	Good thermal stability and oxidation resistance. Excellent compatibility with polyolefin substrates. Customizable tack–shear balance through copolymer design. Recyclable and low-VOC	Poor adhesion to polar substrates unless modified. Narrow tack temperature range.	Packaging laminates, automotive labels, assembly bonding	[17]

2.1 Overview and Mechanistic Fundamentals

Pressure-sensitive adhesives (PSAs) are viscoelastic polymer systems capable of forming immediate bonds under light pressure without external activation such as heat or solvents. Their performance is governed by a delicate balance between adhesion (surface wetting and interfacial interactions) and cohesion (internal resistance to deformation).[10]

From a rheological perspective, effective PSA performance requires the elastic modulus (G′) to remain below approximately 10^6 Pa at room temperature, as described by the Dahlquist criterion. [11-12] Typical PSAs exhibit glass transition temperatures between −60 and −20 °C, ensuring sufficient chain mobility for wetting while maintaining elastic recovery during debonding. Peel strengths generally range from 2 to 10 N/25 mm, depending on polymer architecture, tackifier compatibility, and crosslink density.[13]

Dynamic mechanical analysis (DMA) is widely used to define the “tack window” of PSAs, where the ratio of loss modulus (G″) to storage modulus (G′) enables energy dissipation during debonding without cohesive failure. However, conventional PSAs often rely on solvent-based or emulsion-based processing routes, which introduce environmental burdens related to solvent recovery, drying energy,

and emissions. These challenges motivate the integration of PSA-like viscoelasticity into hot-melt systems, giving rise to HMPSAs. [14-18]

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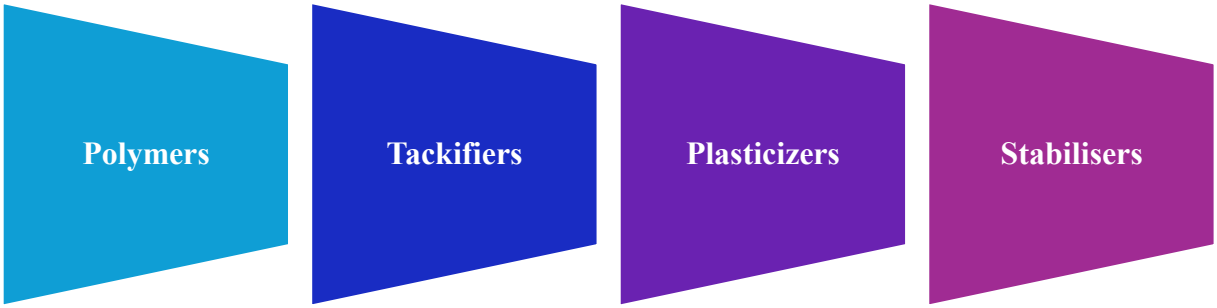
2.2 Chemical Composition and Formulation Strategy

PSA formulations are typically multiphase systems composed of polymers, tackifiers, plasticizers, and stabilizers in precise ratios [13].



2.2.1 Polymer Backbone

2.2.2 Tackifiers and Plasticizers

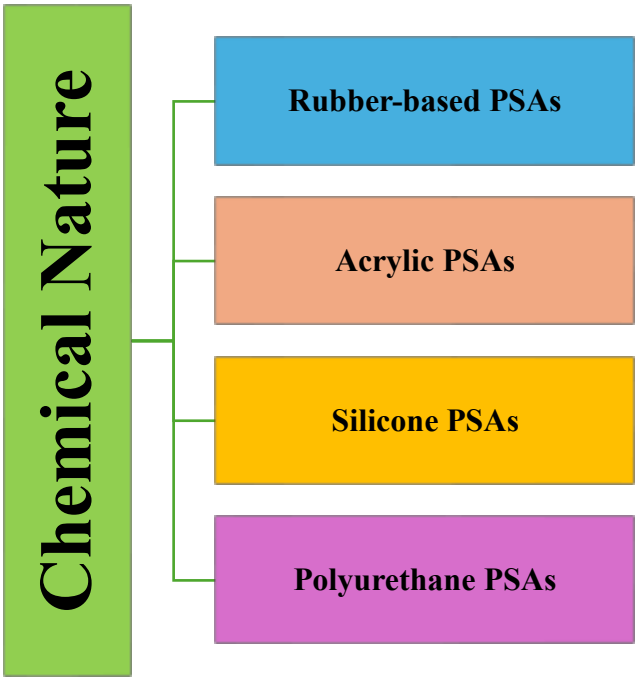
Low-molecular-weight resins called tackers are added to adhesive formulations to change the base polymer's viscoelastic characteristics. Hydrocarbon resins, terpene-phenolic resins, and rosin esters are typical examples. Tackifiers improve initial tack and peel performance by modifying the glass-transition temperature (Tg) and increasing surface wetness through interactions with the polymer matrix [18-22]. Because they affect cohesive strength, adhesive softness, and long-term stability, their choice and concentration are crucial.

Another significant family of modifiers are plasticizers, which are employed to improve chain mobility and soften the polymer matrix, hence promoting immediate adhesion under low pressure. Phthalates, adipates, and bio-based oils are examples of common plasticizers that improve conformability to substrate imperfections while also lowering the adhesive's modulus. Plasticizers allow the balance between adhesion and cohesion to be fine-tuned when used in conjunction with tackifiers, guaranteeing that the adhesive has enough tack without sacrificing mechanical integrity.

2.2.3 Crosslinking and Additives

Adhesives frequently use crosslinking techniques to improve dimensional stability and long-term cohesive strength. UV irradiation, metal chelates, or multifunctional monomers that can create a three-dimensional network inside the polymer matrix can all be used to accomplish crosslinking [19]. The integrity of the adhesive bond is preserved under prolonged mechanical stress or high temperatures because to this network's improved resistance to creep, flow, and thermal softening. To guarantee processing stability, product consistency, and aesthetic appeal, formulation additives such antioxidants, fillers, and pigments are included in addition to crosslinking agents. Pigments offer color consistency or opacity without sacrificing adhesive function, fillers can alter viscosity, hardness, or dimensional stability, and antioxidants guard against thermal and oxidative deterioration during production and use.

2.3 Classification of PSAs



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Functional Application:

- **Permanent PSAs:** Designed for long-term adhesion (e.g., tapes, labels, assembly films).
- **Removable PSAs:** Engineered for clean peel and residue-free removal (e.g., protective films, repositionable notes).
- **Specialty PSAs:** Include optically clear PSAs for displays, conductive PSAs, and biomedical PSAs for skin contact [20].

2.4 Fundamental Adhesion Mechanisms

Pressure-sensitive adhesives' (PSAs') adhesion process can be viewed as a series of connected actions that together dictate the strength and functionality of the bond:

1. **Wetting:**
   The adhesive flows to optimize contact with the substrate when light pressure is applied. The degree of wetting is significantly influenced by the adhesive's surface tension (γ), substrate roughness, and surface energy. Strong adhesive bonding requires intimate contact, which is accomplished by effective wetting [11].

2. **Interdiffusion:**
   The adhesive's polymer chains partially penetrate the substrate surface or suitable coating layers at the molecular level. Chain entanglement and interdiffusion improve mechanical interlocking and increase cohesive strength at the interface.[22-23]

3. **Bond Formation:**
   Secondary interactions such van der Waals forces, hydrogen bonds, or electrostatic attractions sustain the adhesive–substrate interface after wetting and interdiffusion. Certain chemical interactions, such as covalent or ionic bonding, may strengthen the adhesion even more in specialty systems.[24-26]

4. **Energy Dissipation:**
   The adhesive's viscoelastic properties control energy dissipation during debonding. The adhesive's capacity to absorb and release mechanical energy is reflected in its loss modulus (G), which is essential for tack performance, peel resistance, and preventing cohesive failure

[12]. A quantitative guideline for PSA performance is provided by the Dahlquist criterion, which states that the adhesive's elastic modulus (G′) must be less than ~10^6 Pa at the bonding temperature (~25 °C) in order to produce efficient wetting and adhesion [10]. Adhesives with a modulus higher than this are too stiff to adjust to surface imperfections, which leads to poor contact and a weaker bond.[27-30]

2.5 Evaluation Tests and Characterization

Key tests for PSA performance include: (Refer Table 2)

Property	**Standard / Method**	**What It Measures**
Peel adhesion	ASTM D3219	Force to remove adhesive from substrate
Tack (loop/rolling ball)	ASTM D2879	Instantaneous adhesion force
Shear strength	ASTM D3554	Cohesive strength under static load
Creep resistance	ISO 4587	Time-dependent deformation under stress
Thermal aging / UV stability	ASTM D573	Long-term durability and oxidation behaviours

Table 2: Property Testing and Standard Method

Advanced characterization includes **Dynamic Mechanical Analysis (DMA)** to capture viscoelastic spectra and **Atomic Force Microscopy (AFM)** to examine nanoscale interfacial adhesion [18-19;31-35].

2.6 Applications and Industry Relevance

Because of their quick adherence, simplicity of use, and adaptability, pressure-sensitive adhesives (PSAs) are used extensively in a variety of industrial and consumer sectors. Their uses take advantage of the special blend of conformability, peel strength, and tack that PSA compositions offer. Among the main application areas are:

Labelling and Packaging:
PSAs are widely utilized in flexible packaging laminates, pressure-sensitive tapes, labels, and carton sealing, where high throughput and quick bonding are crucial. The adhesives assist effective automated production lines by offering dependable adherence to a range of substrates, such as paper, cardboard, plastics, and metals.[31]

Medical & Healthcare:

Hypoallergenic and biocompatible PSAs are essential parts of transdermal medication administration systems, surgical drapes, wound dressings, and diagnostic equipment in the medical field [16]. These adhesives are essential for clinical applications and patient care because they provide painless removal and maintain a strong bond to skin or **medical** substrates. [32]
Automotive Applications:

PSAs are being used more and more in car interiors for sound absorption, carpeting, headliners, wiring harness tapes, and panel bonding. Their benefits, which support both performance and regulatory compliance, include low volatile organic compounds (VOCs), resistance to vibration and temperature cycling, and compatibility with a variety of interior substrates.[33]

Electronics and Displays:

Touch screens, flexible circuits, display laminations, and electronic device assembly all depend on sophisticated PSAs like optically clear adhesives (OCA). Under repeated use, these adhesives retain their mechanical performance and dimensional stability while offering high transparency, low haze, and consistent adherence. [34]

Intelligent and Responsive Systems:

New developments have produced PSAs that exhibit stimuli-responsive behaviours, allowing for the reversible modulation of adhesion by pH, temperature, light, or magnetic fields [20]. The increasing interest in functional materials and cutting-edge adhesion technologies is reflected in the use of these adhesives in smart packaging, reusable labels, and adaptive devices. [35]

2.7 Research Trends and Challenges

Sustainability, enhanced functionality, and process efficiency are driving more and more PSA research today, meeting both technological and environmental concerns. Among the primary areas of concentration are:

Bio-based PSAs:

There are initiatives to switch from petroleum-based resins to renewable feedstocks such lignin, rosin, or terpene derivatives [18]. These bio-based substitutes minimize environmental effect while reducing dependency on fossil fuels and providing customization options for adhesion, tack, and viscoelastic characteristics.

Recyclable PSAs:

Formulations that enable clean delamination are becoming more and more popular because they make it easier to recycle packaging films and laminated materials. In order to promote circular economy goals, these adhesives are made to maintain enough performance during usage while permitting controlled debonding under particular circumstances.[36]

Stimuli-Responsive PSAs:

In order to demonstrate reversible adhesion in response to external stimuli like temperature, light, magnetic fields, or pH changes, smart or adaptive PSAs are now being developed [20]. The functional range of conventional PSAs could be increased by using these systems in dynamic assembly procedures, adaptive medical devices, and reusable labels.[37]

HMPSA Hybrids:

Recent research combines the fast tack of PSAs with the processability of HMAs to create solvent-free, energy-efficient coatings by integrating pressure-sensitive adhesive behaviours into hot-melt systems. Because they shorten drying periods and lower volatile emissions, these hybrid systems hold great promise for high-speed industrial coating and lamination operations (more on this in Section 3).[38]

3. Hot-Melt Pressure-Sensitive Adhesives (HMPSAs) and Their Applications

3.1 Definition and Unique Features of HMPSAs

Hot-melt pressure-sensitive adhesives (HMPSAs) combine the solvent-free processing of HMAs with the viscoelastic tack and removability of PSAs. Unlike conventional HMAs, which solidify into rigid thermoplastic joints, HMPSAs remain tacky at room temperature and can form bonds under light pressure after cooling. In contrast to traditional PSAs, HMPSAs are applied in the molten state, eliminating solvent evaporation and reducing processing time and emissions.[39]

Chemically, HMPSAs are designed to achieve a controlled viscoelastic balance through polymer architecture, molecular weight distribution, and tackifier compatibility. Typical HMPSAs exhibit melt viscosities between 2,000 and 50,000 cP at application temperatures of 120–160 °C, glass transition temperatures between −50 and −10 °C, and peel strengths comparable to solvent-based PSAs.[40]

3.2 Polymer Systems and Formulation Strategies

The performance of HMPSAs is strongly influenced by polymer molecular weight distribution and crosslink density. Narrow molecular weight distributions promote predictable rheology and coating behavior, while broader distributions enhance energy dissipation and peel strength. Light crosslinking improves shear resistance and creep performance but must be carefully controlled to avoid loss of tack or processability.

Tackifier compatibility plays a critical role in defining the viscoelastic window. Poor compatibility can lead to phase separation, migration, and long-term performance degradation. Sustainable HMPSA design therefore requires not only renewable components but also optimized polymer–tackifier interactions to maintain tack–peel–shear balance.

3.3 Processing Considerations

To address limitations of conventional peel and shear testing, advanced characterization methods are increasingly employed:

- **Probe tack testing** provides direct measurement of instantaneous adhesion and debonding energy.
- **Shear Adhesion Failure Temperature (SAFT)** evaluates thermal creep resistance under load.
- **Time–Temperature Superposition (TTS)** generates master curves describing viscoelastic behavior over extended time and temperature ranges.
- **DMA-based tack windows** correlate G′ and G″ with adhesive performance.
- **Atomic Force Microscopy (AFM) adhesion mapping** reveals nanoscale interfacial heterogeneity and filler dispersion.

These techniques enable quantitative correlation between molecular design, viscoelastic behavior, and adhesive performance.

4. Need for Sustainable Chemistries in HMPSAs

4.1 Sustainability Challenges and Design Strategies in HMPSAs

Although HMPSAs are inherently solvent-free, sustainability challenges persist due to reliance on petrochemical polymers, energy-intensive melt processing, and poor end-of-life compatibility. Residual HMPSAs on paper labels and plastic films can contaminate recycling streams, reducing fiber quality and plastic recyclate purity.

Recent research focuses on renewable polymer backbones, natural tackifiers (rosin esters, terpenes, lignin derivatives), and bio-based plasticizers. However, these materials introduce trade-offs, including reduced thermal stability, oxidative sensitivity, and altered viscoelastic behavior. Lignin-based components, for example, improve renewable content but may increase brittleness unless compatibilized, while terpene tackifiers offer good tack but limited high-temperature resistance.

Sustainable HMPSA design therefore requires holistic optimization of performance, processability, and recyclability rather than simple substitution of raw materials.

4.2 Sustainability Challenges in HMPSAs

Although hot-melt pressure-sensitive adhesives (HMPSAs) are inherently solvent-free systems, offering significant advantages over solvent- or water-based formulations in terms of VOC elimination and processing efficiency, they continue to face several critical sustainability challenges across their material sourcing, manufacturing, and end-of-life stages.

1. **Dependence on Non-Renewable Monomers:**

The majority of commercial HMPSAs are derived from petrochemical-based polymers such as acrylics, styrene block copolymers (SBCs), and polyolefins. These monomers—commonly including 2-ethylhexyl acrylate, styrene, isoprene, and ethylene—are sourced from fossil hydrocarbons, making the adhesive lifecycle highly carbon-intensive and resource-dependent. Transitioning toward bio-based analogues or partially renewable monomers is therefore essential for reducing environmental impact without compromising adhesive performance.

2. **Energy-Intensive Processing:**

HMPSAs are processed primarily through melt extrusion and hot-coating operations, which typically require elevated temperatures (120–180 °C) to achieve the necessary flow and coating uniformity. These energy-intensive conditions contribute significantly to the overall carbon footprint of the adhesive manufacturing process. Advanced polymer architectures—such as low-melting thermoplastic elastomers or reactive macromonomers—offer promising pathways to reduce melt temperatures while maintaining coating quality and mechanical integrity.

3. **End-of-Life Limitations:**

Despite their operational efficiency, most HMPSAs are non-biodegradable and difficult to separate from substrates such as paper, films, or textiles. This limits recyclability and complicates post-consumer waste management, especially in multi-layer packaging and label applications. Furthermore, the presence of residual adhesive residues can compromise recycling streams for paper and plastics. Emerging strategies, including designing depolymerizable or compostable HMPSAs and developing removable or reworkable adhesive systems, are being explored to enhance circularity.

Addressing these challenges demands a holistic approach that integrates:

- Renewable material innovation (bio-based monomers, natural tackifiers, and degradable backbones);
- Process optimization for reduced energy demand and improved melt rheology; and
- Lifecycle management frameworks encompassing recyclability, reusability, and biodegradability [22,29].

Through these combined strategies, the HMPSA industry can move toward sustainable adhesive technologies that balance performance, energy efficiency, and environmental responsibility, aligning with modern regulatory and circular economy objectives.

4.3 Role of Renewable Components

Strategies for Enhancing the Sustainability of HMPSAs

To mitigate the environmental footprint of hot-melt pressure-sensitive adhesives (HMPSAs), recent research has shifted toward the development of bio-based and renewable formulations that reduce fossil dependence while maintaining performance parity with conventional systems. These innovations primarily target the polymer backbone, tackifier system, and plasticizer components, which collectively determine the adhesive's mechanical, rheological, and interfacial properties.

1. Bio-Based Polymers:

The replacement of petroleum-derived monomers with renewable macromonomers has emerged as a key strategy for sustainable HMPSA design. Polymers synthesized from lactic acid, ε-caprolactone, itaconic acid, and sugar-derived monomers provide tunable flexibility and polarity, improving compatibility with conventional acrylic networks.
For instance, Gu et al. demonstrated that partial substitution of 2-ethylhexyl acrylate with L-lactide and ε-caprolactone-based macromonomers significantly increased the biomass content of acrylic HMPSAs while maintaining peel strength and processability suitable for high-speed coating [22,22]. Such systems achieve a balanced combination of renewable content, thermal stability, and adhesive performance, positioning them as promising candidates for next-generation sustainable adhesives.

2. Natural Tackifiers:

Traditional hydrocarbon-based tackifiers, such as C5/C9 resins, are being progressively replaced by plant-derived resins—notably terpenes, rosin esters, and gum resins—which are renewable, biodegradable, and exhibit strong compatibility with both acrylic and elastomeric matrices [28]. These natural tackifiers can enhance initial tack, adhesion to polar substrates, and oxidative stability, while also minimizing VOC emissions and toxic residues. Moreover, their diverse chemical structures allow fine-tuning of softening points and viscoelastic properties, offering a versatile pathway to sustainable formulation design.

3. Renewable Plasticizers:

To replace conventional phthalates and mineral oil-based plasticizers, researchers are increasingly incorporating citrate esters, epoxidized vegetable oils, and fatty acid derivatives derived from renewable sources. These bio-plasticizers improve flexibility and low-temperature adhesion without compromising cohesive strength. Additionally, their low volatility and non-toxicity align with modern regulatory standards and product safety requirements, making them attractive for medical, packaging, and consumer applications.

Together, these strategies allow HMPSAs to retain mechanical integrity, adhesion reliability, and process efficiency, while substantially improving their environmental performance. By integrating renewable feedstocks, green chemistry approaches, and circular economy principles, researchers are paving the way for eco-efficient HMPSAs that support both industrial scalability and regulatory compliance [22,28,29].

4.4 Design Considerations for Sustainable HMPSAs

Developing sustainable hot-melt pressure-sensitive adhesives (HMPSAs) requires a careful balance between adhesive performance, processing feasibility, and environmental impact. While renewable materials and green additives offer clear ecological benefits, their incorporation can significantly influence the adhesive's molecular structure, rheology, and end-use properties. Therefore, a holistic formulation strategy must consider the following critical aspects:

1. Adhesion–Cohesion Balance:

Renewable polymers, tackifiers, and plasticizers often differ in polarity, molecular weight, and compatibility compared to their petrochemical counterparts. These differences can alter the viscoelastic window that governs tack, peel, and shear performance. Maintaining an optimal balance between adhesion and cohesion is essential to ensure high peel strength (surface wetting and bonding) and shear resistance (internal integrity) [22,22]. Advanced formulation tools, such as dynamic mechanical analysis (DMA) and time–temperature superposition, are increasingly used to fine-tune this balance in bio-based HMPSA systems.

2. Processability and Thermal Stability:

Sustainable HMPSAs must remain processable within conventional industrial settings, including extrusion, slot-die coating, and hot-melt application lines. This requires controlling melt viscosity,

softening temperature, and flow characteristics within the operational range of 120–170 °C. Bio-based monomers and additives should not introduce premature crosslinking, degradation, or phase separation during melting or coating. Thermal stabilizers and reactive compatibilizers can help preserve melt integrity and coating uniformity, ensuring scalability with minimal equipment modification [29].

3. Lifecycle Assessment (LCA) and End-of-Life Considerations:

Beyond formulation and processing, the environmental performance of sustainable HMPSAs must be quantified through Lifecycle Assessment (LCA) metrics—encompassing carbon footprint, resource use, toxicity potential, and end-of-life fate. Factors such as biodegradability, recyclability, and ease of adhesive removal directly influence circularity and sustainable product design. Integrating LCA-driven decision-making enables researchers and manufacturers to identify the most impactful substitutions and align HMPSA development with global sustainability frameworks and eco-labeling standards [22].

4.5 Industrial Relevance

Application Potential of Sustainable HMPSAs

The transition toward sustainable chemistries in hot-melt pressure-sensitive adhesives (HMPSAs) is being driven not only by environmental imperatives but also by sector-specific performance and regulatory demands. The integration of renewable raw materials, energy-efficient processing, and optimized viscoelastic design is expanding the application potential of bio-based HMPSAs across multiple high-value industries.

1. Packaging and Labeling Applications:

In the packaging sector, the demand for bio-based, recyclable, and compostable adhesives has risen sharply in response to corporate sustainability commitments and circular economy initiatives. Adhesives used in labels, tapes, and multilayer packaging must maintain strong adhesion to diverse substrates (paper, bioplastics, or films) while being compatible with recycling or repulping processes [22,28]. Renewable HMPSAs based on acrylic or polyester architectures offer promising solutions by combining high tack and peel strength with reduced carbon intensity and minimal VOC emissions.

2. Medical and Wearable Devices:

In biomedical and wearable applications, biocompatibility, non-toxicity, and skin adhesion performance are critical requirements. Conventional HMPSAs often contain residual monomers, plasticizers, or antioxidants that may raise irritation or sensitization concerns. The use of renewable and non-toxic building blocks, such as citrate-based plasticizers, lactic acid–derived polymers, and natural tackifiers, significantly reduces chemical exposure while preserving comfort, breathability, and reusability [29]. Such bio-based HMPSAs are increasingly employed in transdermal patches, wound dressings, and flexible electronic sensors, where soft adhesion and safe removability are vital.

3. Automotive and Electronics:

In the automotive and electronics sectors, adhesives play a crucial role in interior assembly, component mounting, and thermal management. These industries are under growing regulatory pressure to reduce VOC emissions, energy consumption, and carbon footprint [28]. Sustainable HMPSAs formulated with low-emission tackifiers and thermally stable bio-based polymers provide strong adhesion and heat resistance while contributing to cleaner production lines and lightweight design goals.

Collectively, these innovations demonstrate that sustainable HMPSAs are no longer limited to niche applications. Through the combined use of renewable feedstocks, energy-optimized manufacturing, and advanced material design, such systems are increasingly viable for both industrial-scale and consumer applications, offering a realistic pathway toward eco-efficient, high-performance adhesive technologies [22,29].

5. Sustainable HMPSAs

5.1 Definition and Scope

Sustainable HMPSAs are engineered to balance adhesive performance with environmental impact. Key strategies include:

- Partial replacement of petroleum-derived monomers with bio-based macromonomers (e.g., lactide or caprolactone-based acrylates)
- Use of renewable tackifiers with controlled softening points
- Design of clean-removal or debond-on-demand adhesives to facilitate recycling

Quantitatively, sustainable HMPSAs typically exhibit slightly higher Tg and reduced melt flow compared to conventional systems, requiring formulation adjustments to preserve tack and peel strength. Lifecycle assessment approaches are increasingly used to evaluate carbon footprint reduction, resource efficiency, and end-of-life performance.

5.2 Bio-Based Polymer Systems

5.2.1 Acrylic-Based HMPSAs

Acrylic polymers remain a dominant choice for hot-melt pressure-sensitive adhesives (HMPSAs) due to their inherent tack, peel strength, and viscoelastic balance. Recent research has focused on incorporating bio-based macromonomers, such as those derived from l-lactide, ε-caprolactone, or other renewable lactones, to increase biomass content while retaining the desirable adhesive properties of conventional acrylics.

For instance, Gu et al. (2014) demonstrated that partial replacement of 2-ethylhexyl acrylate with renewable macromonomers resulted in peel strength enhancements of 15–20% without compromising cohesive integrity or processing behavior [22,30]. This approach illustrates how sustainable macromonomers can be integrated into HMPSAs to achieve high-performance adhesives with reduced environmental impact, providing a pathway for bio-based, industrially viable formulations.

5.2.2 Polyolefin-Based HMPSAs

Metallocene polyolefins (mPOs) are widely used in HMPSAs due to their excellent thermal stability, processability, and mechanical robustness. However, their intrinsically non-polar nature limits adhesion to polar or high-surface-energy substrates. To overcome this limitation, mPOs can be partially modified with polar functional groups or grafted with bio-based monomers, enhancing interfacial interactions without significantly affecting thermal or mechanical properties [28,31]. Such modifications enable stronger adhesion to metals, fibers, and polar plastics, broadening the applicability of mPO-based HMPSAs while maintaining their high-speed processing and durability.

5.3 Renewable Tackifiers and Plasticizers

In addition to bio-based polymer backbones, the tackifier and plasticizer components of HMPSAs can be sustainably sourced to further reduce environmental impact while maintaining adhesive performance.

- Natural tackifiers: Plant-derived resins, including terpenes, rosin esters, and gum resins, are increasingly used as alternatives to petroleum-based hydrocarbon tackifiers [30]. These materials provide strong initial tack, good peel strength, and compatibility with both acrylic and polyolefin matrices, while contributing to renewable content.
- Bio-based plasticizers: Renewable plasticizers such as citrate esters, vegetable oil derivatives, and lactone-based esters improve flexibility, low-temperature performance, and melt processability without introducing toxic or volatile components [31].

The integration of these renewable additives allows HMPSAs to retain their viscoelastic properties, peel strength, and shear resistance while significantly enhancing the adhesive's bio-based fraction, making them more suitable for sustainable and circular applications.

5.4 Performance Optimization Strategies

Formulation Strategies for Sustainable HMPSAs

Designing sustainable hot-melt pressure-sensitive adhesives (HMPSAs) requires a careful balance between adhesion, cohesion, and processability to ensure performance parity with conventional adhesives while maximizing renewable content. Key formulation strategies include:

- Adhesion optimization: Incorporation of functional monomers or grafted bio-based macromonomers enhances peel strength and shear resistance, enabling durable bonding across diverse substrates while increasing the bio-based fraction of the adhesive [30].
- Thermal and rheological tuning: Melt viscosity, softening temperature, and flow behavior are adjusted through renewable plasticizers, molecular weight control, and polymer architecture. These adjustments ensure smooth high-speed coating, uniform film formation, and dimensional stability during application [31].
- Substrate compatibility: Effective adhesion to polar or challenging surfaces is achieved via surface-active renewable monomers, minor polar grafts, or compatibilizing additives. These modifications enhance interfacial wetting and bonding without compromising processability or the solvent-free nature of HMPSAs [28,31].

By integrating these strategies, formulators can produce eco-efficient HMPSAs that maintain viscoelastic performance, peel and shear strength, and industrial applicability, while substantially reducing environmental impact.

5.5 Case Studies

1. High-Biomass Acrylic HMPSAs:

Incorporation of l-lactide and ε-caprolactone macromonomers enables biomass contents up to 70%, while improving peel strength and maintaining shear resistance comparable to conventional acrylic HMPSAs. These systems combine renewable content with robust adhesive performance [22,30].

2. Metallocene Polyolefin HMPSAs:

Bio-based polyolefin backbones grafted with polar monomers (e.g., acrylic acid) enhance adhesion to polar substrates such as glass and metals. This modification expands eco-friendly packaging applications while preserving the thermal and mechanical stability inherent to polyolefins [28,31].

3. Bottlebrush Elastomer HMPSAs:

Additive-free architectures with renewable side chains naturally widen the tack window, reduce melt viscosity, and improve reworkability without compromising performance. These topologies simplify formulation and minimize potential compatibility or migration issues associated with conventional elastomeric HMPSAs [21,30].

5.6 Environmental and Industrial Impact

Sustainable HMPSAs provide significant environmental and functional benefits compared to conventional petroleum-based adhesives, making them increasingly attractive for next-generation industrial and consumer applications:

- Lower carbon footprint: Use of renewable polymers, tackifiers, and plasticizers reduces dependency on fossil resources and decreases overall CO_2 emissions during production [30].

- Reduced VOC emissions: Solvent-free or low-solvent processing minimizes volatile organic compound (VOC) release, improving workplace safety and environmental compliance [31].
- Enhanced recyclability and potential biodegradability: Formulations designed for easy removal, depolymerization, or compostability support circular economy initiatives in packaging and product life cycles.
- Regulatory compliance: Renewable and non-toxic components help meet stringent safety standards for biomedical, food-contact, and consumer products, reducing exposure to hazardous chemicals [30,31].

Together, these benefits make sustainable HMPSAs suitable for high-performance applications in packaging, biomedical devices, textiles, wearable electronics, and flexible assembly, combining eco-efficiency with reliable adhesive performance.

6. Futuristic Approaches in HMPSAs

6.1 Smart and Stimuli-Responsive HMPSAs

Recent developments in hot-melt pressure-sensitive adhesives (HMPSAs) focus on creating stimuli-responsive systems that adapt adhesion properties in response to external triggers, enabling reworkable, reversible, or remotely controlled bonding for advanced applications. Key approaches include:

- Thermo-responsive HMPSAs: Adhesion strength can be modulated by temperature, allowing controlled reworkability or reversible bonding. For instance, bottlebrush thermoplastic elastomers exhibit tunable tack and flow properties depending on applied heat, making them suitable for repositionable adhesives and high-speed coating processes [21,32].
- Moisture- or pH-responsive adhesives: These systems adjust adhesion in response to skin hydration, wound exudate, or pH changes, making them particularly useful in biomedical applications, such as transdermal drug delivery patches, wound dressings, and wearable sensors [33].
- Light- or electric-field-responsive HMPSAs: Adhesion can be remotely controlled through photo- or electro-responsive mechanisms, enabling dynamic attachment and detachment. Such systems have potential in flexible electronics, robotics, and wearable devices, where precise control of adhesion is critical [32].

By integrating adaptive functionality with sustainable or high-performance chemistries, these next-generation HMPSAs open new possibilities for smart adhesives that respond to environmental or user-defined stimuli while maintaining processability and mechanical reliability.

6.2 Additive-Free and Minimal-Additive Architectures

Reducing reliance on conventional tackifiers, plasticizers, and stabilizers is a key strategy to enhance both the sustainability and performance of HMPSAs.

- Bottlebrush polymer architectures inherently produce soft, tacky surfaces, minimizing the need for traditional additives while maintaining robust adhesive properties [21,32].
- These architectures also provide wide viscoelastic windows, improved cohesive strength, and lower melt viscosity, enabling energy-efficient processing, smooth coating, and uniform film formation.

By leveraging structural design rather than additive loading, bottlebrush-based HMPSAs combine eco-efficiency, processability, and high-performance adhesion, representing a promising pathway for next-generation sustainable adhesives.

6.3 Circular and Recyclable HMPSAs

Next-generation HMPSAs are being engineered to support circular material life cycles, reducing environmental impact and enabling sustainable end-of-life strategies.

- Reprocessable thermoplastic adhesives: These adhesives can be remelted and reused multiple times without significant loss of peel strength, tack, or shear resistance, facilitating material recovery and reuse in industrial processes [33].
- Bio-based and biodegradable backbones: Incorporating renewable monomers and biodegradable polymer architectures allows adhesives to be composted or safely disposed at the end of their service life, minimizing accumulation in landfills [30,33].
- Hybrid sustainable systems: Combining recyclable thermoplastic matrices with bio-based content achieves adhesive performance comparable to conventional HMPSAs while actively supporting circular economy objectives, including material recovery, reduced resource consumption, and minimized environmental footprint.

These strategies position HMPSAs as next-generation sustainable adhesives that integrate high-performance bonding with eco-efficient, circular design principles, aligning with global sustainability and regulatory goals.

6.4 Functional and Multifunctional HMPSAs

Emerging hot-melt pressure-sensitive adhesives (HMPSAs) are being designed to incorporate advanced functionalities beyond conventional adhesion, enabling multifunctional and high-performance applications:

- Electrical conductivity: By integrating conductive fillers or intrinsically conductive polymers, HMPSAs can form adhesive circuits, sensors, or flexible electronic interfaces, expanding their role in wearable electronics and soft robotics [32].
- Self-healing adhesives: Incorporation of dynamic covalent bonds or supramolecular interactions allows HMPSAs to recover adhesion after mechanical damage, extending service life and durability in demanding applications [33].
- Anti-fouling or antimicrobial properties: Functional additives or surface-active monomers impart hygiene and contamination control, which is critical for biomedical devices, wound dressings, and food-contact packaging [32].

These innovations illustrate how next-generation HMPSAs can combine sustainable formulation, stimuli-responsiveness, and multifunctionality, enabling smart adhesives that meet both performance and environmental requirements.

6.5 Advanced Processing Techniques

Futuristic HMPSAs increasingly leverage innovative processing technologies to enhance both performance and sustainability:

- 3D and 4D printing: Enables precise deposition of HMPSAs in customized shapes, patterns, and multilayered structures, expanding their applicability in electronics, biomedical devices, and smart packaging [32].
- Solvent-free melt extrusion with controlled cooling: Optimizes film uniformity, coating thickness, and adhesive microstructure while reducing energy consumption, aligning with eco-efficient manufacturing practices [31,33].

- In-line characterization and real-time rheology monitoring: Facilitates dynamic control of tack, flow, and setting behavior during production, minimizing trial-and-error, ensuring consistent product quality, and improving process efficiency [32].

These advanced processing approaches, when combined with sustainable chemistries and functional HMPSA architectures, enable high-performance, environmentally responsible adhesives suitable for industrial-scale and high-value applications.

6.6 Potential Industrial Impact

Next-generation HMPSAs are being deployed across a wide range of industrial and consumer applications, leveraging their sustainability, functionality, and stimuli-responsive properties:

- Electronics: Smart and conductive HMPSAs enable wearables, flexible circuits, and reconfigurable electronic devices, providing reliable adhesion while integrating additional electrical functionality [32].
- Biomedical applications: Responsive adhesives improve patient comfort, controlled drug delivery, wound care, and reduced skin irritation, making them suitable for transdermal patches, medical dressings, and wearable biosensors [33].
- Sustainable packaging: Circular and recyclable HMPSAs minimize waste and carbon footprint, supporting eco-friendly packaging solutions and helping companies meet global sustainability targets [32,33].
- Advanced textiles: Functional HMPSAs enable seamless bonding, reworkable fabric laminates, and self-healing garments, providing durability, comfort, and multifunctionality in smart textiles and wearable technologies [32,33].

These applications illustrate the broad impact of next-generation HMPSAs, combining performance, sustainability, and adaptability to meet evolving industrial and consumer demands.

Emerging HMPSAs incorporate stimuli-responsive behavior, including thermo-responsive, moisture-responsive, and light-activated adhesion. These systems enable reversible bonding, reworkability, and controlled debonding, particularly relevant for electronics, medical devices, and recyclable packaging.

Additive-free architectures such as bottlebrush polymers offer wide viscoelastic windows without conventional tackifiers, reducing migration risks and simplifying formulation. In parallel, circular design concepts aim to produce reprocessable or biodegradable HMPSAs compatible with closed-loop material systems.

7. Evaluation Tests and Quantitative Characterization of HMPSAs

Hot-melt pressure-sensitive adhesives (HMPSAs) are evaluated using a combination of adhesion, cohesion, and rheological tests to establish structure–property–performance relationships. Unlike conventional structural adhesives, HMPSAs require characterization methods that capture their time- and temperature-dependent viscoelastic behavior, which governs tack, peel strength, and shear resistance.[41]

7.1 Tack Measurement

Tack represents the instantaneous bonding ability of HMPSAs under light pressure and short contact time. Commonly employed methods include probe tack, loop tack, and rolling ball tack tests.[42] Reported probe tack forces for commercial HMPSAs typically range from 0.5 to 5.0 N, depending on polymer Tg, tackifier compatibility, and testing temperature. Increasing tackifier content generally enhances tack by lowering the effective glass transition temperature and increasing chain mobility, although excessive softening can reduce cohesive strength.

7.2 Peel Strength

Peel tests (90° or 180°) are used to quantify interfacial adhesion under steady debonding conditions. Peel strength values for HMPSAs typically fall within 2–10 N/25 mm, comparable to solvent-based PSAs. Peel performance is strongly influenced by molecular weight distribution, polymer–tackifier miscibility, and interfacial wetting behavior. Bio-based HMPSAs have been reported to achieve peel strengths within this range, demonstrating that sustainability improvements do not necessarily compromise adhesion performance.[43]

7.3 Static Shear and Creep Resistance

Static shear tests evaluate the ability of HMPSAs to resist deformation under constant load. Shear holding times for HMPSAs typically range from minutes to several hours, depending on crosslink density and elastic modulus. Light crosslinking improves creep resistance and shear stability but must be carefully controlled to avoid loss of tack or removability.[44]

7.4 Shear Adhesion Failure Temperature (SAFT)

SAFT testing provides a quantitative measure of thermal creep resistance by determining the temperature at which adhesive failure occurs under load. Commercial HMPSAs generally exhibit SAFT values between 70 and 150 °C, depending on polymer backbone and formulation. Higher SAFT values correlate with improved heat resistance but may be accompanied by reduced low-temperature tack.[45]

7.5 Dynamic Mechanical Analysis (DMA) in HMPSA Research

Although Dynamic Mechanical Analysis (DMA) is not routinely employed as an industrial quality-control test for HMPSAs, it is widely used in academic and formulation research to correlate **viscoelastic parameters with adhesive performance**. DMA provides access to storage modulus (G′), loss modulus (G″), and tan δ over a broad temperature and frequency range.[46]

Effective HMPSA performance is typically associated with:

- **Glass transition temperatures (Tg): −50 to −10 °C**
- **Storage modulus (G′) below ~10^6 Pa at room temperature**, enabling substrate wetting and compliance
- A broad **viscoelastic "tack window"**, where energy dissipation (G″) dominates during debonding

DMA-derived master curves generated via time–temperature superposition enable prediction of long-term creep, peel behavior, and thermal stability. Recent studies have demonstrated strong correlations between DMA parameters and peel strength, tack, and SAFT values, positioning DMA as a powerful **structure–property analysis tool** for HMPSA design rather than a routine industrial test.[48-50]

Conclusion:

The evolution of HMPSA technology reflects a convergence of thermoplastic processability and pressure-sensitive responsiveness, enabling high-performance bonding solutions across a spectrum of substrates and operating conditions. While HMAs deliver rapid solidification and recyclability, their thermal sensitivity and limited adhesion to non-polar surfaces necessitate formulation innovations. PSAs, with their viscoelastic tunability, offer clean peel and conformability but demand careful molecular design to balance tack, cohesion, and aging resistance. Advancements in bio-derived polymers, nanofiller integration, and functional group grafting are expanding the operational envelope of HMPSAs, aligning adhesive science with circular economy principles and high-demand engineering applications. Future research should prioritize hybrid architectures, predictive rheological modeling, and adaptive formulation strategies to overcome current limitations and unlock next-generation adhesive systems with enhanced durability, sustainability, and multifunctionality.

The development of sustainable HMPSAs represents a critical intersection of adhesive science, polymer engineering, and environmental responsibility. By integrating quantitative rheological analysis, advanced characterization techniques, and lifecycle-driven design, next-generation HMPSAs can achieve high performance while addressing recyclability, renewable content, and regulatory compliance. Future progress will depend on molecular-level control of viscoelastic behavior and system-level optimization across the adhesive life cycle.

Recent advances in HMPSA research increasingly emphasize quantitative structure–property relationships, renewable polymer architectures, and advanced viscoelastic characterization. Compared to earlier descriptive studies, modern HMPSA design relies on numerical correlations between Tg, rheological moduli, peel strength, tack, and shear resistance to optimize performance and sustainability simultaneously. The growing body of post-2020 literature reflects this shift toward data-driven adhesive design, supporting the development of eco-efficient HMPSAs compatible with industrial processing and circular economy objectives.

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