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Review and Perspectives of End-of-Life Tires Applications for Fuel and Products

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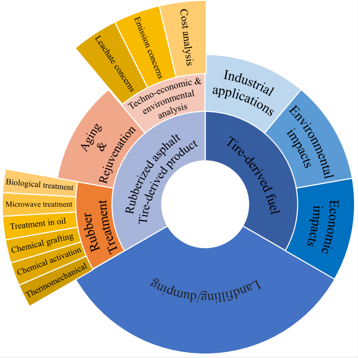
ABSTRACT: Applications of end-of-life tires (ELT) for energy recovery and new products can contribute to a circular economy. This paper reviews the environmental, health, and economic aspects of various applications of ELT, focusing specifically on the use of ELT in fuel, energy, and construction. Impediments to a more widespread application of repurposed ELT include a lack of streamlined handling and recycling processes; a poor understanding of the long-term ecological, human health, and safety consequences; and very few application-specific techno-economic assessments and life-cycle analyses. The prospects for rubberized asphalt as an economical and environmentally benign application of ELT are reviewed in four areas: (1) rubber pretreatment—the most effective techniques are chemical or microwave radiation, and the most sustainable technique is microbial devulcanization; (2) aging behavior—rubber particles delay aging in asphalt through several mechanisms; (3) techno-economic benefits—rubberized asphalt improves the cost-efficiency and energy-efficiency of asphalt; and (4) environmental emissions and leakage concerns—the risk of heavy metals and hazardous pollutants such as polycyclic aromatic hydrocarbons and volatile organic compounds contaminating the ecosystem or causing occupational diseases in workers. There are four priority directions for future research on rubberized asphalt: (1) determine the critical aging point; (2) describe the self-rejuvenating behavior; (3) quantify the long-term leachate under extensive weathering; and (4) compare the ecological aspects of pretreatment techniques in terms of potential greenhouse gas emissions and human health.



1. INTRODUCTION

Transforming waste into a commodity such as chemical feedstock and repurposing it for new applications such as energy production closes the loop on zero waste, promoting a circular economy. A prime example of waste with such potential is end-of-life tires (ELT). Every year, approximately 1−1.8 billion tires reach the end of their lives globally, and the number is projected to increase with the rapid expansion of the automobile industry.[1]

Discarding, stockpiling, and disposing of ELT in landfills would take up valuable space and could trigger spontaneous fires, mar the landscape, and create breeding grounds for mosquitoes and rodents.[2] On the other hand, recycling and reclaiming ELT can yield significant economic, social, health, and environmental benefits. Examples of economic and social benefits include increased industrial activity and employment opportunities and improved allocation of funds for ELT management.[3] In addition, recycling ELT prevents them from becoming homes to rodents and mosquitoes that would otherwise bear and spread various diseases.[2] Furthermore, recycling ELT reduces greenhouse gas emissions, and fabricated products from ELT have a considerably smaller carbon footprint than those made of virgin resins.[4]

The primary recovery routes for ELT can be put into three categories. The first category is material recovery or tire-derived products (TDP), including but not limited to rubber itself. For instance, each passenger car tire contains 1.1 kg of steel wire. Recovering these wries will lead to more than 1 million tons of recycled steel each year globally,[5] helping to conserve 1.1 million tons of iron ore, 0.6 million tons of coking coal, and 0.05 million tons of limestone.[6] More examples of TDP include new tires, rubber walkways, playground mulch, synthetic turf, and sports surfaces.[7−11] The second category is energy recovery or tire-derived fuels (TDF), where ELT are used as an alternative fuel source.[12] The third category of ELT recovery is civil engineering, where ELT are repurposed for applications such as breakwaters and coastal protection,[13] erosion barriers,[14] ground improvement,[15] landfill construction (as a lightweight backfill in gas venting systems, in leachate collection systems and operational liners, and for capping and daily cover),[16] slope stabilization and retaining walls,[17,18] sound barriers and insulation applications,[19] stormwater

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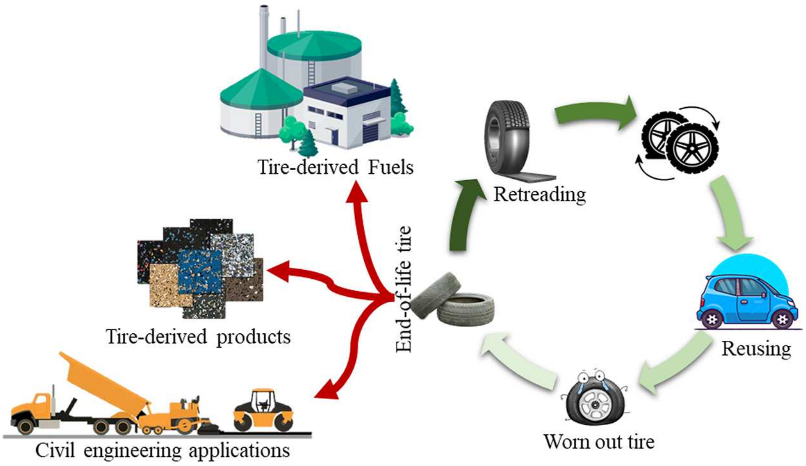


Figure 1. Life-cycle of tires.

Table 1. Comparison of Typical Constituents (wt. %) and Heating Value of TDF and Coal [32]

fuel	carbon	hydrogen	oxygen	nitrogen	sulfur	ash	moisture	heating value (MJ/kg)
TDF	90	8	0	0.3	2	5.0	0.6	39−44
coal	76	5	8	1.5	2	7.8	5.0	31

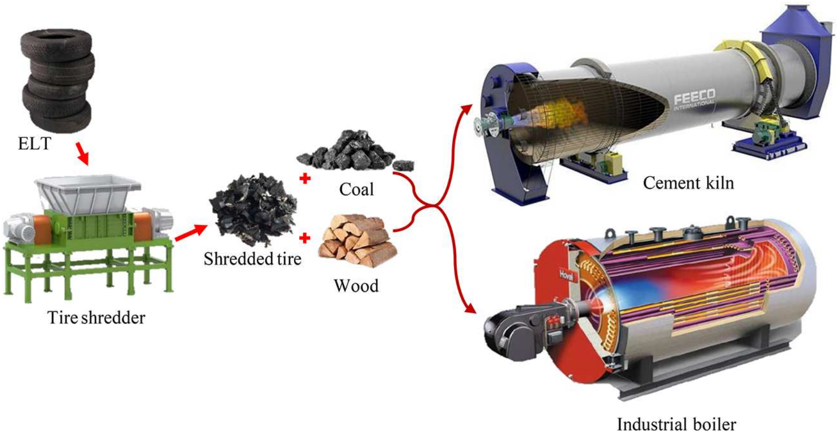


Figure 2. ELT mixed with coal and wood as a fuel.

infiltration galleries,[20] railroad ties,[21] and roads.[22] Figure 1 shows the schematic of the three categories mentioned above.

Tire-derived fuels (TDF) are usually produced via the pyrolysis or incineration of ELT. (1) ELT pyrolysis usually involves thermally depolymerizing scrap tires in an oxygen-free environment (no combustion). This process decomposes scrap tires into smaller molecules, leaving fuel oil, carbon black, char, steel wire, and gas behind. The oil derived from ELT pyrolysis has a high energy density of about 39−44 MJ/kg and is easily transportable and storable under normal conditions.[12,23] A comparison of the constituents and heating value of TDF and coal is presented in Table 1. Pyrolysis oil can be fractionated into kerosene (33%), diesel (34%), heavy vacuum oil (15%), gasoline (12%), and fuel oil (6%), with the concentrations varying by the boiling temperature.[24] Because tire pyrolysis oil is miscible with petroleum diesel, it is a suitable alternative to petroleum-based diesel fuel.[25] The characteristics and structural properties of recovered fuel are sensitive to processing parameters such as temperature, atmosphere, and pressure, so minor changes in these parameters could considerably alter the fuel's combustion properties and emission profile.[7] Despite the heating value of tire oil, its high sulfur concentration (>0.6 wt. %) and the presence of aromatics and heavy molecules with boiling points above 350 °C in it limits its direct application, necessitating additional treatment[26,27] and making its production expensive and energy-intensive.[28,29] For instance, the optimum temperature for producing pyrolytic oil from waste tires is about 550 °C, which in turn reduces the overall energy recovery ratio.[12] In addition to the liquid form, pyrolytic products have a recovered gas phase that can be used as a clean fuel. However, this gas phase contains paraffin, olefins (other hydrocarbons could also appear), carbon oxides, hydrogen, and trace amounts of sulfur and nitrogen compounds.[30] In addition, despite its high heating value (up to 42 MJ/kg), tire-recovered gas fuel has a meager energy recovery ratio of 0.1−0.4.[31]

(2) Incineration of ELT involves burning a mix of ELT with wood and coal to fuel energy-intensive industries such as cement kilns, lime kilns, paper and pulp mills, power plants, industrial boilers, foundries, and smelters (Figure 2). This process utilizes the high calorific value of ELT and coal and the efficient, controllable combustion of wood waste. Every ton of

ELT in kilns replaces 1.25 tons of coal.[33] Regarding emissions, tires generate 85 kg of CO_2 for every gigajoule of energy, while coal, pet coke, and wood generate 90, 100, and 110 kg of CO_2, respectively.[34] In addition, burning ELT releases three to five times less NO_x, similar levels of SO_2, and more polycyclic aromatic hydrocarbons (PAHs) compared with burning coal.[27,35] TDF has also been found to produce almost no ash after combustion and to be mercury-free, which would reduce its leachate potential.[34] However, the concentration of ELT in the coincineration mix used in cement plants should be controlled.[36] Due to its high zinc content, ELT should not exceed 30% of kiln fuel to avoid the generation of a protective coating of zinc compounds on cement grains that would in turn severely influence the cement's hardening process.[33,37,38] As for the economic aspect, the global TDF market was valued at 406 million US dollars in 2022 and is projected to grow to 554 US million dollars by 2028.[39]

Statistics show that only 59% of ELT is recycled globally, with 42% for material recovery, 15% as TDF for energy generation in heavy industries, and 2% for civil engineering applications. The rest ends up in landfills.[40] Figure 3 compares the environmental and economic hierarchy of these management routes,[41] and Figure 4 compares the emission of primary pollutants such as CO_2, CO, NO_x, SO_x, and particulate matter (PM) associated with them. Fuel production via pyrolysis leads to 54.5 kg of CO_2 per ton of waste tire used, an emission mounting to 220 kg via incineration. In addition, recycling one ton of waste tires through devulcanization requires a great deal of energy and produces 170 kg of CO_2. In contrast to the other routes, the construction route causes no significant environmental impact beyond emitting negligible traces of dust into the air during ELT grinding.[1,42] This route also leads to significant carbon sequestration of about 838 kg of every ton of waste tire.[30] Regarding profit margin, using rubber as waste material in asphalt pavement is more advantageous than material recovery.[41] A pyrolysis plant of medium size, capable of recycling 30−40 tons per day, generates $222 per ton. However, the recycling process costs $200 per ton, leaving only a profit of $22 per ton from material recycling.[43] Therefore, given the high cost of producing TDFs, their environmental impacts, and their limited industrial applications, reusing ELT in civil engineering projects and materials may be a more energy-efficient and environmentally friendly option for humans and the environment.

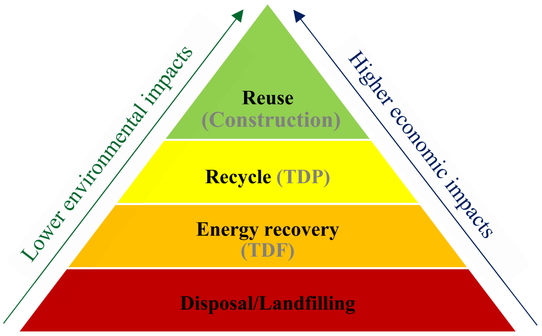
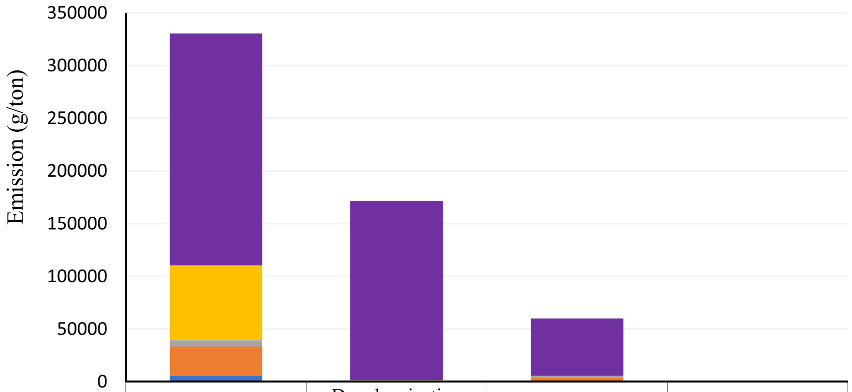


Figure 3. ELT management routes. Reproduced with permission from ref 41. Copyright 2022 American Chemical Society.

As previously noted, one destination of ELT in the construction route is rubberized (rubber-modified) asphalt, in which ground/granulated rubber particles are introduced to the paving mixture via addition to the binder (wet process) or aggregates (dry process). An Arizona Department of Transportation report found that rubber-modified asphalt can reduce tire wear by up to 50%.[44] Rubberized asphalt results in higher carbon sequestration than conventional asphalt pavements using 1,600−2,000 tires per lane-mile.[45] Today, almost 12 million discarded tires are repurposed annually for rubberized asphalt,[46] a number that could substantially increase given the growing network of asphalt paved roads worldwide.[47] It has also been reported that rubberized asphalt enhances durability and performance while reducing noise, thus augmenting economic and social benefits.

Most existing reviews on rubberized asphalt pavements focused on performance indicators such as permanent deformations, fatigue life, noise, and wear.[48−51] However, various applications of ELT have not been compared and



	Incineration	Devulcanization for recycling	Pyrolysis	Ambient grinding
🟪 CO2	220000	170000	54500	0
🟨 CO	71100	210	0	0
■ NOx	6000	385	1400	0
🟧 SOx	28000	720	3550	0
🟦 Particulate matter (PM)	5350	351	580	191

Figure 4. Amount of pollutants emitted by various ELT management methods.

Table 2. Comparison of Dry and Wet Modifications of Rubber and Asphalt

	Dry modification	Wet modification
Production Process	• Crumb rubber is added to hot aggregates and then mixed with binder. • Rubber content ranges from 5 to 20 wt.% of asphalt content. • Key design factors are aggregate gradation, rubber gradation, binder content, and air voids.	• Crumb rubber and asphalt are mixed directly at 176–226 °C. • An extended period of agitation and cooking is required. • Rubber content should be > 15 wt.% of asphalt. • Constant mixing is required to avoid phase separation. • Crumb rubber particles are generally smaller than 2 mm. • Binder type, blending time and temperature, content and size of rubber, and grinding method affect properties.
Equipment needed	• No special equipment or change in the asphalt plant is needed. • Only a steel-drum roller should be used for compaction.	• Special equipment and handling are needed to avoid phase separation. • Grinding rubber to fine particles requires special tools and procedures. • Only a steel-drum roller should be used for compaction.
Mix properties	• Improves resistance to permanent deformation and rutting. • The flexibility of rubber improves low-temperature crack resistance. • Lower oxidative aging due to the presence of fine rubber particles. • Mixture has greater noise reduction.	• Increases viscosity of binder due to absorbing lighter asphalt compounds. • Improves moisture resistance of asphalt mixture due to enhanced adhesion. • Improves flexibility of asphalt matrix due to swelled rubber. • Mixture has greater fatigue resistance.
Pros and cons	• + Costs are 15%–50% lower than wet process and polymer-modified asphalt. • + Higher compaction energy is required. • + Polymer-modified binders can be used. • - Mixture has higher moisture sensitivity. • - Higher asphalt binder content of 0.2%–0.4% is required. • - Mixture adheres to rubber-tired rollers.	• + Mixture has the improved properties listed above. • - Phase separation causes poor workability. • - Process is more energy intensive. • - Overheating transforms rubber's pore structure and disrupts its dynamic viscosity. • - Mixture adheres to rubber-tired rollers.

contrasted, nor have the ecological, health, and emission aspects been thoroughly examined. Accordingly, this paper reviews major studies relevant to rubber treatment technologies and the rejuvenation, health, economic, and environmental aspects of ELT in different applications, including but not limited to pavements. Some mainstream online databases such as Scopus, Web of Science, ACS, and ScienceDirect are surveyed, and only articles published in English peer-reviewed journals, mainly over the past decade, are framed.

It should be noted that there are other recovery routes for scrap tires that are out of the scope of this survey. One of these routes, for instance, is retreading (recapping or remolding), a remanufacturing process that replaces the tread on worn tires and reintroduces them into the distribution circuit. Compared with one-and-done tires, retreaded tires are environmentally and economically more advantageous. For example, a 2016 life-cycle assessment of retreaded tires found them associated with 70% less natural resource extraction, 29% less land use, 24% less carbon emissions, 21% less air pollution, and 19% lower water consumption. In addition, the rubber buffed off old tires' tread surfaces is easier to recycle and repurpose.[52] Retreading, however, is limited mainly to tires made of thicker materials, such as heavy-duty commercial truck tires.

2. CRUMB RUBBER AS A MODIFIER IN ASPHALT

Crumb rubber is the result of grinding waste rubbers into small particles. Crumb rubber has been used in asphalt mix in two ways: (1) dry mixing, where crumb rubber particles partially replace mineral fillers or aggregates,[53] and (2) wet mix, where crumb rubber is dispersed in powder form in asphalt binder to modify its properties.[54] Several aspects of these modifications are shown in Table 2.

3. RUBBER TREATMENT TECHNOLOGIES

Pretreatment is usually required to enhance rubber compatibility with the asphalt binder. Various pretreatment techniques and combinations thereof are in use. Here, some of the primary techniques are reviewed.

3.1. Thermomechanical Treatment. Pretreating waste rubbers with thermomechanical techniques involves devulcanizing them using high shear at elevated temperatures.[55,56] Depending on the rubber's particle size, the rotational speed of blades and the temperature in these machines can be adjusted to disrupt sulfoxide bonds.[57−64]

Wu et al.[65] investigated the influence of the degree of devulcanization and the content of pyrolyzed crumb rubber extruded with a twin-screw extruder on the rheological properties of asphalt binder. They reported that extrusion temperatures exceeding 300 °C severely damaged rubber's cross-linked structure and reduced its molecular weight, increasing sol content in the rubber. (A sol is a colloidal suspension of solid particles in a liquid phase.) In contrast, asphalt containing rubber extruded at low temperatures (e.g., $\leq$ 260 °C) exhibited improved rheological properties at both low and high temperatures. The study mentioned the potential of using modified rubber with a high sol content (extruded at 300 °C or higher) as a proper partial substitute for asphalt binder. Besides improving the rheological properties of modified asphalt binder, it was found that the extruded rubber had improved compatibility with asphalt because of the larger contact area between the extruded rubber and asphalt binder. Modifying asphalt binder with extruded rubber also reduced the temperature sensitivity of the binder. This approach can use up to 30 wt. % rubber without compromising the performance of the asphalt.[66] Low-temperature devulcanized crumb rubber also reduces the moisture susceptibility of hot-mix asphalt and enhances its resistance to rutting, cracking, and fatigue, showing promise to rival styrene−butadiene−styrene as the most common polymer for asphalt modification.[67]

3.2. Chemical Activation Treatment. Digestion by chemicals is another approach to desulfurizing vulcanized rubbers. In this approach, vulcanized rubber is submerged in solvents and stirred for a particular duration. The degree of desulfurization depends heavily on the solvent type and concentration, reaction time and temperature, and rubber particle size.[68]

Various solvents have been used for crumb rubber desulfurization and asphalt modification. A commonly used solvent is hydrogen peroxide (H_2O_2); this has been shown to oxidize, changing the morphology of the rubber surface and decreasing the average pore size of crumb rubber.[69,70] In addition, high concentrations of hydrogen peroxide influence flocks on the crumb rubber surface, making them "fluffier" and increasing the surface porosity. These changes in the morphology of rubber have led to increased adoption of rubber particles with nonpolar components in asphalt. However, H_2O_2 activation of rubber increases the release and dispersion of rubber carbon black in the asphalt matrix, leading to a reduction in the elasticity, viscosity, and rutting resistance of asphalt modified by crumb rubber.[69] Another treatment for crumb rubber desulfurization is to treat rubber in supercritical carbon dioxide ($ScCO_2$) at high pressure (12 MPa) and high temperature (180 °C) for 1 h; this has been reported to extend the degree of devulcanization.[71]

The grinding technique can also influence rubber's chemical activation. Cryogenic grinding involves freezing rubber with liquid nitrogen, then shattering the frozen rubber as it passes through a hammer mill. Binders modified with crumb rubber produced by cryogenic grinding have outperformed those modified with crumb rubber produced by ambient-temperature grinding in terms of creep recovery and elastic-related properties.[72] However, cryogenic-ground rubber has a marginal influence on rheological parameters.

The use of catalysts during the activation process is another factor that influences the efficacy of rubber in improving the properties of asphalt binder. For example, Fenton's reagent ($FeSO_4$) gives rise to the formation of carboxylic sites on the rubber surface, enhancing its compatibility with asphalt binder. Also, cryogenic-ground rubber with furfural sprayed on it significantly improves some properties of the binder, such as storage stability and resistance to rutting and creep.[73] In addition, bitumen modified with crumb rubber activated with sodium hypochlorite (NaClO) exhibits lower rutting susceptibility and higher workability (lower viscosity).[71] These improvements have been attributed to increased energy, polarity, and roughness on the rubber surface.[74] Tetraethyl orthosilicate (TEOS) has also been shown to facilitate the entanglement of molecular chains between asphalt and modified rubber, increasing the storage stability of asphalt.[75]

3.3. Chemical Grafting Treatment. Chemical grafting involves functionalizing the rubber surface to increase its compatibility and bonding with asphalt.[76] Various chemicals and monomers, such as acrylamide, maleic anhydride, octadecyl amine, polystyrene, and acrylic acid, have been used for rubber functionalization. Xie et al.[77,78] comprehensively examined the grafting potential of acrylamide to the waste rubber surface and the effects of using up to 20 wt. % of functionalized rubber on asphalt properties. The process involved mixing 100 g of purified rubber with 40 g of acrylamide and 2.5 g of potassium persulfate in 1000 mL of water and stirring the mix for four hours at 80 °C. The treated rubber showed improved compatibility with asphalt, as evidenced by a 60% lower segregation index after 72 h of storage at high temperatures (over 160 °C). This improvement was attributed to the copolymerization of amide groups with rubber macromolecules and their reaction with acid groups in the asphalt. Also, the functionalized rubber had a rougher surface and exhibited a more uniform dispersion in asphalt than the untreated rubber. Overall, at an optimum concentration of 20 wt. %, the functionalized rubber outperformed regular crumb rubbers in improving the high- and low-temperature properties of asphalt. The group also evaluated the compatibility of asphalt binder with rubber surfaces activated by grafting polyamide 6 and showed its potential in improving the softening point, viscosity, and high-temperature and low-temperature properties of asphalt.[79] According to Hanet al.,[80] the high compatibility between asphalt and rubber grafted with octadecyl amine can be due to chemical bonds formed between the amino group ($-NH_2$) on the octadecyl amine and the interfacial carboxyl group ($-COOH$) of waste rubber. Two additional examples of materials that have been used for rubber functionalization are polystyrene and acrylic acid. Polystyrene improved the rutting resistance and fatigue life of asphalt by 97% and 85%, respectively.[81] Acrylic acid increased the viscosity and reduced the temperature sensitivity of modified asphalt.[82]

Table 3. Summary of Oil-Treated Rubber in Asphalt[89]

	Oil	Asphalt/rubber	Treatment	Major observations	Ref.
Physisorption	waste lubricating by-products (LBP)	#70 asphalt / mesh #40 rubber	rubber mixed with oil for 1 h at 150 °C	• LBP-activated rubber had improved dispersion in asphalt with no signs of rubber agglomeration. • LBP-activated rubber improved the storage stability in modified asphalt binder. • LBP-activated rubber had lower susceptibility to aging, improved low-temperature performance, and reduced rotational viscosity in asphalt.	85
	swine manure bio-oil	#70 asphalt / mesh #40 rubber	bio-oil and rubber powder preheated at 90 °C stirred at a reaction temperature of 120 °C for 1 h	• Pretreatment of rubber with swine manure bio-oil before adding it to asphalt binder is a more efficient method for producing rubber-modified bio-binder than adding untreated rubber to bio-binder. • Rubber treated with swine manure oil improved the high-temperature performance of modified asphalt.	86
	heavy bio-oil (HBO)	#70 asphalt / mesh #40 rubber	rubber submerged in HBO for 45 h	• Bio-modification of rubber reduced the preparation temperature of rubberized asphalt and rubberized asphalt mixture. • Bio-modified rubberized asphalt showed improved rutting and fatigue resistance, workability, and storage stability and showed slightly poorer low-temperature performance.	87
	waste engine oil (WEO)	#50 asphalt / mesh #60 rubber	rubber swelled in oil at 100 °C for 1 h, then extruded at 150–280 °C	• Swelling rubber in WEO enhanced partial degradation of ground tire rubber, enhancing rubber devulcanization in extrusion, per sol fraction and thermogravimetric analyses. • Higher concentrations of WEO for rubber modification decreased the stiffness of modified asphalt but reduced its storage stability and anti-rutting properties. • Higher extrusion temperature led to better dispersion, leading to reduced rubber particle size, higher workability, and improved storage stability and rheological properties.	88
	waste cooking oil residues (WCOR)	PG 64-22 asphalt/ mesh #40 rubber	rubber mixed with oil at 190 °C for 4 h	• Penetration of WCOR molecules increased swelling level of rubber and improved its compatibility. • Bio-modified rubber reduced separation tendency and improved compatibility.	89
	waste cooking oil	PG 82-16 asphalt/ -	one part rubber mixed with four parts oil at 260 °C for 2 h	• Oil-treated rubber rejuvenated aged asphalt. • Oil-treated rubber significantly improved the low-temperature properties of aged binder, but improvement of high-temperature properties was marginal. • Oil-treated rubber outperformed waste cooking oil in improving aging resistance of asphalt binder.	90
	amide-based bio-binder	PG 64-22 asphalt/ -	bio-binder mixed with rubber at 135 °C for 30 min	• There was stronger non-covalent interaction between the bio-modifier's –NH2 site and the rubber's S–S crosslink than with the rubber's C–S part, potentially leading to partial devulcanization. • Amide-rich bio-binder significantly improved the workability and durability of vulcanized rubber when mixed with bitumen.	91
Chemisorption	bio-oil derived from wood sawdust, corn straw, and rice husk	#80 asphalt/ mesh #20 to #100 rubber	bio-oil mixed with rubber at a mass ratio of 1.5:1, then heated in a microwave oven with 600 W power for 2 min	• Regardless of oil treatment, the size of rubber particles significantly influenced asphalt's properties. • Longer fatigue life was associated with larger crumb rubber, but rubber between 40 mesh and 80 mesh was recommended for storage stability.	91
	epoxidized soybean oil (ESO)	#70 asphalt/ mesh #80 rubber	heated in a 600W microwave oven for 2.5 min, then submerged in epoxidized soybean oil for 24 h	• Asphalt modified with oil-treated rubber had improved high-temperature storage stability and ductility.	92
	waste cooking oil	#90 asphalt/ mesh #60 rubber	rubber mixed with waste oil 1:1 at 135°C for 30 min and swelled for 12 h, then irradiated with 400W microwave for 4 min	• Oil-treated rubber improved the aging resistance of asphalt binder (35% and 34% lower carbonyl and sulfoxide indices compared to binder with conventional rubber). • Oil-treated rubber improved softening and deagglomeration. • Oil-treated rubber showed 84% and 106% higher asphalt healing capacity after thermal and UV aging, respectively, than regular rubber.	93
	swine manure bio-oil	PG 64-22 asphalt/ mesh #80 rubber	rubber and waste oil mixed 1:1 at 135°C for 30 min and swelled for 12 h before 4 min irradiation at 400W microwave	• FTIR analyses confirmed dissociation of rubber polymer chains by irradiation and subsequent crosslinking to bio-oil molecules. • Higher workability (60% reduction in asphalt viscosity) was achieved by bio-modifying rubber. • Oil-treated rubber caused 86% less segregation phase in asphalt than regular rubber. • Oil-treated rubber enhanced asphalt's fracture energy by 62% compared to regular rubber.	94
	amide-rich bio modifier	PG 64-22 asphalt/ mesh #80 rubber	rubber submerged in oil 1:1 for 12 h before 4 min irradiation with 400W microwave	• Irradiation of chain backbones of rubber polymer led to the formation of carbon-centered and sulfur-centered radicals. Irradiated molecules of bio-modifier led to the formation of amidyl radicals. • Reaction pathways of amidyl radicals with carbon-centered radicals were energetically more favorable and stable than those with sulfur-centered radicals. • Bio-oil functional groups appearing in treated rubber indicated successful grafting of highly polar molecules of bio-modifier such as hexadecanamide onto the surface of the rubber. • Bio-modification of rubber increased the acid-base component of rubber's surface energy from 2.9 mJ/m^2 to 14.6 mJ/m^2, reducing phase separation.	95
	bio-oil residue of refining fatty acids from castor oil	PG 64-22 asphalt/ mesh #20 rubber	rubber submerged in oil 1:1 for 12 h before 4 min irradiation with 400W microwave	• Adsorption of ester biomolecules increased interactions between rubber and asphalt binder, significantly reducing the segregation of rubber and asphalt. • Oil-treated rubber increased bitumen's elastic recovery by over 2.5 times and improved asphalt's crack resistance better than asphalt containing regular rubber.	84
	bio-oils derived from waste vegetable oil, wood pellet oil, corn stover oil, miscanthus oil, and castor oil	PG 64-22 asphalt/ mesh #60 rubber	rubber submerged in oil 1:1 for 12 h before 4 min irradiation with 400W microwave	• Treating rubber with wood-based and waste vegetable oils reduced the segregation index by 82% and 70%, respectively. • Among surface-activated rubbers, corn stover had the highest percent recovery of 40%, followed by miscanthus (25%) and wood pellet (24%). • Wood pellets and waste vegetable oil both showed the highest adsorption onto the rubber surface. However, waste vegetable oil, which showed good adsorption to rubber, did not interact strongly with the bitumen matrix, leading to a very low percent recovery. • All asphalt binders containing surface-activated rubber except the one with miscanthus oil showed low susceptibility to moisture.	96

3.4. Treatment by Oil. Waste oils and bio-oils have been extensively used to produce biobinders or rejuvenating agents in asphalt recycling.[83] In addition, oils have been used to improve the compatibility of rubber particles with asphalt binder. Pretreating the rubber in oil at elevated temperatures is a physisorption process that involves the diffusion of bio-oil into the rubber particles, making them preswell. This treatment reduces the density of rubber and prevents the rubber from adsorbing asphalt's light components, such as resins, after mixing the rubber and asphalt.[84] Recently, chemisorption has

Table 4. Studies on Microbial Desulfurization of Waste Ground Tire Rubber (GTR)

Microorganism	Rubber type	Amount of rubber/size	Medium	Major observations	Ref.
Gordonia desulfuricans	GTR	150 g	1.5 L mineral salt medium, with 20 g/L glucose, 30 °C, and pH 7	• G. desulfuricans showed further devulcanization than Rhodococcus sp. according to its greater change in sol fraction (from 2.3 to 3.3).	108
Rhodococcus sp. AF21875					
Thiobacillus	GTR	0.5 and 5% (w/v), ≤0.354 mm and 0.18–0.25 mm	DSMZ medium (70ml) without ferrous sulfate, pH 1.4, 30 °C, 10 and 20 days incubation	• Rubber content and size were the most important factors in rubber devulcanization; larger rubber particles showed more devulcanization. • Within 0.5% w/v, all bacteria could desulfurize rubber (by 6% to 21%). • Only Thiobacillus ferroxidans (DSMZ 583) and Acidothiobacillus ferrooxidans kept their efficiency in higher rubber content.	109
Gordonia			Mineral salt medium, pH 1.4, 30 °C, 20 days incubation		
Nocardia					
Amycolaptopsis					
Pseudomonas					
Acidothiobacillus ferrooxidans					
Thiobacillus ferrooxidans	GTR	5% (w/v)	Mineral salt medium (200 ml), pH 2.5, 30 °C, 30 days devulcanization	• Increase in sol fraction was from 4.69% to 7.43% (only superficial devulcanization). • Better mechanical performance of NR filled with devulcanized GTR (DGTR) due to better bonding between two rubbers caused by rupture of interfacial crosslink bonds of DGTR.	110
Candida methanosorbosa BP-6	Ozone-treated GTR	1g,130 µm	Mineral salt medium (40 ml), pH 6.9, 30 °C, 21 days devulcanization	• Although C. methanosorbosa BP-6 yeast strain metabolic conditions were higher in ozonide rubber (20% more devulcanization), the ozone pretreatment hindered proper bacteria growth.	111
Sphingomonas sp.	GTR	117 µm	Mineral salt medium, pH 7, 30 °C, 10 days devulcanization	• Compared to GTR, this had 24.7% lower sulfur and 19.4% lower oxygen content.	112
Gordonia sp.				• Compared to GTR, this had 19.4% lower sulfur and 20.2% lower oxygen content.	
Mixture of both				• This had a 6.7% increase in swelling value and a 9.5% reduction in crosslink density (3.3% and 2.8% more than Sphingomonas sp and Gordonia sp.).	
T. perometabolis	GTR	10 g, 0.1 mm	Mineral salt medium (20 ml), 30 °C, 30 days	• This had a 20% higher reduction of sulfur bonds in 30 days compared to chemical devulcanization with di-(cobenzanidopheny)-disulfide solvent	113

been presented as a more efficient approach to improve rubber's compatibility with oils. This method involves grafting oils' biomolecules onto rubber surfaces with microwave irradiation to create surface-modified rubber.[84] Table 3 summarizes recent studies on the application of physisorption and chemisorption in improving the performance of waste rubber in asphalt.

3.5. Microwave Treatment. Microwave radiation has proved more energy-efficient than conventional thermomechanical approaches in decomposing C−S and S−S bonds in

Table 5. Summary of Advantages and Disadvantages of Methods for Rubber Treatment

Method	Advantages	Disadvantages
Thermomechanical treatment	• convenient equipment • scalable for industrial use • high-degree devulcanization	• high energy consumption • emission of volatile degradation products
Chemical activation	• unsophisticated equipment • low-cost solvents	• use of environmentally harmful solvents and catalyzers • low controllability and low efficiency • need for elevated temperatures to accelerate devulcanization by some solvents • superficial treatment
Grafting treatment	• more effective than devulcanization by chemicals • ability to tailor properties by grafting various functional groups	• need for sophisticated operators • expensive • long processing time
Oil treatment	• application of waste or green material for treatment • low-cost • unsophisticated equipment	• Comparatively time-consuming
Microwave treatment	• short treatment process • solvent-free and environmentally friendly • devulcanization controllability (by controlling output power and irradiation time) • applicable even to thick rubbers	• needs expensive microwave ovens • inability to efficiently mix rubbers during irradiation • emission of volatile degradation products • unfit for non-polar rubbers • needs initial processing to remove metal particles (especially in waste rubbers)
Biological treatment	• low cost • energy efficient • environmentally friendly • simplicity of approach • low requirement for high-tech equipment	• low devulcanization efficiency • time-consuming • need for purification of waste rubber to remove zinc oxide and zinc salts that harm microorganisms and prohibit their cultivation

rubber and creating active sites on the surface.[55,97] This technique uses electromagnetic waves ranging from 300 MHz to 300 GHz that directly interact with polar molecules or ions, heating the rubber from the inside.[97−99] The formation of free radicals on the rubber surface increases its interactions with the surrounding matrix, such as asphalt. For instance, rubbers irradiated in an 800 W microwave oven for 90 s showed increased surface activity, increased specific surface area, and partial swelling, all of which improved the compatibility with asphalt binder. Also, asphalt modified with microwave-treated rubber exhibited lower stress sensitivity, higher aging resistance, and marginal improvement in high-temperature performance. However, compared with untreated crumb rubber, microwaved crumb rubber increases the risk of fatigue cracking caused by age-hardening and degrades the viscoelastic performance at intermediate temperatures.[100] In terms of the environmental impact, compared with conventional rubber, the application of microwave-treated rubber in asphalt is associated with lower emission of toxic gases and lower mixing and compaction temperatures due to its lower viscosity.[101−103]

Like bio-oil chemisorption onto the rubber surface, polymers or coupling agents have been grafted to microwave-activated rubber to further improve its compatibility with the asphalt binder. Adding 4.5% of *trans*-polyoctenamer (TOR) by weight of rubber to microwave-activated rubber has been found to improve viscoelastic functions (G', G'', and $G^*/\sin\delta$) and nonlinear viscoelastic functions such as steady-state flow and strain recovery rate. It also decreases asphalt's susceptibility to heat, increasing its elasticity and making it more resistant to permanent deformation.[104] In addition, TOR grafting alleviates the adverse effects of aging on asphalt binders modified with microwave-activated rubber.[100,103] Grafting microwave-activated large rubber particles (150 mesh) with silane coupling agent KH550 improves the hot-storage stability of the rubberized binder with a 1.8 °C difference in softening point in the tube test. This improve-

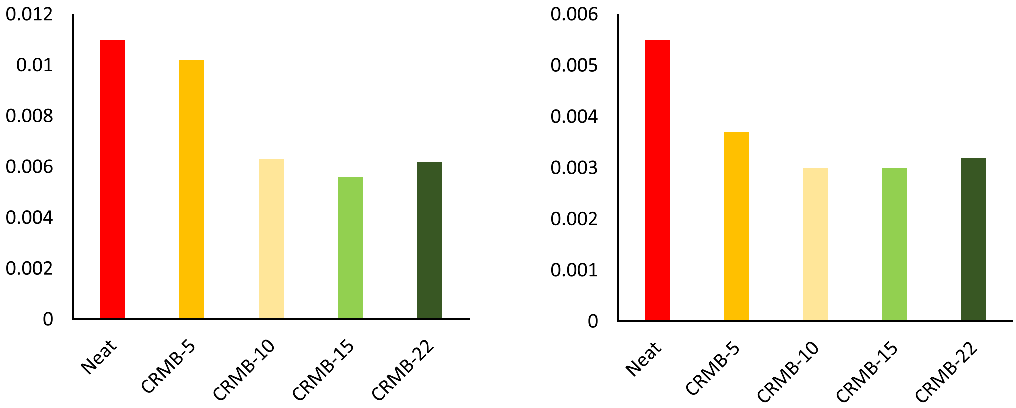


Figure 5. Carbonyl index (left) and sulfoxide index (right) of neat and rubberized binders subjected to long-term aging. CRMB is short for crumb rubber-modified bitumen, and the numbers in the abscissa refer to rubber weight concentration.[127]

ment in compatibilization is due to a large number of active groups such as −OH and −NH on rubber surfaces, resulting in higher resistances to crack and fatigue damage, improved elastic recovery, and higher failure temperature compared with conventional rubberized asphalt and neat asphalt binder.[105]

3.6. Biological Treatment. Biological treatment is a more recent technique for devulcanizing and recycling rubbers. In this technique, microorganisms such as bacteria, fungi, and algae isolated from soil or oceans are cultivated to decompose polymeric wastes such as waste rubber.[106,107] The microorganisms' growth and reproduction require sulfur, which causes the rubber to devulcanize. Table 4 summarizes recent studies on the microbial desulfurization of waste ground tire rubber (GTR).

Table 4 shows various microorganisms used to decompose rubber more efficiently. However, the application of biologically treated rubbers in asphalt is limited to an investigation by Kabir et al.[114] They first devulcanized rubber using waste-activated sludge microbes, which effectively desulfurized almost 34% of crumb rubber. As a result of this devulcanization, the acid−base component of the rubber's surface energy increased by 3.7 mJ/m^2. Then, they introduced 15 wt. % of the treated rubber into asphalt, which promoted interactions between rubber particles and bitumen and reduced their segregation by 68%. As a result, the bitumen containing devulcanized rubber exhibited 10% higher stiffness and 5% greater stress−relaxation capacity than asphalt containing untreated rubber. In addition, there were increases of 6% in elastic recovery, 27% in moisture resistance, and 12.5% in workability.

In general, treated rubber has proved effective at increasing the durability of asphalt pavements. Table 5 summarizes the advantages and disadvantages of the technologies for rubber treatment reviewed above.

4. AGING AND REJUVENATION OF RUBBERIZED ASPHALT

Like any other organic material, asphalt ages as it undergoes volatilization, oxidation, and condensation due to exposure to oxygen, moisture, sunlight, and other environmental conditions. Aging hardens asphalt, accelerating pavement distresses like raveling, potholes, and cracking.[115] Crumb rubber improves the rheology of asphalt binder and retards its aging. Aging retardation by crumb rubber is due to the release of rubber components such as carbon black and silica;[116] carbon black absorbs the light components in asphalt and prevents the light components' volatilization throughout the aging process and reduce oxygen diffusion.[117,118] In addition, the light fractions of asphalt (maltenes) can be absorbed by crumb rubber to protect it from aging.[119,120] Silica's significant steric hindrance effect on asphaltene weakens the asphalt binder's intermolecular interaction and disrupts asphaltene agglomeration, and its release into asphalt prevents asphalt from long-term aging.[121−123] Moreover, stronger interactions between crumb rubber and asphalt are associated with improved rubberized asphalt aging resistance.[120,124] This improvement is due to delays in the volatilization and transformation of asphalt's lightweight components and the hindrance the rubber network places to the intrusion of oxygen molecules, delaying the oxidation of polar groups in asphalt modified by crumb rubber.[125,126] Figure 5 shows the influence of crumb rubber on two long-term aging metrics of asphalt; carbonyl and sulfoxide indexes.[127] The optimum aging resistance, as evidenced by the lowest carbonyl and sulfoxide indices, is associated with 15 wt. % rubber particles.

The aging behavior of rubberized asphalt is affected by variables such as mixing parameters and crumb rubber content.[119,128] For instance, terminal-blended crumb rubber further retards weather aging in asphalt by releasing more carbon black to the asphalt matrix.[129,130] In addition, analysis of thermo-oxidative aging of rubber-modified binders has shown that as crumb rubber ages, its cross-linking network breaks apart, leading to a lower cross-linking density and accelerated dissolution of crumb rubber in asphalt binder.[131] As a result, the breakdown of crumb rubber by aging improves the aging resistance of rubberized asphalt binder, giving rise to a more flexible rubberized asphalt with greater penetration. In addition, a morphological analysis of asphalt modified by crumb rubber and exposed to UV aging indicated that blends with higher rubber content experienced finer cracks, suggesting rubber held the cracked asphalt together.[132]

Combining crumb rubber with polymers, oils, and nanomaterials has proved synergistic in improving rubberized asphalt binders' aging resistance. For instance, waste rubber pyrolyzed with 5.5 wt. % of poly(styrene-butadiene-styrene), or SBS, has improved the rheological characteristics and aging resistance of asphalt binder better than neat waste rubber.[133] Also, a rubber-regenerating agent composed of SBR powder, light oil, and liquid rubber has been found to improve the aging resistance of rubberized asphalt.[134] However, excessive amounts of the rubber regenerating agent (e.g., > 8 wt. %) could adversely

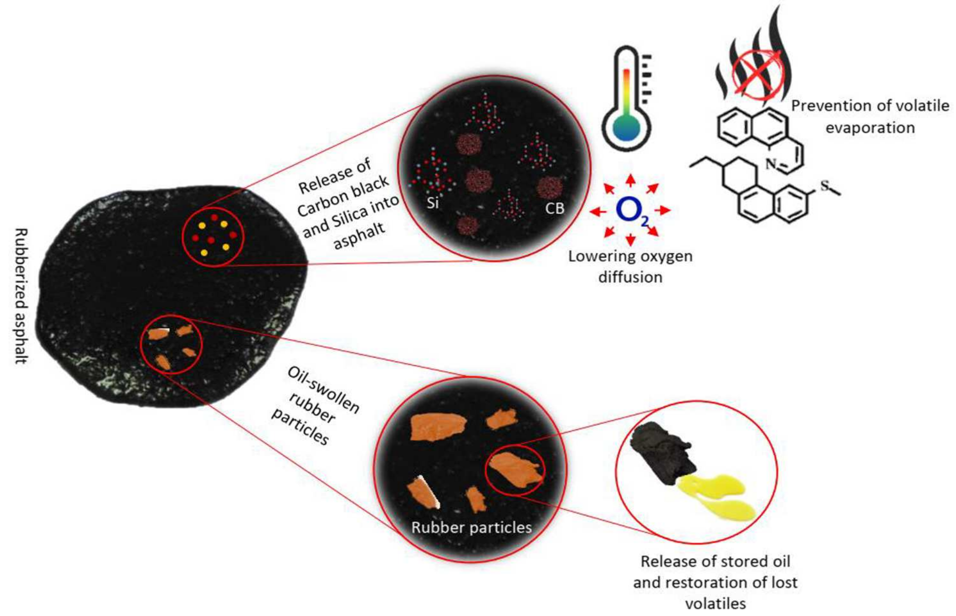


Figure 6. Aging deceleration mechanisms in rubberized asphalt binder

influence the mechanical and aging performance of rubberized asphalt. An example of the oils used with crumb rubber to modify asphalt is amorphous polyalphaolefin (APAO). Adding 4 wt. % APAO in rubberized asphalt improves the rheological properties and the high-temperature and fatigue performance of asphalt without significant sacrifice in low-temperature performance.[134,135] Furthermore, with crumb rubber, APAO reduces the generation of carbonyl groups in the long term, enhancing the aging resistance of asphalt binder. Other materials used in conjunction with crumb rubber to delay aging in asphalt are layered double hydroxides, which are UV-resistant, layered nanomaterials belonging to the anionic clay family.[136]

Besides the mechanisms and factors mentioned above, rubber decelerates aging in asphalt in another way by acting as a rejuvenator (Figure 6). The continuous action of traffic loads on crumb-rubberized pavement causes the crumb rubber particles to exude some of their maltenes (oils and resins) compounds, rejuvenating the mastic in the asphalt mixture.[137] This phenomenon partly explains the formation of less severe and less extensive cracks in driving lanes than road shoulders. However, the rejuvenation of rubberized asphalt has barely been investigated. To the best of our knowledge, the work by Guo et al.[138] is the only work in this area. They analyzed the compositional changes of asphalt fractions and the derived colloidal stability index to investigate the changes in neat asphalt, asphalt modified by crumb rubber, and SBS-modified asphalt after aging and rejuvenation. The authors observed that adding crumb rubber to asphalt did not affect the diminishing stability of its colloidal structure as it aged, indicating that crumb rubber did not prevent asphalt from aging. However, after rejuvenation, the recovery of the stability of the colloidal structure of asphalt modified by crumb rubber was 0.39; this was much greater than the values for neat asphalt and SBS-modified asphalt, which had relative colloidal structural stability recovery values of −0.29 and −5.18, respectively. Also, there appears to be a lack of research on the critical aging point of rubberized asphalt pavement beyond which restoration and rejuvenation are ineffective.

5. ECONOMIC, ENVIRONMENTAL, AND HEALTH IMPACTS

While some benefits of rubber as either a modifier (wet modification) or aggregate (dry modification) in asphalt (e.g., reduced energy consumption, pollution, and cost) have been well established, there is still controversy surrounding rubber's economic, environmental, and health impacts.

Techno-economic advantages are among the top motives for agencies and clients interested in rubberized asphalt mixtures.[139] Zarei et al.[140] analyzed the economic and environmental considerations related to modifying asphalt with up to 20 wt. % crumb rubber to weigh its cost-benefit ratio. Such analyses involve estimating the production and maintenance cost of rubberized asphalt mix and dividing it by the economic gain associated with reduced pollution (e.g., carbon monoxide, carbon dioxide, nitrogen oxides, sulfur dioxide, methane gas, volatile organic compounds [VOCs], and PM10). Cost-benefit ratios of 1.15 and 1.02 were calculated for mixes containing 5 and 15 wt. % crumb rubber, respectively, while mixes with 10 and 20 wt. % were deemed economically unfeasible. The same authors evaluated the influence of adding carbon nanofibers as mechanical reinforcement on the cost-benefit ratio of rubberized mixtures.[141] Results showed that adding 0.5% nanofibers to asphalt modified with 15% rubber powders improved the Marshall stability by 42% and was considered the most cost-effective mixture among the mixtures with 0.5 wt. % to 3 wt. % nanofibers and 5 wt. % to 20 wt. % rubber powder. According to life-cycle assessments and cost analyses, producing one metric ton of aggregate makes 28.8 kg of CO_2 while consuming 9 L of fuel and 8 kWh of electrical energy. In contrast, adding granulated tire rubber to asphalt pavement results in a net energy gain between 310,267 and 566,109 kJ/kg, with 109,314 kJ/kg of it being due to lowering the production of aggregate;[142] this indicates the significant energy-saving potential of rubberized asphalt mixtures. It has also been estimated that using crumb rubber as an alternative polymer to SBS would save 19.95 million tons of coal equivalent (=644.04 $\times 10^6$ GJ = 178,900 $\times 10^6$ kWh) per year, reduce CO_2

emissions by 30.33 million tons, and cut costs by 8.6%−9.4%.[143]

Apart from environmental benefits such as lower energy consumption and less pollution during material preparation, the application of rubber in asphalt can also save valuable space in landfills, reduce the asphalt layer's thickness, and decrease the energy and pollution associated with multiple maintenance and rehabilitation interventions over the asphalt's service life.[140] This section offers a more granular look at some points neglected in the application of waste rubbers, reviews the leachate potential of hazardous components in waste rubber, and compares the emission of toxic gases in rubber-modified and conventional asphalt mixtures.

About 41% of waste rubber is not recycled and usually accumulates in landfills.[143] Hazardous and toxic chemicals such as zinc (used as a vulcanization activator by manufacturers),[144−146] manganese, iron,[143] cadmium,[147] dissolved organic carbon, dissolved nitrogen, inorganic compounds,[148] and tire rubber antioxidants[149] are known to leach from ELT into the environment. Excessive concentrations of these compounds in soil and water resources are a risk to human and aquatic life by causing various diseases (e.g., gastrointestinal and cardiovascular diseases). These compounds can also cause animal death or damage plants' DNA.[150,151] Some factors that affect the leachate potential of ELT toxic chemicals include the ambient pH and temperature,[143,145] the size of rubber particles,[146] and weathering of rubber.[144]

Containing waste rubber in construction materials limits its direct contact with atmospheric conditions such as heat, air, and water, reducing its leachate potential below what would otherwise occur in the landfill. However, the leachate potential of some chemicals in waste rubber could be significant. Azizian et al.[152] investigated the detection of 20 metals in short-term and long-term leachates of asphalt modified by crumb rubber in highway environments. Of these, seven elements (Al, Ca, K, Mg, Sr, P, and Hg) were detected, and the remaining (V, Zn, As, Ba, Ni, Co, Fe, Cr, Cu, Sb, Pb, Cd, and Se) were below detection limits. The highest long-term concentrations of Hg (mercury) and Al (aluminum) were 0.116 mg/L and 1.81 mg/L, respectively. To put these concentrations into perspective, the maximum levels of mercury and aluminum in drinking water are 0.002 mg/L and 0.05−0.2 mg/L, respectively, per the US Environmental Protection Agency (EPA) recommendation.[153] Furthermore, benzothiazole, a highly toxic compound, leached 0.45 and 0.54 mg/L at short-term (24 h) and long-term (7 days). Liu et al.[154] found that containing rubber in asphalt decreased the leakage of Zn (zinc) and other toxic elements (Be, Cr, Cu, As, Se, Cd, Sb, Ba, Pb, and Tl). With 50% asphalt coverage on rubber surfaces, zinc leaching was reduced by 67%; doubling coverage to 100% marginally improved it to 82%. Additionally, a significant reduction in zinc leaching from tires was demonstrated by increasing pH, especially in the pH range between 5 and 8. In this pH range, more free negatively charged surface sites are available to adsorb positively charged zinc species. Moreover, zinc leaching increased with decreasing size of rubber particles since smaller particles have a greater surface area. Fathollahi et al.[155] investigated the PAH leachate from different rubberized asphalts. They reported that the total content of PAHs in rubberized asphalt specimens varied between 0.019 and 4.992 μg/g, depending on the volume of crumb rubber granulates in the asphalt concrete mix design, and the binder type—PAHs are abundant organic compounds in asphalt binders widely distributed in the environment as organic pollutants—according to Fathollahi et al., benzo[*b*]fluoranthene, benzo[*k*]fluoranthene, and naphthalene were the most leached PAHs with 64-day cumulative releases per specimen surface area >1 g/m^2, and the cumulative concentrations of acenaphthylene, fluoranthene, fluorene, and indeno[1,2,3-*c*,*d*]pyrene were between 0.1 and 1 g/m^2.

Besides leaching harmful chemicals, waste rubber in asphalt causes an increase in the emission of toxic volatiles that risks human health, especially the health of asphalt laborers. A 2015 field study in Italy[156] measured the concentration of toxic volatiles such as PAHs and VOCs in gap- and dense-graded asphalt mixes with asphalt binder in regular and modified (18 wt. % rubber) forms and found that crumb rubber increased the emission of PAHs and VOCs. Gap-graded rubberized asphalt mixes were associated with increased hazard quotient (HQ) and increased incremental excess life cancer risk (IELCR). Compared with dense-graded rubberized and regular hot-mix asphalts, gap-graded rubber-modified asphalt caused cancer in workers by factors of 3.5 and 2.9, respectively, and had 1.3 and 1.2 times higher toxicologic effects. This observation was attributed to the greater quantity of binder used in gap-graded mixtures. The same authors showed that paver operators were at higher risk of exposure to toxic volatiles than screed operators, while the carcinogenic risk was higher for workers closer to the screed.[157] An investigation into different paving projects demonstrated that the composition of asphalt fumes depended on material properties such as crumb rubber type, source of asphalt, and mix composition, as well as site-related factors such as layer thickness, laying, air temperature, wind, and pressure.[158] However, the bitumen's quantity, type, and composition were the most critical factors.

In light of a significant increase in emitted VOCs from an asphalt mixture with a high content of rubber (25−45%), Zhang et al.[159] investigated the merit of adding different VOC inhibitors into a rubberized mixture to prevent excessive VOC emissions. All inhibitors, including activated carbon, 4A zeolite, and nano calcium carbonate, effectively reduced VOC emissions, with nano calcium carbonate exhibiting the highest efficacy. In a similar study, Cao et al.[160] showed that deodorants effectively suppressed the escape of toxic volatiles from rubberized asphalt. In addition, deodorant increased the stability of rubberized asphalt during heating, facilitated rubber dispersion in asphalt binder, and improved the aging resistance of asphalt binder. Furthermore, it was reported that adding 1 wt. % deodorant prevented the desulfurization and devulcanization of waste rubber to a certain extent, lessening the emission of harmful components from rubber-modified mixtures. Adding crumb rubber to asphalt has been shown to emit pollutant gases such as xylene and toluene.[161] However, the severity of these emissions is proportional to the temperature. Rubberized warm-mix asphalt emits one-third less pollutant than rubberized hot-mix asphalt, thus the recommendation of rubberizing warm-mix asphalt instead of hot-mix asphalt. Interestingly, activation of the surface of rubber by grafting *trans*-polyoctenamer on it has proved to be effective at reducing the concentrations of aromatic hydrocarbons and PAHs, lowering asphalt fume's toxicological potential.[104] Lv et al. reported a 41.38% reduction in the types of emitted VOCs and a 50.4% reduction in their concentration after adding UiO-66 absorbent with a metal−organic framework to rubberized asphalt.[162]

6. CHALLENGES, GAPS, AND PERSPECTIVES

Despite significant evidence of ELT's advantages in different applications, scrap tires continue to stockpile in the US and worldwide. This review identified five categories for the challenges hindering more effective use of ELT: (1) lack of uniform, simple, and low-cost methods for recycling ELT, as current methods involve time-consuming and costly recycling steps such as solvent recovery, washing, drying, and disposal of associated wastewater, (2) lack of techniques and regulations to handle and recycle products made with ELT, (3) lack of information about long-term consequences of using ELT concerning ecological and human health and safety, (4) lack of clear authority and liability provisions as it comes to ELT production and reuse, and (5) lack of holistic techno-economic and LCA analyses of ELT recovery routs.

In terms of the application of ELT in pavements, the following specific research gaps were identified:

- Proper recycling of a rubberized asphalt mixture requires knowledge regarding its critical aging point (CAP), beyond which rejuvenation of the rubberized asphalt may not be effective. Even though the CAP of conventional asphalt has been established,[161] that of rubberized asphalt is yet to be researched.
- Limited attention has been paid to the self-rejuvenating feature of rubberized asphalt. Multiscale characterization can provide an in-depth understanding of this phenomenon.
- There is a lack of long-term data regarding the leachate of hazardous contaminants from rubberized asphalt binders, especially in extreme weather conditions or after extensive weathering.
- Although rubber pretreatment (surface activation, biotreatment, microbial treatment, preswelling, etc.) can improve the performance of rubberized pavements, a thorough comparison of the ecological and health implications of pretreatment techniques is needed.

7. CONCLUSION

This study reviewed the application of end-of-life tires (ELT) as fuel or a basis for some products and drew a prospect for its potential to promote a circular economy. The pros and cons of each application were highlighted in terms of performance, emission, ecological impact, health, and economy. In addition, various ELT pretreatment technologies, especially for civil engineering applications such as roadways, were compared.

ELT as fuel or products are associated with high emissions of toxic substances such as polycyclic aromatic hydrocarbons and volatile organic compounds.[41] A considerably more economical and environmentally benign destination for ELT is in construction materials such as pavement. However, pretreatment is usually required to improve the compatibility of ELT with the surrounding matrix (i.e., the asphalt mixture). All pretreatment technologies reviewed herein were found effective in enhancing ELT-matrix compatibility, thus improving the mechanical performance of the asphalt mixture. Surface activation via hybrid bio-oil and microwave irradiation appeared most effective because of the controllability of the process and allowing for tailoring the surface chemistry of rubber particles. Microbial devulcanization of rubber has shown great potential as a green approach to rubber treatment. Rubber particles also delay aging in asphalt while enhancing its durability and fatigue life. In addition, rubber-modified asphalt has a lower CO_2 equivalent, making it a smaller greenhouse gas emitter than conventional asphalt.[143,163] However, high concentrations of toxic and occasionally carcinogenic substances such as benzothiazole derivatives and 6PPD-quinone and heavy metals such as zinc, manganese, and cadmium in tires pose a significant risk of the leachate of these contaminants into soil and water resources, especially as weathering continues to age and erode pavements. This concern is particularly acute in open-graded mixtures, which show 1.2 times more toxicological effects and 2.9 times more carcinogenicity than conventional hot-mix asphalt. Besides the environmental concerns, the rubber dosage in the asphalt should be optimized to make it economically viable.

■ ASSOCIATED CONTENT

Data Availability Statement

All data used in this study have been included in the manuscript.

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■ REFERENCES

(1) Li, X.; Xu, H.; Gao, Y.; Tao, Y. Comparison of end-of-life tire treatment technologies: A Chinese case study. *Waste management* **2010**, *30* (11), 2235−2246.

(2) Mohammed, B. S.; Adamu, M.; Shafiq, N. A review on the effect of crumb rubber on the properties of rubbercrete. *International Journal of Civil Engineering and Technology* **2017**, *8* (9), 599−615.

(3) Liang, S.; Zhang, T. Comparing urban solid waste recycling from the viewpoint of urban metabolism based on physical input−output model: a case of Suzhou in China. *Waste management* **2012**, 32 (1), 220−225.

(4) Valentini, F.; Pegoretti, A. End-of-life options of tyres. A review. *Advanced Industrial and Engineering Polymer Research* **2022**, *5*, 203.

(5) *Recycled Tire Wire*; https://libertytire.com/Products/Commercial-Products/Wire/ (accessed 04.07.2023).

(6) *Mineral Commodity Summaries*; US Geological Survey [Electronic resource]: (accessed 19.05.2022), 2022; https://pubs.usgs.gov/periodicals/mcs2022/mcs2022.pdf.

(7) Campuzano, F.; Abdul Jameel, A. G.; Zhang, W.; Emwas, A.-H.; Agudelo, A. F.; Martínez, J. D.; Sarathy, S. M. Fuel and chemical properties of waste tire pyrolysis oil derived from a continuous twin-auger reactor. *Energy & Fuels* **2020**, *34* (10), 12688−12702.

(8) Jonusas, A.; Miknius, L. Effect of the pressure and feedstock state on the yield and properties of waste tire thermolysis products. *Energy & fuels* **2014**, *28* (4), 2465−2471.

(9) Campuzano, F.; Cardona-Uribe, N.; Agudelo, A. F.; Sarathy, S. M.; Martínez, J. D. Pyrolysis of Waste Tires in a Twin-Auger Reactor Using CaO: Assessing the Physicochemical Properties of the Derived Products. *Energy & Fuels* **2021**, *35* (10), 8819−8833.

(10) Muelas, A. l.; Aranda, D.; Callén, M. a. S.; Murillo, R. n.; Veses, A.; Asrardel, M.; Ballester, J. Properties and combustion characteristics of bio-oils from catalytic co-pyrolysis of grape seeds, polystyrene, and waste tires. *Energy & Fuels* **2020**, *34* (11), 14190−14203.

(11) *RUBBER SIDEWALKS, WALKWAYS, AND PATHS*; https://sustainablesurfacing.com/rubber-sidewalks (accessed 04/10/2023).

(12) Wądrzyk, M.; Janus, R.; Rządzik, B.; Lewandowski, M.; Budzyń, S. Pyrolysis oil from scrap tires as a source of fuel components: manufacturing, fractionation, and characterization. *Energy & Fuels* **2020**, *34* (5), 5917−5928.

(13) Joshi, P.; Goyal, P.; Goyal, P. K., Coastline Protection Using a Rubble Mound Seawall: A Case Study. In *Advances in Construction Materials and Sustainable Environment*; Springer: 2022; pp 321−330.

(14) Leknoi, U.; Likitlersuang, S. Good practice and lesson learned in promoting vetiver as solution for slope stabilisation and erosion control in Thailand. *Land use policy* **2020**, *99*, 105008.

(15) Liu, L.; Cai, G.; Zhang, J.; Liu, X.; Liu, K. Evaluation of engineering properties and environmental effect of recycled waste tire-sand/soil in geotechnical engineering: A compressive review. *Renewable and Sustainable Energy Reviews* **2020**, *126*, 109831.

(16) Liu, L.; Cai, G.; Liu, X. Investigation of thermal conductivity and prediction model of recycled tire rubber-sand mixtures as lightweight backfill. *Construction and Building Materials* **2020**, *248*, 118657.

(17) Fareghian, M.; Afrazi, M.; Fakhimi, A. Soil Reinforcement by Waste Tire Textile Fibers: Small-Scale Experimental Tests. *Journal of Materials in Civil Engineering* **2023**, *35* (2), 04022402.

(18) Modalavalasa, U.; Hatiwala, S. A.; Agarwal, B. K.; Pawar, S.; Patel, J. B., Dynamic Behavior of Retaining Wall Back Paneled by Waste Tire Shredded Rubber Fiber—An Experimental Study. In *Construction in Geotechnical Engineering*; Springer: 2020; pp 465−482.

(19) Thai, Q. B.; Chong, R. O.; Nguyen, P. T.; Le, D. K.; Le, P. K.; Phan-Thien, N.; Duong, H. M. Recycling of waste tire fibers into advanced aerogels for thermal insulation and sound absorption applications. *Journal of Environmental Chemical Engineering* **2020**, *8* (5), 104279.

(20) Deng, Y.; Morris, C.; Rakshit, S.; Landa, E.; Punamiya, P.; Sarkar, D. Water treatment residuals and scrap tire rubber as green sorbents for removal of stormwater metals. *Water Environment Research* **2016**, *88* (6), 500−509.

(21) Kaewunruen, S.; Li, D.; Chen, Y.; Xiang, Z. Enhancement of dynamic damping in eco-friendly railway concrete sleepers using waste-tyre crumb rubber. *Materials* **2018**, *11* (7), 1169.

(22) Hallmark-Haack, B. L.; Hernandez, N. B.; Williams, R. C.; Cochran, E. W. Ground tire rubber modification for improved asphalt storage stability. *Energy & Fuels* **2019**, *33* (4), 2659−2664.

(23) dos Santos, R. G.; Rocha, C. L.; Felipe, F. L. S.; Cezario, F. T.; Correia, P. J.; Rezaei-Gomari, S. Tire waste management: an overview from chemical compounding to the pyrolysis-derived fuels. *Journal of Material Cycles and Waste Management* **2020**, *22* (3), 628−641.

(24) Gamboa, A. R.; Rocha, A. M.; dos Santos, L. R.; de Carvalho, J. A., Jr Tire pyrolysis oil in Brazil: Potential production and quality of fuel. *Renewable and Sustainable Energy Reviews* **2020**, *120*, 109614.

(25) Martínez, J. D.; Lapuerta, M.; García-Contreras, R.; Murillo, R.; García, T. Fuel properties of tire pyrolysis liquid and its blends with diesel fuel. *Energy & Fuels* **2013**, *27* (6), 3296−3305.

(26) Hita, I.; Arabiourrutia, M.; Olazar, M.; Bilbao, J.; Arandes, J. M.; Castaño, P. Opportunities and barriers for producing high quality fuels from the pyrolysis of scrap tires. *Renewable and Sustainable Energy Reviews* **2016**, *56*, 745−759.

(27) Hita, I.; Rodríguez, E.; Olazar, M.; Bilbao, J.; Arandes, J. M.; Castaño, P. Prospects for obtaining high quality fuels from the hydrocracking of a hydrotreated scrap tires pyrolysis oil. *Energy & Fuels* **2015**, *29* (8), 5458−5466.

(28) Sathiskumar, C.; Karthikeyan, S. Recycling of waste tires and its energy storage application of by-products−a review. *Sustainable Materials and Technologies* **2019**, *22*, e00125.

(29) Sołowski, G.; Shalaby, M.; Özdemir, F. A. Plastic and Waste Tire Pyrolysis Focused on Hydrogen Production—A Review. *Hydrogen* **2022**, *3* (4), 531−549.

(30) Czajczyńska, D.; Krzyżyńska, R.; Jouhara, H.; Spencer, N. Use of pyrolytic gas from waste tire as a fuel: A review. *Energy* **2017**, *134*, 1121−1131.

(31) Oboirien, B. O.; North, B. C. A review of waste tyre gasification. *Journal of environmental chemical engineering* **2017**, *5* (5), 5169−5178.

(32) Karell, M. A.; Blumenthal, M. H. Air regulatory impacts of the use of tire-derived fuel. *Environmental progress* **2001**, *20* (2), 80−86.

(33) Pipilikaki, P.; Katsioti, M.; Papageorgiou, D.; Fragoulis, D.; Chaniotakis, E. Use of tire derived fuel in clinker burning. *Cement and Concrete Composites* **2005**, *27* (7−8), 843−847.

(34) Liberty Tire Recycling *Tire-Derived Fuel*. https://libertytire.com/Products/Commercial-Products/TireDerived-Fuel/#:~:text=Tire%2Dderived%20fuel%20(TDF),to%20disposing%20them%20in%20landfills. (accessed 11/30/2022).

(35) Caponero, J.; Tenório, J. A.; Levendis, Y. A.; Carlson, J. B. Emissions of batch combustion of waste tire chips: The afterburner effect. *Energy & Fuels* **2003**, *17* (1), 225−239.

(36) Singh, S.; Nimmo, W.; Gibbs, B.; Williams, P. Waste tyre rubber as a secondary fuel for power plants. *Fuel* **2009**, *88* (12), 2473−2480.

(37) Chatziaras, N.; Psomopoulos, C. S.; Themelis, N. J. Use of waste derived fuels in cement industry: a review. *Management of Environmental Quality: an international journal* **2016**, *27*, 178.

(38) Fernández Olmo, I. F.; Chacon, E.; Irabien, A. Influence of lead, zinc, iron (III) and chromium (III) oxides on the setting time and strength development of Portland cement. *Cement and concrete research* **2001**, *31* (8), 1213−1219.

(39) Market-Data-Forecast *Tire Derived Fuel Market Research Report - Segmentation By End Use Industry (Cement Kilns, Pulp and Paper Mills and Utility and Industrial Boilers) and Region - Industry Forecast 2023 to 2028*; 2023; p 175.

(40) Alfred Charles, C. *Global ELT Management − A global state of knowledge on regulation, management systems, impacts of recovery and*; World Business Council for Sustainable Development: Switzerland, 2019-12-03, 2019.

(41) Rahman, M. M.; Yu, Y.; Wu, H. Valorisation of Waste Tyre via Pyrolysis: Advances and Perspectives. *Energy & Fuels* **2022**, *36* (20), 12429−12474.

(42) Ćetković, J.; Lakić, S.; Žarković, M.; Vujadinović, R.; Knežević, M.; Živković, A.; Cvijović, J. Environmental Benefits of Air Emission Reduction in the Waste Tire Management Practice. *Processes* **2022**, *10* (4), 787.

(43) Goksal, F. P. An economic analysis of scrap tire pyrolysis, potential and new opportunities. *Heliyon* **2022**, *8* (11), e11669.

(44) *6PPD AND TIRE MANUFACTURING*; https://web.archive.org/web/20220325003324/https://www.ustires.org/6ppd-and-tire-manufacturing.

(45) *CalRecycle Rubberized Asphalt Concrete (RAC)*. https://calrecycle.ca.gov/tires/rac/ (accessed Dec.02,2022).

(46) Ghabchi, R.; Arshadi, A.; Zaman, M.; March, F. Technical challenges of utilizing ground tire rubber in asphalt pavements in the united states. *Materials* **2021**, *14* (16), 4482.

(47) Zapata, P.; Gambatese, J. A. Energy consumption of asphalt and reinforced concrete pavement materials and construction. *Journal of infrastructure systems* **2005**, *11* (1), 9−20.

(48) Zhang, L.; Zhang, C.; Zhang, Z.; Wang, H.; Wang, S. Characterization, Properties and Mixing Mechanism of Rubber Asphalt Colloid for Sustainable Infrastructure. *Polymers* **2022**, *14* (20), 4429.

(49) Duan, K.; Wang, C.; Liu, J.; Song, L.; Chen, Q.; Chen, Y. Research progress and performance evaluation of crumb-rubber-modified asphalts and their mixtures. *Construction and Building Materials* **2022**, *361*, 129687.

(50) Xu, N.; Wang, H.; Wang, H.; Kazemi, M.; Fini, E. Research progress on resource utilization of waste cooking oil in asphalt materials: A state-of-the-art review. *Journal of Cleaner Production* **2023**, *385*, 135427.

(51) Milad, A.; Ahmeda, A. G.; Taib, A. M.; Rahmad, S.; Solla, M.; Yusoff, N. I. M. A review of the feasibility of using crumb rubber derived from end-of-life tire as asphalt binder modifier. *Journal of Rubber Research* **2020**, *23* (3), 203−216.

(52) Mugnier, E.; Farhangi, C.; Kley, S.; Aurez, V.; Chhang, A., *The Socio-Economic Impact of Truck Tyre Retreading in Europe*, 2016.

(53) Tahami, S. A.; Mirhosseini, A. F.; Dessouky, S.; Mork, H.; Kavussi, A. The use of high content of fine crumb rubber in asphalt mixes using dry process. *Construction and Building Materials* **2019**, *222*, 643−653.

(54) Poovaneshvaran, S.; Zheng, L. W.; Hasan, M. R. M.; Yang, X.; Diab, A. Workability, compactibility and engineering properties of rubber-modified asphalt mixtures prepared via wet process. *International Journal of Pavement Research and Technology* **2021**, *14* (5), 560−569.

(55) Asaro, L.; Gratton, M.; Seghar, S.; Ait Hocine, N. Recycling of rubber wastes by devulcanization. *Resources, Conservation and Recycling* **2018**, *133*, 250−262.

(56) Soni, V. K.; Singh, G.; Vijayan, B. K.; Chopra, A.; Kapur, G. S.; Ramakumar, S. Thermochemical recycling of waste plastics by pyrolysis: a review. *Energy & Fuels* **2021**, *35* (16), 12763−12808.

(57) Diaz, R.; Colomines, G.; Peuvrel-Disdier, E.; Deterre, R. Thermo-mechanical recycling of rubber: Relationship between material properties and specific mechanical energy. *Journal of Materials Processing Technology* **2018**, *252*, 454−468.

(58) Maridass, B.; Gupta, B. Effect of extruder parameters on mechanical properties of revulcanized ground rubber tire powder. *Polimery* **2007**, *52* (6), 456−460.

(59) Zhang, X.; Zhu, X.; Liang, M.; Lu, C. Improvement of the properties of ground tire rubber (GTR)-filled nitrile rubber vulcanizates through plasma surface modification of GTR powder. *Journal of applied polymer science* **2009**, *114* (2), 1118−1125.

(60) Formela, K.; Cysewska, M.; Haponiuk, J.; Stasiek, A. The influence of feed rate and shear forces on the devulcanization process of ground tire rubber (GTR) conducted in a co-rotating twin screw extruder. *Polimery* **2013**, *58*, 906−912.

(61) Movahed, S. O.; Ansarifar, A.; Karbalaee, S.; Far, S. A. Devulcanization and recycling of waste automotive EPDM rubber powder by using shearing action and chemical additive. *Progress in Rubber Plastics and Recycling Technology* **2015**, *31* (2), 87−116.

(62) Liu, H.; Wang, X.; Jia, D. Recycling of waste rubber powder by mechano-chemical modification. *Journal of Cleaner Production* **2020**, *245*, 118716.

(63) Shi, J.; Jiang, K.; Ren, D.; Zou, H.; Wang, Y.; Lv, X.; Zhang, L. Structure and performance of reclaimed rubber obtained by different methods. *Journal of applied polymer science* **2013**, *129* (3), 999−1007.

(64) Seghar, S.; Asaro, L.; Rolland-Monnet, M.; Ait Hocine, N. Thermo-mechanical devulcanization and recycling of rubber industry waste. *Resources, Conservation and Recycling* **2019**, *144*, 180−186.

(65) Wu, X.; Wang, S.; Dong, R. Lightly pyrolyzed tire rubber used as potential asphalt alternative. *Construction and Building Materials* **2016**, *112*, 623−628.

(66) Ma, Q.; Guo, Z.; Guo, P.; Yang, F.; Li, H. Research on the Viscosity-Temperature Properties and Thermal Stability of Stabilized Rubber Powder Modified Asphalt. *Sustainability* **2021**, *13* (24), 13536.

(67) Dong, R.; Li, J.; Wang, S. Laboratory evaluation of pre-devulcanized crumb rubber−modified asphalt as a binder in hot-mix asphalt. *Journal of Materials in Civil Engineering* **2011**, *23* (8), 1138−1144.

(68) Zhang, H.; Zhang, Y.; Chen, J.; Liu, W.; Wang, W. Effect of desulfurization process variables on the properties of crumb rubber modified asphalt. *Polymers* **2022**, *14* (7), 1365.

(69) Li, B.; Li, H.; Wei, Y.; Zhang, X.; Wei, D.; Li, J. Microscopic properties of hydrogen peroxide activated crumb rubber and its influence on the rheological properties of crumb rubber modified asphalt. *Materials* **2019**, *12* (9), 1434.

(70) Lei, Y.; Wei, Z.; Wang, H.; You, Z.; Yang, X.; Chen, Y. Effect of crumb rubber size on the performance of rubberized asphalt with bio-oil pretreatment. *Construction and Building Materials* **2021**, *285*, 122864.

(71) Li, X.; Xu, X.; Liu, Z. Cryogenic grinding performance of scrap tire rubber by devulcanization treatment with ScCO2. *Powder Technology* **2020**, *374*, 609−617.

(72) Shatanawi, K. M.; Biro, S.; Naser, M.; Amirkhanian, S. N. Improving the rheological properties of crumb rubber modified binder using hydrogen peroxide. *Road Materials and Pavement Design* **2013**, *14* (3), 723−734.

(73) Shatanawi, K. M.; Biro, S.; Geiger, A.; Amirkhanian, S. N. Effects of furfural activated crumb rubber on the properties of rubberized asphalt. *Construction and Building Materials* **2012**, *28* (1), 96−103.

(74) Li, B.; Li, Q.; Zhu, X.; Wei, Y.; Li, Z.; Wei, D. Effect of Sodium Hypochlorite-Activated Crumb Rubber on Rheological Properties of Rubber-Modified Asphalt. *Journal of Materials in Civil Engineering* **2020**, *32* (11), 04020326.

(75) Wang, Z.; Xu, X.; Wang, X.; Jinyang, H.; Guo, H.; Yang, B. Performance of modified asphalt of rubber powder through tetraethyl orthosilicate (TEOS). *Construction and Building Materials* **2021**, *267*, 121032.

(76) Kazemi, M.; Fini, E. H. State of the art in the application of functionalized waste polymers in the built environment. *Resources, Conservation and Recycling* **2022**, *177*, 105967.

(77) Xie, J.; Yang, Y.; Lv, S.; Peng, X.; Zhang, Y. Investigation on preparation process and storage stability of modified asphalt binder by grafting activated crumb rubber. *Materials* **2019**, *12* (12), 2014.

(78) Xie, J.; Yang, Y.; Lv, S.; Zhang, Y.; Zhu, X.; Zheng, C. Investigation on Rheological Properties and Storage Stability of Modified Asphalt Based on the Grafting Activation of Crumb Rubber. *Polymers* **2019**, *11* (10), 1563.

(79) Xie, J.; Zhang, Y.; Yang, Y.; Ma, Y.; Li, J.; Huang, M. The effect of activation method of rubber on the performance of modified asphalt binder. *Materials* **2020**, *13* (17), 3679.

(80) Han, M.; Zeng, X.; Muhammad, Y.; Li, J.; Yang, J.; Yang, S.; Wei, Y.; Meng, F. Preparation of octadecyl amine grafted over waste rubber powder (ODA-WRP) and properties of its incorporation in SBS-modified asphalt. *Polymers* **2019**, *11* (4), 665.

(81) Liu, B.; Li, J.; Han, M.; Zhang, Z.; Jiang, X. Properties of polystyrene grafted activated waste rubber powder (PS-ARP) composite SBS modified asphalt. *Construction and Building Materials* **2020**, *238*, 117737.

(82) Kocevski, S.; Yagneswaran, S.; Xiao, F.; Punith, V.; Smith, D. W., Jr; Amirkhanian, S. Surface modified ground rubber tire by grafting acrylic acid for paving applications. *Construction and Building Materials* **2012**, *34*, 83−90.

(83) Kazemi, M.; Wang, H.; Fini, E. Bio-based and Nature Inspired Solutions: A Step Toward Carbon-neutral Economy. *Journal of Road Engineering* **2022**, *2* (3), 221−242.

(84) Zhou, T.; Kabir, S. F.; Cao, L.; Luan, H.; Dong, Z.; Fini, E. H. Comparing effects of physisorption and chemisorption of bio-oil onto rubber particles in asphalt. *Journal of Cleaner Production* **2020**, *273*, 123112.

(85) Kong, P.; Xu, G.; Yang, J.; Chen, X.; Zhu, Y. Study on Storage Stability of Activated Reclaimed Rubber Powder Modified Asphalt. *Materials* **2021**, *14* (16), 4684.

(86) Wang, H.; Jing, Y.; Zhang, J.; Cao, Y.; Lyu, L. Preparation and performance evaluation of swine manure bio-oil modified rubber asphalt binder. *Construction and Building Materials* **2021**, *294*, 123584.

(87) Yu, J.; Ren, Z.; Gao, Z.; Wu, Q.; Zhu, Z.; Yu, H. Recycled heavy bio oil as performance enhancer for rubberized bituminous binders. *Polymers* **2019**, *11* (5), 800.

(88) Li, Y.; Shen, A.; Lyu, Z.; Wang, S.; Formela, K.; Zhang, G. Ground tire rubber thermo-mechanically devulcanized in the presence of waste engine oil as asphalt modifier. *Construction and Building Materials* **2019**, *222*, 588−600.

(89) Ma, J.; Hu, M.; Sun, D.; Lu, T.; Sun, G.; Ling, S.; Xu, L. Understanding the role of waste cooking oil residue during the preparation of rubber asphalt. *Resources, Conservation and Recycling* **2021**, *167*, 105235.

(90) Yi, X.; Dong, R.; Tang, N. Development of a novel binder rejuvenator composed by waste cooking oil and crumb tire rubber. *Construction and Building Materials* **2020**, *236*, 117621.

(91) Faisal Kabirb, S.; Zakertabrizi, M.; Hosseini, E.; Fini, E. H. Effects of amide-based modifiers on surface activation and devulcanization of rubber. *Comput. Mater. Sci.* **2021**, *188*, 110175.

(92) Yin, J.; Wang, S.; Lv, F. Improving the short-term aging resistance of asphalt by addition of crumb rubber radiated by microwave and impregnated in epoxidized soybean oil. *Construction and Building Materials* **2013**, *49*, 712−719.

(93) Lyu, L.; Pei, J.; Hu, D.; Fini, E. H. Durability of rubberized asphalt binders containing waste cooking oil under thermal and ultraviolet aging. *Construction and Building Materials* **2021**, *299*, 124282.

(94) Hosseinnezhad, S.; Kabir, S. F.; Oldham, D.; Mousavi, M.; Fini, E. H. Surface functionalization of rubber particles to reduce phase separation in rubberized asphalt for sustainable construction. *Journal of cleaner production* **2019**, *225*, 82−89.

(95) Mousavi, M.; Hosseinnezhad, S.; Kabir, S. F.; Burnett, D. J.; Fini, E. H. Reaction pathways for surface activated rubber particles. *Resources, Conservation and Recycling* **2019**, *149*, 292−300.

(96) Kabir, S. F.; Mousavi, M.; Fini, E. H. Selective adsorption of bio-oils' molecules onto rubber surface and its effects on stability of rubberized asphalt. *Journal of Cleaner Production* **2020**, *252*, 119856.

(97) Formela, K.; Hejna, A.; Zedler, L.; Colom, X.; Cañavate, J. Microwave treatment in waste rubber recycling−recent advances and limitations. *Express polymer letters* **2019**, *13* (6), 565−588.

(98) Siores, E.; Do Rego, D. Microwave applications in materials joining. *Journal of materials processing technology* **1995**, *48* (1−4), 619−625.

(99) Hirayama, D.; Saron, C. Chemical modifications in styrene−butadiene rubber after microwave devulcanization. *Ind. Eng. Chem. Res.* **2012**, *51* (10), 3975−3980.

(100) Liang, M.; Xin, X.; Fan, W.; Ren, S.; Shi, J.; Luo, H. Thermo-stability and aging performance of modified asphalt with crumb rubber activated by microwave and TOR. *Materials & Design* **2017**, *127*, 84−96.

(101) Yang, X.; Shen, A.; Li, B.; Wu, H.; Lyu, Z.; Wang, H.; Lyu, Z. Effect of microwave-activated crumb rubber on reaction mechanism, rheological properties, thermal stability, and released volatiles of asphalt binder. *Journal of Cleaner Production* **2020**, *248*, 119230.

(102) Li, B.; Zhou, J.; Zhang, Z.; Yang, X.; Wu, Y. Effect of short-term aging on asphalt modified using microwave activation crumb rubber. *Materials* **2019**, *12* (7), 1039.

(103) Zhang, B.; Chen, H.; Zhang, H.; Wu, Y.; Kuang, D.; Guo, F. Laboratory investigation of aging resistance for rubberized bitumen modified by using microwave activation crumb rubber and different modifiers. *Materials* **2020**, *13* (19), 4230.

(104) Liang, M.; Ren, S.; Fan, W.; Wang, H.; Cui, W.; Zhao, P. Characterization of fume composition and rheological properties of asphalt with crumb rubber activated by microwave and TOR. *Construction and Building Materials* **2017**, *154*, 310−322.

(105) Liu, W.; Xu, Y.; Wang, H.; Shu, B.; Barbieri, D. M.; Norambuena-Contreras, J. Enhanced storage stability and rheological properties of asphalt modified by activated waste rubber powder. *Materials* **2021**, *14* (10), 2693.

(106) Bosco, F.; Antonioli, D.; Casale, A.; Gianotti, V.; Mollea, C.; Laus, M.; Malucelli, G. Biodegradation of unvulcanized natural rubber by microorganisms isolated from soil and rubber surface: A preliminary study. *Bioremediation Journal* **2018**, *22* (1−2), 43−52.

(107) Meyer-Cifuentes, I. E.; Werner, J.; Jehmlich, N.; Will, S. E.; Neumann-Schaal, M.; Öztürk, B. Synergistic biodegradation of aromatic-aliphatic copolyester plastic by a marine microbial consortium. *Nature communications* **2020**, *11* (1), 1−13.

(108) Tatangelo, V.; Mangili, I.; Caracino, P.; Bestetti, G.; Collina, E.; Anzano, M.; Branduardi, P.; Posteri, R.; Porro, D.; Lasagni, M.; et al. Microbial desulfurization of ground tire rubber (GTR): Characterization of microbial communities and rheological and mechanical properties of GTR and natural rubber composites (GTR/NR). *Polym. Degrad. Stab.* **2019**, *160*, 102−109.

(109) Ghavipanjeh, F.; Ziaei Rad, Z.; Pazouki, M. Devulcanization of ground tires by different strains of bacteria: optimization of culture condition by taguchi method. *Journal of Polymers and the Environment* **2018**, *26* (8), 3168−3175.

(110) Li, Y.; Zhao, S.; Wang, Y. Microbial desulfurization of ground tire rubber by Thiobacillus ferrooxidans. *Polym. Degrad. Stab.* **2011**, *96* (9), 1662−1668.

(111) Marchut-Mikołajczyk, O.; Drożdżyński, P.; Januszewicz, B.; Domański, J.; Wrześniewska-Tosik, K. Degradation of ozonized tire rubber by aniline−Degrading Candida methanosorbosa BP6 strain. *Journal of hazardous materials* **2019**, *367*, 8−14.

(112) Cui, X.; Zhao, S.; Wang, B. Microbial desulfurization for ground tire rubber by mixed consortium-Sphingomonas sp. and Gordonia sp. *Polym. Degrad. Stab.* **2016**, *128*, 165−171.

(113) Kim, J. K.; Park, J. W. The biological and chemical desulfurization of crumb rubber for the rubber compounding. *Journal of applied polymer science* **1999**, *72* (12), 1543−1549.

(114) Kabir, S. F.; Zheng, R.; Delgado, A. G.; Fini, E. H. Use of microbially desulfurized rubber to produce sustainable rubberized bitumen. *Resources, Conservation and Recycling* **2021**, *164*, 105144.

(115) Hou, X.; Xiao, F.; Wang, J.; Amirkhanian, S. Identification of asphalt aging characterization by spectrophotometry technique. *Fuel* **2018**, *226*, 230−239.

(116) Ghavibazoo, A.; Abdelrahman, M.; Ragab, M. Evaluation of oxidization of crumb rubber−modified asphalt during short-term aging. *Transportation Research Record* **2015**, *2505* (1), 84−91.

(117) Zhong, K.; Li, Z.; Fan, J.; Xu, G.; Huang, X. Effect of carbon black on rutting and fatigue performance of asphalt. *Materials* **2021**, *14* (9), 2383.

(118) Cong, P.; Xu, P.; Chen, S. Effects of carbon black on the anti aging, rheological and conductive properties of SBS/asphalt/carbon black composites. *Construction and Building Materials* **2014**, *52*, 306−313.

(119) Billiter, T.; Chun, J.; Davison, R.; Glover, C.; Bullin, J. Investigation of the curing variables of asphalt-rubber binder. *Petroleum science and technology* **1997**, *15* (5−6), 445−469.

(120) Ghavibazoo, A.; Abdelrahman, M. Effect of crumb rubber modification on short term aging susceptibility of asphalt binder. *International Journal of Pavement Research and Technology* **2014**, *7* (4), 297.

(121) Long, Z.; You, L.; Tang, X.; Ma, W.; Ding, Y.; Xu, F. Analysis of interfacial adhesion properties of nano-silica modified asphalt mixtures using molecular dynamics simulation. *Construction and Building Materials* **2020**, *255*, 119354.

(122) Tang, N.; Lv, Q.; Huang, W.; Lin, P.; Yan, C. Chemical and rheological evaluation of aging characteristics of terminal blend rubberized asphalt binder. *Construction and Building Materials* **2019**, *205*, 87−96.

(123) Yusoff, N. I. M.; Breem, A. A. S.; Alattug, H. N.; Hamim, A.; Ahmad, J. The effects of moisture susceptibility and ageing conditions on nano-silica/polymer-modified asphalt mixtures. *Construction and Building Materials* **2014**, *72*, 139−147.

(124) Zhou, T.; Zhou, J.; Li, Q.; Li, B. Aging properties and mechanism of microwave-activated crumb rubber modified asphalt binder. *Frontiers in Materials* **2020**, *7*, 603938.

(125) Yin, L.; Yang, X.; Shen, A.; Wu, H.; Lyu, Z.; Li, B. Mechanical properties and reaction mechanism of microwave-activated crumb rubber-modified asphalt before and after thermal aging. *Construction and Building Materials* **2021**, *267*, 120773.

(126) Chen, Z.; Wang, T.; Pei, J.; Amirkhanian, S.; Xiao, F.; Ye, Q.; Fan, Z. Low temperature and fatigue characteristics of treated crumb rubber modified asphalt after a long term aging procedure. *Journal of Cleaner Production* **2019**, *234*, 1262−1274.

(127) Wang, H.; Liu, X.; Apostolidis, P.; van de Ven, M.; Erkens, S.; Skarpas, A. Effect of laboratory aging on chemistry and rheology of crumb rubber modified bitumen. *Materials and Structures* **2020**, *53*, 1−15.

(128) Wang, L.; Cui, S.; Feng, L. Research on the influence of ultraviolet aging on the interfacial cracking characteristics of warm mix crumb rubber modified asphalt mortar. *Construction and Building Materials* **2021**, *281*, 122556.

(129) Wang, Q.; Li, S.; Wu, X.; Wang, S.; Ouyang, C. Weather aging resistance of different rubber modified asphalts. *Construction and Building Materials* **2016**, *106*, 443−448.

(130) Wang, S.; Huang, W.; Kang, A. Laboratory evaluation of the properties of high-cured crumb rubber modified asphalt containing sulfur and polymer after the oxidative aging procedure. *Construction and Building Materials* **2021**, *304*, 124611.

(131) Wang, S.; Wang, Q.; Li, S. Thermooxidative aging mechanism of crumb-rubber-modified asphalt. *J. Appl. Polym. Sci.* **2016**, *133* (16), 43323.

(132) Zadshir, M.; Ploger, D.; Yu, X.; Sangiorgi, C.; Yin, H. Chemical, thermophysical, rheological, and microscopic characterisation of rubber modified asphalt binder exposed to UV radiation. *Road Materials and Pavement Design* **2020**, *21* (sup1), S123−S139.

(133) Wang, R.; Xu, G.; Chen, X.; Zhou, W.; Zhang, H. Evaluation of aging resistance for high-performance crumb tire rubber compound modified asphalt. *Construction and Building Materials* **2019**, *218*, 497−505.

(134) Geng, J.; Chen, M.; Xia, C.; Liao, X.; Chen, Z.; Chen, H. Aging characteristics of crumb rubber modified asphalt binder and mixture with regenerating agent. *Construction and Building Materials* **2021**, *299*, 124299.

(135) Wang, S.; Gao, Y.; Yan, K.; You, L.; Jia, Y.; Dai, X.; Chen, M.; Diab, A. Effect of long-term aging on waste tire rubber and amorphous poly alpha olefin compound modified asphalt binder and its mixtures. *Construction and Building Materials* **2021**, *272*, 121667.

(136) Pang, L.; Liu, K.; Wu, S.; Lei, M.; Chen, Z. Effect of LDHs on the aging resistance of crumb rubber modified asphalt. *Construction and Building Materials* **2014**, *67*, 239−243.

(137) Noorvand, H.; Kaloush, K.; Medina, J.; Underwood, S. Rejuvenation Mechanism of Asphalt Mixtures Modified with Crumb Rubber. *CivilEng* **2021**, *2* (2), 370−384.

(138) Guo, M.; Liang, M.; Sreeram, A.; Bhasin, A.; Luo, D. Characterisation of rejuvenation of various modified asphalt binders based on simplified chromatographic techniques. *International Journal of Pavement Engineering* **2022**, *23*, 4333.

(139) Salehi, S.; Arashpour, M.; Kodikara, J.; Guppy, R. Sustainable pavement construction: A systematic literature review of environmental and economic analysis of recycled materials. *Journal of Cleaner Production* **2021**, *313*, 127936.

(140) Zarei, M.; Rahmani, Z.; Zahedi, M.; Nasrollahi, M. Technical, economic, and environmental investigation of the effects of rubber powder additive on asphalt mixtures. *Journal of Transportation Engineering, Part B: Pavements* **2020**, *146* (1), 04019039.

(141) Zarei, M.; Mirbaha, B.; Akbarinia, F.; Rahmani, Z.; Zahedi, M.; Zarei, A. Application of concordance analysis method (CA) for optimal selection of asphalt mixtures reinforced with rubber powder and carbon fiber. *Electronic Journal of Structural Engineering* **2020**, *20*, 53−62.

(142) Wang, T.; Xiao, F.; Zhu, X.; Huang, B.; Wang, J.; Amirkhanian, S. Energy consumption and environmental impact of rubberized asphalt pavement. *Journal of Cleaner Production* **2018**, *180*, 139−158.

(143) Wang, Q.-Z.; Wang, N.-N.; Tseng, M.-L.; Huang, Y.-M.; Li, N.-L. Waste tire recycling assessment: Road application potential and carbon emissions reduction analysis of crumb rubber modified asphalt in China. *Journal of cleaner production* **2020**, *249*, 119411.

(144) Halsband, C.; Sørensen, L.; Booth, A. M.; Herzke, D. Car tire crumb rubber: does leaching produce a toxic chemical cocktail in coastal marine systems? *Frontiers in Environmental Science* **2020**, *8*, 125.

(145) Dillon, E. The effect of temperature on zinc leaching from rubber tire mulch. *Journal of the South Carolina Academy of Science* **2017**, *15* (2), 15.

(146) Rhodes, E. P.; Ren, Z.; Mays, D. C. Zinc leaching from tire crumb rubber. *Environmental science & technology* **2012**, *46* (23), 12856−12863.

(147) Ishchenko, V. Heavy metals in municipal waste: the content and leaching ability by waste fraction. *Journal of Environmental Science and Health, Part A* **2019**, *54* (14), 1448−1456.

(148) Selbes, M.; Yilmaz, O.; Khan, A. A.; Karanfil, T. Leaching of DOC, DN, and inorganic constituents from scrap tires. *Chemosphere* **2015**, *139*, 617−623.

(149) Tian, Z.; Zhao, H.; Peter, K. T.; Gonzalez, M.; Wetzel, J.; Wu, C.; Hu, X.; Prat, J.; Mudrock, E.; Hettinger, R.; et al. A ubiquitous tire rubber−derived chemical induces acute mortality in coho salmon. *Science* **2021**, *371* (6525), 185−189.

(150) Sankhla, M. S.; Kumar, R.; Prasad, L. Zinc impurity in drinking water and its toxic effect on human health. *Indian Internet Journal of Forensic Medicine & Toxicology* **2019**, *17* (4), 84−87.

(151) Bleackley, M. R.; MacGillivray, R. T. Transition metal homeostasis: from yeast to human disease. *Biometals* **2011**, *24* (5), 785−809.

(152) Azizian, M. F.; Nelson, P. O.; Thayumanavan, P.; Williamson, K. J. Environmental impact of highway construction and repair materials on surface and ground waters: Case study: crumb rubber asphalt concrete. *Waste management* **2003**, *23* (8), 719−728.

(153) U.S. EPA *Drinking Water Regulations and Contaminants*. https://www.epa.gov/sdwa/drinking-water-regulations-and-contaminants (accessed Dec. 17, 2022).

(154) Liu, X.; Wang, J.; Gheni, A.; ElGawady, M. A. Reduced zinc leaching from scrap tire during pavement applications. *Waste Management* **2018**, *81*, 53−60.

(155) Fathollahi, A.; Makoundou, C.; Coupe, S. J.; Sangiorgi, C. Leaching of PAHs from rubber modified asphalt pavements. *Sci. Total Environ.* **2022**, *826*, 153983.

(156) Zanetti, M. C.; Farina, A. Life Cycle Risk Assessment Applied to Gaseous Emissions from Crumb Rubber Asphalt Pavement Construction. *Sustainability* **2022**, *14* (9), 5716.

(157) Zanetti, M. C.; Santagata, E.; Fiore, S.; Ruffino, B.; Dalmazzo, D.; Lanotte, M. Evaluation of potential gaseous emissions of asphalt rubber bituminous mixtures. Proposal of a new laboratory test procedure. *Construction and Building Materials* **2016**, *113*, 870−879.

(158) Zanetti, M. C.; Fiore, S.; Ruffino, B.; Santagata, E.; Lanotte, M. Assessment of gaseous emissions produced on site by bituminous mixtures containing crumb rubber. *Construction and Building Materials* **2014**, *67*, 291−296.

(159) Zhang, J.; Chen, M.; Wu, S.; Zhou, X.; Zhao, G.; Zhao, Y.; Cheng, M. Evaluation of vocs inhibited effects and rheological properties of asphalt with high-content waste rubber powder. *Construction and Building Materials* **2021**, *300*, 124320.

(160) Cao, L.; Yang, C.; Li, A.; Wang, P.; Zhang, Y.; Dong, Z. Flue gas composition of waste rubber modified asphalt (WRMA) and effect of deodorants on hazardous constituents and WRMA. *J. Hazard. Mater.* **2021**, *403*, 123814.

(161) Yang, X.; You, Z.; Perram, D.; Hand, D.; Ahmed, Z.; Wei, W.; Luo, S. Emission analysis of recycled tire rubber modified asphalt in hot and warm mix conditions. *Journal of hazardous materials* **2019**, *365*, 942−951.

(162) Lv, Y.; Wu, S.; Li, N.; Cui, P.; Wang, H.; Amirkhanian, S.; Zhao, Z. Performance and VOCs emission inhibition of environmentally friendly rubber modified asphalt with UiO-66 MOFs. *Journal of Cleaner Production* **2023**, *385*, 135633.

(163) Lushinga, N.; Dong, Z.; Cao, L.; Yang, C.; Ullah, S. Fatigue characteristics and environmental sustainability of rubber asphalt composite modified asphalt mixtures incorporating nanosilica. *Construction and Building Materials* **2022**, *346*, 128382.