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REVIEW

4R of rubber waste management: current and outlook

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Abstract

Excessive accumulation of rubber waste necessitates the need to revisit the effectiveness of the existing rubber waste management system. This review provides an overview of the legislative frameworks, techniques, challenges, and trends of rubber waste management in various countries. The 4R (reduce, reuse, recycle and recover) framework applied in waste management system in some countries appears to be viable for the processing of rubber waste. Certain countries especially some of the European Union (EU) members have implemented extended producer responsibility (EPR) system to manage the collection of rubber waste, particularly used tires. The processing of rubber waste in each level of the 4R hierarchy was then discussed, with detailed elaboration on the most practiced 'R', recycling which encompasses the direct recycling of products, as well as material recycling via physical and/or chemical means. The challenges faced in the implementation of rubber waste management system in different countries were highlighted and recommendations for a more sustainable rubber consumption were provided at the end of this review.

Keywords Rubber waste treatment · Waste management system · 4R policy · Sustainable rubber consumption

Introduction to 4R concept of rubber waste management

Rubber manufacturing and production remain strong throughout the years due to high market demand. Based on the statistic, the amount of natural rubber (NR) produced in 2015 has doubled compared to that in 2000. Thailand was the major NR exporting country in 2020, with export values totaling 3.5 billion U.S. dollars, and this accounted for about 30 percent of the global value of NR shipments that year [1]. Demand for rubber is expected to remain strong in the near future as rubber products are still very relevant and in need for various niche applications. An increase in the rubber production and consumption would inevitably increase the production of rubber waste, and improper handling or processing of the waste could harm the environment. This can be perceived as an obstacle to achieve Goal 12 of the United Nations' Sustainable Development Goals (SDG12) on "Responsible Consumption and Production" which aims to achieve good management of waste and reduce its accumulation through the practice of 3R waste management [2]. Rubber products, especially the synthetic ones are quite resistant to natural degradation due to the vulcanization process [3]. The sulfur crosslinks formed between the polymer chains are meant to improve the mechanical strength, as well as the physical and chemical properties of the rubber. However, the complex three-dimensional structure of the vulcanized rubber also makes it more difficult to be decomposed via biodegradation. Besides, the presence of additives, antioxidants, and fillers also fosters its resistance towards biodegradation [3, 4].

The used tires are one of the largest sources of rubber waste in the environment due to the large proportion of raw rubber that goes into the tire production sector, which is about 70% of the annual NR production [5, 6]. It was forecasted that the amount of discarded waste tires each year could reach 1.2 billion pieces by the year 2030 [7]. Besides, the usage of rubber gloves is also on surge in light of the SARS-CoV-2 pandemic situation beginning March 2020

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when personal protective equipment (PPE), such as masks and gloves are widely used. As of June 2020, it was estimated that approximately 129 billion pieces of face masks which ear loops are made of polyisoprene rubber [8], and 65 billion pieces of rubber gloves are disposed each month globally [9]. Other major sources of rubber waste include automotive components, such as shock absorbers and conveyor belts, and municipal solid wastes, such as clothing, shoe soles, toys, electric wires, sofas, and cushions [10].

Traditional methods of rubber waste management are usually landfill disposal or incineration. However, several negative environmental implications may result from the landfill disposal of rubber waste, and this includes the leaching of toxic substances and heavy metals into the groundwater. Besides, the stockpile may cause inextinguishable fire as the rubber can serve as excellent fuel to sustain combustion [11]. An unstoppable fire incident that lasted for 18 days took place at a landfill site containing about 20.5 million kg of tires in Iowa City, Iowa, USA in 2012. The fire caused the release of excessive hazardous air pollutants, such as carbon dioxide, sulfur dioxide, fine particulate mass (PM2.5), polycyclic aromatic hydrocarbons (PAH), etc. [12, 13]. Incineration of rubber waste without proper control will also contribute to the emission of greenhouse gases (GHGs) [14].

To improve the handling of rubber waste, 4Rs (reduce, reuse, recycle, and recover) waste management was introduced by the European Commission on 19 November 2008 to be adopted as the common practice in waste processing [15, 16]. The distinguishable difference between the 4R Framework and the 3R Framework is the fourth R (Recover), which re-defines the incineration of waste materials in terms of the energy recovery efficiency [15]. The most prioritized item in the 4R hierarchy is "Reduce", which refers to the practices that decrease the generation of waste, in this case, rubber waste [17, 18]. "Reuse" helps to prolong the lifetime of the rubber products for the same purpose of usage. "Recycling" may involve grinding of the rubber waste into small particles (ground rubber), or devulcanization that breaks the crosslinks between polymer chains, and re-purposed for other applications [19]. The last R, "Recover" usually indicates the process of decomposing the waste by heat into fuels and/or other valuable substances with lower molecular weights [19, 20].

Legislation framework and regulations on rubber waste management in different countries

Intervention from federal and/or local governments is essential for effective rubber waste management, and many countries have implemented various policies and regulations on issues related to waste management. Malaysia, being the fifth largest country in the production and export of NR products has a clear legislation framework on waste management [21], shown in Fig. 1. The Solid Waste Management and Public Cleansing Act (Act 672) was implemented in 2011 to improve the waste management service in seven states in the country [23]. Bursa Malaysia has also mandated its members to perform Corporate Social Responsibility Reporting (CSRR) annually from 2007 onwards, which includes concerns on pollution control and waste management, with the hope to better manage the environmental health [24]. In 2010, approximately 245,000 tons of used tire was generated, but its flow distribution was not recorded and there is lack of update in recent years [22].

In the European Union (EU), the member states were obliged to manage rubber waste, specifically waste tires under two EU policies which are the 1999 Directive on the Landfill of Waste 1000/31/EC, and the end-of-life vehicle directive 2000/53/EC [25, 26]. Under these policies, stockpiling of waste tires other than bicycle tires and those with an external diameter > 1.4 m were prohibited, and the tires must be removed from the end-of-life vehicles for recycling [26]. The policy is still in operation and updated to raise the reuse and recycling rate from 80% in 2006 to 85% beginning January 2015, and the rate of reuse and recovery was raised from 85 to 95% [27]. However, the outline of the recycling procedure of the tires was not provided in these policies. Consequently, each member state of the EU had developed its regulations for its operations [26]. This in turn leads to the development of three different systems: extended producer responsibility (EPR), tax system, and free market system [28]. Amongst the three systems, the EPR system which requires the producers or importers to bear the responsibility for waste-tire recycling is adopted in most of the member states in the EU [28]. Implementation of EPR in Portugal has successfully increased the waste recycling rates from 69 to 98%, although improvement in consistency of the management was needed [29]. A comparative study on waste tire management between Italy and Romania showed that different pathways of management were attributed to the different regulations and economic context of the two countries [30]. EPR system adopted in both countries had led to the collection of used tires amounting to 403,000 tons for Italy and 46,000 tons for Romania in 2012. 18.1% of the collected tires in Italy were reused, while the remaining was distributed to recycling (36.1%), energy recovery (57.9%), and landfilling (6%). The end-of-life tires (ELTs) in Romania were channeled to recycling (43.5%) and energy recovery (56.5%) [30]. A study on the comparison of the EPR system between Belgium, The Netherlands, and Italy was also conducted, and the findings showed that EPR helps to increase resource efficiency, reduce illegal stockpiling, and move up the waste hierarchy [31]. For Belgium, a report published by the Public Waste Agency of Flanders stated that 51,375 tons

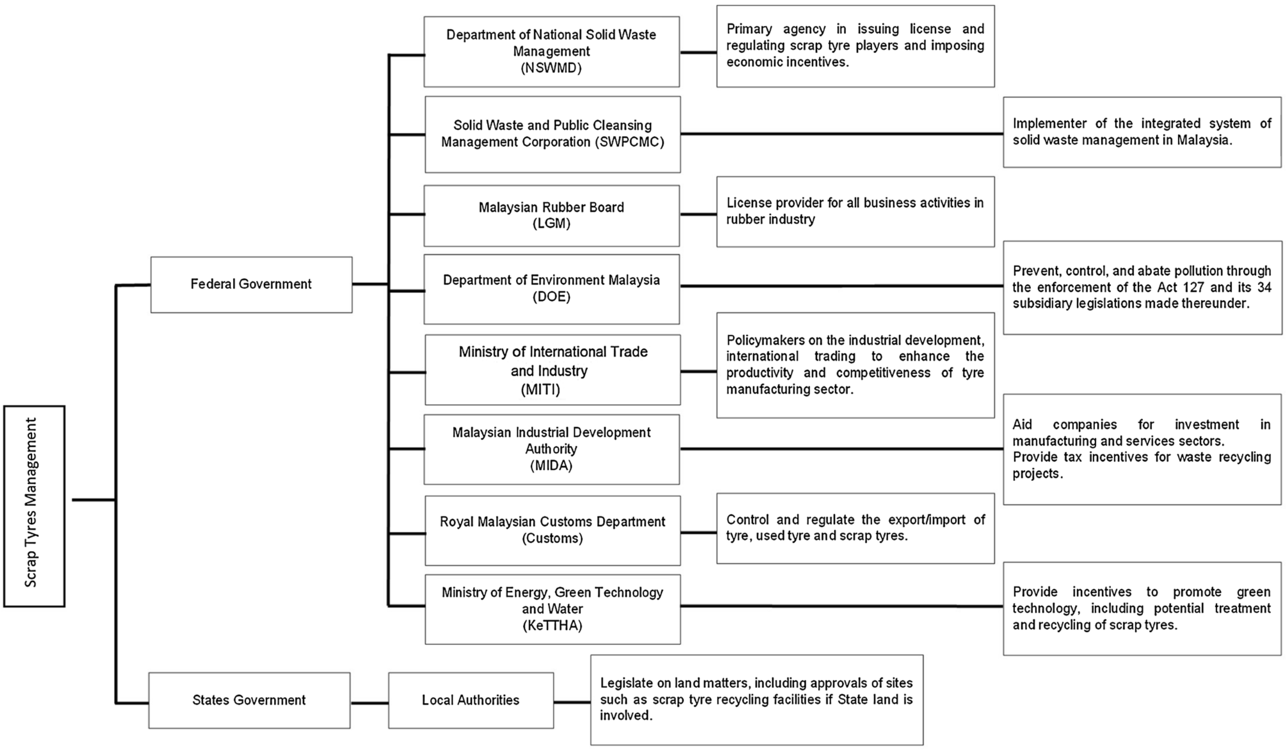

Fig. 1 Government agencies regulations of rubber waste management (particularly tire products) [22]

of waste were collected in 2012 in the Flanders region, plus a collection rate of 88% was attained in the previous year. Of all the collected tires in Belgium, 85.0% were directed for recycling and reuse, while 15.0% were intended for energy recovery [32]. Poland which adopted a free market system in waste management implemented the Act of 11 May 2001 which required the entrepreneurs to manage their wastes by means of recovery and recycling [33, 34]. Waste tires were included under this act and the percentage of recovery and recycling was set at 75 and 15%, respectively in 2011 [33, 35]. A recent report in 2017 found that in 2014, the recovery rate of the collected tires had reached 76.3% while the recycling rate achieved 24.6%, which satisfied the set requirements [36]. It was; however, unclear if the recovery rate reported is inclusive of the recycling of rubber materials or exclusive to energy recovery.

Japan has implemented various laws for environmental protection, laws to reduce the consumption of natural resources and its environmental burden, as well as laws for proper waste management by practicing 3Rs in treating municipal and industrial wastes [37, 38]. End-of-life vehicle (ELV) recycling act was implemented in Japan in 2005 [37, 39], and the recycling of ELV follows the EPR concept whereby the responsibilities of ELV processing are on the manufacturers and importers using the fee collected from the vehicle owners [39]. A report in 2020 stated that Japan has collected 937,000 tons of ELTs, in which energy recovery was the dominant process of managing the waste tires, amounting to about 65% from the total [40]. In South Korea, the Plastic Waste Control Plan (PWCP) was established in 2018 by the Korean Ministry of Environment based on the Resource Circulation Act (RCA) and Resource Circulation Master Plan (RCMP). The objective of the plan was to reduce at least 50% of the plastic waste and recycle over 70% of the plastic waste produced by 2030, wherein synthetic rubber waste was included in the term 'plastic waste' [41]. South Korea recycled and recovered 70% of the waste tires generated per annum (500,000 metric tons) as tire-derived fuel (TDF) to emphasize energy recovery and control emissions. Collaboration from the relevant authorities with Sumitomo Heavy Industries was made to set up two TDF co-firing Circulating Fluidized Bed Combustion (CFBC) boiler plants to rely less on the use of coal while encouraging the involvement of TDF for power and heat generation [42].

In Turkey, Environmental Law (EL), and Regulation on Control of the End-of-Life Tires (RCELT) are two regulations that are responsible to control and manage the ELTs [34]. The RCELT helps to set up a transport system to collect the ELT. Under this regulation, proper management of ELTs was practiced in which recovery is strongly encouraged while import, disposal at landfill, or open burning were prohibited [34]. Recorded statistics showed that 72.4

kilotons (kt) of ELTs were collected, of which 33.4 kt was recycled and 33.9 kt was recovered in cement plants [34]. Colombia is one of the countries in South America that has adopted EPR as well, since 2007. The implementation of EPR was only partially successful due to governance issues, although the amount of recycled tire wastes did increase following the implementation. The responsibilities of waste tire management, both operational and financial were imposed on the tire producers alone, causing the lack of participation from other players in the tire production chain [43]. A total of 45,800 tons ELTs was collected in the year 2016, of which the main processing method was recycling (70%) followed by recovery (22%) and reusing (8%) [43].

Canada collected an estimate of 508 kt waste rubber tires in 2019, and about 450 kt of ELTs (date not specified) was recycled [44]. EPR was practiced in Canada, and there were more than ten provincial and territorial EPR provincial tire stewardship programs. Waste Diversion Act (WDA) was also enacted to supervise the diversion of target waste streams away from landfills [45]. Such scheme and the cooperation among local authorities managed to divert bulk of the waste tires from landfills, with the average diversion rate of about 98% over the past ten years [46]. In Australia, 69% of 466,000 metric tons of ELTs was managed through reusing (26.3%), recycling into tire-derived products (58.8%), and recovery process (14.9%), respectively [47]. Although the figures reported are rather impressive, Belgium had however outperformed Australia in terms of environmental performance by a factor of 7.9 [48]. Two reasons were provided for the observation; first, the regulatory framework in Australia favors the profit-driven automotive recycling industries which paid more attention to low costing materials, and secondly, the voluntary-based waste policy caused price competition between legitimate and illegitimate sectors. In turn, the non-adherence to the environmental standards and competitive prices offered by some of the illegitimate recyclers could demotivate the legitimate recyclers in the movement of proper ELT treatment [48]. To mitigate this problem, the enactment of ELV legislation was refined in 2021 to prohibit the exportation of unprocessed ELTs and maintain the exportation right exclusive to the licensed exporters [49].

Moving the focus to Brazil, only 10% recycling rate was achieved for annual tire disposal weighing 300 tons despite various enacted laws and regulations [50]. One of the enacted regulations is The Normative Instruction N °001/2010 from the National Council of the Environmental of Brazil (CONAMA), under which the manufacturers and importers are obligated to recycle 100% of the outstanding tires in Brazil, and companies are to bear the responsibilities of ELTs and their destinations [51]. Besides, Brazil has also enacted Publication of Resolution No. 416/09 which specifies the installation of collection points at a frequency of 1 to 100,000 inhabitants in cities to aid the collection of wastes. It however did not produce a practical effect as there were only 4 out of 120 proposed collection points being set up in São Paulo [51]. There is no denying that problems like poor infrastructure, insufficient funding, and attitudinal behavior of the citizen and corporates that are driven by profitability have limited the effectiveness of waste management in many countries, including Brazil [52].

Waste tire management in mainland China appears to be somewhat effective with a recycling rate of about 30%, comparable with countries such as Japan and Poland. However, a report by W. Huang in 2020 suggested that that the figure could have been higher in China and pointed out to several factors that may have retarded the recycling rate, and this includes the lack of regulations, government support, and relevant agencies. It was stated that the recycling system of car tires worked more effectively than that for tires from bicycles and electric motorbikes. The popularity of bicycles and motorbikes in China could have therefore contributed to the country's lower than expected rubber recycling rate [53]. The increase in disposable income and the reduction in the price of tires contributed to the surge in waste tires from bicycles and electric motorbikes, which exceeded the number of waste tires from cars. The author commented that Vietnam also experienced a similar problem due to higher usage of motorbikes than cars [53]. There is a lack of intervention from government agencies and the recycling of rubber waste is mostly managed by the private sector [54]. In Anh's findings in 2015, Vietnam generated about 400,000 tons of waste tires per annum. 50% of it was disposed or landfilled, while 40% was converted into thermal energy, and 10% for recycling and reuse [54]. In Taiwan, the Environmental Protection Agency established the Recycling Fund Management Board in 1998 which is responsible for inspection and management of the waste tire recycling process [55]. Collected waste tires were converted through shredding and grinding into rubber chips or powder for recycling or recovery purposes. It was recorded that in 2012, 80% of the rubber pieces from the waste tires was channeled as the auxiliary fuel source, while 15% was for the rubber materials, and 5% was directed to pyrolysis [55].

Techniques in rubber waste management

There are several techniques that have been reported in rubber waste management, and this includes ways to reuse rubber, rubber product and material recycling, as well as recovering energy from the rubber waste. A comparison of the pros and cons of each reported technique is shown in Table 1.

Table 1 Comparison of various techniques in rubber waste management [26, 55–60]

Techniques	Pros	Cons
Retreading	Reduce the utilization of rubber resources Reduce rubber waste generation	Require high operator's technical level Require high investment on retreading equipment
Product recycling	Recycling of entire tires without any treatments	Possible risk of leaching of additives and degraded materials into the environment
Pyrolysis	Economically competitive Produce wide range of products (hydrocarbons, CB, steel wire, etc.)	Yield fuel may lead to engine performance problem (due to high sulfur, ash, and char content)
Material recycling	Retrieve rubber material for the use in production of composite material/blending	Requires shredding and granulating equipment
Downsizing		Requires controlled conditions to prevent degradation of properties
Reclamation/Devulcanization		Chemical usage may cause toxicity or pollution

Retreading and reuse

Retreading technology is applied on the end-of-life tires where the worn treads are replaced with new treads to allow the tires to be reused, and such technology prevents the piling up of waste tires in the environment [61, 62]. The general process involved in the retreading of a used tire is shown in Fig. 2. The carcass of the waste tire will first be inspected for reusability, after which the grinding and repairing procedure will follow to remove the tire crown from the carcass. After repairing, a piece of buffer rubber will be attached prior to the pasting of pre-vulcanized tread rubber. The last step of the process is the vulcanization of the pasted rubber tread. Finally, the products are inspected before they are released into the market [57].

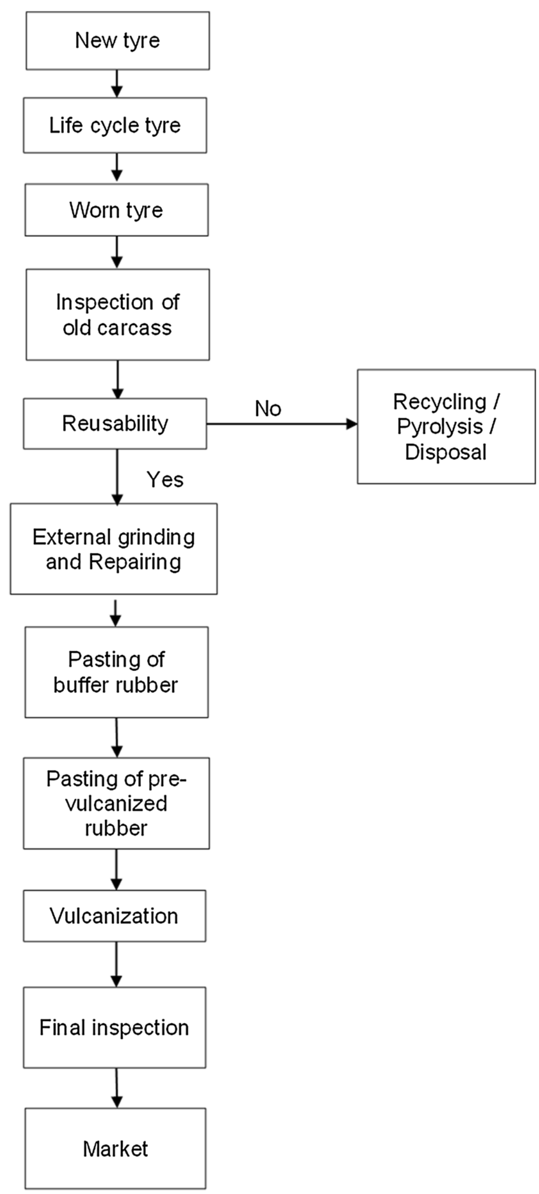

Fig. 2 General flow of tire retreading process [57]

Product recycling

Direct recycling of used tires while retaining their original forms was seen in various applications. These include the use in boats as fenders, scrap tires for the construction of artificial reef, as insulation for the foundation of buildings, etc. [26]. Some of the applications are however not widely practiced in recent times due to the adverse impacts they had on the environment. For instance, the initiative of using waste tires to construct an artificial reef in Florida in 1967, with the intention to create a marine habitat was unsuccessful. The dispersion of the waste tires brought by the water currents and storms damaged the coral reef, leading to an extra cost of up to 30 million USD to remove 2 million pieces of waste tires from the coast [58].

Pyrolysis

Pyrolysis normally refers to the breakdown of the rubber waste via incineration in an anaerobic condition, into smaller compounds such as fuel oil, gas, carbon black (CB), sulfur, and metal [63, 64]. The fuel oil produced could be refined to filter out the sulfur, char, and ash to ensure better engine performance. The gas can be used to drive the generation of heat and electricity in power plants [65], meanwhile the CB can be blended into plastic, EVA foam, or converted into activated carbon [55].

Material recycling

(a) Downsizing rubber particles

Description of different methods of downsizing rubber material and the characteristics of the ground rubber particles produced from each method are given in Table 2. Each of the methods has its own pros and cons. Dry ambient grinding of rubber is a relatively simple process that is carried by physical grinding of the rubber at ambient temperature, and the crumbs produced have high surface area to volume ratio. However, the friction between the rubber surface and grinder generates high amount of heat, and the temperature of the rubber may reach as high as 130 °C, resulting in oxidation of the rubber surface. Another relatively simple method, wet ambient grinding is also able to produce ground particles with a high surface area to volume ratio, but such method requires a long post-grinding drying period. Rubber can also be downsized using high water pressure jet, and such technique is environmentally friendly and cost-effective. However, highly trained personnel are needed to ensure smooth operation of the process. Berstoff's method is an effective method to produce rubber particles with small grain size, but such method requires invested facilities such as an industrial miller and twin-screw extruder. Finally, the cryogenic method has also been adopted to produce ground rubber particles, and one apparent advantage of the technique is the ability to prevent surface oxidation on the rubber. This method is however quite costly due to the high consumption of liquid nitrogen in the process [59].

The crumb rubber produced could regain its application in different industries. Oil absorptivity of a blend produced from mixing crumb rubber with 4-tert-butylstyrene (tBS) was reported by Wu and Zhou. Results from their study showed that the blend ratio of 60% waste tire crumb rubber to 40% tBS could attain a maximum oil sorption capacity of 24 g/g, a value which is slightly lower than the commercial counterpart used for benchmarking (30 g/g) [66]. A separate study was conducted by Odeh and Okpaire in 2020 to investigate the effect of rubber particle size on the oil absorptivity. They concluded that the optimized mesh size of 0.15 mm could achieve oil absorptivity of 4.71 g/g due to its great surface area to volume ratio [67]. Considering the reusability of the waste tire powder of up to 100 times, the total amount of oil that can be absorbed throughout the service time of the waste tire powder could be more than 200 g/g, a value which is higher than some of the single use commercially available oil sorbent [68]. A barrier made of recycled tires performed the best in soundproofing among all other materials such as concrete, metal, and wood. The barrier effectively reduced the noise level of a residential area near highways from 90 to 53 dBA [69]. Preparation of stone mastic asphalt added with crumb rubber and limestone yielded similar volumetric and mechanical properties, and a superior feature of reducing tire-road noise level as compared to the standard mixture [70]. Incorporation of crumb rubber was also proven to constrict the emission of CO and CH_4 significantly [71].

(b) Reclamation and devulcanization of rubber waste

The principle of rubber reclamation and devulcanization lies in the breaking of crosslinks between the polymeric chains of the rubber. It should be made clear that reclamation and devulcanization are distinct from each other in terms of definition. Devulcanization refers to the scission of crosslinks, such as C-S and S–S bonds, whereas reclamation refers to the scission of both the crosslinks and the main chain bonds [72]. The physical method of rubber devulcanization takes the advantage of overcoming the lower bond energies of C-S bond (310 kJ mol^{-1}) and the S–S bond (270 kJ mol^{-1}), while rubber reclamation involves an additional C–C bond which have higher bond energy, 370 kJ mol^{-1} [73]. In

Table 2 Different downsizing methods used in recycling rubber waste [59]

Methods	Description	Characteristics of the ground rubber particles
Dry ambient	Repeated grinding of rubber waste until crumb rubber is obtained	Size approximately 300 µm rough and irregular surface
Wet ambient	Grind suspension of shredded rubber in the presence of water as coolant	Size approximately 100 µm rough and irregular surface
Water jet	Uses pressurized water jet (> 2000 bar) to strip and breaks rubber	Rough and irregular surface
Berstoff's method	Uses the combination of rolling mill and twin-screw extruder	Rough and irregular surface
Cryogenic	Rubber cooled in liquid nitrogen and shattered using impact type mill	Size approximately 75 µm sharp edge, flat/ smooth surface

reality however, the main chain scission could still occur in the devulcanization process due to the fact that the carbon–carbon bonds are present in higher abundance than the crosslinking bonds [74]. Different techniques of rubber reclamation, as well as their advantages and disadvantages, are summarized in Table 3.

(i) Thermomechanical method In thermomechanical devulcanization, the breaking of crosslinks is accomplished using a combination of high temperature and shear force. Many different equipment and techniques used for thermomechanical devulcanization have been reported. Formela et al. in 2014 found out that an increase in the treatment temperature decreases the screw torque required to reclaim ground tire rubber (GTR), and the properties of the reclaimed rubber are also affected by the temperature used. Out of three treatment temperatures (60, 120, and 180 °C), the one produced at 60 °C had the best mechanical properties, suggesting it experienced the least main chain scission. GTR which were reclaimed at 180 °C however incorporates better and more homogeneously in other polymeric matrices such as in styrene-butadiene rubber (SBR) blend to give better mechanical properties [74]. In 2019, Seghar et al. studied the effect of temperature, from 80 to 220 °C on the devulcanization of NR by feeding it at a rate of 5 kg/h into an industrial screw extruder with a screw speed of 240 rpm. They discovered that the degree of reclamation can achieve approximately 90% irrespective of the temperatures, but 80–100 °C range was reported to yield the best devulcanized rubber quality, presumably due to the highest tendency of sulfur bond scission in that temperature range. It was reported that NR has the tendency to undergo self-heating when the shear strain was applied to it, leading to the cleavage of the sulfur bonds. They concluded that relatively low energy consumption is needed to promote the recycling of NR [82]. Another study by Piszczyk et al. in 2015 was conducted to investigate the effect of GTR produced by thermomechanical reclamation on the properties of polyurethane foam. Results showed that the incorporation of reclaimed GTR by up to 30 wt% had resulted with an increase in apparent density, from 290 to 315 kg/m^3, and increased the initial degradation temperature of the polyurethane composite by 14 °C. Blending the polyurethane foam with GTR however, did not affect the compressive strength of the composite [83].

(ii) Microwave method This method utilizes the heating effect of the microwave to cleave the crosslinks in the rubber, provided that the rubber is sufficiently polar to absorb the microwave radiation [60]. For rubber with a poor absorptivity of the radiation such as NR, SBR, and ethylene-propylene-diene rubber (EPDM), polar fillers which absorbs microwave radiation such as silica and CB could be incorporated to improve the devulcanization process [78, 84]. This was reported in a study carried out by de Sousa et al. in 2015 to determine the effect of CB content and length of exposure time to microwave on the degree of devulcanization. It turned out that the gel content of the devulcanized NR showed negative correlations to both variables [84]. Moreover, the efficiency of devulcanization via microwave irradiation also depends on the devulcanizing agent and types of oil used in the process. In 2016, Molanorouzi and Mohaved published their studies on the reclamation of rubber waste using various devulcanizing agents and 2 types of oils (paraffinic and aromatic). Of all the agents, compositions, and conditions experimented, the best result was obtained by using 30 phr of aromatic oil, 6 phr of diphenyl disulfide, DPDS and a temperature of 240 °C. Aromatic oil is preferred probably because it enhanced the solubility of DPDS in it. Interestingly, the polysulfidic crosslinks decreased greatly, but the monosulfidic linkage increased [85].

(iii) Ultrasonic method The scission of crosslinks (C-S and S–S bonds) in vulcanized rubber can be accomplished by the vibrations induced by wave energy that leads to the formation of cavities in the polymer matrix [77]. A study by Isayev et al. in 2014 established a relationship between the

Table 3 Comparison of different reclamation or devulcanization techniques [74–81]

Techniques	Advantages	Disadvantages
Physical		
Thermomechanical	High selectivity for crosslink scission Easy to scale up for industrial application	Potential main chain scission due to high abundancy of carbon–carbon bond
Microwave	Homogenous heating environmentally friendly	Restricted to polar rubber molecule which absorbs microwave, otherwise requires suitable additives
Ultrasound	Efficient process that can takes place within a few seconds	High energy input required Costly
Chemical	Wide selection of devulcanizing chemicals and high efficiency	Harmful to environment (secondary pollution)
Biological	High selectivity for crosslink bond by desulfurization enzymes	Microbial degradation is very slow. Devulcanization only happen on rubber surface

particle size or surface area of the rubber and the efficiency of devulcanization. It was found that under the same amplitude of ultrasonic wave and temperature 250 °C, rubber of size 30 mesh experienced greater extent of devulcanization that led to lower gel content compared to that of size 10 mesh [77]. Another interesting research by Sun and Isayev in 2008 revealed a positive correlation between the ultrasonic amplitude with the degree of devulcanization. It was found that the addition of processing oil could aid the devulcanization of isoprene rubber (IR) and NR. They also carried out a comparison on the degree of devulcanization of CB-filled IR and NR, and the results obtained showed that the effect of CB loadings on devulcanization of both rubbers was different, suggesting the stereoregular structure of rubber could play a role in affecting the extent of devulcanization [86].

(iv) Chemical method The chemical method of rubber reclamation involves the use of chemicals to break the crosslinks between rubber chains and/or to block the successful recombination of sulfur linkage between the polymer chains. Many types of chemicals have been reported to serve as effective devulcanizing agents for rubber, and they include sulfides, peroxides, amines, deep eutectic solvents, and ionic liquids.

The use of diphenyl disulfide, DPDS as a devulcanizing agent was reported by Vega et al. in 2007, during which they performed devulcanization of rubber using a microwave coupled with DPDS. The incorporation of DPDS facilitated the devulcanization process, evidenced by the evolution of a high amount of squalene, and triterpene [87]. In 2012, Jiang et al. devulcanized butyl rubber with different amounts of DPDS, from 0 to 5 g, in combination with supercritical CO_2 to swell the rubber vulcanizate. It was found that the sol fraction increased from about 15 to 98.5% when the amount of DPDS was raised from 0 to 4 g, indicating that the DPDS was involved in the devulcanization process rather than being the result of thermal degradation alone [88]. Thiosalicylic acid was used by Thaichaoroen et al. to reclaim the NR vulcanizate through the mechano-chemical method. The NR vulcanizate was heated at 140 °C for 30 min with different loading of thiosalicyclic acid after it was milled at room temperature. The result indicated that 1 phr thiosalicylic acid produced the optimal devulcanization effect and they claimed that the thiosalicylic acid had comparable devulcanization efficiency as the DPDS [80].

Zhang et al. in 2021 reported the use of hydrogen peroxide (H_2O_2) to reclaim SBR. The SBR was turned into powder and blended with soybean oil and H_2O_2 at different ratios. The mixed sample was then heated in a drying oven at 100 °C for 4 h with consistent air supply. After 4 h, the experiment yielded 100% sol fraction, suggesting complete devulcanization of the rubber [89]. Another study by Sabzekar et al. in 2015 investigated the effect of reaction time, temperature, and the amount of benzoyl peroxide as devulcanizing agent on the devulcanization efficiency of sulfur-cured NR. It was shown that cleavage of the crosslink bonds was achieved at lower concentrations of the benzoyl peroxide, from 2 to 8 phr, and a further increase in the concentration of the peroxide led to non-selective cleavage. A shorter reaction time of 2 h was recommended to prevent main chain scission, and it was found that a reaction temperature of 110 °C with low benzoyl peroxide content of up to 4 phr could drastically reduce the crosslink density of the rubber [90].

Walvekar et al. in 2018 performed the devulcanization of waste tire rubber with amines in combination with ultrasonic treatment at different temperatures and rubber: amine ratios. They found that tertiary amine (3-aminopropyltrimethoxysilane) produced better outcome compared to primary amine [(n-diethyl-3-aminopropyl) trimethoxysiloxane] at sonication temperature of 50 °C, with gel content of the treated rubber at 63–77% and 75–87%, respectively [91]. Walvekar et al. also investigated the use of deep eutectic solvent, DES as a devulcanizing agent. Rubber waste was devulcanized via ultrasonic method using zinc chloride: urea at the mole ratio of 2: 7 and 1: 4 at temperatures 30, 130, 150, and 180 °C. DES with $ZnCl_2$: urea in (2: 7) ratio requires temperature of 130 °C for optimal devulcanization, and the higher temperature resulted in bond reformation that decreased the devulcanization efficiency. Temperature higher than 130 °C was however required for $ZnCl_2$: urea ratio (1:4) mixture for better devulcanization effect. Based on the high sol fraction produced, > 85%, it was concluded that DES comprising of $ZnCl_2$ and urea is very effective for desulfuration of rubber [92]. Pyrrolidinium hydrogen sulfate ionic liquid, IL was used in combination with microwave treatment by Seghar et al. in 2015 to devulcanize SBR. Different microwave energy from 0 to 440 Wh/kg and 10 wt % of the IL was used in the experiment. It was observed that the sol fraction is positively correlated to the microwave energy, and the addition of the IL further increased the sol fraction at microwave energy > 220Wh/kg, confirming the role of the IL as devulcanizing agent [93].

(v) Biological method Unlike chemical method which deals with a variety of harmful chemical solvents, the biological degradation of rubber polymer offers a safer alternative as the devulcanization process is catalyzed by enzymes produced from microorganisms [76]. In 2012, Li et al. used Thiobacillus sp. for devulcanization of GTR and investigated the properties of NR/devulcanized GTR (NR/dGTR) composite. An increase in the wettability of the composite was observed in which the water contact angle decreased from 120.5 to 93.5°. This is attributed to about 30% increase in oxygen content in the dGTR after oxidative desulfurization by the Thiobacillus sp. The tear strength of the composite improved when the rubber waste content was below 30 phr due to better homogeneity, as well as due to the presence of high concentration of active sites for revulcanization to takes place [94]. Yao et al. reported the ability of Alicyclobacillus sp. to devulcanize rubber latex waste (prevulcanized) at a concentration of 5% (w/v) with the aid of Tween 80 as surfactant. A 62.5% decrement in the sulfur content and a 34.9% increment in oxygen content after 10 days of co-culturing were reported, pointing to successful oxidative cleavage of the sulfur crosslinks. This outcome was supported by the change of water contact angle from 104.3 to 85.0° which means an increase in the hydrophilicity of the latex rubber was achieved after desulfurization [95].

(vi) Supercritical devulcanization Recent devulcanization technology uses CO_2 at supercritical state ($scCO_2$) as the reaction medium for devulcanization. CO_2 is known for its non-flammability, non-toxicity, and chemical inactivity, all of which are desirable properties to ensure the safety of the process [96]. It was also reported to have a critical point that is easily achieved and good diffusivity which helps the swelling process for the diffusion of devulcanization agent into the rubber material [97]. $scCO_2$ was used as a medium to devulcanize unfilled NR/SBR blend under non-isothermal conditions at 120 °C set-point temperature and 8 MPa pressure, with the use of bulb crude extract (BCE) from *Tulbaghia violacea* as the devulcanizing agent. It was reported that the use of 1.0 wt % of BCE managed to increase the sol content of the vulcanizate to a maximum of about 45% at reaction time $t = 17$ min, and this is a big improvement compared to the untreated control (~ 19%) [97]. Devulcanization of GTR was conducted by Mangili et al. in 2014 using DPDS in $scCO_2$ at temperature 180 °C and pressure 15 MPa. The sol fraction of the GTR increased from 1.1 to 8.3 wt %, accompanied by a reduction in the crosslink density from 0.082 to 0.037. The changes were attributed to the crosslink scission whereby the sulfur crosslinks were broken and reacted with radicals produced from DPDS [98]. Liu et al. in 2015 investigated the parameters involved in devulcanization of waste tread rubber by using $scCO_2$ as the medium. Their findings suggested that DPDS concentration of at least 10 g L^{-1}, pressure below 7.38 MPa, and temperature of 140 °C are suitable for the scission of sulfur crosslinks in the waste tread rubber. $scCO_2$ increased the cleavage of the crosslinks as it facilitated the devulcanization agent DPDS to reach the crosslink bonds [99]. In another study, GTR was produced by jet pulverization of $scCO_2$, along with the addition of DPDS as the devulcanization agent. Application of $scCO_2$ led to the swelling of the waste tire rubber, enhancing the penetration of the DPDS into the rubber for efficient devulcanization. Result from FTIR analysis showed that the use of $scCO_2$ allowed the radical reactions to incorporate benzene rings and cleave the sulfur crosslinks [96].

Biodegradability of NR and its usefulness in biological recycling of waste rubber

NR, in chemistry context, is the polymeric product of the isoprene unit connected through carbon-1 and -4 at cis-configuration [100]. The raw material of NR can be identified from over 2000 plant species, but the major contributor of NR in the rubber industry comes from *Hevea brasiliensis*. The naturally occurring polymer has been reported to be degradable by native microbes.

Kasai et al. published an article in 2017 regarding the biodegradability of deproteinized natural rubber, DPNR by a soil bacterium, *Rhizobacter gummiphilus* NS21. NS21 was incubated on the DPNR-overlay agar plate for 3 days, followed by Schiff's staining and gel permeation chromatography (GPC) analysis. The color development indicated the cleavage of the long poly(cis-1,4-isoprene) chain to produce short-chain products with terminal aldehyde groups. Result from GPC analysis agreed with the suggestion that *R. gummiphilus* degraded the DPNR, with notable decrease in the peak height at MW = 1300 kDa and increase in peak height at MW = 110 kDa [101, 102]. Degradation of various NR products inclusive of fresh latex stripes, latex condom, latex glove, and car tires was also observed after incubation with *Streptomyces* sp. CFMR7 which was isolated from rubber plantation site. Detection of the carbonyl group in the fungus-treated fresh latex pieces using FTIR-ATR spectroscopy underpinned the oxidative cleavage of the C = C bond by the *Streptomyces* sp. CFMR7. The transition of the MW distribution from higher to lower value was verified through GPC analysis, and collectively the results suggests that NR could be biodegraded by the microorganisms found in the native environment [103, 104].

Nguyen et al. in 2020 performed a study on the bacteria consortia enriched from rubber-processing factory's waste in Vietnam to compare the biodegradation of NR and DPNR film. The consortia were enriched using the soil and wastewater samples from the factory and incubated with NR or DPNR with constant shaking at 150 rpm, and temperature 30 °C for 2 weeks. It was found that the highest weight loss achieved for NR was 35.4% which was lower than DPNR's 48.4%, and this is due to the presence of protein as the natural antioxidant to protect the NR from oxidative degradation. The degradation of proteins by Proteobacteria and Bacteroidetes caused the NR to be susceptible to the chain cleavage reaction from microorganisms. Among the genus in the consortia, *Gordonia* and *Mycobacterium* in the phylum *Actinobacteria* were identified to be the main bacteria mediating the biodegradation reaction of the NR [105].

18 actinobacterial strains were reported by Basik et al. in 2021 to have the NR degradation capability through

random screening on NR latex agar. Surprisingly, two rare strains related to *Microtetraspora glauca* (strain AC03309) and *Dactylosporangium sucinum* (strain AC04546) were discovered amongst the 18 strains. Both strains were tested for their capability to degrade latex gloves and tire granules by incubation in 50 mL of mineral salts medium, MSM and 0.5% (w/v) rubber material, with constant shaking at 180 rpm and temperature 28 °C for 30 days. SEM analysis showed that both strains were able to colonize and utilize the rubber material, among which the AC04546 strain showed a good utilization of rubber from tire samples. The authors suspected that the rubber-degrading ability of these strains may be stimulated by their exposure to the rubber particles deposited in the environment [106].

Streptomyces sp. AC04842 identified from a soil sample was found to be able to attach and colonize latex pieces, latex glove strips, and tire granules, a deduction made based on SEM images that shows the extension of mycelia to the surface of the materials. Degradation on the latex pieces and glove strips by *Streptomyces* sp. was recognized through the presence of holes and cavities in the polymer under SEM images after they were cultivated in the fungal culture for 60 days at 28 °C. Besides, the formation of oxygen-carrying bonds after incubation with *Streptomyces* sp. supported the authors' claim that degradation process had taken place. [107]. *Lactobacillus plantarum* (strain LOCK 1145) is another bacterium that is capable of degrading NR, discovered by Olejnik et al. in 2022. In the experiment, various rubber materials were cultured with the bacteria at 30 °C for 14 days, and the average carbon content of the samples has reduced from the range of 89.1–95.4% to 56.5–65.7%. Pore formation observed in the NR vulcanizate along with the recorded mass loss of 1–5% strongly implies that the bacterial strain could degrade the rubber material [108].

Collectively, the findings summarized above suggest that natural rubber can be biodegraded by selected microbial strains. The ability of the microbes to degrade NR should be improved further through biotechnological research to enhance the biodegradation of waste rubber. While Hevea rubber offers the advantage of being an environmentally friendly source for rubber, the sole dependence on this tropical plant might not be the best option. Rapid expansion of Hevea rubber tree plantation may leave a negative environmental impact such as biodiversity loss and climate change [109]. Other sources of NR should be exploited and among the promising ones include those from Guayule or Kazakh dandelion. Although there are various challenges to the industrial production of NR from these plants, they are nevertheless candidates with great potential to increase the global natural rubber supply. These plants are native to temperate regions which potentially are the emerging sources of NR other than Hevea rubber [110].

Future outlook on rubber waste issues: prospects and challenges

The generation of rubber waste will continue to increase as the demand for rubber products continues to remain strong in the near future. Although the most common rubber product being discussed was rubber tires, the volume of rubber products in the medical field such as rubber gloves, ear-loop bands, and catheters are increasing significantly in light of the COVID-19 pandemic. Effective implementation of rubber waste management to resolve the growing amount of rubber waste requires close cooperation between the government and the industry stakeholders. Such cooperation is also crucial to ensure that sustainable development can take place parallel with economic development. Unfortunately, it remains a challenge to maintain a balance between economy and the environmental protection as these two aspects are often of causal relationship [111]. Poorer countries may not have much option but to prioritize the economic development of their respective countries over environmental preservation, especially during this time when the global economy is badly affected by the ongoing Covid-19 pandemic. The use of face mask and other rubber products such as rubber gloves led to the generation of additional rubber waste, which is normally incinerated or sent to licensed landfill sites after sterilization [112, 113]. It is likely that the current waste management system inclusive of rubber waste will remain as it is, and any improvement to the system will only take place when the global economy has recovered from the pandemic.

To develop an effective and efficient rubber waste management system, there are a few challenges that need to be considered. First, it is undeniable that the legislative framework in a country is important for good rubber waste management. The lack of laws and regulations in controlling the disposal of rubber waste, along with the lack of action in the waste management industry and support from the government are believed to be some of the main reasons behind the poor recycling and recovery of rubber from the waste. The case study in Colombia had shown that the government did not incentivize the waste processors and the end-users in their EPR model, and this has resulted in a lack of participation from the industry players and consumers in recycling the waste tires [43]. In contrast, the rubber waste management system in the EU seems to be relatively well managed, with the presence of EPR system in managing the collection and recycling of rubber waste. It is plausible that the system will continue to support the recycling of rubber waste products beyond waste tires in the future.

Rubber waste management is an industry where the initial capital needed is very high as it requires the construction of the recycling and recovery facilities such as the pyrolytic treatment plant which are often very costly. Since a huge amount of capital must be invested for the setting of the treatment plant, giant reactors, efficient rubber waste collection system, etc., not many entrepreneurs have the capacity to venture into the waste management business. In addition to the cost factor, the risk from the uncertain return of investment, ROI could shy away investors too. It is noteworthy that the quality of the recycled products may vary due to the differences in the composition of the rubber wastes, as well as due to the processing condition such as the temperature, pressure, catalysts, and types of reactors used [114, 115]. The inconsistency may affect the properties of the recycled products, and this makes commercialization much more difficult.

Conclusion and recommendations

4R waste management framework (Reduce, Reuse, Recycling, and Recover) seems to be a viable and practical approach to address the excessive rubber waste accumulation issue and relieve the environmental consequences brought by it. Out of the 4R, 'Recycling' is the most adopted practice, and material recycling which encompasses grinding of rubber waste into smaller particles, and devulcanization techniques to retrieve the rubber component appear to be some of the most applied techniques in the rubber waste management industry. This is probably because the techniques allow greater utilization of waste material, and the retrieved rubber can be used to generate added value products with decent qualities.

Based on our findings from reviewing the current development in rubber waste management and its related issues, the followings are some of the recommendations to address the issues and promote more sustainable rubber consumption:

(1) The policy for rubber waste management should be carefully devised to increase the recycling rate. The rubber waste management model and relevant legislations that have been successfully implemented in some of the EU member countries should be adopted by others to improve the rubber recycling rate. From economic standpoint, rubber waste management is a challenging industry that requires high capital, high operating cost, and uncertain return on investment, leading to a lack of participation from the industry players and investors. Government should incentivize the industry to promote it and encourage more participation. Studies on the cost return of rubber waste management could be helpful for the government to devise a framework for rubber waste management system to maximize the profit from rubber waste recycling.

(2) From a long-term perspective, it is important to establish rubber recycling and recovery systems in all countries. Considering the effectiveness and encouraging results reported in many countries, these initiatives should be prioritized in rubber waste management. However, in some of the developing countries which currently lack the necessary basic infrastructures for rubber recycling and recovery exercise, the use of NR may be considered over inert synthetic rubbers, where possible. As elaborated in the earlier section, natural rubber has the potential to be degraded by various native microbes leading to wide range of weight loss, while synthetic ones such as nitrile rubber tend to be highly resistant to biodegradation. Besides, prioritizing naturally derived products could help to reduce the dependency on petrochemicals which are nonrenewable and known to add carbon footprint to the environment. However, more detailed studies on the effectiveness of biodegradation of NR in landfill and its potential impact to the surrounding ecosystem are necessary before any recommendation can be made on its suitability to be part of mainstream rubber waste management system. The effect of wide range of additives formulated in commercial NR products on their biodegradability need to be carefully evaluated too. For now, recycling and recovery remains as the best options when it comes to treatment of rubber waste.

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Declarations

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