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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.

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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: 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.<sup>2,3</sup>

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

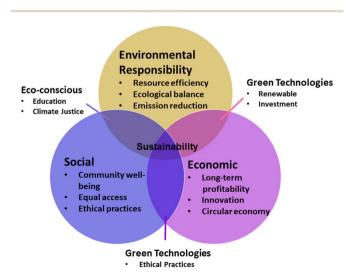


Fig. 1 Components of sustainability showing the interconnection between environmental, social, and economic pillars essential for sustainable development.

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". 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

Firstly, improperly disposed tyres can release chemicals into the air, ground, and water, contributing to pollution and altering ecosystems.<sup>8</sup> Tyres also do not decompose and can release methane gas and toxic smoke when burned.<sup>9,10</sup> Secondly, sustainability is crucial for reducing the financial and logistical burden of tyre disposal.<sup>11</sup> 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.<sup>12,13</sup>

The tyre industry is raw material intensive, accounting for 65% of production costs. <sup>14</sup> Sustainable materials are naturally abundant, easy to extract with minimal energy spent, and easy to recycle. <sup>15</sup> 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. <sup>16</sup>

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. <sup>17</sup> Natural rubber (NR) is a commonly used rubber in the tyre industry, but there is an imbalance between demand and supply. <sup>18</sup> 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. <sup>19,20</sup> Global tyre raw suppliers and manufacturers are currently researching sustainable materials to reduce petroleum-based raw material usage by 30%. <sup>14,15</sup>

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. <sup>14,21,22</sup> 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. <sup>16,23</sup> Tyre

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Classification of various sustainable raw materials used in the tyre industry

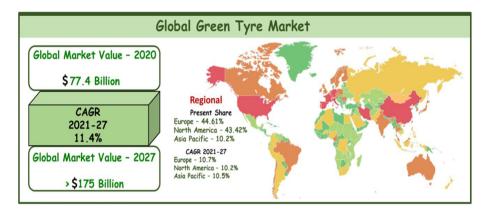


Fig. 3 Regional dynamics of the global green tyre market showing market shares and growth projections from 2021 to 2027. [Reproduced from ref. 20 with permission from Springer Nature, copyright 2022].

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

#### 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.<sup>32</sup>

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 highquality 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, Nova-BioRubber, and Sumitomo, among others use T. kok-saghyz as a source of rubber.<sup>36</sup> 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  $\times$  10<sup>5</sup> to 2.4  $\times$ 10<sup>5</sup> g mol<sup>-1</sup> 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.<sup>47</sup> While bio-based polymers like polylactide, polyhydroxyalkanoates, and poly(butylene succinate) have succeeded in various industrial applications,<sup>48–50</sup> their suitability for tyre production is not yet fully established.

Researchers have investigated using soybean oil-based elastomers, polyester elastomers, and poly(diisoamyl itaconate-coisoprene) elastomers. 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. 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

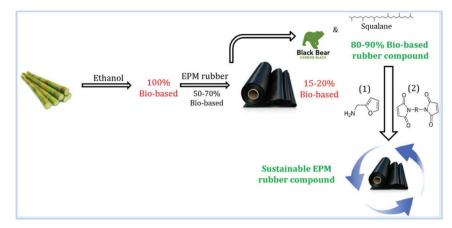


Fig. 6 The cross-linking reaction of the EPM rubber compound with carbon black and squalene. [Reproduced from ref. 71 with permission from Taylor & Francis, copyright 2019].

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.<sup>55,56</sup> 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. <sup>59,60</sup> 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.<sup>64</sup> 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.<sup>67</sup> These biobased 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<sup>-3</sup>. 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 crosslinked 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<sup>74–81</sup>

Property	Conventional silica	Green silica
Туре	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<sub>2</sub>) 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<sub>2</sub> 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 p-anhydroglucopyranose units and  $\beta$ -(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. <sup>100</sup> 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. <sup>101</sup> The molecular structure of cellulose is composed of repeating units of  $\beta$ -D-glucopyranose, connected by  $\beta$ -1,4-glycosidic linkages at the C1 and C4 positions. <sup>102–107</sup>

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". 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  $\alpha$ -cellulose concentration (5 parts per hundred rubber (phr)), but then decreased as the  $\alpha$ -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 ecofriendly 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 reagglomeration, 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. 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. 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. Laggshell 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. Late 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 petroleumbased oils in the rubber industry.<sup>154</sup> 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 ecofriendly tyre production. 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. Studies have

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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 oilbased 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.<sup>166</sup> 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

Comparison of virgin carbon black (vCB) vs. recovered carbon black (rCB)<sup>185–198</sup>

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<sup>-1</sup> compared to 9.7 N kN<sup>-1</sup> 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.<sup>173</sup>

#### 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. 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.<sup>184</sup> Straightforward manufacturing processes of rayon and lyocell and favourable properties make them attractive choices, as shown by their successful application in high performance tyres.

#### 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. The tyre industry generates a substantial volume of waste tyres, presenting significant environmental challenges. 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.<sup>209</sup> 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).<sup>217,218</sup> 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].

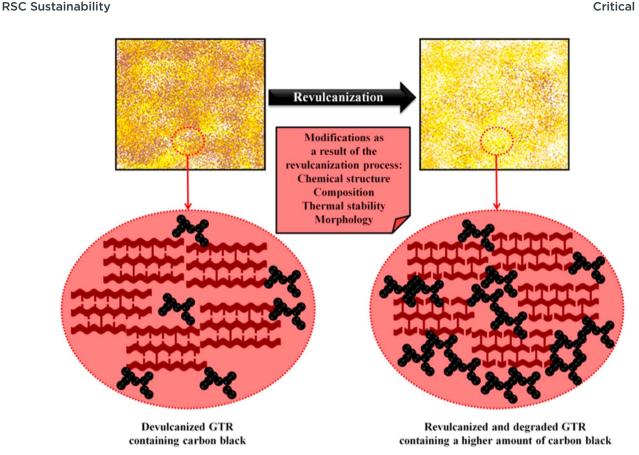
Reclaimed rubber, a valuable and sustainable material, has garnered significant attention as a key component in various innovative recycling approaches aimed at recycling waste tyre rubber.<sup>219</sup> Methods to transform reclaimed rubber encompass using reclaiming agents, advanced manufacturing techniques, and sustainable additives, offering promising solutions to enhance rubber properties, reduce waste, and promote ecofriendly practices within the rubber industry.<sup>220–223</sup> 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.<sup>224</sup> 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.<sup>225</sup> Incorporating silica into natural rubber reclaims rubber blends via sol-gel techniques, and enhanced reinforcement with increased reclaim rubber content.<sup>226</sup> 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.<sup>227</sup>

Rubber devulcanization involves the breaking of sulfursulfur 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.<sup>228</sup> 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].



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.<sup>233</sup> 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.<sup>230</sup> 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.<sup>241</sup> 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, 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. 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. <sup>186,187</sup> 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. 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<sup>191</sup> 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<sub>2</sub> produced, with further optimizations expected to reduce this to 18–20 kWh per kg of hydrogen.<sup>192</sup>

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. Costeffective regular production of circular carbon blacks is expected by 2041. In line with circular economy goals, tyre pyrolysis oil, a valuable byproduct of end of life tyre pyrolysis, contains key

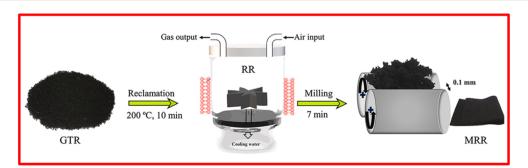


Fig. 9 Thermo oxidative exfoliation of carbon black from ground tyre rubber. [Reproduced from ref. 190 with permission from Elsevier, copyright 2021].

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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<sup>193</sup>

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 preconsumer 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 CO2 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. 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. <sup>253</sup> 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. <sup>254</sup>

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.<sup>255</sup> 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 ( <i>Hevea brasiliensis</i> ), synthetic rubbers (petrochemicalderived, <i>e.g.</i> , SBR)	Natural rubber (Russian dandelion, guayule, lettuce, goldenrod, <i>Euphorbia characias</i> L., <i>Ficus</i> L. spp., <i>Alstonia boonei</i> De Wild, <i>Taraxacum mongolicum</i> HandMazz., and <i>Helianthus annuus</i> L.), bio-based synthetic rubbers (soybean oilbased elastomers, polyester elastomers, poly(diisoamyl itaconate- <i>co</i> -isoprene), β-myrcenederived elastomers, lignin-derived elastomers, poly(dimethyl itaconate- <i>co</i> -butadiene)s, and <i>trans</i> -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 <sub>2</sub> 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 (petroleumbased, <i>e.g.</i> , 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 zincfree 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 <sub>2</sub> emissions in steel production

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

## 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 selfhealing 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 CO2 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 biobased 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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#### References

- 1 A. Patel and S. Singh, Implementing Circular Economy Strategies in the Automobile Industry – A Step toward Creating Sustainable Automobiles, *Benchmarking*, 2023, 30(7), 2225–2233, DOI: 10.1108/BIJ-08-2021-0486.
- 2 V. Toteva and K. Stanulov, Waste Tyres Pyrolysis Oil as a Source of Energy: Methods for Refining, *Prog. Rubber, Plast. Recycl. Technol.*, 2020, 36(2), 143–158, DOI: 10.1177/1477760619895026.
- 3 A. B. Kharissova, O. V. Kharissova, B. I. Kharisov and Y. Peña Méndez, Carbon negative footprint materials: a review, *Nano-Struct. Nano-Objects*, 2024, 37, 101100, DOI: 10.1016/j.nanoso.2024.101100.
- 4 F. Valentini and A. Pegoretti, End-of-Life Options of Tyres. A Review, *Adv. Ind. Eng. Polym. Res.*, 2022, 5(4), 203–213, DOI: 10.1016/j.aiepr.2022.08.006.
- 5 X. Yuan, C. W. Su, M. Umar, X. Shao and O. R. Lobonţ, The Race to Zero Emissions: Can Renewable Energy Be the Path to Carbon Neutrality?, *J. Environ. Manage.*, 2022, **308**, 114648, DOI: **10.1016/j.jenvman.2022.114648**.
- 6 A. Maevsky, L. Zhang, M. T. Nguyen and P. R. Jensen, Advanced climate modeling frameworks: state-of-the-art techniques, uncertainties, and the principle of responsibility, *Modeling Earth Systems and Environment*, 2025, 11, 331, DOI: 10.1007/s40808-025-02525-6.
- 7 Q. Tushar, J. Santos, G. Zhang, M. A. Bhuiyan and F. Giustozzi, Recycling Waste Vehicle Tyres into Crumb Rubber and the Transition to Renewable Energy Sources: A Comprehensive Life Cycle Assessment, *J. Environ. Manage.*, 2022, 323, 116289, DOI: 10.1016/ j.jenvman.2022.116289.
- 8 T. Weldeslassie, H. Naz, B. Singh and M. Oves, Chemical Contaminants for Soil, Air and Aquatic Ecosystem, in *Modern Age Environmental Problems and Remediation*, 2018, vol. 1, pp. 1–22, DOI: 10.1007/978-3-319-64501-8\_1.
- 9 A. T. Hoang, P. Murugesan, E. Pv, D. Balasubramanian, S. Parida, C. Priya Jayabal, M. Nachippan, M. A. Kalam, T. H. Truong, D. N. Cao and V. V. Le, Strategic Combination of Waste Plastic/Tyre Pyrolysis Oil with Biodiesel for Natural Gas-Enriched HCCI Engine: Experimental Analysis and Machine Learning Model, Energy, 2023, 280, 128233, DOI: 10.1016/ j.energy.2023.128233.
- 10 I. Glushankova, A. Ketov, M. Krasnovskikh, L. Rudakova and I. Vaisman, End of Life Tyres as a Possible Source of Toxic Substances Emission in the Process of Combustion, *Resources*, 2019, 8(2), 113, DOI: 10.3390/resources8020113.
- 11 J. Park, N. Díaz-Posada and S. Mejía-Dugand, Challenges in Implementing the Extended Producer Responsibility in an Emerging Economy: The End-of-Life Tyre Management in

- Colombia, *J. Cleaner Prod.*, 2018, **189**, 754–762, DOI: **10.1016/j.jclepro.2018.04.058**.
- 12 D. DeGroot and P. Hobart, Mexico-US Cross-Border Resolution of Waste Tyre Disposal, in *Restoring Our Natural Habitat*, 2007, pp. 1–13, DOI: 10.1061/40927(243)301.
- 13 C. Avilés-Palacios and A. Rodríguez-Olalla, The Sustainability of Waste Management Models in Circular Economies, *Sustainability*, 2021, 13, 7105, DOI: 10.3390/ su13137105.
- 14 S. Das, H. Satpathi, S. Roopa and S. Das Gupta, Sustainability of the Tyre Industry: Through a Material Approach, in *Applied Biopolymer Technology and Bioplastics*, Apple Academic Press, Waretown, NJ, 2021, pp. 53–98.
- 15 J. Thomas and R. Patil, The Road to Sustainable Tyre Materials: Current State-of-the-Art and Future Prospectives, *Environ. Sci. Technol.*, 2023, 57, 2209–2216, DOI: 10.1021/acs.est.2c07642.
- 16 J. Araujo-Morera, R. Verdejo, M. A. López-Manchado and M. H. Hernández Santana, Sustainable Mobility: The Route of Tyres through the Circular Economy Model, Waste Manage., 2021, 126, 309–322, DOI: 10.1016/ j.wasman.2021.03.025.
- 17 P. Sarkar and A. K. Bhowmick, Sustainable Rubbers and Rubber Additives, *J. Appl. Polym. Sci.*, 2018, 135, 45701, DOI: 10.1002/app.45701.
- 18 A. A. Khin, R. L. Leh Bin, O. C. Keong, F. Win Yie and N. J. Liang, Critical Factors of the Natural Rubber Price Instability in the World Market, *Humanities & Social Sciences Reviews*, 2019, 7, 199–208, DOI: 10.18510/ hssr.2019.7124.
- 19 W. Li, W. Peng, S. Ren and A. He, Synthesis and Characterization of Trans-1,4-Poly(Butadiene-co-Isoprene) Rubbers (TBIR) with Different Fraction and Chain Sequence Distribution and Its Influence on the Properties of Natural Rubber/TBIR/Carbon Black Composites, *Ind. Eng. Chem. Res.*, 2019, 58, 10609–10617, DOI: 10.1021/acs.iecr.9b01447.
- 20 V. Bijina, P. J. Jandas, S. Joseph, J. Gopu, K. Abhitha and H. John, Recent Trends in Industrial and Academic Developments of Green Tyre Technology, *Polym. Bull.*, 2022, 1–30, DOI: 10.1007/s00289-022-04445-2, published online.
- 21 N. R. Singha, M. Mahapatra, M. Karmakar and P. K. Chattopadhyay, Processing, Characterization and Application of Natural Rubber Based Environmentally Friendly Polymer Composites, in *Sustainable Polymer Composites and Nanocomposites*, 2019, pp. 855–897, DOI: 10.1007/978-3-030-05399-4\_29.
- 22 C. Luca, P. Francesco, A. Marco, H. Thomas and A. Enrico, Tyres for vehicles and elastomeric compositions for tyres comprising particular silicas from rice husk ash, *EP Pat.*, EP3802152B1, Pirelli Tyre Co., Ltd, 2018.
- 23 C. Luca, H. Thomas, Z. Luca, O. Marco and B. Davide, Tyre, and processes for preparing a predispersion of natural rubber and lignin and for making a cross-linkable

- elastomeric composition, *Br. Pat.*, BR112018012094B1, Pirelli Tyre Co., Ltd, 2015.
- 24 P. J. Martin, P. Brown, A. V. Chapman and S. Cook, Silica-Reinforced Epoxidized Natural Rubber Tyre Treads—Performance and Durability, *Rubber Chem. Technol.*, 2015, 88, 390–411, DOI: 10.5254/rct.15.85940.
- 25 B. P. Chang, A. Gupta, R. Muthuraj and T. H. Mekonnen, Bioresourced Fillers for Rubber Composite Sustainability: Current Development and Future Opportunities, *Green Chem.*, 2021, 23, 5337–5378, DOI: 10.1039/D1GC01115D.
- 26 G. F. C. Rodrigues and N. P. Oliveira, Impacts of Green Tyre Technology: Case Study of Environmental and Customer Perspectives, in *Water, Energy and Food Nexus in the Context of Strategies for Climate Change Mitigation*, 2020, pp. 271–285, DOI: 10.1007/978-3-030-57235-8 21.
- 27 K. P. Nair, Rubber (Hevea brasiliensis), in *Tree Crops*, Springer, Cham, 2021, pp. 287–332, DOI: 10.1007/978-3-030-62140-7 8.
- 28 C. C. Ho, The Production of Natural Rubber from Hevea brasiliensis Latex: Colloidal Properties, Preservation, Purification and Processing, in *Natural Rubber Materials*, *Vol. 1: Blends and IPN*, Royal Society of Chemistry, 2013, pp. 73–106, DOI: 10.1039/9781849737647-00073.
- 29 D. Rasutis, K. Soratana, C. McMahan and A. E. Landis, A Sustainability Review of Domestic Rubber from the Guayule Plant, *Ind. Crops Prod.*, 2015, **70**, 383–394, DOI: **10.1016/j.indcrop.2015.03.042**.
- 30 K. S. Chow, K. L. Wan, M. N. Isa, A. Bahari, S. H. Tan, K. Harikrishna and H. Y. Yeang, Insights into Rubber Biosynthesis from Transcriptome Analysis of Hevea brasiliensis Latex, *J. Exp. Bot.*, 2007, **58**, 2429–2440, DOI: **10.1093/jxb/erm093**.
- 31 S. Malavika and S. Sambhudevan, Natural Rubber and Gutta-Percha Rubber: Applications, in *Handbook of Biopolymers*, Springer Nature Singapore, Singapore, 2023, pp. 1–35.
- 32 W. Pootakham, C. Sonthirod, C. Naktang, P. Ruang-Areerate, T. Yoocha, D. Sangsrakru, K. Theerawattanasuk, R. Rattanawong, N. Lekawipat and S. Tangphatsornruang, De Novo Hybrid Assembly of the Rubber Tree Genome Reveals Evidence of Paleotetraploidy in Hevea species, *Sci. Rep.*, 2017, 7, 41457, DOI: 10.1038/srep41457.
- 33 S. Musto, V. Barbera, G. Guerra and M. Galimberti, Processing and Strain Induced Crystallization and Reinforcement under Strain of Poly(1,4-cis-Isoprene) from Ziegler–Natta Catalysis, Hevea brasiliensis, Taraxacum kok-saghyz and Parthenium argentatum, Adv. Ind. Eng. Polym. Res., 2019, 2, 1–12, DOI: 10.1016/j.aiepr.2018.08.003.
- 34 R. Zhao, G. Liu, R. Fu, J. Zhang, X. Jie, Y. Dong, Z. He and Q. Nie, Process Optimization of Green Aqueous-Based Extraction Technology of Taraxacum kok-saghyz Rubber, *Rubber Chem. Technol.*, 2022, **95**, 425–449, DOI: **10.5254/rct.22.77883**.
- 35 D. Rasutis, K. Soratana, C. McMahan and A. E. Landis, A Sustainability Review of Domestic Rubber from the Guayule Plant, *Ind. Crops Prod.*, 2015, **70**, 383–394, DOI: **10.1016/j.indcrop.2015.03.042**.

- 36 F. M. Jara, K. Cornish and M. Carmona, Potential Applications of Guayulins to Improve Feasibility of Guayule Cultivation, *Agronomy*, 2019, **9**, 1–11, DOI: 10.3390/agronomy9120804.
- 37 S. Cherian, S. B. Ryu and K. Cornish, *Plant Biotechnol. J.*, 2019, 17, 2041–2061, DOI: 10.1111/pbi.13181.
- 38 P. L. Eranki and A. E. Landis, Pathway to Domestic Natural Rubber Production: A Cradle-to-Grave Life Cycle Assessment of the First Guayule Automobile Tyre Manufactured in the United States, *Int. J. Life Cycle Assess.*, 2019, 24, 1348–1359, DOI: 10.1007/s11367-018-1572-3.
- 39 P. L. Eranki and A. E. Landis, Pathway to Domestic Natural Rubber Production: A Cradle-to-Grave Life Cycle Assessment of the First Guayule Automobile Tyre Manufactured in the United States, *Int. J. Life Cycle* Assess., 2019, 24, 1348–1359, DOI: 10.1007/s11367-018-1572-3.
- 40 C. L. Swanson, R. A. Buchanan and F. H. Otey, Molecular Weights of Natural Rubbers from Selected Temperate Zone Plants, *J. Appl. Polym. Sci.*, 1979, 23, 743–748, DOI: 10.1002/app.1979.070230309.
- 41 R. A. Sessa, M. H. Bennett, M. J. Lewis, J. W. Mansfield and M. H. Beale, Metabolite profiling of sesquiterpene lactones from Lactuca species. Major latex components are novel oxalate and sulfate conjugates of lactucin and its derivatives, *J. Biol. Chem.*, 2000, 275, 26877–26884, DOI: 10.1074/jbc.M000244200.
- 42 M. Kwon, C. L. Hodgins, E. M. Salama, K. R. Dias, A. Parikh, A. V. Mackey, K. F. Catenza, J. C. Vederas and D. K. Ro, New insights into natural rubber biosynthesis from rubberdeficient lettuce mutants expressing goldenrod or guayule cis-prenyltransferase, *New Phytol.*, 2023, 239, 1098–1111, DOI: 10.1111/nph.18994.
- 43 S. Reyes-Chin-Wo, Z. Wang, X. Yang, A. Kozik, S. Arikit, C. Song, L. Xia, L. Froenicke, D. O. Lavelle, M. J. Truco, R. Xia, S. Zhu, C. Xu, H. Xu, X. Xu, K. Cox, I. Korf, B. C. Meyers and R. W. Michelmore, Genome assembly with in vitro proximity ligation data and whole-genome triplication in lettuce, *Nat. Commun.*, 2017, 8, 14953, DOI: 10.1038/ncomms14953.
- 44 G. J. Seiler, M. E. Carr and M. O. Bagby, Renewable resources from wild sunflowers (Helianthus spp., Asteraceae), *Econ. Bot.*, 1991, 45, 4–15, DOI: 10.1007/BF02860045.
- 45 H. Badouin, J. Gouzy, C. J. Grassa, F. Murat, S. E. Staton, L. Cottret, C. Lelandais-Brière, G. L. Owens, S. Carrère, B. Mayjonade, L. Legrand, N. Gill, N. C. Kane, J. E. Bowers, S. Hubner, A. Bellec, A. Bérard, H. Bergès, N. Blanchet, M. C. Boniface, D. Brunel, O. Catrice, N. Chaidir, C. Claudel, C. Donnadieu, T. Faraut, G. Fievet, N. Helmstetter, M. King, S. J. Knapp, Z. Lai, M. C. Le Paslier, Y. Lippi, L. Lorenzon, J. R. Mandel, G. Marage, G. Marchand, E. Marquand, E. Bret-Mestries, E. Morien, S. Nambeesan, T. Nguyen, P. Pegot-Espagnet, N. Pouilly, F. Raftis, E. Sallet, T. Schiex, J. Thomas, C. Vandecasteele, D. Varès, F. Vear, S. Vautrin, M. Crespi, B. Mangin,

- J. M. Burke, J. Salse, S. Muños, P. Vincourt, L. H. Rieseberg and N. B. Langlade, The sunflower genome provides insights into oil metabolism, flowering and asterid evolution, *Nature*, 2017, **546**, 148–152, DOI: **10.1038/nature22380**.
- 46 T. Lin, X. Xu, H. L. Du, X. L. Fan, Q. W. Chen, C. Y. Hai, Z. J. Zhou, X. Su, L. Q. Kou, Q. Gao, L. W. Deng, J. S. Jiang, H. L. You, Y. H. Ma, Z. K. Cheng, G. D. Wang, C. Z. Liang, G. M. Zhang, H. Yu and J. Y. Li, Extensive sequence divergence between the reference genomes of Taraxacum kok-saghyz and Taraxacum mongolicum, *Sci. China: Life Sci.*, 2022, 65, 515–528, DOI: 10.1007/s11427-021-2033-2.
- 47 M. Burelo, A. Martínez, J. D. Hernández-Varela, T. Stringer, M. Ramírez-Melgarejo, A. Y. Yau, G. Luna-Bárcenas and C. D. Treviño-Quintanilla, Recent developments in synthesis, properties, applications and recycling of bio-based elastomers, *Molecules*, 2024, 29(2), 387, DOI: 10.3390/molecules29020387.
- 48 A. Corma, S. Iborra and A. Velty, Chemical routes for the transformation of biomass into chemicals, *Chem. Rev.*, 2007, **107**, 2411–2502, DOI: **10.1021/cr050989d**.
- 49 Z. Li and X. J. Loh, Water Soluble Polyhydroxyalkanoates: Future Materials for Therapeutic Applications, *Chem. Soc. Rev.*, 2015, 44(10), 2865–2879, DOI: 10.1039/c5cs00089k.
- 50 M. Habib Ullah, M. Mahadi Dusselier, P. Van Wouwe, A. Dewaele, P. A. Jacobs and B. F. Sels, Green chemistry. Shape-selective zeolite catalysis for bioplastics production, *Science*, 2015, 349, 78–80, DOI: 10.1126/science.aaa7169.
- 51 M. H. Ullah and T. A. Latef, Aerogel poly(butylene succinate) biomaterial substrate for RF and microwave applications, *Sci. Rep.*, 2015, 5, 12868, DOI: 10.1038/srep12868.
- 52 Z. Wang, X. Zhang, R. Wang, H. Kang, B. Qiao, J. Ma, L. Zhang and H. Wang, Synthesis and characterization of novel soybean-oil-based elastomers with favorable processability and tunable properties, *Macromolecules*, 2012, 45, 9010–9019, DOI: 10.1021/ma301938a.
- 53 R. van Lith, E. K. Gregory, J. Yang, M. R. Kibbe and G. A. Ameer, Engineering biodegradable polyester elastomers with antioxidant properties to attenuate oxidative stress in tissues, *Biomaterials*, 2014, 35, 8113–8122, DOI: 10.1016/j.biomaterials.2014.06.004.
- 54 W. Lei, X. Zhou, T. P. Russell, K.-c. Hua, X. Yang, H. Qiao, W. Wang, F. Li, R. Wang and L. Zhang, High performance bio-based elastomers: energy efficient and sustainable materials for tyres, *J. Mater. Chem. A*, 2016, 4, 13058–13062, DOI: 10.1039/C6TA05001H.
- 55 E. Breitmaier, Terpenes: Flavors, Fragrances, Pharmaca, Pheromones, *Appl. Organomet. Chem.*, 2006, **21**, 377, DOI: **10.1002/aoc.1209**.
- 56 U. Biermann, U. Bornscheuer, M. A. R. Meier, J. O. Metzger and H. J. Schäfer, Oils and fats as renewable raw materials in chemistry, *Angew. Chem., Int. Ed. Engl.*, 2011, 50, 3854–3871, DOI: 10.1002/anie.201002767.

- 57 A. Behr and L. Johnen, Myrcene as a natural base chemical in sustainable chemistry: a critical review, *ChemSusChem*, 2009, 2, 1072–1095, DOI: 10.1002/cssc.200900186.
- 58 P. Sarkar and A. K. Bhowmick, Synthesis, characterization and properties of a bio-based elastomer: polymyrcene, *RSC Adv.*, 2014, 4, 61343–61354, DOI: 10.1039/C4RA09475A.
- 59 P. Sarkar and A. K. Bhowmick, Green approach toward sustainable polymer: synthesis and characterization of poly(myrcene-co-dibutyl itaconate), ACS Sustain. Chem. Eng., 2016, 4, 2129–2141, DOI: 10.1021/ acssuschemeng.5b01591.
- 60 P. Sarkar and A. K. Bhowmick, Terpene based sustainable elastomer for low rolling resistance and improved wet grip application: synthesis, characterization and properties of poly(styrene-co-myrcene), *ACS Sustain. Chem. Eng.*, 2016, **4**, 5462–5474, DOI: **10.1021/acssuschemeng.6b01038**.
- 61 P. Sarkar and A. K. Bhowmick, Terpene based sustainable methacrylate copolymer series by emulsion polymerization: synthesis and structure-property relationship, *J. Polym. Sci., Part A: Polym. Chem.*, 2017, 55, 2639–2649, DOI: 10.1002/pola.28661.
- 62 M. Li, H. Chen, C. Liu, J. Guo, X. Xu, H. Zhang, R. Nian and M. Xian, Improvement of isoprene production in Escherichia coli by rational optimization of RBSs and key enzymes screening, *Microb. Cell Fact.*, 2019, 18, 4, DOI: 10.1186/s12934-018-1051-3.
- 63 L. Yuan, Y. Zhang, Z. Wang, Y. Han and C. Tang, Plant oil and lignin-derived elastomers via thermal azide–alkyne cycloaddition click chemistry, ACS Sustain. Chem. Eng., 2019, 7, 2593–2601, DOI: 10.1021/acssuschemeng.8b05617.
- 64 J. Zhang, J. Lu, K. Su, D. Wang and B. Han, Bio-based β-myrcene-modified solution-polymerized styrene-butadiene rubber for improving carbon black dispersion and wet skid resistance, *J. Appl. Polym. Sci.*, 2019, **136**, 48159, DOI: **10.1002/app.48159**.
- 65 A. Khan, L. K. Kian, M. Jawaid, A. A. P. Khan, M. M. Alotaibi, A. M. Asiri and H. M. Marwani, Preparation of styrene– butadiene rubber (SBR) composite incorporated with collagen-functionalized graphene oxide for green tyre application, *Gels*, 2022, 8, 161, DOI: 10.3390/gels8030161.
- 66 X. Zhou, H. Ji, G. H. Hu, R. Wang and L. Zhang, A solvent-less green synthetic route toward a sustainable bio-based elastomer: design, synthesis, and characterization of poly(dibutyl itaconate-co-butadiene), *Polym. Chem.*, 2019, **10**, 6131–6144, DOI: **10.1039/C9PY01393H**.
- 67 H. Ji, H. Yang, X. Zhou, C. Sun, L. Li, S. Zhao, J. Yu, S. Li, R. Wang and L. Zhang, Preparation of bio-based elastomer and its nanocomposites based on dimethyl itaconate with versatile properties, *Composites, Part B*, 2023, 248, 110383, DOI: 10.1016/j.compositesb.2022.110383.
- 68 M. J. Syu, Biological production of 2,3-butanediol, *Appl. Microbiol. Biotechnol.*, 2001, 55, 10–18, DOI: 10.1007/s002530000486
- 69 J. P. Nirmala, B. Kumar and S. Ananda Kumar, Development and characterization of polysaccharides/

- carrageenan based biofilms, *J. Biorem. Biodegrad.*, 2017, **8**, 6, DOI: 10.4172/2155-6199-C1-011.
- 70 S. Fang, S. Wu, J. Huang, D. Wang, Z. Tang, B. Guo and L. Zhang, Notably improved dispersion of carbon black for high-performance natural rubber composites via triazolinedione click chemistry, *Ind. Eng. Chem. Res.*, 2020, 59, 21047–21057, DOI: 10.1021/acs.iecr.0c04242.
- 71 L. M. Polgar, N. Migliore, F. Picchioni and M. Van Duin, Sustainable EPM rubber compounds, *Polym.-Plast. Technol. Mater.*, 2020, **59**, 1379–1385, DOI: **10.1080**/ 25740881.2020.1738480.
- 72 P. Sahu, P. Sarkar and A. K. Bhowmick, Design of a molecular architecture via a green route for an improved silica reinforced nanocomposite using bioresources, ACS Sustain. Chem. Eng., 2018, 6, 6599–6611, DOI: 10.1021/acssuschemeng.8b00383.
- 73 S. Steven, I. C. Sophiana, Z. Murti, M. Mulyono, R. Y. H. Sinaga, N. Nadirah, E. S. A. Soekotjo, Y. Ramli, P. Hernowo, P. Pasymi, E. Restiawaty and Y. Bindar, A simple material and energy input–output performance in evaluating silica production from conventional, fume, and biomass thermochemical conversion routes, *Waste Biomass Valorization*, 2024, 15, 2705–2720, DOI: 10.1007/s12649-023-02348-5.
- 74 S. He, Y. Huang, G. Chen, M. Feng, H. Dai, B. Yuan and X. Chen, Effect of heat treatment on hydrophobic silica aerogel, J. Hazard. Mater., 2019, 362, 294–302, DOI: 10.1016/j.jhazmat.2018.08.087.
- 75 Y. Shen, Rice husk silica-derived nanomaterials for battery applications: a literature review, *J. Agric. Food Chem.*, 2017, 65, 995–1004, DOI: 10.1021/acs.jafc.6b04777.
- 76 J. Chun and J. H. Lee, Recent progress on the development of engineered silica particles derived from rice husk, Sustainability, 2020, 12, 10683, DOI: 10.3390/su122410683.
- 77 N. Sapawe and M. F. Hanafi, Production of silica from agricultural waste, *Archives of Organic and Inorganic Chemical Sciences*, 2018, 3, 342–343, DOI: 10.32474/AOICS.2018.03.000160.
- 78 R. Abolghasemi, M. Haghighi, M. Solgi and A. Mobinikhaledi, Rapid synthesis of ZnO nanoparticles by waste thyme (Thymus vulgaris L.), *Int. J. Environ. Sci. Technol.*, 2019, **16**, 6985–6990, DOI: **10.1007/s13762-018-2112-1**.
- 79 M. Nazar, A. Yasar, S. A. Raza, A. Ahmad, R. Rasheed, M. Shahbaz and A. B. Tabinda, Techno-economic and environmental assessment of rice husk in comparison to coal and furnace oil as a boiler fuel, *Biomass Convers. Biorefin.*, 2023, 13, 1671–1679, DOI: 10.1007/s13399-020-01238-3.
- 80 A. L. Jembere and S. W. Fanta, Studies on the synthesis of silica powder from rice husk ash as reinforcement filler in rubber tyre tread part: replacement of commercial precipitated silica, *Int. J. Mater. Sci. Appl.*, 2017, **6**, 37–44, DOI: 10.11648/j.ijmsa.20170601.16.
- 81 M. Lolage, P. Parida, M. Chaskar, A. Gupta and D. Rautaray, Green silica: industrially scalable & sustainable approach towards achieving improved "nano filler-elastomer"

- interaction and reinforcement in tyre tread compounds, Sustainable Mater. Technol., 2020, 26, e00232, DOI: 10.1016/j.susmat.2020.e00232.
- 82 N. S. Chundawat, B. S. Parmar, A. S. Deuri, D. Vaidya, S. Jadoun, P. Zarrintaj, M. Barani and N. P. S. Chauhan, Rice husk silica as a sustainable filler in the tyre industry, Arabian J. Chem., 2022, 15, 104086, DOI: 10.1016/ j.arabjc.2022.104086.
- 83 B. Shoul, Y. Marfavi, B. Sadeghi, E. Kowsari, P. Sadeghi and S. Ramakrishna, Investigating the potential of sustainable use of green silica in the green tyre industry: a review, Environ. Sci. Pollut. Res. Int., 2022, 29, 51298-51317, DOI: 10.1007/s11356-022-20894-8.
- 84 K. Wudy, D. Drummer, F. Kühnlein and M. Drexler, Influence of degradation behavior of polyamide 12 powders in laser sintering process on produced parts, AIP Conf. Proc., 2014, 691-695, DOI: 10.1063/1.4873873.
- 85 B. Yoon, S. Kim, A. Lang, C. Egelkamp, J. Meier, U. Giese, B. Kim, J. H. Kim, J. W. Bae, G. Y. Um, S. H. Kim, D. I. Kim, S. J. Kim and J. Suhr, Friction behaviors of rice husk silica-reinforced elastomer composites in contact with rough self-affine surfaces, Polym. Test., 2022, 116, 107764, DOI: 10.1016/j.polymertesting.2022.107764.
- 86 D. Miyashiro, R. Hamano and K. Umemura, A review of applications using mixed materials of cellulose, nanocellulose and carbon nanotubes, Nanomaterials, 2020, 10, 186, DOI: 10.3390/nano10020186.
- 87 M. Mehdinia, M. Farajollah Pour, H. Yousefi, A. Dorieh, A. J. Lamanna and E. Fini, Developing bio-nano composites using cellulose-nanofiber-reinforced epoxy, J. Compos. Sci., 2024, 8, 250, DOI: 10.3390/jcs8070250.
- 88 J. Shojaeiarani, D. Bajwa and A. Shirzadifar, A review on cellulose nanocrystals as promising biocompounds for the synthesis of nanocomposite hydrogels, Carbohydr. Polym., 2019, 216, 247-259, DOI: 10.1016/ j.carbpol.2019.04.033.
- 89 C. M. Obele, M. I. Ejimofor, C. U. Atuanya and M. E. Ibenta, Cassava stem cellulose (CSC) nanocrystal for optimal methylene blue biosorption with response surface design, Curr. Res. Green Sustainable Chem., 2021, 4, 100067, DOI: 10.1016/j.crgsc.2021.100067.
- 90 S. H. Hassan, L. H. Voon, T. S. Velayutham, L. Zhai, H. C. Kim, J. Kim, J. W. Bae, G. Y. Um, S. H. Kim, D. I. Kim, S. J. Kim and J. Suhr, Review of cellulose smart material: biomass conversion process and progress on cellulose-based electroactive paper, J. Renewable Mater., 2018, 6, 1-25, DOI: 10.7569/JRM.2017.634173.
- 91 S. Ummartyotin and H. Manuspiya, A critical review on cellulose: from fundamental to an approach on sensor technology, Renewable Sustainable Energy Rev., 2015, 41, 402-412, DOI: 10.1016/j.rser.2014.08.050.
- 92 V. K. Thakur and M. K. Thakur, Processing and characterization of natural cellulose fibers/thermoset polymer composites, Carbohydr. Polym., 2014, 109, 102-117, DOI: 10.1016/j.carbpol.2014.03.039.
- 93 H. L. Teo and R. A. Wahab, Towards an eco-friendly deconstruction of agro-industrial biomass

- preparation of renewable cellulose nanomaterials: a review, Int. J. Biol. Macromol., 2020, 161, 1414-1430, DOI: 10.1016/j.ijbiomac.2020.08.076.
- 94 W. Jiang, P. Shen, J. Yi, L. Li, C. Wu and J. Gu, Surface modification of nanocrystalline cellulose and application in natural rubber composites, J. Appl. Polym. Sci., 2020, 137(39), e49163, DOI: 10.1002/app.49163.
- 95 N. M. F. Hakimi, S. H. Lee, W. C. Lum, S. F. Mohamad, S. S. Osman Al Edrus, B. D. Park and A. Azmi, Surface modified nanocellulose and its reinforcement in natural rubber matrix nanocomposites: a review, Polymers, 2021, 13(19), 3241, DOI: 10.3390/polym13193241.
- 96 O. Somseemee, P. Saeoui, F. T. Schevenels, et al., Enhanced interfacial interaction between modified cellulose nanocrystals and epoxidized natural rubber via ultraviolet irradiation, Sci. Rep., 2022, 12, 6682, DOI: 10.1038/s41598-022-10558-5.
- 97 S. Singh, G. L. Dhakar, B. P. Kapgate, P. K. Maji, C. Verma, M. Chhajed, K. Rajkumar and C. Das, Synthesis and chemical modification of crystalline nanocellulose to reinforce natural rubber composites, Polym. Adv. Technol., 2020, 31(12), 3059-3069, DOI: 10.1002/pat.5030.
- 98 L. Cao, Z. Gong, C. Xu and Y. Chen, Mechanically strong and recyclable rubber nanocomposites with sustainable cellulose nanocrystals and interfacial exchangeable bonds, ACS Sustainable Chem. Eng., 2021, 9(28), 9409-9417, DOI: 10.1021/acssuschemeng.1c02581.
- 99 S. R. Karnati, B. Høgsaa, L. Zhang and E. H. Fini, Developing carbon nanoparticles with tunable morphology and surface chemistry use construction, Constr. Build. Mater., 2020, 262, 120780, DOI: 10.1016/j.conbuildmat.2020.120780.
- 100 K. Roy, A. Pongwisuthiruchte, S. C. Chandra Debnath and P. Potiyaraj, Application of cellulose as green filler for the development of sustainable rubber technology, Green Sustainable Chem., 2021, 4, 100140, DOI: 10.1016/ j.crgsc.2021.100140.
- 101 J. George and S. N. Sabapathi, Cellulose nanocrystals: synthesis, functional properties, and applications, Nanotechnol., Sci. Appl., 2015, 8, 45-54, DOI: 10.2147/ NSA.S64386.
- 102 A. Mandal and D. Chakrabarty, Isolation of nanocellulose waste sugarcane bagasse (SCB) and its characterization, Carbohydr. Polym., 2011, 86(3), 1291-1299, DOI: 10.1016/j.carbpol.2011.06.030.
- 103 L. Ludueña, D. Fasce, V. A. Alvarez and P. M. Stefani, Nanocellulose from rice husk following alkaline treatment to remove silica, BioResources, 2011, 6(2), 1440-1453, DOI: 10.15376/biores.6.2.1440-1453.
- 104 F. Jiang and Y. L. Hsieh, Cellulose nanocrystal isolation from tomato peels and assembled nanofibers, Carbohydr. Polym., 2015, 122, 60-68, DOI: 10.1016/ j.carbpol.2014.12.064.
- 105 E. Kalita, B. K. Nath, F. Agan, V. More and P. Deb, Isolation and characterization of crystalline, autofluorescent, cellulose nanocrystals from saw dust wastes, Ind. Crops

*Prod.*, 2015, 65, 550–555, DOI: 10.1016/j.indcrop.2014.10.004.

**RSC Sustainability** 

- 106 M. K. M. Haafiz, A. Hassan, H. P. S. Abdul Khalil, A. F. Owolabi, M. M. Marliana, I. Arjmandi Inuwa, M. R. N. Fazita and M. H. Hussin, Cellulose nanowhiskers from oil palm empty fruit bunch biomass as green fillers, in *Cellulose-Reinforced Nanofibre Composites: Production, Properties and Applications*, 2017, pp. 241–259, DOI: 10.1016/B978-0-08-100957-4.00010-3.
- 107 N. Muhd Julkapli and S. Bagheri, Nanocellulose as a green and sustainable emerging material in energy applications: a review, *Polym. Adv. Technol.*, 2017, 28(12), 1583–1594, DOI: 10.1002/pat.4074.
- 108 M. Dominic, R. Joseph, P. M. Sabura Begum, B. P. Kanoth, J. Chandra and S. Thomas, Green tyre technology: effect of rice husk derived nanocellulose (RHNC) in replacing carbon black (CB) in natural rubber (NR) compounding, *Carbohydr. Polym.*, 2020, 230, 115620, DOI: 10.1016/ j.carbpol.2019.115620.
- 109 M. Haghighat, A. Zadhoush and S. N. Khorasani, Physicomechanical properties of alpha-cellulose-filled styrene-butadiene rubber composites, *J. Appl. Polym. Sci.*, 2005, **96**(6), 2203–2211, DOI: **10.1002/app.21691**.
- 110 K. Pal, S. G. Chowdhury, D. Mondal, D. Chattopadhyay, S. K. Bhattacharyya and R. Mukhopadhyay, Impact of α-cellulose as a green filler on physico-mechanical properties of a solution grade styrene-butadiene rubber based tyre-tread compound, *Polym. Eng. Sci.*, 2021, 61(12), 3017–3028, DOI: 10.1002/pen.25814.
- 111 Z. Yang, C. Savari and M. Barigou, Numerical and experimental investigations of horizontal turbulent particle-liquid pipe flow, *Ind. Eng. Chem. Res.*, 2022, 61(32), 12040–12051, DOI: 10.1021/acs.iecr.2c02183.
- 112 C. Xu, R. A. Arancon, J. Labidi and R. Luque, Lignin depolymerisation strategies: towards valuable chemicals and fuels, *Chem. Soc. Rev.*, 2014, 43(22), 7485–7500, DOI: 10.1039/c4cs00235k.
- 113 B. Khoury, T. Lecomte, G. Fortin, M. Masse, P. Therien, V. Bouchard, M.-A. Chapleau, K. Paquin S. Hofmann, Mindfulness-based therapy: comprehensive meta-analysis, ClinicalPsychology a Review, 2013, 33(6), 763-771, DOI: 10.1016/ j.cpr.2013.05.005.
- 114 K. P. R. Dandamudi, T. Murdock, P. J. Lammers, S. Deng and E. H. Fini, Production of functionalized carbon from synergistic hydrothermal liquefaction of microalgae and swine manure, *Resour., Conserv. Recycl.*, 2021, 170, 105564, DOI: 10.1016/j.resconrec.2021.105564.
- 115 H. Guo, B. Zhang, Z. Qi, C. Li, J. Ji, T. Dai, A. Wang and T. Zhang, Valorization of lignin to simple phenolic compounds over tungsten carbide: impact of lignin structure, *ChemSusChem*, 2017, **10**(3), 523–532, DOI: **10.1002/cssc.201601326**.
- 116 T. J. Szalaty, L. Klapiszewski and T. Jesionowski, Recent developments in modification of lignin using ionic liquids for the fabrication of advanced materials—

- a review, *J. Mol. Liq.*, 2020, **301**, 112417, DOI: **10.1016**/j.molliq.2019.112417.
- 117 H. Lange, S. Decina and C. Crestini, Oxidative upgrade of lignin recent routes reviewed, *Eur. Polym. J.*, 2013, 49(6), 1151–1173, DOI: 10.1016/j.eurpolymj.2013.03.002.
- 118 L. A. Zevallos Torres, A. Lorenci Woiciechowski, V. O. de Andrade Tanobe, S. G. Karp, L. C. Guimarães Lorenci, C. Faulds and C. R. Soccol, Lignin as a potential source of high-added value compounds: a review, *J. Cleaner Prod.*, 2020, 263, 121499, DOI: 10.1016/j.jclepro.2020.121499.
- 119 G. de Gonzalo, D. I. Colpa, M. H. M. Habib and M. W. Fraaije, Bacterial enzymes involved in lignin degradation, *J. Biotechnol.*, 2016, 236, 110–119, DOI: 10.1016/j.jbiotec.2016.08.011.
- 120 J. H. Lora and W. G. Glasser, Recent industrial applications of lignin: a sustainable alternative to non-renewable materials, *J. Polym. Environ.*, 2002, **10**(1/2), 39–48, DOI: **10.1023/A:1021070006895**.
- 121 M. Mariana, T. Alfatah, H. P. S. Abdul Khalil, E. B. Yahya, N. G. Olaiya, A. Nuryawan, E. M. Mistar, C. K. Abdullah, S. N. Abdulmadjid and H. Ismail, A current advancement on the role of lignin as sustainable reinforcement material in biopolymeric blends, *J. Mater. Res. Technol.*, 2021, 15, 2287–2316, DOI: 10.1016/j.jmrt.2021.08.139.
- 122 M. Lawoko, L. Berglund and M. Johansson, Lignin as a renewable substrate for polymers: from molecular understanding and isolation to targeted applications, *ACS Sustain. Chem. Eng.*, 2021, **9**(16), 5481–5485, DOI: **10.1021**/ **acssuschemeng.1c01741**.
- 123 J. Qiu, S. Yuan, H. Xiao, J. Liu, T. Shen, Z. Tan, W. Zhuang, H. Ying, M. Li and C. Zhu, Study on lignin amination for lignin/SiO<sub>2</sub> nano-hybrids towards sustainable natural rubber composites, *Int. J. Biol. Macromol.*, 2023, 233, 123547, DOI: 10.1016/j.ijbiomac.2023.123547.
- 124 E. Adler, Lignin chemistry—past, present and future, *Wood Sci. Technol.*, 1977, 11(3), 169–218, DOI: 10.1007/BF00365615.
- 125 S. Hait, D. De, P. Ghosh, J. Chanda, R. Mukhopadhyay, S. Dasgupta, A. Sallat, M. Al Aiti, K. W. Stöckelhuber, S. Wießner, G. Heinrich and A. Das, Understanding the coupling effect between lignin and polybutadiene elastomer, *J. Compos. Sci.*, 2021, 5(6), 154–169, DOI: 10.3390/jcs5060154.
- 126 R. Shorey, A. Gupta and T. H. Mekonnen, Hydrophobic modification of lignin for rubber composites, *Ind. Crops Prod.*, 2021, 174, 114189, DOI: 10.1016/ j.indcrop.2021.114189.
- 127 S. Hait, D. De, A. K. Ghosh, M. Al Aiti, P. Ghosh, J. Chanda, R. Mukhopadhyay, S. Dasgupta, S. Wießner, G. Heinrich and A. Das, Treasuring waste lignin as superior reinforcing filler in high cis-polybutadiene rubber: a direct comparative study with standard reinforcing silica and carbon black, *J. Cleaner Prod.*, 2021, 299, 126841, DOI: 10.1016/j.jclepro.2021.126841.
- 128 Z. He, Y. Li, C. Liu, Y. Li, M. Qian, Y. Zhu and X. Wang, Controllable conversion of biomass to lignin–silica hybrid nanoparticles: high-performance renewable dual-phase

- fillers, Waste Manage., 2021, 135, 381–388, DOI: 10.1016/j.wasman.2021.09.025.
- 129 M. G. A. Vieira, M. A. Da Silva, L. O. Dos Santos and M. M. Beppu, Natural-based plasticizers and biopolymer films: a review, *Eur. Polym. J.*, 2011, 47(3), 254–263, DOI: 10.1016/j.eurpolymj.2010.12.011.
- 130 D. Barana, S. D. Ali, A. Salanti, M. Orlandi, L. Castellani, T. Hanel and L. Zoia, Influence of lignin features on thermal stability and mechanical properties of natural rubber compounds, ACS Sustain. Chem. Eng., 2016, 4(10), 5258–5267, DOI: 10.1021/acssuschemeng.6b00774.
- 131 H. C. Chenette and S. M. Husson, Membrane adsorbers comprising grafted glycopolymers for targeted lectin binding, *J. Appl. Polym. Sci.*, 2015, **132**(21), 1–7, DOI: **10.1002**/app.41437.
- 132 N. A. Mohamad Aini, N. Othman, M. H. Hussin, K. Sahakaro and N. Hayeemasae, Lignin as alternative reinforcing filler in the rubber industry: a review, *Front. Mater.*, 2020, 6, 329, DOI: 10.3389/fmats.2019.00329.
- 133 G. A. Carpenedo, N. B. Guerra, M. Giovanela, M. A. D. De Paoli and J. Crespo, Evaluation of lignin as stabilizer in vulcanized natural rubber formulations, *Polímeros*, 2022, 32(3), e2022036, DOI: 10.1590/0104-1428.20220077.
- 134 D. N. Bikiaris, Nanocomposites of aliphatic polyesters: an overview of the effect of different nanofillers on enzymatic hydrolysis and biodegradation of polyesters, *Polym. Degrad. Stab.*, 2013, 98, 1908–1928, DOI: 10.1016/j.polymdegradstab.2013.05.016.
- 135 S. K. Kim, J. Kang, Y. Choe and Y. Chang, Structure and properties of the organoclay filled NR/BR nanocomposites, *Macromol. Res.*, 2006, **14**(2), 187–193, DOI: **10.1007/BF03218507**.
- 136 M. Kodal, N. Yazıcı Çakır, R. Yıldırım, N. Karakaya and G. Özkoç, Improved heat dissipation of NR/SBR-based tyre tread compounds via hybrid fillers of multi-walled carbon nanotube and carbon black, *Polymers*, 2023, 15(23), 4503, DOI: 10.3390/polym15234503.
- 137 S. Hou, Z. Xie, D. Zhang, B. Yang, Y. Lei and F. Liang, High-purity graphene and carbon nanohorns prepared by base-acid treated waste tyres carbon via direct current arc plasma, *Environ. Res.*, 2023, 238(1), 117071, DOI: 10.1016/j.envres.2023.117071.
- 138 A. Diekmann, M. C. V. Omelan, U. Giese and V. Rose, Carbon nanohorn-based NBR hybrid nanocomposites, *Rubber Chem. Technol.*, 2020, 93(4), 615–632, DOI: 10.5254/rct.20.79958.
- 139 J. R. Potts, D. R. Dreyer, C. W. Bielawski and R. S. Ruoff, Graphene-based polymer nanocomposites, *Polymer*, 2011, 52(1), 5–25, DOI: 10.1016/j.polymer.2010.11.042.
- 140 C. Wang, D. Li, T. Zhai, H. Wang, Q. Sun and H. Li, Direct conversion of waste tyres into three-dimensional graphene, *Energy Storage Mater.*, 2019, 23, 499–507, DOI: 10.1016/ j.ensm.2019.04.014.
- 141 K. P. Jibin, V. Prajitha and S. Thomas, Silica–graphene oxide reinforced rubber composites, *Mater. Today: Proc.*, 2021, 34(2), 502–505, DOI: 10.1016/j.matpr.2020.03.100.

- 142 V. Prajitha, K. P. Jibin, V. K. Abitha, K. S. Sisanth, M. Huskic, A. P. Meera, J. S. George and S. Thomas, Advancing mechanical performance in sustainable engineering: synergistic effects of graphene oxide/nanosilica hybrid nanofiller via latex coagulation in natural rubber composites, *Polym. Compos.*, 2024, 45(7), 5980–5991, DOI: 10.1002/pc.28174.
- 143 L. Jong, Improved natural rubber composites reinforced with a complex filler network of biobased nanoparticles and ionomer, *Mater. Chem. Phys.*, 2018, **203**, 156–165, DOI: **10.1016/j.matchemphys.2017.09.067**.
- 144 C. S. Barrera and J. L. Tardiff, Static and dynamic properties of eggshell filled natural rubber composites for potential application in automotive vibration isolation and damping, *J. Cleaner Prod.*, 2022, 353, 131656, DOI: 10.1016/j.jclepro.2022.131656.
- 145 A. Bodaghi, An overview on the recent developments in reactive plasticizers in polymers, *Polym. Adv. Technol.*, 2020, 31(3), 355–367, DOI: 10.1002/pat.4790.
- 146 F. van Elburg, F. Grunert, C. Aurisicchio, M. di Consiglio, R. di Ronza, A. Talma, P. Bernal-Ortega and A. Blume, Exploring the impact of bio-based plasticizers on the curing behavior and material properties of a simplified tire-tread compound, *Polymers*, 2024, 16, 1880, DOI: 10.3390/polym16131880.
- 147 S. Dasgupta, S. L. Agrawal, S. Bandyopadhyay, S. Chakraborty, R. Mukhopadhyay, R. K. Malkani and S. C. Ameta, Characterisation of eco-friendly processing aids for rubber compound: part II, *Polym. Test.*, 2008, 27(3), 277–283, DOI: 10.1016/j.polymertesting.2007.11.004.
- 148 M. Alexander and E. T. Thachil, A comparative study of cardanol and aromatic oil as plasticizers for carbon-black-filled natural rubber, *J. Appl. Polym. Sci.*, 2006, **102**(5), 4835–4841, DOI: **10.1002/app.24811**.
- 149 M. Alexander and E. T. Thachil, A comparative study of cardanol and aromatic oil as plasticizers for carbon-black-filled natural rubber, *J. Appl. Polym. Sci.*, 2006, **102**(5), 4835–4841, DOI: **10.1002/app.24811**.
- 150 C. Bueno-Ferrer, M. C. Garrigós and A. Jiménez, Characterization and thermal stability of poly(vinyl chloride) plasticized with epoxidized soybean oil for food packaging, *Polym. Degrad. Stab.*, 2010, 95(11), 2207–2212, DOI: 10.1016/j.polymdegradstab.2010.01.027.
- 151 W. G. D. Jayewardhana, G. M. Perera, D. G. Edirisinghe and L. Karunanayake, Study on natural oils as alternative processing aids and activators in carbon black filled natural rubber, *J. Natl. Sci. Found. Sri Lanka*, 2009, 37(3), 187–193, DOI: 10.4038/jnsfsr.v37i3.1212.
- 152 J. Yang, S. R. Lu, L. L. Pan, Q. Y. Luo, L. F. Song, L. Y. Wu and J. H. Yu, Effect of epoxidized soybean oil grafted poly(12-hydroxy stearate) on mechanical and thermal properties of microcrystalline cellulose fibers/polypropylene composites, *Polym. Bull.*, 2017, 74(4), 911–930, DOI: 10.1007/s00289-016-1753-9.
- 153 T. Hayashi and N. Kato, Rubber composition for studless tyre and studless tyre using the same, *US Pat.*, 9132698B2, Sumitomo Rubber Industries, Ltd, 2008.

154 C. Hayichelaeh and K. Boonkerd, Utilization of palm oil as an alternative processing oil in carbon black-filled natural rubber compounds, *Ind. Crops Prod.*, 2023, **194**, 116270, DOI: **10.1016/j.indcrop.2023.116270**.

**RSC Sustainability** 

- 155 Z. Wang, Y. Peng, L. Zhang, Y. Zhao, R. Vyzhimov, T. Tan and H. Fong, Investigation of palm oil as green plasticizer on the processing and mechanical properties of ethylene propylene diene monomer rubber, *Ind. Eng. Chem. Res.*, 2016, 55(10), 2784–2789, DOI: 10.1021/acs.iecr.5b04527.
- 156 C. K. Lee, J. G. Seo, H. J. Kim and S. H. Song, Novel green composites from styrene butadiene rubber and palm oil derivatives for high performance tyres, *J. Appl. Polym. Sci.*, 2019, **136**(25), 47672, DOI: **10.1002/app.47672**.
- 157 K. Sahakaro and A. Beraheng, Epoxidized natural oils as the alternative safe process oils in rubber compounds, *Rubber Chem. Technol.*, 2011, **84**(2), 200–214, DOI: **10.5254**/1.3577518.
- 158 N. Aprianti, A. Kismanto, N. K. Supriatna, L. K. T. Nainggoaln, R. I. Purawiardi, O. Fariza, F. J. Ermada, P. Zuldian, A. A. Rakosdewanto and R. Alamsyah, Prospect and challenges of producing carbon black from oil palm biomass: a review, Bioresour. 101587, DOI: 10.1016/ Technol. Rep., 2023, 23, j.biteb.2023.101587.
- 159 Y. M. Shashidhara and S. R. Jayaram, Vegetable oils as a potential cutting fluid—an evolution, *Tribol. Int.*, 2010, 43(5-6), 1073–1081, DOI: 10.1016/j.triboint.2009.12.065.
- 160 S. Moresco, M. Giovanela, L. N. Carli and J. S. Crespo, Development of passenger tyre treads: reduction in zinc content and utilization of a bio-based lubricant, *J. Cleaner Prod.*, 2016, 117, 199–206, DOI: 10.1016/ j.jclepro.2016.01.013.
- 161 C. Siriwong, P. Khansawai, S. Boonchiangma, C. Sirisinha and P. S. Sae-Oui, The influence of modified soybean oil as processing aids in tyre application, *Polym. Bull.*, 2021, 78(7), 3589–3606, DOI: 10.1007/s00289-020-03296-z.
- 162 J. Tardiff, C. M. Flanigan and L. Beyer, Evaluation of soy oils and fillers in automotive rubber, in *Soy-Based Chemicals and Materials*, ACS Symposium Series, 2014, ch. 14, vol. 1178, pp. 315–356, DOI: 10.1021/bk-2014-1178.ch014.
- 163 C.-H. Kang, W.-B. Jung, H.-J. Kim and H.-T. Jung, Highly enhanced tyre performance achieved by using combined carbon nanotubes and soybean oil, *J. Appl. Polym. Sci.*, 2021, 138(10), e49945, DOI: 10.1002/app.49945.
- 164 H. Xu, T. Fan, N. Ye, W. Wu, D. Huang, D. Wang, Z. Wang and L. Zhang, Plasticization effect of bio-based plasticizers from soybean oil for tyre tread rubber, *Polymers*, 2020, 12(3), 623, DOI: 10.3390/polym12030623.
- 165 P. Nun-Anan, C. Hayichelaeh and K. Boonkerd, Effect of a natural processing aid on the properties of acrylonitrilebutadiene rubber: study on soybean oil fatty acid from seed crop, *Polymers*, 2021, 13(20), 3459, DOI: 10.3390/ polym13203459.
- 166 S. Sahoo, D. Basu, A. Kumar, M. Nawale, S. Kadam,
  A. Bhujbal, K. Rajkumar, A. Bhowmick and
  S. Chattopadhyay, Bio-based oil derived from waste coconut shell: a potential additive for enhancing

- silanization in silica filled styrene butadiene copolymer, *J. Polym. Res.*, 2022, **29**(8), 311, DOI: **10.1007/s10965-022-03168-2**.
- 167 K. J. S. Brito, C. J. Mauss, P. Cofferri and M. M. de Camargo Forte, Sustainable plasticizer from agroindustrial waste for natural rubber compounds: influence on the curing system and compound properties, *J. Elastomers Plast.*, 2023, 55(3), 409–425, DOI: 10.1177/00952443221150762.
- 168 A. Bardha, S. Prasher, J. Villarta, M. S. Francis, C. Y. Khirpin, J. J. Mehlem and M.-J. Dumont, Nut shell and grain husk waste biochar as carbon black replacements in styrene-butadiene rubber composites and improvements through steam activation, *Ind. Crops Prod.*, 2023, 203, 117180, DOI: 10.1016/j.indcrop.2023.117180.
- 169 D. Lee and S. H. Song, A study of silica reinforced rubber composites with eco-friendly processing aids for pneumatic tyres, *Macromol. Res.*, 2019, 27(9), 850–856, DOI: 10.1007/s13233-019-7125-1.
- 170 N. C. Kim and S. H. Song, Effects of zinc-free processing aids on silica-reinforced tread compounds for green tyres, *Int. J. Polym. Sci.*, 2019, **2019**, 1–9, DOI: **10.1155/2019/9123635**.
- 171 C. J. Brinker, Hydrolysis and condensation of silicates: effects on structure, *J. Non-Cryst. Solids*, 1988, **100**(1–3), 31–50, DOI: **10.1016**/0022-3093(88)90005-1.
- 172 B. K. Coltrain and L. W. Kelts, The chemistry of hydrolysis and condensation of silica sol–gel precursors, *Adv. Chemother.*, 1994, 403–418, DOI: 10.1021/ba-1994-0234.ch019.
- 173 J. G. Seo, C. K. Lee, D. Lee and S. H. Song, High-performance tyres based on graphene coated with Zn-free coupling agents, *J. Ind. Eng. Chem.*, 2018, **66**, 78–85, DOI: **10.1016/j.jiec.2018.04.015**.
- 174 C. Nakason, A. Kaesaman and K. Eardrod, Cure and mechanical properties of natural rubber-g-poly(methyl methacrylate)-cassava starch compounds, *Mater. Lett.*, 2005, 59(29–30), 4020–4025, DOI: 10.1016/j.matlet.2005.07.057.
- 175 C. Liu, Y. Shao and D. Jia, Chemically modified starch reinforced natural rubber composites, *Polymer*, 2008, 49(8), 2176–2181, DOI: 10.1016/j.polymer.2008.03.005.
- 176 H. Tang, Q. Qi, Y. Wu, G. Liang, L. Zhang and J. Ma, Reinforcement of elastomer by starch, *J. Mater. Eng.*, 2006, **291**(6), 629–637, DOI: **10.1002/mame.200600033**.
- 177 J. H. Lora, M. J. Trojan and W. H. Klingensmith, Rubber compositions containing high purity lignin derivatives, *US Pat.*, 5196460A, Repap Technologies Inc., 1993.
- 178 C. Jiang, H. He, H. Jiang, L. Ma and D. M. Jia, Nano-lignin filled natural rubber composites: Preparation and characterization, *eXPRESS Polym. Lett.*, 2013, 7(5), 480–493, DOI: 10.3144/expresspolymlett.2013.44.
- 179 G. Wypych, *Encyclopedia of Polymer and Rubber Additives*, ChemTech Publishing, Toronto, 2024, ISBN 978-1-77467-028-6 (print), ISBN 978-1-77467-029-3 (eBook).
- 180 S. Bhadra, N. Mohan and S. Nair, Suitability of different biomaterials for the application in tyre, *SN Appl. Sci.*, 2019, 1(12), 1554, DOI: 10.1007/s42452-019-1625-7.

- 181 J. Jung and H. A. Sodano, Cellulose nanocrystal functionalized aramid nanofiber reinforced rubber compounds for tyre tread application, Cellulose, 2022, 29(14), 7735-7749, DOI: 10.1007/s10570-022-04716-1.
- 182 X. Jiang, Y. Bai, X. Chen and W. Liu, A review on raw materials, commercial production and properties of Lyocell fiber, J. Bioresour. Bioprod., 2020, 5(1), 16-25, DOI: 10.1016/j.jobab.2020.03.002.
- 183 I.-H. Kwon, S.-M. Choi, W.-S. Yang, et al., Lyocell multifilament, EP1500724A1, https:// EPPat., patents.google.com/patent/EP1500724A1/en.
- 184 W. Huang, Y. Li, H. Zhao, W. Wang, B. Yu, N. Ning, M. Tian and L. Zhang, A new eco-friendly dipping system for PA66 fiber cords/rubber composites with strong interfacial adhesion and good fatigue stability, Composites, Part B, 2023, 253, 110541, DOI: 10.1016/ j.compositesb.2023.110541.
- 185 A. Jain and S. K. Tripathi, Fabrication and characterization of energy storing supercapacitor devices using coconut shell based activated charcoal electrode, Mater. Sci. Eng., B, 2014, 183, 54-60, DOI: 10.1016/j.mseb.2013.12.004.
- 186 S. Khodabakhshi, P. F. Fulvio and E. Andreoli, Carbon black reborn: Structure and chemistry for renewable energy harnessing, Carbon, 2020, 162, 604-649, DOI: 10.1016/j.carbon.2020.02.058.
- 187 W. Urrego-Yepes, N. Cardona-Uribe, C. A. Vargas-Isaza and J. D. Martínez, Incorporating the recovered carbon black produced in an industrial-scale waste tyre pyrolysis plant into a natural rubber formulation, J. Environ. Manage., 2021, 287, 112292, DOI: 10.1016/j.jenvman.2021.112292.
- 188 S. Balbay, Effects of recycled carbon-based materials on tyre, J. Mater. Cycles Waste Manage., 2020, 22(6), 1768-1779, DOI: 10.1007/s10163-020-01064-9.
- 189 A. A. Hassan, Z. Zhang, K. Formela and S. Wang, Thermooxidative exfoliation of carbon black from ground tyre rubber as potential reinforcement in green tyres, Compos. Technol., 2021, 214, 108991, DOI: 10.1016/ j.compscitech.2021.108991.
- 190 M. Sagar, K. Nibedita, N. Manohar, K. R. Kumar, S. Suchismita, A. Pradnyesh, A. B. Reddy, E. R. Sadiku, U. N. Gupta, P. Lachit and J. Jayaramudu, A potential utilization of end-of-life tyres as recycled carbon black in EPDM rubber, Waste Manage., 2018, 74, 110-122, DOI: 10.1016/j.wasman.2018.01.003.
- 191 P. Toth, T. Vikström, R. Molinder and H. Wiinikka, Structure of carbon black continuously produced from biomass pyrolysis oil, Green Chem., 2018, 20(17), 3981-3992, DOI: 10.1039/C8GC01539B.
- 192 F. Laurent, R. Vandad-Julien, W. Elliott, H. Ned and D. Enoch, An energy-efficient plasma methane pyrolysis process for high yields of carbon black and hydrogen, Int. J. Hydrogen Energy, 2023, 48(8), 2920-2928, DOI: 10.1016/ j.ijhydene.2022.10.144.
- 193 F. Campuzano, J. D. Martínez, A. F. Agudelo Santamaría, S. M. Sarathy and W. L. Roberts, Pursuing the end-of-life tyre circularity: an outlook toward the production of secondary raw materials from tyre pyrolysis oil, Energy

- DOI: Fuels, 2023, 37(13), 8836-8866, 10.1021/ acs.energyfuels.3c00847.
- 194 R. Kumar, R. Kumar Singh and D. Pratap Singh, Natural and waste hydrocarbon precursors for the synthesis of carbon-based nanomaterials: graphene and CNTs, Renewable Sustainable Energy Rev., 2016, 58, 976-1006, DOI: 10.1016/j.rser.2015.12.120.
- 195 P. McKendry, Energy production from biomass (part 2): conversion technologies, Bioresour. Technol., 2002, 83(1), 47-54, DOI: 10.1016/S0960-8524(01)00119-500119-5.
- 196 O. Das, A. K. Sarmah and D. Bhattacharyya, A novel approach in organic waste utilization through biochar addition in wood/polypropylene composites, Manage., 2015, 38, 132-140, DOI: 10.1016/ j.wasman.2015.01.015.
- 197 S.-H. Lee, J.-H. Kim and H.-H. Park, Upcycling green carbon black as a reinforcing agent for styrene-butadiene rubber materials, RSC Adv., 2022, 12(47), 30480-30486, DOI: 10.1039/d2ra05299g.
- 198 J. Choi, J. Kang, H. Yang, S. Yoon, J.-H. Kim and H.-H. Park, Enhancing uptake capability of green carbon black recycled from scrap tyres for water purification, Coatings, 2024, 14, 389, DOI: 10.3390/coatings14040389.
- 199 S. M. Cadwell, R. A. Merrill, C. M. Sloman and F. L. Yost, Rubber in the automotive industry, from the viewpoint of the rubber technologist, Rubber Chem. Technol., 1941, 14(2), 378-385, DOI: 10.5254/1.3540034.
- 200 G. Gaustad, M. Krystofik, M. Bustamante and K. Badami, Circular economy strategies for mitigating critical material supply issues, Resour., Conserv. Recycl., 2018, 135, 24-33, DOI: 10.1016/j.resconrec.2017.08.002.
- 201 S. Jadoun, Synthesis, mechanism, and applications of selfhealing materials, Biomed. Mater. & Devices, 2024, 2, 225-240, DOI: 10.1007/s44174-023-00107-7.
- 202 K. Choi, A. Noh, J. Kim, P. H. Hong, M. J. Ko and S. W. Hong, Properties and applications of self-healing polymeric materials: a review, Polymers, 2023, 15, 4408, DOI: 10.3390/polym15224408.
- 203 S. Mandal, M. Malanin, B. Ghanti, S. Banerjee, J. Sawada, T. Tada, G. Heinrich, S. Wießner and A. Das, Design of sacrificial network in modified natural rubber leads to strikingly improved mechanical performance with selfhealing capability, Chem. Eng. J., 2023, 474, 145838, DOI: 10.1016/j.cej.2023.145838.
- 204 S. Utrera-Barrios, R. Verdejo, M. A. López-Manchado and M. Hernández Santana, Evolution of self-healing elastomers, from extrinsic to combined intrinsic mechanisms: a review, Mater. Horiz., 2020, 7(11), 2882-2902, DOI: 10.1039/D0MH00535E.
- 205 S. J. Garcia, Effect of polymer architecture on the intrinsic self-healing character of polymers, Eur. Polym. J., 2014, 53, 118-125, DOI: 10.1016/j.eurpolymj.2014.01.026.
- 206 C. Xu, L. Cao, B. Lin, X. Liang and Y. Chen, Design of selfhealing supramolecular rubbers by introducing ionic crosslinks into natural rubber via a controlled vulcanization, ACS Appl. Mater. Interfaces, 2016, 8(27), 17728-17737, DOI: 10.1021/acsami.6b05941.

- 207 A. Fazli and D. Rodrigue, Recycling waste tyres into ground tyre rubber (GTR)/rubber compounds: a review, *J. Compos. Sci.*, 2020, 4(3), 103, DOI: 10.3390/jcs4030103.
- 208 Y. Hou and W. Wu, Derived from corn straw cellulose: modified used tyre rubber powder composites, *Cellulose*, 2022, 29(7), 3935–3945, DOI: 10.1007/s10570-022-04549-y.
- 209 X. Colom, M. Marín-Genescà, R. Mujal, K. Formela and J. Cañavate, Structural and physico-mechanical properties of natural rubber/GTR composites devulcanized by microwaves: influence of GTR source and irradiation time, *J. Compos. Mater.*, 2018, 52(22), 3099–3108, DOI: 10.1177/0021998318761554.
- 210 J. Araujo-Morera, S. Utrera-Barrios, R. Doral Olivares, M. de los Reyes Verdugo Manzanares, M. Ángel López-Manchado, R. Verdejo and M. H. Santana, Solving the dichotomy between self-healing and mechanical properties in rubber composites by combining reinforcing and sustainable fillers, *Mater. Eng.*, 2022, 307, 2200261, DOI: 10.1002/ mame.202200261.
- 211 I. B. Joohari and F. Giustozzi, Waste tyres crumb rubber as a sustainability enhancer for polymer-modified and hybrid polymer-modified bitumen, *Int. J. Pavement Eng.*, 2022, 23(12), 4357–4371, DOI: 10.1080/10298436.2021.1943745.
- 212 M. Sienkiewicz, H. Janik, K. Borzędowska-Labuda and J. Kucińska-Lipka, Environmentally friendly polymerrubber composites obtained from waste tyres: a review, *J. Cleaner Prod.*, 2017, **147**, 560–571, DOI: **10.1016**/j.jclepro.2017.01.121.
- 213 K. F. Abo Elenien, N. A. Azab, G. Bassioni and M. H. Abdellatif, The effect of tyre rubber particles on the mechanical and physical properties of polyester, *IOP Conf. Ser.: Mater. Sci. Eng.*, 2020, 973(1), 1–11, DOI: 10.1088/1757-899X/973/1/012019.
- 214 J. Araujo-Morera, R. Verdugo-Manzanares, S. González, R. Verdejo, M. A. Lopez-Manchado and S. Hernández, On the use of mechano-chemically modified ground tyre rubber (GTR) as recycled and sustainable filler in styrene-butadiene rubber (SBR) composites, *J. Compos. Sci.*, 2021, 5(3), 68, DOI: 10.3390/jcs5030068.
- 215 M. H. Hernández Santana, M. Huete, P. Lameda, J. Araujo, R. Verdejo, M. A. López-Manchado and S. Hernández, Design of a new generation of sustainable SBR compounds with good trade-off between mechanical properties and self-healing ability, *Eur. Polym. J.*, 2018, **106**, 273–283, DOI: **10.1016/j.eurpolymj.2018.07.040**.
- 216 S. Utrera-Barrios, J. Araujo-Morera, L. Pulido de Los Reyes, R. Verdugo Manzanares, R. Verdejo, M. Á. López-Manchado and M. H. Hernández Santana, An effective and sustainable approach for achieving self-healing in nitrile rubber, *Eur. Polym. J.*, 2020, 139, 110032, DOI: 10.1016/j.eurpolymj.2020.110032.
- 217 B. E. Rogachuk and J. A. Okolie, Waste tyres based biorefinery for biofuels and value-added materials production, *Chem. Eng. J. Adv.*, 2023, **14**, 100476, DOI: **10.1016/j.ceja.2023.100476**.
- 218 S. Luo and Y. Feng, The production of fuel oil and combustible gas by catalytic pyrolysis of waste tyre using

- waste heat of blast-furnace slag, *Energy Convers. Manage.*, 2017, **136**, 27–35, DOI: **10.1016/j.enconman.2016.12.076**.
- 219 Z. Tang, Y. Liu, Q. Huang, J. Zhao, B. Guo, L. Zhang and A. Real, Recycling loop of sulfur-cured rubber through transalkylation exchange of C-S bonds, *Green Chem.*, 2018, 20(24), 5454–5458, DOI: 10.1039/C8GC02932F.
- 220 K. Aoudia, S. Azem, N. A. Aït Hocine, M. Gratton, V. Pettarin and S. Seghar, Recycling of waste tyre rubber: microwave devulcanization and incorporation in a thermoset resin, *Waste Manage.*, 2017, 60, 471–481, DOI: 10.1016/j.wasman.2016.10.051.
- 221 T. Garima and S. P. Singh, Part III strategies: application of bioremediation on solid waste management: a review, *J. Biorem. Biodegrad.*, 2014, 5, 248, DOI: 10.4172/2155-6199.1000248.
- 222 K. Formela, M. Klein, X. Colom and M. R. Saeb, Investigating the combined impact of plasticizer and shear force on the efficiency of low temperature reclaiming of ground tyre rubber (GTR), *Polym. Degrad. Stab.*, 2016, 125, 1–11, DOI: 10.1016/j.polymdegradstab.2015.12.022.
- 223 S. Ghorai, S. Bhunia, M. Roy and D. De, Mechanochemical devulcanization of natural rubber vulcanizate by dual function disulfide chemicals, *Polym. Degrad. Stab.*, 2016, 129, 34–46, DOI: 10.1016/j.polymdegradstab.2016.03.008.
- 224 H. Tseng, Z. Y. L. Lin, S.-H. Chen, W.-H. Lai and M.-Y. Wey, Reuse of reclaimed tyre rubber for gas-separation membranes prepared by hot-pressing, *J. Cleaner Prod.*, 2019, 237, 117739, DOI: 10.1016/j.jclepro.2019.117739.
- 225 D. De, P. K. Panda, M. Roy, S. Bhunia and A. I. Jaman, Reinforcing effect of nanosilica on the properties of natural rubber/reclaimed ground rubber tyre vulcanizates, *Polym. Eng. Sci.*, 2013, 53(2), 227–237, DOI: 10.1002/pen.23255.
- 226 A. A. Hassan, K. Formela and S. Wang, Reclaimed rubber in situ grafted with soybean oil as a novel green reactive plasticizer in SBR/silica compounds, *ACS Sustain. Chem. Eng.*, 2019, 7(17), 14991–15001, DOI: 10.1021/acssuschemeng.9b03339.
- 227 E. Akca, A. Gursel and N. Sen, A review on devulcanization of waste tyre rubber, *Nat. Sci.*, 2018, **6**(1), 154–160, DOI: **10.21533/pen.v6i1.167**.
- 228 R. Ghosh, C. Mani, R. Krafczyk, R. Schnell, A. Paasche, A. Talma, A. Blume and W. K. Dierkes, New route of tyre rubber devulcanization using silanes, *Polymers*, 2023, 15(13), 2848, DOI: 10.3390/polym15132848.
- 229 D. Á. Simon and T. Bárány, Microwave devulcanization of ground tyre rubber and its improved utilization in natural rubber compounds, *ACS Sustain. Chem. Eng.*, 2023, **11**(5), 1797–1808, DOI: **10.1021/acssuschemeng.2c05984**.
- 230 F. D. B. de Sousa, A. Zanchet and C. H. Scuracchio, From devulcanization to revulcanization: challenges in getting recycled tyre rubber for technical applications, *ACS Sustain. Chem. Eng.*, 2019, 7(9), 8755–8765, DOI: 10.1021/acssuschemeng.9b00655.
- 231 J. S. Oh, S. Ghose and A. I. Isayev, Effects of ultrasonic treatment on unfilled butadiene rubber, *J. Polym. Sci.*,

Critical Review

- Part B: Polym. Phys., 2003, 41(22), 2959-2968, DOI: 10.1002/ polb.10606.
- 232 R. Pérez-Campos, J. Fayos-Fernández, J. Monzó-Cabrera, F. Martín Salamanca, J. López Valentín, J. M. Catalá-Civera, P. Plaza-González and J. R. Sánchez-Marín, Dynamic permittivity measurement of ground-tyre rubber during microwave-assisted devulcanization, Polymers, 2022, 14(17), 3543, DOI: 10.3390/polym14173543.
- 233 L. E. Alonso Pastor, K. C. Núñez Carrero, J. Araujo-Morera, M. Hernández Santana, J. M. Pastor, J. López Valentín, J. M. Catalá-Civera, P. Plaza-González and J. R. Sánchez-Marín, Setting relationships between structure and devulcanization of ground tyre rubber and their effect on self-healing elastomers, Polymers, 2021, 14(1), 11, DOI: 10.3390/polym14010011.
- 234 F. Cataldo, Thermochemistry of sulfur-based vulcanization and of devulcanized and recycled natural rubber compounds, Int. J. Mol. Sci., 2023, 24(3), 2623, DOI: 10.3390/ijms24032623.
- 235 S. Ghorai, D. Mondal, S. Hait, A. K. Ghosh, S. Wiessner, A. Das and D. De, Devulcanization of waste rubber and generation of active sites for silica reinforcement, ACS Omega, 2019, **4**(18), 17623–17633, DOI: **10.1021**/ acsomega.9b01424.
- 236 Y. Chen, S. Ibrahim, S. Zheng, L. Wittenberg, S. Chapple, G. LaChapelle, C. H. Iao, A. Bourke and M. A. Brook, Cleaning steel by devulcanizing rubber from used automotive tyres, RSC Sustainability, 2023, 1(8), 2006-2013, DOI: 10.1039/D3SU00218G.
- 237 A. J. Bowles, G. D. Fowler, C. O'Sullivan and K. Parker, Sustainable rubber recycling from waste tyres by waterjet: a novel mechanistic and practical analysis, Sustainable Mater. Technol., 2020, 25, e00173, DOI: 10.1016/ j.susmat.2020.e00173.
- 238 M. Chen, H. Zhong, H. Wang and M. Zhang, Behaviour of recycled tyre polymer fibre reinforced concrete under dynamic splitting tension, Cem. Concr. Compos., 2020, 114, 103764, DOI: 10.1016/j.cemconcomp.2020.103764.
- 239 B. Husson-Tissier, P. Russo, B. Gros and C. Clauzade, A new scrap grade for the steel industry: steel wire recycling from the treatment of end-of-life tires, Rev. Metall., 2010, 107(9), 345-351, DOI: 10.1051/metal/2010062.
- 240 W. McBride and S. Keys, Application of a vertical venturi separator for improved recycling of automotive tyres, Chem. Eng. Process., 2005, 44(2), 287-291, DOI: 10.1016/ j.cep.2004.02.023.
- 241 A. Michalik, F. Chyliński, J. Zychowicz and W. Pichór, Challenges of a circular economy: the example of raw recycled tyre steel fibres added to concrete, Materials, 2024, 17, 4554, DOI: 10.3390/ma17184554.
- 242 A. A. Kruth, S. L. Dean-Sioss, J. Y. Che, R. V. Dennis-Pelcher, A. M. Baldan, D. A. Till, B. Zielinski and K. C. Lin, Sustainable tire produced with high bio-content, US Pat.,

- US20240326513A1, 2024, https://patents.google.com/ patent/US20240326513A1.
- 243 Ö. Onay and H. Koca, Determination of synergetic effect in co-pyrolysis of lignite and waste tyre, Fuel, 2015, 150, 169-174, DOI: 10.1016/j.fuel.2015.02.041.
- 244 M. R. Islam, M. Akter and M. A. H. Akhand, Utilization of agricultural waste as fillers in elastomeric materials: a sustainable approach, Polymers, 2023, 15(4), 812, DOI: 10.3390/polym15040812.
- 245 P. Sae-Oui, C. Sirisinha and P. Thaptong, Utilization of limestone dust waste as filler in natural rubber, J. Mater. Cycles Waste Manage., 2009, 11(2), 166-171, DOI: 10.1007/ s10163-008-0230-4.
- 246 T. Tharmaratnam, Investigation on the uses of coconut shell powder replacing carbon black as filler in natural rubber, in Proceedings of the 5 Research Symposium of Uva Wellassa University, Sri Lanka, 2015, pp. 44-46.
- 247 M. R. Snowdon, A. K. Mohanty and M. Misra, A study of carbonized lignin as an alternative to carbon black, ACS Sustain. Chem. Eng., 2014, 2(5), 1257-1263, DOI: 10.1021/ sc500086v.
- 248 S. A. Riyajan, Robust and biodegradable polymer of cassava starch and modified natural rubber, Carbohydr. Polym., 2015, 134, 267-277, DOI: 10.1016/j.carbpol.2015.07.038.
- 249 A. Jain and S. K. Tripathi, Fabrication and characterization of energy storing supercapacitor devices using coconut shell based activated charcoal electrode, Mater. Sci. Eng., B, 2014, 183, 54-60, DOI: 10.1016/j.mseb.2013.12.004.
- 250 A. Fazli and D. Rodrigue, Sustainable reuse of waste tire textile fibers (WTTF) as reinforcements, Polymers, 2022, 14(19), 3933, DOI: 10.3390/polym14193933.
- 251 S. F. Kabir, S. V. Sundar, A. Robles, E. M. Miranda, A. G. Delgado and E. H. Fini, Microbially mediated rubber recycling to facilitate the valorization of scrap tyres, Polymers, 2024, **16**, 1017, DOI: polym16071017.
- 252 M. Tamborski, I. Rojek and D. Mikołajewski, Revolutionizing tire quality control: AI's impact on research, development, and real-life applications, Appl. Sci., 2023, 13, 8406, DOI: 10.3390/app13148406.
- 253 W. Wu, X. Cao, J. Zou, Y. Ma, X. Wu, C. Sun, M. Li, N. Wang, Z. Wang and L. Zhang, Triboelectric nanogenerator boosts smart green tyres, Adv. Funct. Mater., 2019, 29(41), 1806331, DOI: 10.1002/adfm.201806331.
- 254 L. Porath, B. Soman, B. B. Jing and C. M. Evans, Vitrimers: using dynamic associative bonds to control viscoelasticity, assembly, and functionality in polymer networks, ACS Macro Lett., 2022, 11(4), 475-483, DOI: 10.1021/ acsmacrolett.2c00038.
- 255 J. M. Jafferson and H. Sharma, Design of 3D printable airless tyres using NTopology, Mater. Today: Proc., 2021, 46, 1147-1160, DOI: 10.1016/j.matpr.2021.02.058.