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Advancements in sustainable materials for environmentally responsible tyre production: a comprehensive review

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The tyre industry is at crossroads, grappling with the dual challenges of waste tyre disposal and the environmental impacts of production. In an era where sustainability is paramount, manufacturers are intensifying efforts to minimize raw material consumption, maximize renewable resource utilization, and innovate tyre recycling. This review critically examines these initiatives, revealing a promising shift towards sustainable practices. Key findings include the industry's adoption of bio-based polymers, fabrics, processing aids, and renewable fillers, which offer greener alternatives to traditional materials. Integrating recycled rubber, bio-based oils, and silica from agricultural waste highlights a significant move towards a circular economy. Advanced technologies such as AI, life cycle assessment, triboelectric tyres, and 3D printing are revolutionizing tyre design and production, further reducing environmental footprints. Despite these advancements, significant knowledge gaps remain concerning the long-term performance and sustainability of these materials. Research into self-healing rubber, vitrimers, and advanced recycling techniques underscores the industry's commitment to environmental stewardship. Nevertheless, the full impact of these innovations on environmental lifecycles remains unclear. This review emphasizes the critical need for further research to bridge these gaps, ensuring that the tyre industry can meet future demands sustainably.

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Sustainability spotlight

This review paper highlights how the use of bio-based polymers, textiles, processing aids, and renewable fillers has led to sustainable improvements in tire manufacturing. The industry is transitioning to a circular economy by including silica from agricultural waste, bio-based oils, and recycled rubber. The design and manufacture of tires is being revolutionized by cutting-edge technologies like artificial intelligence (AI), life cycle assessment, triboelectric tires, and 3D printing, which also drastically lower environmental footprints. By encouraging sustainable industrial practices and innovation, this activity supports UN SDGs 9 (Industry, Innovation, and Infrastructure) and 12 (Responsible Consumption and Production).

1. Introduction

The global automotive industry, a fundamental pillar of modern infrastructure, continues its rapid expansion, supporting a global population that reached 8 billion in 2023.[1] It is, indeed, impossible to envision a world without the tyre and automobile industries. Since its invention, the automotive industry has been the most rapidly expanding industry. However, this growth is intrinsically linked to escalating environmental challenges, particularly within the tyre manufacturing sector. The global tyre industry finds itself at a critical juncture, confronted by the dual and pressing challenges of managing an ever-increasing volume of waste tyres and mitigating the significant environmental impacts inherent in conventional production processes. Annually, nearly 1.7 billion tons of waste tyres are generated, leading to severe environmental pollution.

Historically, tyre manufacturing has been deeply entrenched in a reliance on materials predominantly derived from non-renewable, petroleum-based sources. This long-standing dependence on conventional materials carries substantial environmental costs. These conventional raw materials are not only synthesized from finite fossil fuels but are also largely non-biodegradable, thereby contributing significantly to both the carbon footprint of tyre production and the persistent problem of end-of-life waste accumulation in landfills. The critical need for sustainable alternatives stems directly from these inherent environmental shortcomings, driving the industry to seek solutions that align with ecological stewardship.

In an era where sustainability has transcended a mere buzzword to become a global imperative, manufacturers are compelled to intensify their efforts across multiple fronts:

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minimizing raw material consumption, maximizing the utilization of renewable resources, and pioneering innovative tyre recycling methodologies. Sustainability is paramount in the tyre industry due to environmental impacts, financial burdens, and public health concerns. Therefore, efforts are underway to develop sustainable tyre materials, including synthetic rubbers derived from plant sources, fillers from non-petroleum-based origins, and cellulose-based synthetic fibers. Moreover, the global market for sustainable tyre materials is projected to experience significant growth, reflecting the increased focus on sustainability.[2,3]

One important but often overlooked component in sustainable tyre design is the fabric reinforcement used in the carcass and belts. Currently, fabrics such as nylon and polyester dominate the industry due to their strength and heat resistance. However, these materials are derived from petroleum-based sources, are non-biodegradable, and contribute significantly to the environmental footprint of tyre production. Replacing them with bio based alternatives such as cellulose derived rayon, lyocell, or other engineered natural fibers offers a renewable and potentially biodegradable option. In addition to reducing fossil fuel dependence, bio based fabrics can help lower greenhouse gas emissions during production and disposal, contributing to a circular and sustainable tyre ecosystem. Further research into enhancing their mechanical performance and moisture resistance is ongoing to meet industrial requirements. This comprehensive review critically examines these ground breaking initiatives, revealing a discernible and promising paradigm shift towards more environmentally responsible practices within the sector.

Fig. 1 illustrates the interconnections between social, economic, and environmental aspects of sustainability, highlights the integration of these dimensions, and emphasizes the importance of balance for sustainability development. Each component, including business ethics, energy efficiency, and environmental justice, contributes to the primary goal of meeting present needs without compromising the ability of future generations to meet their own needs and underscores the need for comprehensive strategies that address social, economic, and environmental considerations. Currently, 4 billion used tyres are accumulating in landfills, causing severe "black pollution".[4] To tackle this, tyre industries globally have implemented several strategies, such as decreasing the consumption of raw materials, lowering tyre weights, and augmenting the utilization of natural and renewable resources.[5–7]

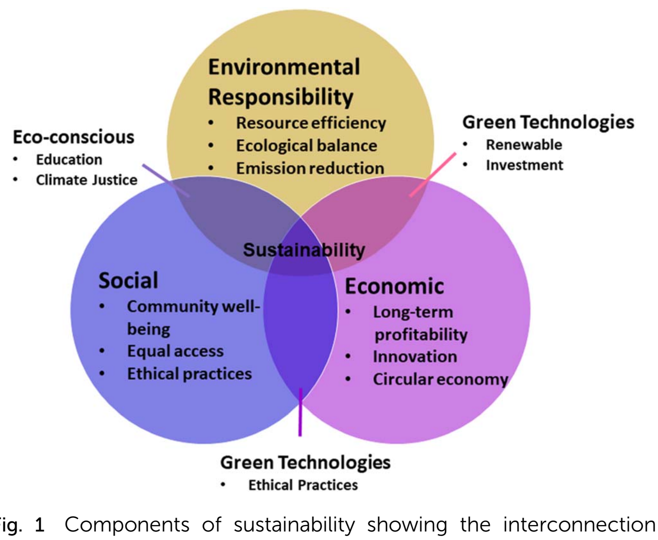

Fig. 1 Components of sustainability showing the interconnection between environmental, social, and economic pillars essential for sustainable development.

Firstly, improperly disposed tyres can release chemicals into the air, ground, and water, contributing to pollution and altering ecosystems.[8] Tyres also do not decompose and can release methane gas and toxic smoke when burned.[9,10] Secondly, sustainability is crucial for reducing the financial and logistical burden of tyre disposal.[11] Laws and regulations have been established to regulate tyre disposal, and cleaning up old tyres can be costly. Lastly, sustainability is essential for public health and safety. Improperly stored tyres can become breeding grounds for disease-carrying mosquitoes, and tyre fires cause air pollution.[12,13]

The tyre industry is raw material intensive, accounting for 65% of production costs.[14] Sustainable materials are naturally abundant, easy to extract with minimal energy spent, and easy to recycle.[15] There are two classes of sustainable materials: renewable/biomaterials (wood, natural fibers, and polymers) which are natural resources that, over time, can replenish themselves or revert to their original reserves through natural growth or replenishment after their depletion; and recycled materials, which are obtained through waste conversion using any recovery process that returns substances or materials used to perform a specific function.[16]

Tyres are composite structures made from elastomers like natural rubber (NR) and styrene–butadiene rubber (SBR) combined with curing accelerators, antioxidants, fillers, and process aids.[17] Natural rubber (NR) is a commonly used rubber in the tyre industry, but there is an imbalance between demand and supply.[18] Synthetic polymers, rubber process oils, polymer cord fabrics, and carbon black (CB) are derived from non-renewable petrochemical resources, whereas steel cords though produced through extractive metallurgy are recyclable and widely reused in the industry.[19,20] Global tyre raw suppliers and manufacturers are currently researching sustainable materials to reduce petroleum-based raw material usage by 30%.[14,15]

The tyre industry is focusing on developing new sustainable materials like synthetic rubbers using plant-derived materials or biomass, fillers from non-petroleum-based origins like silica, clay, rice husk ash, cornflower starch, bagasse, and eggshell-derived nano-calcium carbonate, and coupling agents like baker's yeast are also under investigation for tyre application.[14,21,22] Fig. 2 shows cellulose-based synthetic fibers, and anti-oxidants from flowers, fruits peels, and food wastes to replace petro-based antioxidants. To meet the tyre industry's "magic triangle" criterion, sustainable materials must fulfill superior rolling resistance to reduce fuel consumption, enhanced dry and wet traction to improve vehicle safety, and improved tyre wear resistance to extend tyre life.[16,23] Tyre

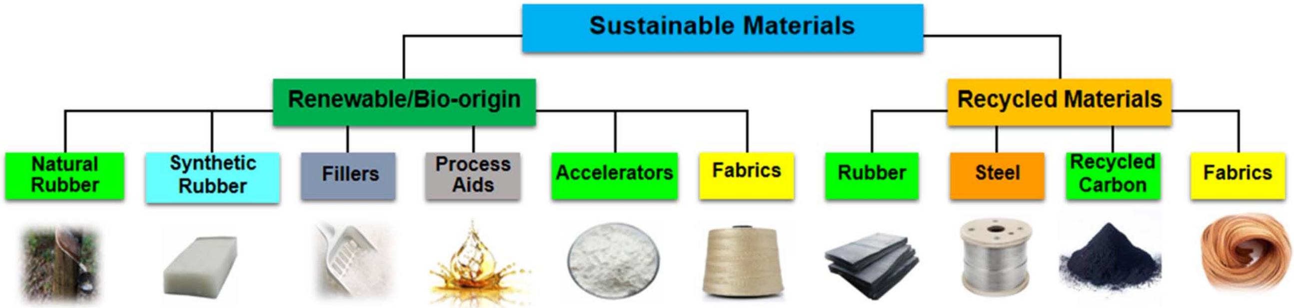

Fig. 2 Classification of various sustainable raw materials used in the tyre industry.

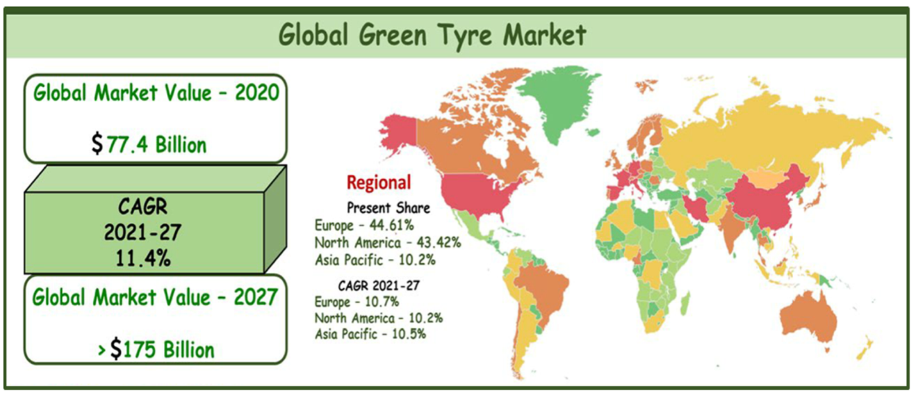

Fig. 3 Regional dynamics of the global green tyre market showing market shares and growth projections from 2021 to 2027. [Reproduced from ref. 20 with permission from Springer Nature, copyright 2022].

designers and material scientists work together to achieve targeted performance properties.[24]

Green tyre technology, aimed at reducing carbon emissions, enhancing fuel efficiency, and promoting sustainability across the tyre industry, is gaining momentum.[21,22,25] In 2022, the worldwide market for sustainable tyre materials was expected to be 35.5 million USD and is projected to reach 700 million USD by 2029, with a compound annual growth rate (CAGR) of 31% from 2023 to 2030.[26] In this study, we compiled studies on sustainable tyre materials to date. The global green tyre market is anticipated to experience significant growth, positioning itself as the dominant force in the industry by 2027, as depicted in Fig. 3 which presents regional dynamics including current market shares and projected growth rates. Europe leads with a 44.6% market share, followed by North America (43.4%) and Asia Pacific (10.2%). The darker colour shading in the figure indicates regions with higher green tyre adoption and projected market expansion, providing a visual representation of the global momentum towards sustainability.

Although this review centres on sustainable tyre technologies for automotive applications, emerging innovations in aerospace such as non-pneumatic tyres and advanced polymeric materials also reflect the broader potential of sustainable materials in mobility systems. In response to the challenges, the tyre industry's latest advancements highlight a robust and proactive adoption of bio-based polymers, fabrics, processing aids, and renewable fillers. These innovative materials offer demonstrably greener and more responsible alternatives to their traditional counterparts. Furthermore, the strategic integration of recycled rubber, bio-based oils, and silica sourced from agricultural waste underscores a significant and necessary transition towards a more circular economic model within the industry. This fundamental shift not only reduces the reliance on virgin fossil resources but also actively addresses the burgeoning problem of industrial and agricultural waste, transforming liabilities into valuable inputs. While these advancements represent significant progress, it is crucial to acknowledge that knowledge gaps persist, particularly concerning the long-term performance, comprehensive degradability, and full environmental interactions of these novel materials across their entire lifecycle. This review, therefore, emphasizes the critical need for continued, rigorous research to bridge these remaining gaps, ultimately ensuring that the tyre industry can meet escalating global demands in a truly sustainable manner for generations to come.

2. Renewable/bio-origin

2.1. Natural rubber

Natural rubber, derived from plant sources, is critical in various industries due to its unique properties and versatility. *Hevea brasiliensis* [Fig. 4A], known as the rubber tree, is an essential source of natural rubber.[27–29] This tree synthesizes *cis*-1,4-polyisoprene through the mevaloic acid pathway, extracting latex

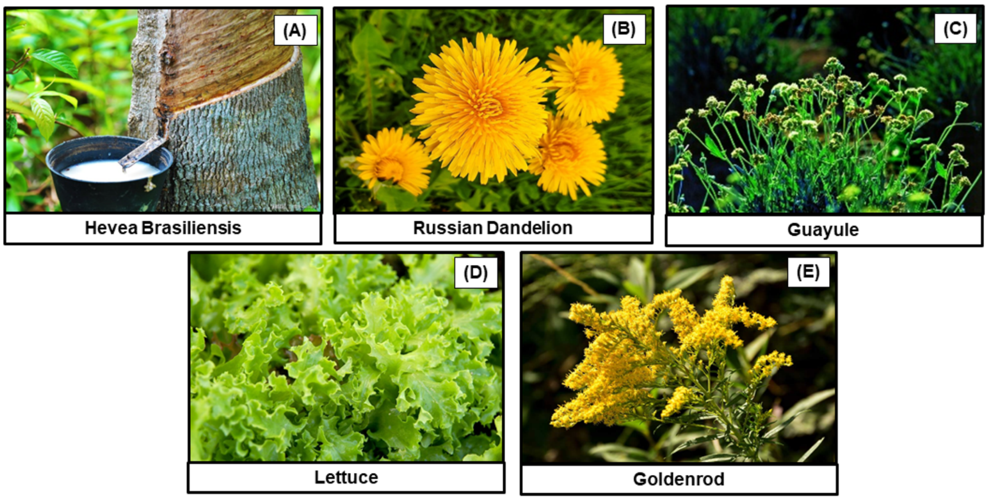

Fig. 4 Various natural rubber plants: (A) *Hevea brasiliensis*, (B) Russian dandelion, (C) guayule, (D) lettuce, and (E) goldenrod.

from rubber particles on its surface.[30] This latex is a crucial raw material in the manufacture of over 40 000 commercial products, underlining its global significance.[31] It possesses unique properties, including flexibility, elasticity, and abrasion resistance, making it irreplaceable for various applications.[31] *Hevea brasiliensis* plays a vital role in the tyre industry, accounting for more than 98% of the world's rubber production.[32]

Taraxacum koksaghyz [Fig. 4B], commonly known as Russian dandelion, is a promising alternative source of natural rubber with high molecular weight due to its capacity to produce high-quality poly(*cis*-1,4-isoprene). It can develop crystallinity and axial orientation, enhancing its mechanical properties.[33,34] Natural rubber (NR) is a crucial raw material in producing various rubber and latex goods, making the Russian dandelion a noteworthy contender for rubber production diversification. Preliminary findings show that tyres made from Russian dandelion rubber exhibit comparable resilience to those made from *Hevea brasiliensis*.[33]

Parthenium argentatum [Fig. 4C], commonly called guayule perennial shrub, synthesizes natural rubber in its bark parenchyma cells, which is subsequently extracted as latex. Guayule rubber's hypoallergenic nature offers an advantage by being safer for individuals with latex allergies. Furthermore, the residual guayule plant material, or bagasse, holds the potential for conversion into valuable co-products such as bioenergy.[35]

Several companies, including Apollo, Bridgestone, Cooper, Energy Ene, Ford, Goodyear, Nokian, PanAridus, and Guayule Australia use alternative rubber crops, *P. argentatum*, as a source of rubber. American Sustainable Rubber, Bridgestone, Continental, Ford, Goodyear, Key Gene, Kulti ava, Ling-long, NovaBioRubber, and Sumitomo, among others use *T. kok-saghyz* as a source of rubber.[36] The first guayule automobile tyre was produced in 2017, and the life cycle energy consumption was 13.7 GJ per tyre, less than that of traditional rubber (16.4 GJ per tyre).[37]

Lettuce (*Lactuca sativa*) [Fig. 4D], a valued leafy vegetable, produces natural rubber (NR) with a molecular weight exceeding 1 million Da.[38–40] Beyond its agricultural significance, lettuce stands out for its ability to synthesize NR and sesquiterpene lactones (STLs), making it a subject of interest in specialized metabolite studies.[37] As an NR producer, lettuce presents an opportunity for molecular genetic exploration due to its amenable characteristics, such as self-pollination and ease of transformation.[41] Its annual life cycle and genetic accessibility through tools like CRISPR/Cas9 genome editing further enhance its appeal for understanding NR biosynthesis at the molecular level.[42]

The plants from the Asteraceae family are diverse rubber producers, and several of them synthesize *cis*-polyisoprenes similar to NR. Examples include dandelion (*Taraxacum* spp.), lettuce (*Lactuca sativa*), and guayule (*Parthenium argentatum*), which all produce NR with considerable molecular weights. Plants like sunflower (*Helianthus* spp.) and goldenrod (*Solidago canadensis*) synthesize *cis*-polyisoprenes with varying molecular weights. Studying the biosynthesis in these plants offers valuable insights.[43]

Solidago altissima (goldenrod) [Fig. 4E], improved through research efforts, demonstrates a latex yield of 12% rubber production potential.[39] The rubber extracted from goldenrod has a low molecular weight ranging from 1.6×10^5 to 2.4×10^5 g mol^{-1} and is used in tyre production.[44] Additional species such as *Euphorbia characias* L., *Ficus* L. spp., and *Alstonia boonei* De Wild demonstrate significant rubber production. However, there is presently little information available on their practical use. Furthermore, *Taraxacum mongolicum* Hand.-Mazz. and *Helianthus annuus* L. possess the ability to produce rubber.[45] Natural rubber certified by the Global Platform for Sustainable Natural Rubber (GPSNR) is sourced from plantations that comply with the organization's sustainability frameworks. These commitments ensure sustainable and ethical practices in the rubber industry.[46]

2.2. Biobased synthetic rubbers

The conventional approach of synthetic rubbers derived from petrochemicals poses significant sustainability challenges. In 2014, the production of synthetic rubber alone consumed

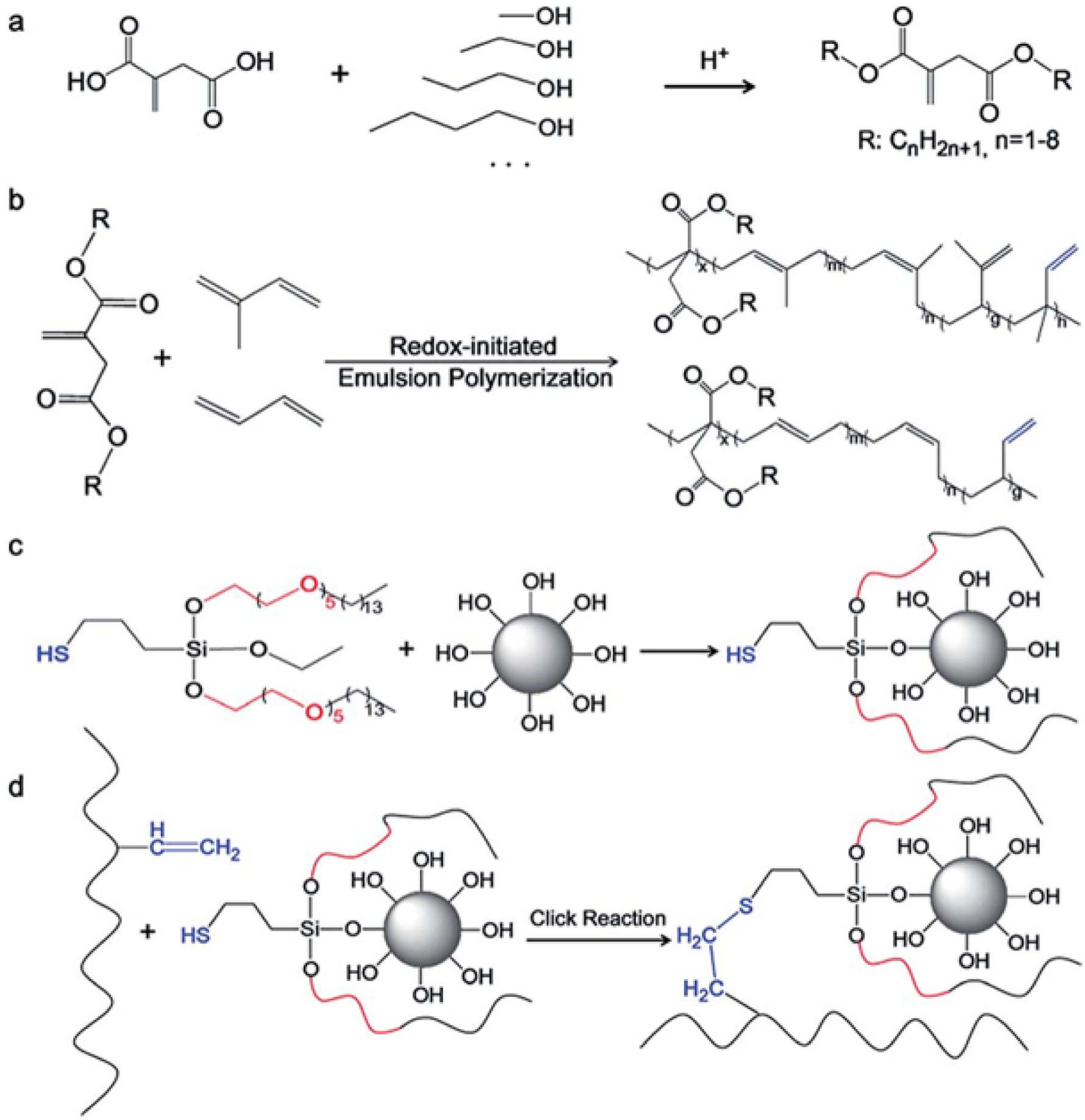

Fig. 5 (a) Synthesis of di-*n*-alkyl itaconates. (b) Synthesis of poly(di-*n*-alkyl itaconate-*co*-isoprene) and poly(di-*n*-butyl itaconate-*co*-butadiene). (c) *In situ* modification and (d) chemical linking reaction between bio-based macromolecular chains and nanoparticles. [Reproduced from ref. 54 with permission from Royal Society of Chemistry, copyright 2016].

a staggering 35.5 million tonnes of crude oil, contributing to carbon dioxide emissions. Given the urgent global concerns, researchers are actively exploring using bio-based polymers as alternatives to traditional synthetic rubber in tyre manufacturing.[47] While bio-based polymers like polylactide, polyhydroxyalkanoates, and poly(butylene succinate) have succeeded in various industrial applications,[48–50] their suitability for tyre production is not yet fully established.

Researchers have investigated using soybean oil-based elastomers, polyester elastomers, and poly(diisoamyl itaconate-*co*-isoprene) elastomers.[51–53] These materials, derived from renewable resources, show potential for tyre applications, and the major players are also interested in transitioning towards sustainable resources for tyre production. To combat CO_2 emissions and air pollution, researchers are developing low roll-resistance "green" tyre elastomers using bio-based chemicals such as itaconic acid, mono-alcohols, and conjugated dienes. One notable innovation comes from Weiwei Lei and collaborators, who successfully manufactured silica/poly(di-*n*-butyl itaconate-*co*-butadiene) nanocomposite-based green tyres. These tyres exhibit low rolling resistance, excellent wet skid resistance, and good wear resistance.[54] The elastomer formulation and processing approach (Fig. 5) resulted in tyres with performance indicators comparable to premium commercial grades. Although early classifications suggested high performance in standardised parameters like wet grip and fuel efficiency, further independent validation and regulatory alignment with updated EU labelling standards (Regulation EU

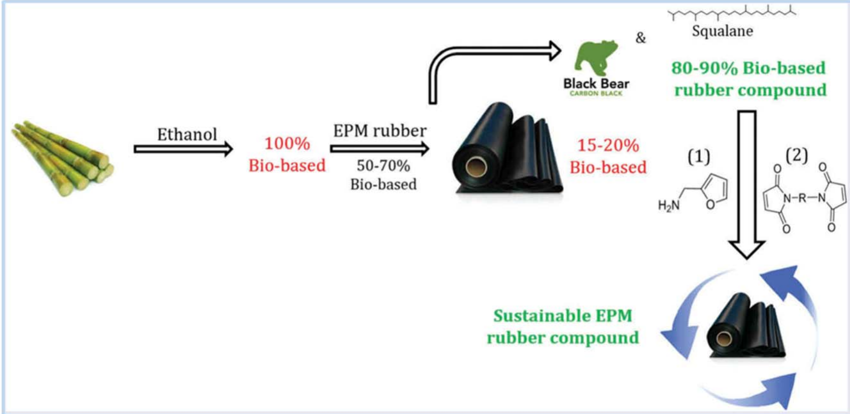

Fig. 6 The cross-linking reaction of the EPM rubber compound with carbon black and squalene. [Reproduced from ref. 71 with permission from Taylor & Francis, copyright 2019].

2020/740) would strengthen the claims. This innovation showcases the potential of bio-based elastomers in meeting key tyre performance criteria for sustainability and safety.

The synthesis of terpenes by conifers and various plants has gained attention as potential building blocks for synthetic polymers.[55,56] β-myrcene (terpene) has shown promise in elastomer design and is considered a natural base chemical in sustainable chemistry.[57] Sarkar and Bhowmick *et al.* synthesized elastomers from β-myrcene through emulsion polymerization.[58] Further studies led to the development of biobased copolymers of β-myrcene with different renewable synthons.[59,60] Another interesting avenue of research is the synthesis of sustainable methacrylate copolymers with β-myrcene through emulsion polymerization.[61] This approach offers versatility in designing bio-based elastomers for various applications.

The quest for sustainable isoprene production is also underway.[62] Researchers are optimizing ribosome binding site (RBS) sequences and enzyme screening to increase isoprene production in *Escherichia coli*.[62] Soybean oil and lignin-derived elastomers were prepared using thermal azide–alkyne cycloaddition click chemistry.[63] These elastomers demonstrated excellent mechanical strength and elasticity, retaining 96–100% of their elasticity after the first cycle of use. Zhang *et al.* reported a series of solution-polymerized styrene–myrcene–butadiene rubber (S-SMBR) with 100% conversion, high molecular weight of 15 000–200 000 Da, low polydispersity, and uniform composition. Furthermore, the wet skid resistance of the rubber could be improved without compromising its low rolling resistance.[64] Anish Khan *et al.* developed a biodegradable SBR rubber composite by incorporating collagen-functionalized nanographene oxide sheets (GO). The SBR/GO/1.5COL sample demonstrated effective embedding of the 1.5 wt% collagen (COL) filler within the SBR/GO matrix, indicating good interfacial interaction. However, at a higher COL filler content (2.5 wt%), the SBR/GO/2.5COL sample exhibited aggregated structures due to unreacted interfaces between the COL filler and SBR/GO.[65] Zhou *et al.* extended this work by preparing poly(dibutyl itaconate-*co*-butadiene) elastomers, substituting isoprene with butadiene.[66] This substitution led to significant improvements in the elastomers' mechanical and dynamic properties.

Haijun Ji *et al.* introduced poly(dimethyl itaconate-*co*-butadiene)s (PDMIBs) synthesized through emulsion copolymerization of dimethyl itaconate (DMI) and butadiene, showcasing remarkable advancements in the tyre industry.[67] These bio-based elastomers exhibit exceptional mechanical properties, including a tensile strength of 10.2 MPa, elongation at break of 1146%, and impressive toughness of 37.6 MJ m^{-3}. The versatility of PDMIB properties extends to nanosilica-filled formulations, offering robust mechanical attributes, excellent abrasion resistance, and heightened oil resistance. Syu investigated the synthesis of bio-based traditional rubbers, including polyisoprene rubber, ethylene–propylene rubber, isobutylene isoprene rubber (IIR), and styrene butadiene rubber (SBR), from monomers like isoprene, ethylene, propylene, isobutylene, and butadiene.[68] The commercialization of a bio-based route for producing *trans*-β-farnesene, derived from sugar feedstocks through fermentation, offers unique low molecular weight polymers with distinctive structure–property relationships.[69] Fang *et al.* introduced high-performance rubber composites by incorporating urazole groups into the NR chain backbone, enhancing the modulus, wet traction, and low rolling resistance.[70] Polgar *et al.* compounded thermoreversible cross-linked ethylenepropylene rubber (EPM) with pyrolysis carbon black and squalane as a sustainable filler and plasticizer,[71] as shown in Fig. 6. Sarkar and Bhowmick conducted emulsion polymerization to synthesize poly-myrcene (PMY).[61] Pranabesh Sahu explored copolymers of β-myrcene with glycidyl methacrylate (GMA), resulting in poly(MY90GMA10) with enhanced wet skid resistance and mechanical properties.[72]

2.3. Bio-based fillers

The incomplete combustion of coal tar, ethylene tar, and a small quantity of vegetable oil generates carbon black. Nevertheless, the industry trend is shifting toward "green tyres," in which silica nanoparticles are increasingly used as substitutes for carbon black. Crystalline silica is a plentiful natural

Table 1 Conventional silica *vs.* green silica[74–81]

Property	Conventional silica	Green silica
Type	Amorphous (precipitated)	Plant-based (*e.g.* rice husk)
Silica content	85–95%	Varies (*e.g.* 92.5% in rice husk)
Environmental impact	Energy intensive	Low cost, utilization of agricultural waste, reduced energy consumption
Mechanical properties	May improve modulus, hardness, and abrasion resistance	Improved modulus, hardness, abrasion resistance, and lower Mooney viscosity
Dispersibility	May require surface modification	Better dispersion and an enhanced reinforcement factor

substance found in quartz, sand, and other minerals. In contrast, green silica derived from agricultural residues such as rice husk ash (RHS), bamboo leaves, sugarcane bagasse, and corn stalks offers a sustainable alternative. Silica occurs in nature in various forms, such as amorphous, crystalline, diatomaceous (fossil-derived), and microcrystalline structures. Precipitated silica refers to silicon dioxide (SiO_2) with a particle size ranging from 1 to 40 nanometers. Reinforcing fillers enhance the tensile strength, tear resistance, abrasion resistance, and hardness of composites. Silica particles have distinct chemical and physical properties compared to green silica (Table 1). While crystalline silica is indeed a plentiful natural resource, its extraction and processing involve significant energy consumption and carbon emissions. A recent material and energy input–output analysis revealed that producing 1 tonne of silica from rice husk requires only 238.38 kg of coal, compared to 1194.08–1954.99 kg for conventional and fume silica routes. Moreover, the CO_2 equivalent emissions for rice husk silica production are significantly lower, 0.85 tonnes, *versus* 10.09 and 18.62 tonnes for conventional and fume routes, respectively.[73] Although rice husk silica production generates wastewater (24.76 tonnes per tonne of silica), it remains more sustainable overall due to its reliance on agricultural waste and lower energy input.

Rice husk silica (RHS) has garnered significant attention due to its potential as a sustainable filler in rubber compounds, particularly for tyre applications.[82–85] However, the quality and quantity of RHS can vary significantly based on factors such as its source, processing method, and storage conditions, affecting particle size, shape, surface area, porosity, purity, and chemical composition, which, in turn, influence its performance. The use of RHS in tyre production faces several challenges. Compatibility with other rubber compound components, such as polymers, vulcanizing agents, antioxidants, and other fillers, also requires careful consideration. The interaction between RHS and these components can impact other polymer parameters.

Cellulose, a natural polymer has gained attention due to its potential for application within the tyre manufacturing sector.[86,87] Cellulose is a green filler, sourced from woody materials and agricultural waste.[88–91] This biopolymer is composed of unbranched polysaccharide chains with long D-anhydroglucopyranose units and β-(1,4)-glycosidic bonds.[92,93] Its unique structure incorporates both intramolecular and intermolecular hydrogen bonding interactions, contributing to its crystalline properties. Despite its environmentally friendly attributes, incorporating cellulose into hydrophobic rubber matrices presents a complex challenge due to its inherent hydrophilic nature. To make cellulose filled rubber composites feasible, surface modification of cellulose to overcome issues related to cellulose natural agglomeration and hydrophilic characteristics is needed.[94,95] Several surface modification methods have been explored in the literature, including chemical grafting, esterification, and silane treatment. For example, modification of cellulose nanocrystals (CNCs) with maleic anhydride (M-CNCs) significantly enhances their interaction with rubber matrices. This results in improved mechanical properties of epoxidized natural rubber (ENR) composites due to the formation of covalent bonds.[96] Similarly, surface modifications with organosilanes have shown benefits in enhancing the mechanical properties and dispersion of CNCs in natural rubber (NR) composites.[97] Epoxy-modified cellulose nanocrystals have also demonstrated enhanced dispersibility and interfacial interactions with rubber matrices, contributing to improved mechanical performance and recyclability of carboxylated styrene–butadiene rubber nanocomposites.[98] Biochars exhibit specific functional groups on their surface, potentially promoting their application as free-radical scavengers in manufacturing and construction.[99]

Roy *et al.* (2021) highlighted several crucial points regarding the use of cellulose in rubber technology. It emphasizes that use of cellulose as a primary filler in large-scale industrial applications is still a subject of ongoing research and that cellulose does not significantly enhance the thermal stability of rubber compounds. It also highlights the potential application of surface-modified cellulose nanocrystals as a reinforcing filler.[100] The degree of polymerization of cellulose varies depending on its origin, with wood derived cellulose having about 600–1500 glucose units and cotton derived cellulose having around 9 000–15 000 glucose units.[101] The molecular structure of cellulose is composed of repeating units of β-D-glucopyranose, connected by β-1,4-glycosidic linkages at the C1 and C4 positions.[102–107]

Nanocrystalline cellulose, due to its exceptional properties, has been investigated as a potential alternative to traditional fillers such as carbon black in the production of "green tyres".[108] Midhun Dominic reported the combined use of carbon black (CB) and rice husk nanocellulose (RHNC) for the purpose of developing environmentally friendly tyres. Composites of NR-CB/RHNC were created using different ratios of CB/RHNC. The NRCB25NC5 composite has the lowest value of $\tan \delta$ at

60 °C among the other produced composites, indicating a reduced rolling resistance.[108] Haghighat *et al.* incorporated α-cellulose powder into styrene–butadiene rubber (SBR) which resulted in an increase in Young's modulus, hardness, and compression set, as well as a decrease in elongation and resilience. However, they found that the tensile strength, tear strength, and abrasion resistance initially increased at a low α-cellulose concentration (5 parts per hundred rubber (phr)), but then decreased as the α-cellulose content increased.[109] In the study by Koushik Pal *et al.*, α-cellulose was investigated as a potential reinforcing filler in S-SBR and PBR tyre-tread compounds and an increased cure rate was found, particularly at a 10% replacement level, without compromising wet grip properties.[110]

By hybridizing pyrolysis carbon black (CBp) with cellulose through ball milling, a novel CBp/nanocellulose hybrid (CNCH) was created. CNCH had a bead chain like structure, tight attachment of CBp to cellulose, and strong hydrophobic and hydrogen bond interactions. NR reinforced with CNCH exhibits low hysteresis and improved anti-abrasion properties, making it an ideal filler for tyre production.[111,112] Recent studies have shown success in functionalizing biochars using a multisource approach, such as blending microalgae and swine manure. Similarly, functionalizing cellulose with tailored surface groups could facilitate better interaction with rubber matrices, providing improved performance in rubber composites.[113,114]

Lignin, found in lignocellulosic materials like wood, constitutes 10% to 35% of these sources.[115,116] It consists mainly of aromatic alcohols with varying degrees of methoxy groups, impacting crosslinking. Lignin is derived from the paper and pulp industry and classified based on the pulping processes, including kraft, sulfite, soda, and organosolv. Recent innovations include lignin extraction using ionic liquids and deep eutectic solvents.[117–119] Lignin is primarily used for specialty products such as dispersants, adhesives, surfactants, and antioxidants in plastics and rubber.[120–123] Lignin is an aromatic biopolymer with a complex structure composed of various functional groups, including methoxy, phenolic, hydroxyl, and carbonyl groups. However, the high polarity of lignin poses a challenge when incorporating it into nonpolar rubber matrices, necessitating various chemical modifications. Different isolation techniques and sources of lignin can result in diverse physical properties, influencing its performance in rubber composites.[123–126] Rubber composites with 50 phr of lignin loading exhibit remarkable properties, including a tensile strength of approximately 10 MPa, an elongation at break of around 276%, and a rubber modulus M100 of about 3.51 MPa. The study's innovative methods for achieving a fine and homogeneous lignin dispersion provide the foundation for enhancing various commercial rubber materials.[127]

Researchers have employed several approaches to overcome this challenge, including high temperature dynamic heat treatment, latex co-precipitation, and surface modification using coupling agents and compatibilizers. These methods aim to improve lignin's compatibility with rubber.[128–131] Despite these challenges, lignin fillers hold significant potential for various applications, particularly in the rubber industry. Lignin can serve as a reinforcing filler in rubber compounds, contributing to improved mechanical properties, reduced rolling resistance, and potential use in "green" tyres. Additionally, lignin can act as a stabilizer in rubber formulations.[132,133] Clay minerals, such as montmorillonite and sepiolite, offer an eco-friendly alternative to conventional fillers. Their natural abundance and compatibility with rubber matrices make them attractive for tyre applications.[134] Recent studies indicate that the incorporation of clay in tyre formulations improves mechanical properties, such as tensile strength and modulus, leading to enhanced tyre performance.[135]

Carbon nanotubes (CNTs) applied in tyre treads enhance grip, diminish rolling resistance, and extend durability.[136] Multi-walled carbon nanotubes (MWCNTs), with their high aspect ratio, act as effective rubber fillers, reinforcing tyres. CNT-enhanced tyres exhibit significant improvements in mechanical properties, and exhibit a nearly 600% increase in tensile strength, a 250% increase in tear strength, and a 70% enhancement in hardness compared to pure styrene–butadiene rubber (SBR) composites. Comparative analyses highlight the superior heat dissipation of CNT-reinforced tyre compounds, addressing concerns related to thermal build up during prolonged use.[136] The market, largely dominated by multi-walled carbon nanotubes, is projected to exceed 50 000 tonnes annually by around 2035, while the value of CNT-enabled products is expected to reach USD 60–100 billion. Despite the performance benefits of MWCNTs, safety concerns remain due to their structural similarity to asbestos and associated carcinogenic potential. While these nanotubes are typically embedded in a rubber matrix reducing immediate exposure risk there is ongoing concern about their potential release during tyre wear, recycling, or combustion. Regulatory agencies such as the U.S. EPA have noted that the risk is lower in encapsulated forms, but a comprehensive life cycle assessment is essential to fully evaluate environmental and human health implications of CNT enabled tyre technologies.

The production of N-doped carbon nanohorns (CNHs) characterized by a hollow structure and high specific surface area, represents high value added carbon nanomaterials with unique properties.[137] These CNHs, derived from recycled waste tyre CBp, contribute to sustainable practices. The hollow structure and high specific surface area of CNHs present opportunities to improve mechanical properties, reinforcing tyre rubber and enhancing wear resistance. Hybrid nanocomposites that contained less than one weight percent CNH demonstrated enhanced dynamic-mechanical and physical properties as a result of unique interactions between the filler and polymer.[138] However, higher CNH amounts led to filler re-agglomeration, resulting in the deterioration of elastomer properties. Despite the intrinsic conductivity of pristine CNHs, their addition did not yield significant increases in electrical conductivity in hybrid compounds.

Graphene, a two-dimensional carbon allotrope, has transformed tyre compounds, offering exceptional mechanical strength and thermal conductivity.[139] The utilization of a direct current (DC) arc plasma method facilitates the transformation of high purity carbon black powder (CBp) into few layer graphene (FLG). FLG, characterized by exceptional properties, serves as a high value added carbon material with diverse applications. FLG derived from waste tyre CBp provides a sustainable solution for the tyre industry, promoting environmental conservation and resource recycling.[140] Synergistic effects of combining graphene with carbon black and silica as nanofillers in natural rubber nanocomposites demonstrate that the hybrid filler composition not only enhances these properties but also offers the potential to reduce reliance on carbon black.[141] The study explores a hybrid nanofiller comprising graphene oxide and amine-modified nanosilica in natural rubber *via* the latex stage coagulation method.[142]

Combining soy particles (SP) and carboxylated styrene–butadiene (CSB) nanoparticles significantly improved the modulus of natural rubber (NR), particularly at an optimal CSB concentration of around 20%. The addition of 10% SP substantially increased tensile strength. Swelling studies indicated reduced swelling with up to 20% CSB. Tensile properties of NR composites with 10–20% CSB and SP closely resembled those of carbon black reinforced NR composites, showcasing their potential for tyre tread applications.[143] Eggshell powder, derived from food industry waste, has attracted attention as a sustainable filler and contributes to improved tensile strength and impact resistance in tyre formulations.[144] Evaluations show that incorporating eggshell powder in tyre compounds not only enhances mechanical properties but also reduces the environmental impact of waste disposal.

2.4. Comparison of sustainable and traditional tyre materials

Sustainable tyre materials, exemplified by silica-based fillers such as ULTRASIL® 4000 GR and Hi-Sil® Reinforcing Silica, offer notable advancements in key performance metrics like rolling resistance, grip, and wear resistance when compared to their traditional counterparts. Balancing sustainability and performance remains a priority for tyre manufacturers, prompting extensive research and development efforts. By optimizing formulations and adopting innovative technologies, manufacturers strive to mitigate the trade-offs typically associated with bio-based or recycled raw materials, while enhancing environmental benefits.

Several leading tyre manufacturers have launched next generation products incorporating high proportions of sustainable materials ranging from bio-based oils and renewable silica to recycled rubber and polymers. These tyres often integrate mass balance certified components, recycled carbon black from end-of-life tyres, and materials derived from plant-based sources or agricultural waste. Many companies have announced targets to increase the share of sustainable materials in their product portfolios to 40% or more by 2030, while some have pledged to transition entirely to renewable and recycled feedstock by 2045. Such initiatives reflect a broader industry trend toward reducing carbon footprints and promoting circularity in material sourcing.

2.5. Renewable/bio based processing aids

Rubber process oils (RPOs) play an essential role in the rubber industry, particularly in enhancing the processability and state-of-mix in high filler content rubber compounds.[145–147] They offer a range of environmental benefits, including reduced carbon emissions and lower carcinogenicity.[148,149] Various vegetable oils, including tea oil (TO), rice bran oil (RBO), soybean oil (SBO), palm oil (PO), sunflower oil (SFO), coconut oil (CO), castor oil (CAO), jatropha oil (JO) and others, have been the subject of investigation for their suitability as replacements for petroleum-based oils in rubber processing.[150,151] These vegetable oils exhibit improved processing characteristics, better interactions with polymers and fillers, and enhanced filler dispersion without significantly compromising the properties of rubber compounds.[152] The patent innovation introduces a rubber composition comprising 0.5 to 5.0 parts by mass of a fatty acid and/or its derivative per 100 parts by mass of rubber. Additionally, synthetic aliphatic carboxylic acids or specific acids like stearic acid are suitable options. Among these, metal salts of these acids, particularly zinc salts, are preferred for their reversion resistance.[153]

Palm oil, derived from the oil palm tree, has emerged as a promising sustainable alternative to traditional petroleum-based oils in the rubber industry.[154] The utilization of palm oil (PO) as an alternative to toxic distillate aromatic extract (DAE) in rubber compounds has displayed remarkable potential. The substitution of DAE with PO exhibited not only comparable filler interactions but also improved rubber–filler interactions, leading to superior rubber properties. Additionally, the change from DAE to PO reduced crosslink density, emphasizing the influence of PO on modifying the crosslinking types within the rubber matrix. In another study, PO was employed as a green plasticizer in EPDM rubber, and the results demonstrated that it performs similarly to conventional paraffin oil in terms of processing efficiency and mechanical performance.[155] Another significant advancement is the development of an amphiphilic palm oil derivative aimed at improving mechanical properties in rubber composites.[156] The successful integration of this derivative in pneumatic tyres led to remarkable outcomes in high-speed endurance, braking performance, and viscoelastic properties, indicating its potential as a replacement for conventional aromatic oils in tyre engineering applications. Additionally, palm oil contributes to improved elastic properties and reduced rolling resistance in rubber compounds, making it an eco-friendly and efficient choice, especially for tyre applications.[157] Aprianti's investigation provides a significant resource for the manufacture of carbon black using only renewable raw materials.[158] In addition, the research extensively analyses the relative benefits of using pyrolysis oil and syngas as opposed to traditional fossil fuels.[159]

Moresco *et al.* explored the feasibility of SO in rubber processing. These oils offer a sustainable and biodegradable alternative to petroleum-based equivalents. While their incorporation may lead to cure retardation and reduced mechanical properties, they hold potential for enhancing wet grip, processability, and filler dispersion, contributing to more eco-friendly tyre production.[160] Soybean oil, when modified into MSO, exhibits significant potential for improving rubber processing and properties. MSO serves as a rubber processing oil (RPO) in tyre tread compound formulations.[161] Studies have shown that the incorporation of MSO results in improved crosslinking density and modulus, especially when compared to unmodified soybean oil.[162] Moreover, MSO-based tyre tread rubber demonstrates better aging resistance and wear resistance properties compared to petroleum-based alternatives, suggesting a longer tyre lifespan. Kang *et al.* focused on elevating tyre performance through the combination of soybean oil and carbon nanotubes (CNTs).[163] Actual vehicle tests have validated the usefulness and effectiveness of this soybean oil-based composite, positioning it as a promising solution for the tyre industry.

Xu *et al.* reported that modified soybean oil (MSO) is produced by combining soybean oil (SO) with sulphur in order to decrease the number of double bonds in SO and prevent any negative impact on the crosslink density and mechanical qualities of rubber. The crosslink density and modulus of MSO-plasticized rubber exhibit considerable enhancements in comparison to those of SO-plasticized tread rubber. The plasticization impact of MSO-6% on tread rubber was found to be the most effective. Subsequently, the Mooney viscosity, Payne effect, and mechanical properties of tread rubber plasticized with varying amounts of MSO-6% are examined to analyse the impact of plasticization. The ageing resistance and wear resistance properties of MSO-6% plasticized rubber exceed those of AO-plasticized rubber, making them more significant.[164]

Nun-Anan conducted a study on the impact of soybean oil fatty acid (SBOFA) on the characteristics of acrylonitrile–butadiene rubber (NBR) in comparison to dioctyl phthalate (DOP). Initially, it was seen that the inclusion of SBOFA enhanced the capacity of the NBR compound to flow, as shown by the gradual reduction in Mooney viscosity with higher amounts of SBOFA. Generally, the addition of SBOFA at a concentration of 4 parts per hundred (phr) improved the ability of the NBR vulcanizate to withstand high temperatures by causing the thermal breakdown to occur at a higher temperature. The research demonstrates that SBOFA is a feasible and environmentally friendly processing aid for NBR, offering a sustainable alternative option.[165]

Suchismita Sahoo explored the use of coconut shell extract (CSE) derived from coconut shell biomass as an eco-friendly alternative to traditional silane coupling agents (CA) in rubber compounding. CSE serves as a dual CA for modifying silica surfaces, addressing filler–filler aggregation.[166] Cardanol oil (Cdn) and chemically modified Cdn-A have been evaluated as plasticizers. Cdn acts as a co-activator for rubber vulcanization, while Cdn-A shows minimal effects on this process. Both oils enhance abrasion and tear resistance, with Cdn-added compounds demonstrating high elongation at break and toughness. The research suggests that Cdn and Cdn-A could replace petroleum-based oils in rubber compounds, emphasizing their potential for wet traction improvements.[167] Through a slurry based steam activation process, the resulting biochars were evaluated for their mechanical properties in rubber composites. Furthermore, the study highlighted the correlation between low ash content in the feedstocks and the improved reinforcement potential in the resulting biochar filled rubber composites.[168]

Dongju Lee's *et al.* replaced conventional carbon black fillers with silica and introduced zinc-free processing aids (ZFAs) to enhance tyre performance.[169] Particularly, when ZFAs replaced zinc containing processing aids (ZCAs) in silica rubber composites, significant improvements in tensile strength and elongation were observed. Furthermore, rubber compounds

Table 2 Comparison of virgin carbon black (vCB) *vs.* recovered carbon black (rCB)[185–198]

Property	Virgin carbon black (vCB) (N550)	Recovered carbon black (rCB)
Composition	Pure carbon with minimal impurities	Contains inorganic and carbonaceous deposits
Elemental composition	High carbon content	Lower carbon content
Volatile matter	Low	High
Ash content	Very low	High
Surface functional groups	Presence of carboxyl groups and ketones	Lower presence of acidic functional groups
BET surface area	Higher	Lower
Particle size distribution	More uniform particle sizes	Wider particle size distribution with larger particle size
Specific gravity	Lower	Slightly higher
Morphological characteristics	Spherical, well-defined and homogeneous particles	Coarse aggregates and heterogeneous particles
Rheology during mixing	No major technical challenges	No major technical challenges
Rheology during vulcanization	Higher cross-linking density	Lower cross-linking density
Thermal behavior	Higher residual content	Lower residual content
Density	Higher density	Lower density
Structural parameters	Higher MCS (indicating better cross-linking)	Lower MCS (indicating reduced cross-linking)
Tensile strength	Strong reinforcing effect	Non-negligible reinforcing effect
Compression strength and compression set	Improved strength with increasing filler loadings	Lower recovery capacity with lower rCB loading
Hardness	Higher hardness	Comparable hardness with greater rCB proportions
Cost-benefit analysis	More expensive	Often lower cost, independent of oil prices

with ZFAs exhibited remarkable enhancements in fatigue properties, contributing to more durable and high performing tyres. The study introduced zinc free processing aids (ZFAs) for rubber formulations in tyre treads, replacing zinc containing processing aids (ZCAs) to address environmental concerns. Integrating ZFAs significantly enhanced rubber composite properties, demonstrating up to a 31% increase in tensile strength and a 20% improvement in elongation compared to ZCA filled formulations. The improved performance was attributed to better silica dispersion within the rubber matrix, facilitated by ZFAs. The research focuses on enhancing "green tyre" development by integrating zinc free coupling agents (ZFCs) and ZFC functionalized graphene into silica tread formulations. Through a novel catalytic synthesis, ZFCs were tailored to interact with silica and graphene, improving dispersion and interfacial adhesion in the rubber matrix. Incorporating these compounds significantly improved the mechanical, thermal, and electrical properties of rubber composites. The study highlights the potential of low loadings of ZFC functionalized graphene (GZFCs) in improving rubber composite properties for tyre engineering, offering a promising pathway for "green tyre" development.[170–172]

The mechanical properties, including the modulus and tensile strength, showed considerable improvement with GZFCs/SBR (up to 4% and 13%, respectively) compared to the control, without compromising elongation. Additionally, dynamic mechanical analysis (DMA) results revealed improved wet grip and rolling resistance in GZFCs/SBR composite tyres. These tyres demonstrated about 4% and 3% better dry and wet braking abilities, respectively, and exhibited a lower rolling resistance coefficient (9.21 N kN^{-1} compared to 9.7 N kN^{-1} in the control). Furthermore, high speed endurance testing, adhering to ECE and DOT guidelines, confirmed the superior performance of the GZFCs/SBR composite tyres in comparison to the control.[173]

2.6. Bio accelerators

In recent years, alternatives to conventional accelerators are researched, with a focus on minimizing environmental impact. Nakason *et al.* investigated cassava starch's impact on natural rubber, noting a decrease in certain mechanical properties with increased starch levels.[174] Liu *et al.* demonstrated improved mechanical properties in natural rubber composites using polybutylacrylate-modified starch.[175] Tang *et al.* presented enhanced properties in SBR compounded with starch modified with resorcinol-formaldehyde and silane.[176] Lora *et al.* emphasized lignin's dual role as a tackifier and antioxidant,[177] while Jiang *et al.* explored nano-lignin composites with natural rubber, showing improved dispersion and mechanical properties.[178] Additionally, soy protein, wheat starch, cassava, lignin, and soya were identified as bio-accelerators accelerating vulcanization and offering potential for sustainable tyre manufacturing. LANXESS introduced a universally suitable vulcanization accelerator, VP Vulkacit TZ, based on aromatic amines, offering an eco-friendly alternative with an extended cure time for efficient rubber production.[179]

2.7. Bio based fabrics

Sustainable fabric choices, including aramid fibers, lyocell, and aramid nanofibers (ANFs), are highlighted in Table 3. Surface modification techniques for aramid fibers further contribute to improved fatigue behavior and mechanical properties, highlighting their sustainable potential. Aramid fibers, renowned for their strength and heat resistance, play a pivotal role in sustainable tyre technology.[180] The integration of functionalized aramid nanofibers and cellulose nanocrystals (fACs) as a hybrid filler in rubber compounds exhibits sustainable advancements.[181] Lyocell, a regenerated cellulose fiber, emerges as an environmentally friendly option for tyre reinforcement.[182] Its recycling production minimizes environmental pollution, positioning lyocell as a sustainable choice for high performance tyres.[183] The development of an eco-friendly dipping system for polyamide 66 (PA66) fiber cords offers a sustainable alternative in the production process.[184] Straightforward manufacturing processes of rayon and lyocell and favourable properties make them attractive choices, as shown by their successful application in high performance tyres.

3. Recycled materials

3.1. Recycled rubber

The challenge arises when aligning rubber materials with circular economy principles by extending their lifespan.[199,200] To address this challenge, researchers have investigated different elastomeric matrices suitable for automotive applications. Crumb rubber (ground tyre rubber-GTR) is a versatile material that can be used in various industries. However, it faces environmental concerns due to the leaching of chemicals and performance limitations. Self-healing materials have gained increasing attention due to their ability to autonomously repair damage, thereby extending product life and reducing waste. The self-healing mechanism involves hydrogen bonds, which are weak attractions between hydrogen atoms and electronegative atoms, capable of being reversed to heal damage. Ionic interactions, where charged groups within the polymer matrix form ionic bonds, also contribute to self-healing. Other mechanisms include reversible chemical reactions and supramolecular interactions, which facilitate the restoration of the material's original structure.[201–203]

Incorporation of self-healing agents can increase production costs, and the manufacturing process can be more complex compared to that of traditional materials. In the tyre industry, incorporating self-healing materials into tyre compounds involves adding functional moieties through processes such as epoxidation, carboxylation, and halogenation. The polymer architecture, including cross-link density, plays a crucial role in determining the effectiveness of these self-healing properties.[204–206] The tyre industry generates a substantial volume of waste tyres, presenting significant environmental challenges.[207,208] The impact of irradiation time on the structure, mechanical properties, and thermal behavior of NR/GTR composites was a central focus of their investigation. Longer irradiation times were found to induce significant alterations in the composite properties.[209] These changes were predominantly attributed to variations in adhesion and cross-link density. Araujo-Morera *et al.* developed self-healing rubber composites by incorporating recycled ground tyre rubber (mGTR) and carbon black (CB) into styrene–butadiene rubber (SBR). This achieved an impressive 80% healing efficiency while increasing tensile strength by 300%, demonstrating a balance between self-healing and mechanical properties.[210] Binti Joohari *et al.* tackled waste tyre disposal by using recycled crumb rubber (CR) with polymers like SBS and EVA to create hybrid recycled polymer modified bitumen (PMB). This improved bitumen viscosity and offered a sustainable solution for waste management.[211] Recent advancements in tyre recycling involve grinding used tyres into rubber granulate, aligning with the circular economy model and reducing environmental impact. Optimizing composite formulations for a balance between strength and healing aligns with circular economy.[212–216]

Reclaimed rubber is a critical component in addressing the environmental challenges posed by the disposal of end of life tyres (ELTs).[217,218] These discarded tyres are composed of various materials, including natural rubber, synthetic rubbers, carbon black, fabric, antioxidants, and steel wires. The need for effective tyre recycling solutions has prompted industry initiatives, such as the Black Cycle Project in Europe, and efforts to enhance the circular economy in the tyre industry [Fig. 7].

Reclaimed rubber, a valuable and sustainable material, has garnered significant attention as a key component in various innovative recycling approaches aimed at recycling waste tyre rubber.[219] Methods to transform reclaimed rubber encompass using reclaiming agents, advanced manufacturing techniques, and sustainable additives, offering promising solutions to enhance rubber properties, reduce waste, and promote eco-friendly practices within the rubber industry.[220–223] Ghorai *et al.* investigated the use of bis[3-(triethoxysilyl)propyl]tetrasulfide (TESPT) as a reclaiming agent, enabling silica dispersion in reclaimed rubber and substituting costly carbon black with low cost silica fillers.[224] Tseng *et al.* utilized a hot pressing method to create gas separation membranes from reclaimed tyre rubber, offering both a sustainable waste tyre management solution and competitive gas separation properties.[225] Incorporating silica into natural rubber reclaims rubber blends *via* sol–gel techniques, and enhanced reinforcement with increased reclaim rubber content.[226] Low temperature oxidation of GTR with soybean oil was demonstrated as an eco-friendly method to achieve a highly efficient RP, contributing to improved rubber performance and sustainable material development in the rubber industry.[227]

Rubber devulcanization involves the breaking of sulfur–sulfur cross-links within vulcanized rubber. The addition of organic disulfides, such as diaryl disulfide, plays a crucial role in breaking sulfidic bonds and facilitating devulcanization. At its core, devulcanization entails the breaking of cross-links within rubber materials, which allows for the reutilization of rubber in various applications. This process has attracted significant attention in recent years, owing to the pressing need to reduce the environmental footprint of rubber waste.[228] The mechanism of devulcanization also involves the careful manipulation of various factors such as temperature, pressure, and chemical agents. By effectively disrupting these bonds, devulcanization enables the material to regain its flexibility and functionality, making it suitable for reuse.[229] Silanes, used as devulcanization aids, initiate the devulcanization process by generating free radicals that break down broken cross-links, preserving the rubber structural integrity.[230,231] Devulcanized rubber has attributes like residual additive retention, preserved original cross-linkings, and decreased polymer chain molar

Fig. 7 Different stages and sizes of shredded tyres. [Reproduced from ref. 217 with permission from Elsevier, Creative Commons CC-BY-NC-ND, 2019].

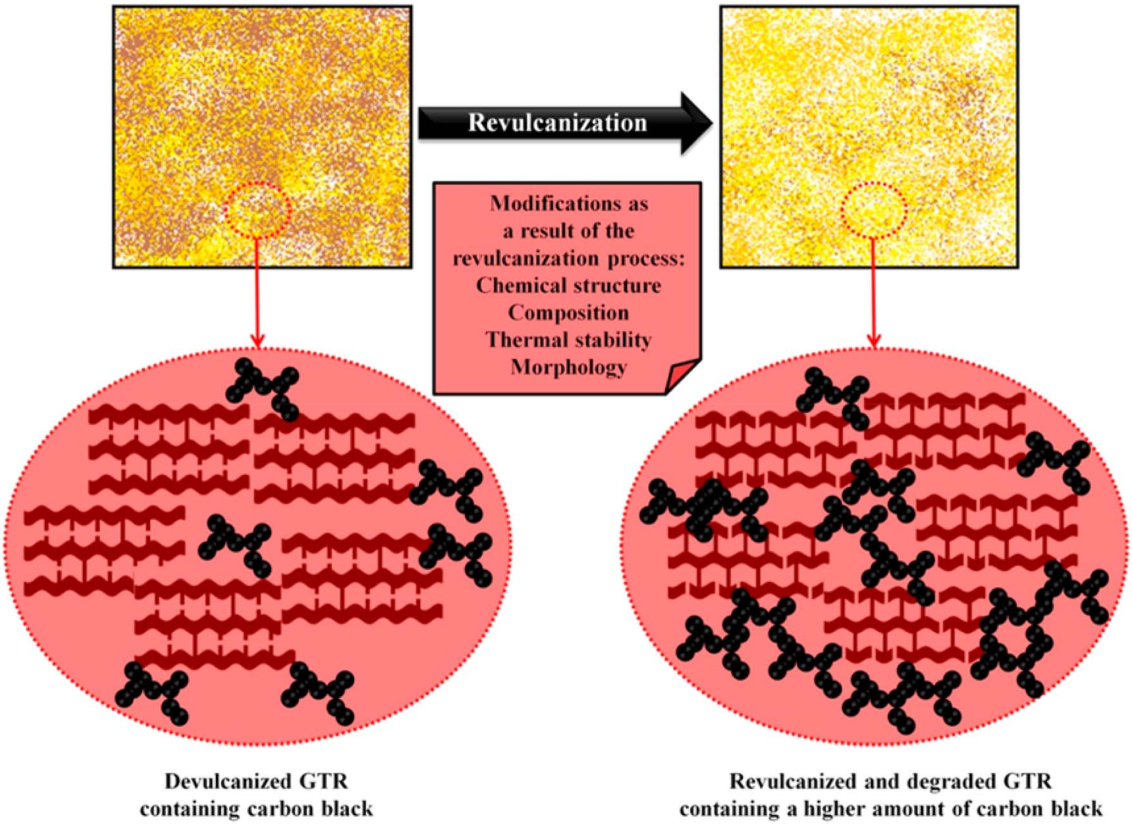

Fig. 8 Modifications in revulcanized samples. [Reproduced from ref. 230 with permission from American Chemical Society, copyright 2019].

mass, resulting in shorter cure times and quicker vulcanization rates compared to raw rubber (Fig. 8).

Oh and Isayev investigated ultrasonic devulcanization on unfilled butadiene rubber, providing insights into devulcanization methods.[232] Pérez-Campos *et al.* investigated the use of microwave devulcanized industrial waste of styrene butadiene rubber in rubber composites specifically designed for automotive applications.[233] Simon *et al.* investigated microwave devulcanization of ground tyre rubber (GTR) at varied temperatures and heating rates. Introducing devulcanized GTRs into natural rubber based mixtures *via* two step mixing significantly improves mechanical properties.[230] Mechanochemical devulcanization allows for re-vulcanization of recycled rubber, and an analytical method distinguishes truly devulcanized rubber from reclaimed rubber.[234] Sulfur vulcanized natural rubber is mechanochemically devulcanized using a silane based tetrasulfide, creating silica based rubber composites. These composites, with 30% replaced by devulcanized NR and varying silica content, exceed natural rubber silica composites in mechanical and dynamic properties. Devulcanized rubber based silica composites show ~20 MPa tensile strength and ~921% maximum elongation strain.[234]

Revulcanization further complicates matters, requiring precise control over parameters for desired product properties. Additionally, there is a lack of a consolidated market for devulcanized and vulcanized materials, alongside limited research and development efforts.[235,236] Devulcanization and vulcanization, crucial processes for rubber recycling, encounter multifaceted challenges. Furthermore, the lack of standardized practices and research limits progress in these technologies. Overcoming these hurdles necessitates collaborative efforts across disciplines to enhance efficiency, cost-effectiveness, and sustainability in rubber recycling.

3.2. Recycled steel

The integration of recycled steel into tyre manufacturing represents a significant stride towards sustainable tyre production. End-of-life tyres (ELTs) generate vast quantities of waste annually. More than 500 million tyres are stockpiled in landfills worldwide, posing serious environmental and health risks.[237] Recycling the steel components, which make up approximately 13–27% of a tyre's weight,[238] mitigates these risks while providing a valuable secondary raw material.[239] Steel recovery from ELTs is typically achieved through ambient or cryogenic mechanical shredding, followed by magnetic separation to extract embedded cords.[240] The recovered steel, once purified to limit non-metallic content below 4%, is processed *via* the energy efficient electric arc furnace (EAF) route, offering substantial reductions in energy use and greenhouse gas emissions.[241] Compared to conventional manufacturing, recycled tyre steel fibers (RTSFs) consume less energy and can be 30–70% less costly, although additional treatments to remove residual rubber and textile contaminants may increase processing costs and the carbon footprint. Recycled steel has already found successful application in tire bead wires and cords; for instance, Bridgestone has incorporated bead wire produced from ELT derived steel in its BWSC tires, processed through collaboration with Nippon Steel and Sanyo Special Steel. Beyond reinforcement, recycled steel is also being utilized in other structural tyre components such as the sidewall and tread.[242] However, variability in quality, contamination, and logistics remain barriers, and market viability depends on steady demand and regulatory support.[243] Encouragingly, Michelin's partnership with Enviro exemplifies how large-scale deployment of pyrolysis and steel recovery technologies can close the material loop; their joint venture envisions recovering up to 90% of materials from ELTs for reuse in tyres and other rubber products.[244] The broader adoption of such strategies not only aligns with circular economy principles but also contributes to resource conservation, reduced emissions, and economic circularity in tyre manufacturing.

3.3. Recycled carbon black

Recycled carbon materials, often derived from waste tyre rubber and other sources, have gained increasing attention as sustainable alternatives in various industries. Currently, the global production of carbon black stands at approximately 8.1 million metric tons, ranking it among the top 50 industrial chemicals manufactured worldwide. However, the manufacturing process of carbon black is associated with a substantial carbon footprint. These "green" fillers are derived from potential recyclable waste materials and industrial/agricultural by products such as peanut shells, rice husks, and fly ash, among others.[245–249] The adoption of these "green" fillers offers the potential to significantly reduce the reliance on fossil fuels in carbon black production, thereby improving the environmental impact on the ecosystem.

Different types of recycled carbon blacks

Renewable carbon blacks. Renewable carbon blacks represent a promising change towards sustainability in the carbon black industry, being entirely bio-based and fossil-free.[185] ECORAX® Nature 200 with lower yield still adheres to ASTM N326 standards.

Recovered carbon black (rCB). Recovered carbon black (rCB) offers another sustainable option, being entirely recycled and exhibiting a significantly improved carbon footprint. Presently, rCBs can effectively replace semi reinforcing carbon blacks like N700, N600, and N500, with industrial volumes expected by 2024.[186,187] The characteristic properties of rCB and virgin Carbon Black (vCB) (ref. N550) are tabulated in Table 2. Notably, 65% of tyre manufacturers view rCB as the ideal material to achieve sustainability goals. By recycling carbon from waste tyres, we can reduce the support on virgin carbon black production, conserving resources and modifying the environmental impact associated with its production.

Research efforts have led to the development of innovative methods for reclaiming carbon materials from waste tyres [Fig. 9]. Understanding and optimizing these processes are crucial steps toward harnessing the potential of recycled carbon materials.[188–190] However, the performance of recycled carbon materials may differ from that of conventional carbon black due to factors like particle size, surface area, and chemical composition. Balancing these properties is essential to optimize their performance in specific applications.

Methane pyrolysis carbon blacks. Generated through methane decomposition in a plasma arc reactor, they offer a further avenue for sustainability, and are able to replace semi reinforcing carbon blacks and could potentially develop into highly reinforcing carbon blacks (N300, N200, and N100) within the next 5–10 years[191] This novel thermal plasma process demonstrates promising selectivity and yield of both solid carbon and gaseous hydrogen, achieving methane feedstock conversions of over 99% and a hydrogen selectivity of over 95%, while also recovering more than 90% of solid carbon. The process is currently more energy efficient than water electrolysis, using around 25 kWh per kg of H_2 produced, with further optimizations expected to reduce this to 18–20 kWh per kg of hydrogen.[192]

Circular carbon blacks. Derived from recycled carbon black oil sources like tyre or plastics pyrolysis, circular carbon blacks are also in development. Collaborative projects, such as the EU-funded Black Cycle project, aim to enhance their production. Cost-effective regular production of circular carbon blacks is expected by 2041.[191] In line with circular economy goals, tyre pyrolysis oil, a valuable byproduct of end of life tyre pyrolysis, contains key

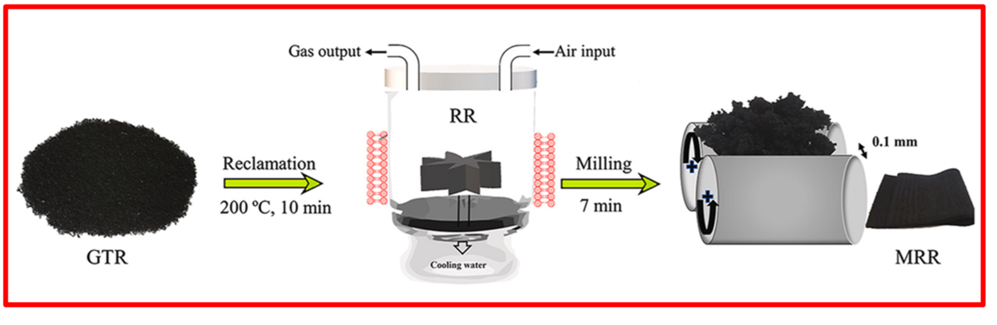

Fig. 9 Thermo oxidative exfoliation of carbon black from ground tyre rubber. [Reproduced from ref. 190 with permission from Elsevier, copyright 2021].

petrochemical building blocks like benzene, toluene, and limonene. Processes such as distillation and fluid catalytic cracking enable tyre pyrolysis oil derived products to meet hydrocarbon market demands, helping the tyre industry transition toward a circular economy and reduced reliance on fossil fuels[193]

Green carbon black (GCB). A scalable process was developed for producing "Green Carbon Black" (GCB) from biomass based pyrolysis oil (PO) using a spray pyrolysis method. At the optimum process temperature of 1300 °C, GCB exhibited structural similarities to medium disperse CB grades, including a cluster form macrostructure and paracrystalline, turbostratic microstructure. GCB produced at 1700 °C exhibited signs of graphitization, indicating a transition towards a graphitic structure. GCB's ability to mimic the structural properties of traditional carbon black, particularly its cluster macrostructure and paracrystalline microstructure, offers the potential to replace conventional CB in tyre applications without sacrificing key performance metrics such as durability and traction. These efforts could align the carbon black industry more closely with circular economy principles, fostering a more sustainable production process that leverages renewable resources effectively.[194–198,250]

3.4. Recycled fabrics

Recycled fabrics are playing an increasingly vital role in sustainable tyre manufacturing, offering both environmental and performance benefits. Among these, post-consumer recycled (PCR) polyester, derived from PET bottles, is the most widely used. Continental's ContiRe.Tex technology exemplifies this innovation, converting up to 15 PET bottles per tyre into high-strength polyester yarns without intermediate chemical steps, ensuring durability under tyre stress. Major tyre companies are working toward using 100% sustainable materials by 2050, and some have already started using these technologies in their factories across Europe. Other recycled fibers include pre-consumer recycled (PIR) polyester from industrial textile waste, recycled nylon from fishing nets and carpets, and recycled aramid from used tyre cords and body armor, all of which are used in structural components such as carcass plies, belt edge strips, and sidewall inserts. These materials contribute to reduced landfill waste and lower raw material costs while maintaining mechanical integrity. However, challenges remain in ensuring consistent quality and processing performance. Innovations such as high modulus low shrinkage PET (HMLS-PET) yarns produced from 10–100% recycled PET through solid state polymerization and controlled spinning have shown promise in meeting the stringent strength and shrinkage requirements of elastomeric products like tyres, while also reducing CO_2 emissions and conserving resources.[251]

4. Tyre manufacturing and engineering: towards sustainability

The tyre manufacturing industry, spearheaded by prominent companies such as Goodyear, Michelin, Bridgestone, and Nokian, is currently undergoing a significant shift towards sustainability. Emerging methods such as microbial etching further enhance the recyclability of scrap rubber by modifying the surface of rubber particles for better integration into various applications, such as bitumen. These innovations minimize energy consumption and promote resource efficiency. Additionally, bio-based oils derived from renewable sources are being employed as substitutes for traditional petroleum based oils in tyre formulations, further decreasing the environmental impact.

Artificial intelligence (AI) is playing a crucial role in this sustainable revolution, serving as a tool for reducing the carbon footprint at every stage of the tyre lifecycle. It assists in choosing eco-friendly materials, optimizing designs, improving manufacturing efficiency, managing supply chains, and handling tyre disposal. By leveraging AI, companies can streamline operations, reduce energy consumption, and improve the overall sustainability of their products.[252] Furthermore, the integration of life cycle assessment (LCA) principles is paramount in sustainable tyre production. LCA evaluates the environmental impact of the entire tyre production process, guiding design choices and optimization strategies to minimize ecological footprint while maximizing performance and safety.

Innovative technologies continue to drive sustainability in tyre engineering. Triboelectric tyres, equipped with triboelectric nanogenerators (TENGs), capture energy from friction to improve fuel efficiency and extend the range of electric vehicles.[253] Triboelectric tyres promise to enhance energy efficiency, reduce emissions, and boost vehicle performance sustainably. Vitrimers offer the potential for recyclable, reprocessable, and self-healing tyre materials, aligning perfectly with the industry's goals of waste reduction and improved material efficiency. As tyre manufacturers strive to meet ambitious sustainability targets, the adoption of vitrimers underscores their commitment to innovation and environmental protection.[254]

Innovations such as Michelin's Uptis (Unique Puncture-Proof Tyre System) exemplify the potential of 3D printed sustainable tyres to transform the automotive industry. Manufacturers like Goodyear, BigRep, Polaris, and Kenda Rubber are actively exploring airless tyre technology for various applications. These advancements in 3D printing and airless tyre technology represent a significant leap.[255] Utilizing nitrogen gas for tyre inflation is a practice that offers multiple benefits. Nitrogen's larger molecular size compared to oxygen leads to slower rates of diffusion through the tyre, maintaining optimal pressure for extended periods.

Tubeless tyres improve fuel efficiency through lower rolling resistance and reduce vehicle weight. Their durability is enhanced by better heat dissipation and increased resistance to punctures, which extends their service life and decreases the frequency of replacements. Innovations in tyre manufacturing and engineering underscore the industry's commitment to environmental sustainability. To complement the discussion on sustainable innovations, Table 3 presents a comparative overview of key material categories used in tyre manufacturing rubbers, fillers, fabrics, processing aids, recycled materials, and other components. It highlights conventional sources, renewable or sustainable alternatives, and the key benefits of

Table 3 Comparison of conventional and sustainable materials in tyre manufacturing

Material category	Currently used materials (source/type)	Renewable/sustainable alternatives (source/type)	Key benefits of alternatives
Rubbers	Natural rubber (*Hevea brasiliensis*), synthetic rubbers (petrochemical-derived, *e.g.*, SBR)	Natural rubber (Russian dandelion, guayule, lettuce, goldenrod, *Euphorbia characias* L., *Ficus* L. spp., *Alstonia boonei* De Wild, *Taraxacum mongolicum* Hand.-Mazz., and *Helianthus annuus* L.), bio-based synthetic rubbers (soybean oil-based elastomers, polyester elastomers, poly(diisoamyl itaconate-*co*-isoprene), β-myrcene-derived elastomers, lignin-derived elastomers, poly(dimethyl itaconate-*co*-butadiene)s, and *trans*-β-farnesene from sugar feedstocks)	Reduced reliance on petrochemicals, potentially lower carbon footprint, hypoallergenic options (guayule), and comparable performance
Fillers	Carbon black (incomplete combustion of coal tar, ethylene tar, and vegetable oil), crystalline silica (quartz, sand, and other minerals)	Recovered carbon black, green silica (agricultural residues: rice husk ash, bamboo leaves, sugarcane bagasse, and corn stalks), cellulose (woody materials and agricultural waste), lignin (paper and pulp industry), clay minerals (montmorillonite and sepiolite), carbon nanotubes (MWCNTs – though safety concerns exist for release during wear), N-doped carbon nanohorns (recycled waste tyre CBp), graphene (waste tyre CBp), soy particles, and eggshell powder (food industry waste)	Lower energy consumption and CO_2 emissions in production (green silica), utilization of waste materials, improved mechanical properties, and reduced rolling resistance
Fabrics	Nylon (petroleum-based) and polyester (petroleum-based)	Cellulose-derived rayon lyocell (regenerated cellulose fiber), engineered natural fibers, aramid fibers and aramid nanofibers (ANFs)	Renewable, potentially biodegradable, lower greenhouse gas emissions during production and disposal, and contributes to a circular economy
Processing aids	Rubber process oils (petroleum-based, *e.g.*, DAE)	Vegetable oils (tea oil, rice bran oil, soybean oil, palm oil, sunflower oil, coconut oil, castor oil, jatropha oil, modified soybean oil (MSO)), coconut shell extract (CSE), cardanol oil (Cdn) and Cdn-A zinc-free processing aids (ZFAs)	Reduced carbon emissions, lower carcinogenicity, improved processing characteristics, better filler dispersion, and enhanced aging and wear resistance
Recycled materials	Crumb rubber (ground tyre rubber – GTR), reclaim rubber in very small proportions	Devulcanized rubber synthesized *via* environmentally friendly and bio-based technologies (currently in nascent stages of development)	Reduces landfill accumulation, extends product lifespan, and promotes a circular economy
Other materials	Steel wire (conventional production)	Recycled steel (steel recovered from end of life products) added during the manufacturing process	Reduces demand for virgin resources, and lowers energy consumption and CO_2 emissions in steel production

adopting these alternatives in terms of environmental impact, performance, and circularity.

5. Challenges and future opportunities

The tyre industry deals with generation of vast quantities of waste tyres and heavy reliance on non-renewable resources. Additionally, the imbalance between natural rubber demand and supply poses obstacles to industry diversification and sustainability. Key challenges include maintaining rubber quality, scaling cultivation, optimizing resin utilization, and overcoming limited output. Furthermore, the development of bio-based polymers for tyre production requires extensive research to meet industry standards, alongside challenges in scaling up production and assessing performance in varied applications. Ensuring consistency in the quality and quantity of renewable fillers like rice husk silica (RHS) and addressing issues with cellulose and lignin utilization are vital for enhancing tyre sustainability. Additionally, compatibility and performance issues with bio-based processing and recycled rubber need attention for holistic sustainability in tyre manufacturing.

The tyre industry has the opportunity to drive sustainability by reducing waste, conserving resources, and exploring alternative materials. Utilizing renewable and recycled materials such as plant derived rubbers and non-petroleum based fillers presents a promising avenue. Furthermore, exploring alternative sources of natural rubber and embracing bio-based elastomers can mitigate environmental impact while meeting industry demands. Opportunities abound for innovation in tyre manufacturing, from utilizing RHS as a filler to exploring cellulose and lignin's potential. Integrating renewable processing aids and exploring hybrid fillers offer avenues for enhancing tyre performance and reducing the environmental footprint. Enhancing rubber material durability through self-healing mechanisms and optimizing tyre recycling methods are critical steps towards sustainability. Collaborations with stakeholders across the automotive industry are essential for driving sustainable practices and achieving shared goals. The tyre industry's shift towards sustainability not only aligns with global sustainability objectives but also presents opportunities for market differentiation and competitive advantage.

6. Conclusion

This comprehensive review critically explores and synthesizes the significant advancements and persistent challenges confronting the tyre industry in its critical transition towards sustainability. We have systematically explored a diverse array of renewable and bio-based materials, driven by the urgent need to diminish dependence on finite, petroleum derived inputs that contribute substantially to environmental and end of life challenges. This includes novel sources of natural rubber (*e.g.*, *Taraxacum koksaghyz* and *Parthenium argentatum*), bio-based synthetic rubbers (*e.g.*, derived from soybean oil, itaconic acid, β-myrcene, and *trans*-β-farnesene), and bio-based processing aids (*e.g.*, various vegetable oils like palm oil, soybean oil, and coconut shell extract). Furthermore, bio-based fabrics, derived from renewable cellulose (*e.g.*, rayon and lyocell) or engineered natural fibers, offer demonstrably greener and potentially biodegradable options that align seamlessly with circular economy principles, directly addressing the unsustainability of conventional nylon and polyester.

The review has also highlighted the innovative application of a wide spectrum of sustainable fillers. This encompasses the development of green silica from agricultural waste (*e.g.*, rice husk ash and bamboo leaves), the utilization of biopolymers like cellulose and lignin, and the integration of advanced carbon-based nanomaterials such as carbon nanotubes (MWCNTs), N-doped carbon nanohorns, graphene (often derived from waste tyre carbon black powder), and soy particles. The potential of eggshell powder as a sustainable filler has also been discussed. These materials not only offer environmental benefits through waste valorization and reduced energy consumption but also contribute to enhanced mechanical properties and reduced rolling resistance, crucial for meeting the tyre industry's "magic triangle" performance criteria.

A significant portion of our analysis focused on the strategic integration and technological evolution of recycled materials. This encompasses the increased utilization of recycled rubber in various forms (*e.g.*, crumb rubber and reclaim rubber), with ongoing research into novel compounding approaches and environmentally friendly technologies for its enhanced reincorporation. The critical role of recycled steel is detailed, demonstrating its re-integration into bead wires and tyre cords to reduce demand for virgin steel production and its associated high energy and CO_2 emissions. Crucially, advancements in recycled carbon black (rCB), including renewable carbon blacks, methane pyrolysis carbon blacks, and circular carbon blacks derived from tyre pyrolysis oil, represent a transformative shift towards a more sustainable carbon black industry, significantly improving the carbon footprint compared to conventional production. The development of "Green Carbon Black" (GCB) from biomass based pyrolysis oil further underscores this progress. Moreover, the increasing adoption of recycled fabrics, such as recycled polyester from PET bottles and recycled nylon from fishing nets, demonstrates a commitment to closing material loops.

Beyond material innovation, this review has underscored the pivotal role of advanced technologies in optimizing sustainability across the entire tyre lifecycle. Artificial Intelligence (AI) is increasingly applied to enhance material selection, optimize design, improve manufacturing efficiency, streamline supply chain management, and facilitate end-of-life handling, thereby reducing the overall carbon footprint. Life Cycle Assessment (LCA) principles are paramount, guiding design choices to minimize ecological impact while maximizing performance and safety. Emerging technologies such as triboelectric tyres, which capture energy from friction to improve fuel efficiency, and the increasing adoption of 3D printing for innovative tyre structures, promise enhanced energy efficiency and reduced material waste. We also highlighted innovative approaches such as using self-healing rubber and vitrimers, which offer the potential for recyclable, reprocessable, and self-repairing tyre materials, extending product durability and reducing waste. The use of nitrogen gas for tyre inflation and the development of tubeless tyres further contribute to improved fuel efficiency and extended tyre life.

Despite these promising developments, several significant knowledge gaps and challenges persist, which necessitate continued, rigorous research. These include:

(1) Long-term performance and durability: ensuring that novel bio-based and recycled materials maintain comparable or superior long-term mechanical performance, durability, and safety characteristics under diverse operational conditions.

(2) Comprehensive degradability and environmental interactions: a deeper understanding of the comprehensive degradability profiles and full environmental interactions of these new materials across their entire lifecycle, including potential microparticle release during wear and their ultimate fate in the environment.

(3) Supply-demand balance and quality consistency: addressing the imbalance between natural rubber demand and supply, and ensuring consistent quality and quantity of renewable fillers and recycled inputs, as feedstock variability can impact final product performance.

(4) Scalability and cost-effectiveness: overcoming challenges in scaling up the production of many bio-based alternatives and advanced recycled materials to meet industrial demands while maintaining cost-effectiveness compared to conventional materials.

(5) Processing and compatibility: further optimizing compounding techniques and surface modification methods to enhance the compatibility and dispersion of hydrophilic bio-based fillers (*e.g.*, cellulose and lignin) within hydrophobic rubber matrices, and improving the processability of recycled rubber.

(6) Standardization and regulation: developing standardized testing protocols and regulatory frameworks for new sustainable materials and recycled content to facilitate their broader adoption and market acceptance.

In conclusion, this review serves as a consolidated knowledge base, illuminating the tyre industry's determined progress towards sustainability. By embracing alternative materials, advancing recycling technologies, and leveraging cutting-edge innovations, the industry is well positioned to lead in environmental stewardship. Addressing the existing limitations through targeted research, fostering cross-disciplinary collaboration, and supporting conducive policy frameworks will be paramount to accelerate the journey toward a truly circular, sustainable, and environmentally responsible future. The commitment demonstrated by leading manufacturers to increase sustainable material content in their products signals a tangible shift, paving the way for a greener, more resilient global tyre economy.

Conflicts of interest

There are no conflicts of interest to declare.

Data availability

All data supporting this study are included within the main manuscript and/or the SI.

Fig. 4: various natural rubber plants (A) *Hevea brasiliensis* – image sourced from Eco Terra Beds blog (**https://ecoterrabeds.com/blogs/eco-terras-healthy-sleep-blog/all-about-hevea-brasiliensis**), used under open-source terms. (B) Russian dandelion – image accessed *via* Google Images from publicly available sources (**https://images.app.goo.gl/LfpPy**). (C) Guayule – image accessed *via* Google Images from publicly available sources (**https://images.app.goo.gl/azjJk**). (D) Lettuce – image accessed *via* Google Images from publicly available sources (**https://images.app.goo.gl/dgSni**). (E) Goldenrod – image accessed *via* Google Images from publicly available sources (**https://images.app.goo.gl/HaQEM**). See DOI: **https://doi.org/10.1039/d5su00177c**.

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