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Review

Recycling of Rubber Wastes as Fuel and Its Additives

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Abstract: Economic, social, and urban developments generally require improvements in the transportation sector, which includes automobiles such as trucks, buses, trailers, airplanes, and even bicycles. All these vehicles use rubber tires. After consumption, these tires become waste, leading to enlarged landfill areas for used tires and implying additional harm to the environment. This review summarizes the growth of rubber recycling application and the sustainability of using waste rubber in the construction field. Furthermore, we provide methods to convert rubber waste to fuel or fuel additives by using tire-derived fuel and concentrate to pyrolysis, which are environmentally friendly and efficient ways. The related parameters such as temperature, pressure, and feedstock composition were studied. Most research papers observed that 500 °C is the optimal temperature at atmospheric pressure in the presence of a specific type of catalyst to improve pyrolysis rate, oil yield, and quality.

Keywords: rubber waste; fuel; fuel additives; sustainability; pyrolysis; recycling

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1. Introduction

1.1. Ecosystem Limitation

Ecosystem services are the advantages people receive from nature. Human survival and well-being depend on these services. Thus, humans must provide the protection and best management to ecosystems [1,2]. The contamination of seas by polymer products is expected to cause a yearly loss of 1–5% in ecosystem services, resulting in financial losses of roughly USD 2.5 billion per year [3,4]. Plastic from (<1 m to less than 1 µm) have the ability to interact with aquatic creatures from all trophic levels and thus have various harmful impacts [5–7].

Extensive and rising quantities of polymer pollution exist in the aquatic environment, which we henceforth refer to as "marine plastic" [8]. Approximately 4.8–12.7 million metric tons of plastic entered the oceans water from land-based sources in 2010 alone, and the flux of plastics to the seas is forecast to increase by an order of level within the next decade [9]. Over time, these plastics may disintegrate into little parts, called "microplastics" (0.1 µm–5 mm), whose massive majority is expected to persist in the environment in several forms over geological periods [10].

1.2. Rubber-Application Growth

As we mentioned previously, there is a current global concern about the environment, with fuel emission consequences to the atmosphere on one hand, and how to gain advantage from other items which cause different types of pollution on other hand.

Rubber is an example of an elastomer class of polymer with the ability to return to its original shape after being stretched or deformed, called natural rubber [11]. Figure 1 shows the natural rubber structure.

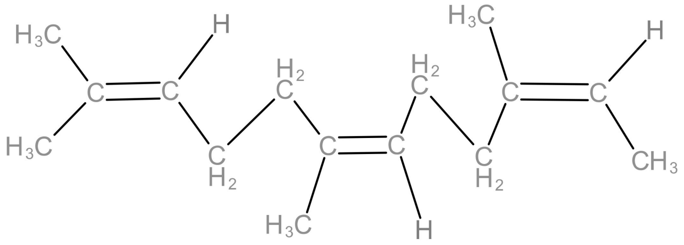

Figure 1. Natural rubber structure.

Conversely, synthetic rubber (also called commercially styrene–butadiene rubber (SBR)), is a copolymer of 1,3-butadiene and styrene mixed in a 3:1 ratio [12]. Rubber has a deep effect on the environment and takes multiple forms. We use it in daily activities such as protective gloves, electrical instruments, and plastics, and one of the most crucial is tires. Figures 2 and 3 shows the consumption of natural and synthetic rubber worldwide (2017–2020).

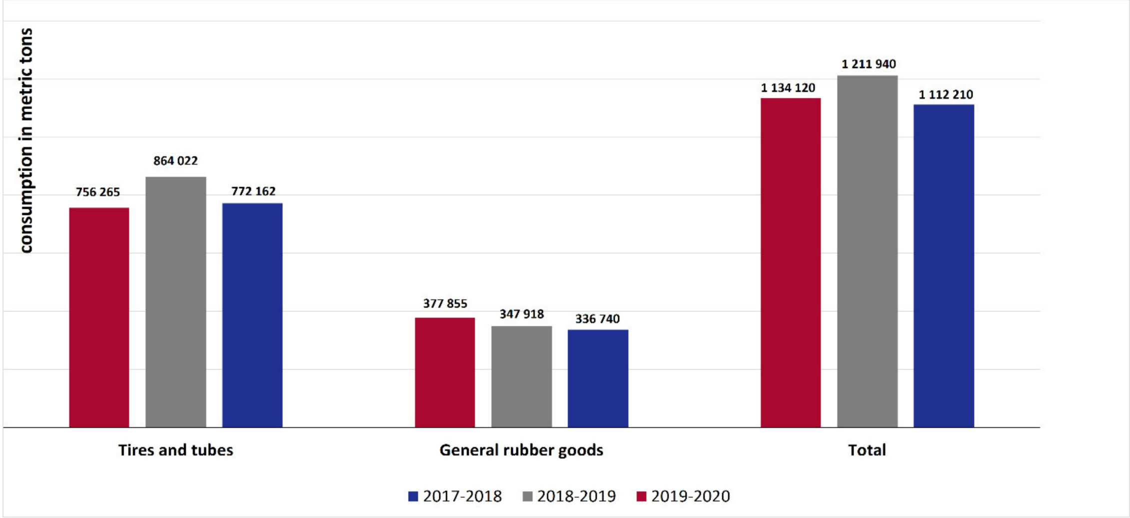

Figure 2. Consumption of natural rubber. Source: statista.com, 27 May 2021.

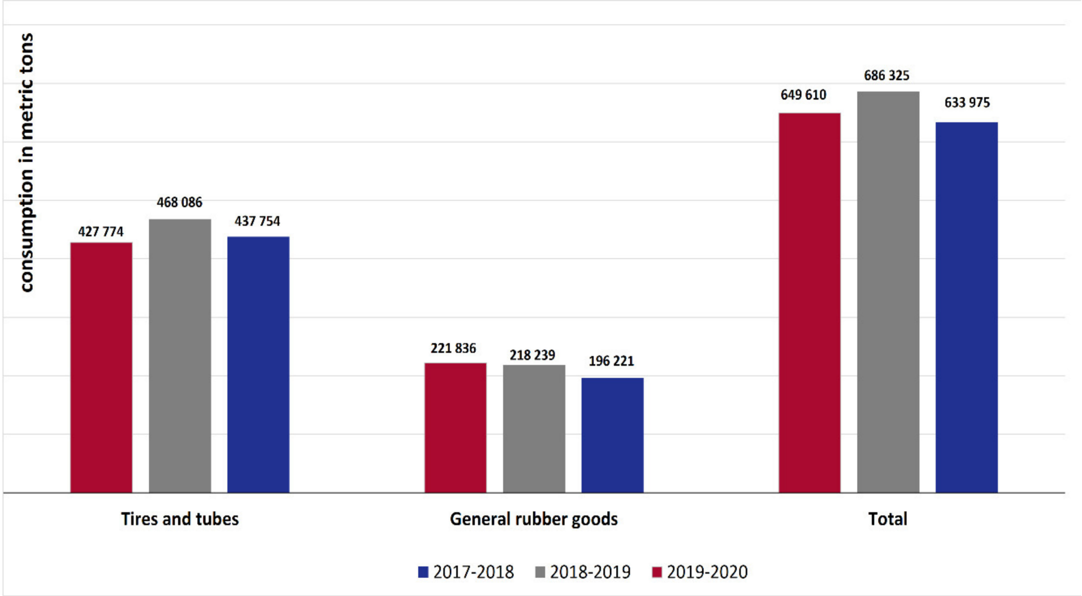

Figure 3. Consumption of synthetic rubber. Source: statista.com, 27 May 2021.

2. Current Usage of Waste Rubber

2.1. Waste-Tire Recycling Industry

The global demand for tires will reach 3.2 billion units in 2022 [13]. A total of about 223 million replacement passenger tires were shipped to the USA in 2019 [14]. Scrap tires could take up space in landfills, or serve as breeding grounds for mosquitoes and rodents when stockpiled or illegally dumped [15]. The U.S. Tire Manufacturers Association published its 2019 Scrap Tire Management Report on 14 October. According to the report, tires remain one of the most recycled products in the U.S., but end-of-life markets are not keeping pace with the annual generation of scrap tires. The report reveals that nearly 76% of scrap tires have been recycled in products such as rubber-modified asphalt, automotive products, mulch for landscaping, and tire-derived fuel in 2019. This value is down from 96% in 2013, when scrap tire recycling peaked [16]. Tires are composed of various elements, including rubber/elastomers (45–47%), carbon black (21.5–22%), metals (12–25%), textile (0–10%), zinc oxide (1–2%), sulfur (1%), and additives (5–7.5%) [17].

Landfilling requires huge capacities of space, as tires cannot be compressed to decrease space taken during disposal. To reduce the space occupied by waste tires in landfills, shredding waste tires prior to disposal is an option, but the high operating costs make this process impractical. Dispatching materials to landfills represent a lost chance in terms of a circular economy and deriving more value from waste. Some solutions can enable this substantial stream of rubber wastes to be converted into energy or new polymer materials [18]. Brown of Watson Brown HSM Ltd. (London, UK) [19] provided the next data for global estimates linked to rubber manufacturing life cycle [20]:

Total amount of rubber recycled at its end-of-life: typically, 3–15%;
Total of waste rubber re-used in some way (e.g., retreading, new products, etc.): 5–23%;
Total of waste rubber used for energy recovery: 25–60%;
Total of waste rubber send out to landfill or stored: 20–30%.

A report released in 2014 from the Recycling and Economic Development Initiative of South Africa mentioned that the tire waste in South Africa was 177,385 tons, and 32% was diverted from landfill [21]. Figure 4 shows the general life cycle of a tire.

Figure 4. Life cycle of a tire.

2.2. Recycling Advantage

The high consumption of rubber prompts us to identify a method of recycling to decrease this undesirable effect. The significant objective of recycling waste rubber is to protect the environment and to produce energy from waste rubber, such as the following: retrieving energy as fuel, cleaning leaking oil, dealing with the same product after some changes, reuse of products of thermal decomposition, regenerative rubber, and powdered rubber [22]. Moreover, waste polyethylene terephthalate bottles and waste rubber tires can be effective additives after certain treatments, which can increase the performance to asphalt and build firm pavements [23].

These benefits from recycling rubber differ regionally. For example, European countries' tire-recycling rates are almost 100%, whereas the ratio is less than 20% in South Africa. Accordingly, the European Union is four times more efficient than South Africa [17]. Table 1 illustrates the current usage of rubber waste.

Table 1. Review of the current usage of rubber waste.

Type of Product Waste	Type of Rubber (Natural/Synthetic)	Method of Reuse/Recycling	Reference
Glove (nitrile)	Synthetic	Recycling	[24]
Tire	Combination	Both	[25,26]
Tubes	Combination	Both	[27,28]
Mattresses	Natural	Recycling	[29]

2.3. Construction Field

Some studies have shown that waste or recyclable rubber can be used as building aggregates or fillers that reduce the thermal conductivity for cementitious composites up to 50% [30]. A study shows that adding 30% ripped rubber to concrete mixture yields the maximum results of compressive strength and modulus of elasticity test [31]. Moreover, the first research in South Korea about tire-derived fuel ash (TDFA) has shown that it may work as a binder material, and that adding TDFA to concrete can improve durability and mechanical performance owing to packing impact [32]. An article published in December 2020 shows that waste-rubber powder and thermoplastic polyurethane have waterproofing elements with an estimated percentage that can support subgrade and sub-base layers for protecting against damage from moisture, especially on high-speed railways [33].

2.4. Sustainable Treatment

Research has shown that using several devulcanization methods for waste tire rubber (WTR) can essentially and selectively split vulcanizate's crosslinks, whereas maintaining the polymeric chains can provide new sustainability pathway(s) for WTR. The generated compound has equivalent physical properties to that of the original source. This advanced release opens new possibilities for innovative economic opportunities worldwide [34]. Additional experiments have shown that waste ground tire rubber can exert an excellent reinforcing influence on SBR solution, owing to the existence of the core–shell structured carbon black after being degraded by a heterogeneous process to less than 150 °C by soybean oil [35]. Rubber wastes also originate from the footwear industry sector, wherein 10–20 parts per hundred (phr) ethylene–vinyl acetate elastomer waste is mixed with natural rubber, styrene-butadiene rubber or acrylonitrile-butadiene rubber, then the composites are vulcanized; no significant effects were observed on the crosslinking formation of mixture [36].

3. Various Techniques for Using Waste Rubber as Fuel and Its Additive

Pyrolysis oil from waste rubber is receiving great interest as alternative fuels. It is a complex mixture of hydrocarbons such as kerosene, gasoline and diesel. Meanwhile, fuel additives can generally be defined as any combination formulated to enhance the quality and effectiveness of fuel [37]. Examples of fuel additives are acetone, hydrogen, ether, nitromethane, and methanol. These chemicals could be synthesized from carbon dioxide, carbon monoxide, hydrogen, and methane, which could be generated from pyrolysis process.

3.1. Tire-Derived Fuel (TDF)

TDF is the most extensively used method to obtain energy from waste tires by mixing shredded rubber with coal or other fuels and then using it to power plant boilers or factories. Additionally, it is a crucial way to lower the levels of NO release by up to 75% [38]. Recently, Choi et al. [39] described an application of TDF fly ash as filler in a heated mixture of asphalt by evaluating three substance fillers, namely, stone dust, hydrated lime, and cement.

More than 7500 tons of waste tires were recovered with TDF in Canada and used as an auxiliary fuel in a cement plant in 2005 [40]. Figure 5 illustrates the process flow diagram for TDF.

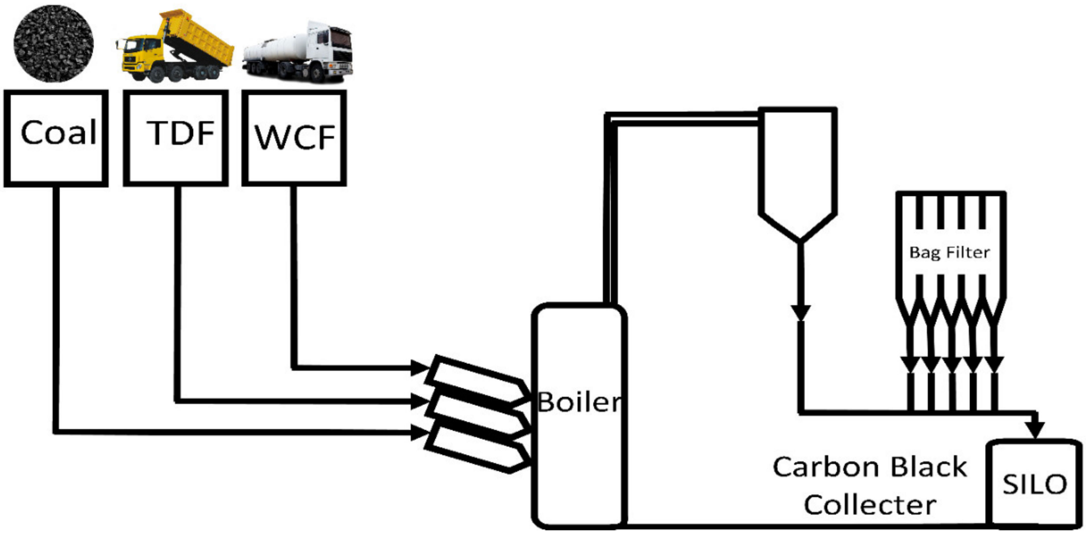

Figure 5. Tire-derived fuel PFD.

3.2. Pyrolysis

3.2.1. Definition

This section focuses on how rubber waste converted to secondary oil fuel has the same capability of the fuel itself through a process called pyrolysis. This technique for recycling adopts heat (>400 °C) for shredding tires in a reactor container under oxygen-free conditions [41]. Moreover, it is an alternative method dealing with vulcanized rubber to recover a useful product [42]. Substantial gases called pyrogas or syngas are released from pyrolysis (condensable and non-condensable compounds) and typically comprise paraffins and olefins [43]. Compounds such as methane and carbon dioxide are also released, but the major ones are a mixture of carbon monoxide and hydrogen (H_2 and CO). The presence of large quantities of hydrocarbons (C_2H_6), (C_2H_4), (C_3H_8), (C_3H_6), (C_4H_{10}), (C_4H_8), and (C_4H_6), nitrogen compounds (NH_3), and sulphur compounds (H_2S, SO_2, COS, and CS_2) at low concentrations has also been confirmed [44,45]. Figure 6 shows the process flow diagram for pyrolysis.

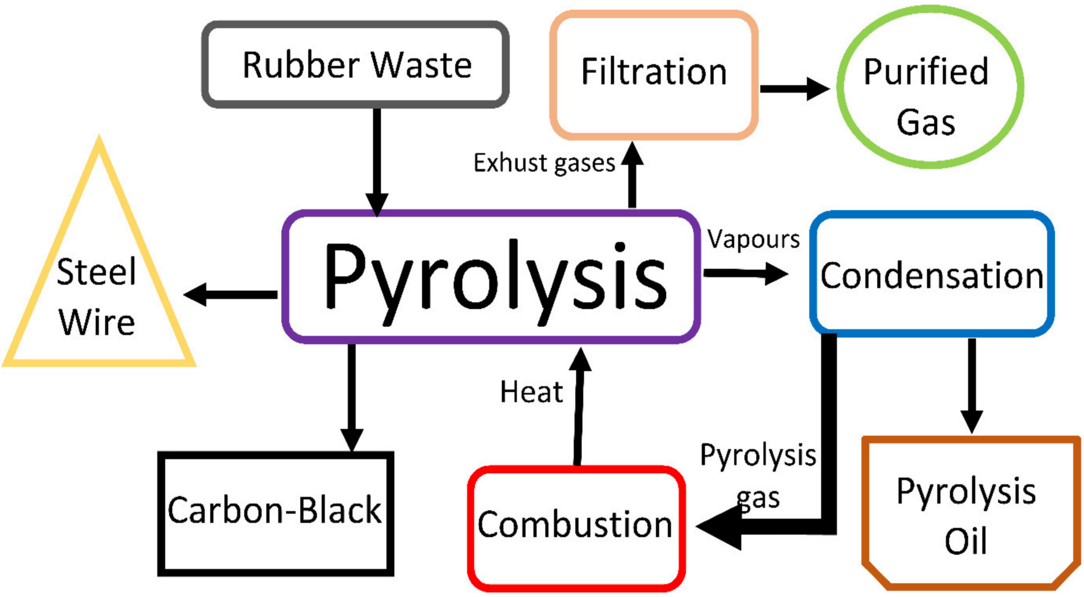

Figure 6. Pyrolysis PFD.

3.2.2. Pyrolysis Reactors

Scheirs mentions that reactor's shape and scale varies depending on usage and volume, from 3 m^3 designed for laboratory use to 20 m^3 used for manufacturing plants. Each kind has advantages and disadvantages in terms of technical, ecological and economic factors [46]. The most commonly used reactors are screw kiln, fixed bed (batch), rotary kiln, fluidized bed, and vacuum [47].

Additionally, Table 2 shows the ability of rubber waste to become a fuel or fuel enhancement.

Table 2. Various synthesis methods of fuel from rubber waste.

Type of Product Waste	Method of Production of Fuel	Fuel/Fuel Additives Product	Reference
Glove (nitrile)	co-pyrolysis	Fuel Additives	[48]
Tire	pyrolysis	Fuel	[49]
Tubes	pyrolysis	Fuel	[50]
Mattresses	pyrolysis	Fuel	[51]

3.2.3. Types of Pyrolysis

We can classify pyrolysis to two main types depending on the operational conditions, such as temperature, heating rate, and volatile residence time. Thus, general simple classifications of slow and fast are possible [52].

Slow Pyrolysis

Slow pyrolysis is also called carbonization. This kind of pyrolysis works at low temperatures and reflects a slow pyrolytic decomposition. Slow pyrolysis is characterized by low heating rates (a temperature range of 400–450 °C) and fairly long solid and vapor residence times (minutes to hours) which lead to higher yields of coke, tar, and char (the main compound) [53]. Fixed-bed reactors (FBRs) are usually used for slow pyrolysis [49].

Fast Pyrolysis

Fast pyrolysis occurs at 500–600 °C [54]. Higher heating rates and short residence times (milliseconds to seconds) represent this type of pyrolysis. Fluidized beds, fixed beds, and vacuum are typically used to operate fast pyrolysis, which requires small particle sizes [55].

4. Pyrolysis Parameters

Waste tire pyrolysis is a heat-absorbing process supported by temperature in a reactor. Thus, temperature has the strongest influence on pyrolysis products and conversion rates [52].

Other influential factors include particle size, feedstock composition, and atmospheric pressure. The presence of a catalyst also has a remarkable effect on the process [56].

4.1. Effect of Reactor Temperature and Pressure

The main variable involved in tire pyrolysis is temperature [57,58]. Several researchers such as de Marco Rodriguez et al. and Uçar et al. [59,60] stated that 500 °C is the optimum temperature at atmospheric pressure to achieve total tire conversion. Some studies have focused on the effect of pyrolysis–reactor temperature for the range of 200–600 °C, wherein char yield markedly decreases with increased temperature. The liquid-fuel yield increases with increased temperature from 280 °C to 350 °C. With further increased temperature, the liquid-fuel yield decreases. Conversely, gas yield increases with increased temperature from 280 °C to 400 °C [61]. Ilkiliç and Aydin [62] found that when the temperature reaches 400 °C, liquid yield increases with increased temperature and reaches its peak value at 500 °C before slightly decreasing to become sluggish. By contrast, Alvarez [63] noted a decline in pyrolysis liquid yield when the process is carried out at similar temperatures in the range of 425–575 °C.

Meanwhile, gaseous yield grows at a roughly linearly rate with increased temperature [64]. Processes at very high temperatures in fluidized and spouted bed reactors normally face a phenomenon of increased solid fraction with increased pyrolysis temperature, as higher heating rates and the vigorous gas–solid contact can boost additional secondary reactions involving the solid pyrolytic carbon black particles [65,66]. Conesa [67] reported that at higher temperatures, solid fractions increase from 20.4 wt.% at 600 °C to 32.5 wt.% at 900 °C by using bubbling FBR. However, in another publication, he used an FBR but did not find liquid-fraction production at 1000 °C, although a higher solid fraction comprising pyrolytic carbon black is observed [68].

4.2. Effect of Reactor Pressure

Another essential factor affecting pyrolysis is pressure. According to Buekens, increased pressure on feedstock pyrolysis that contain plastics leads more viscous liquid products with a higher coking tendency and more auxiliary and dehydrogenation reactions [53]. Ma et al. [69] examined the effects of pressure on limonene conversion during waste-tire pyrolysis. They found that yields of limonene substantially increase by about 20% during high-pressure pyrolysis with decreased residence time from 60 s to 15 s. They also revealed that the mean yield of limonene at 1.0 MPa is roughly 1.7 times larger than that of 0.1 MPa at the same residence time. Additionally, vacuum pyrolysis lowers the incidence of secondary reactions in the vapor period compared with pyrolysis at atmospheric pressure. The residence time of volatiles in vacuum pyrolysis imposes a limitation to secondary reactions [70]. Bikane et al. [71] reported Morupule coal behavior during pyrolysis in a high-pressure wire-mesh reactor. The effect of pressure is clear at 600 and 800 °C, i.e., total volatile yields decrease at higher pressures owing to the repolymerization of volatiles.

4.3. Rubber-Waste Particle Size

Previous studies have shown that the particle size of materials below 5 mm has no effect on reaction rates, as the temperature gradients within the particle are not considerable [72,73]. Furthermore, according to Beaumont and Schwob [74], heat transfer is difficult for larger particle sizes compared with small ones. As a result, the pyrolysis reaction is undertaken at a lower temperature. The size of tire particles and pyrolysis conversion are inversely related [75]. Dai et al. [76] used a CFBR at 500 °C and noticed a higher tire conversion rate and a drop in the pyrolytic CB yield from 45 wt.% to 30 wt.% when the tire particle size is reduced from 0.8 mm to 0.32 mm.

Barbooti et al. [77] obtained greater liquid yield for lower particle sizes when using an FBR at 0.35 m^3/h of N2 and heat at around 400–460 °C. Oyedun et al. [78] found a significant consequence of the particle size on pyrolysis time. At 500 °C and 5 °C/min, the pyrolysis time is around 1.5 h for particles with a radius of 10 mm and 2.5 h for 50 mm. Rofiqul Islam et al. [79] used an FBR at 475 °C and found reduced liquid yield for particles 2 cm^3 in size compared with that for 4 cm^3 particles. Similarly, the pyrolytic CB and the gas fraction are higher for 2 cm^3 than for 4 cm^3. We can assume that secondary reactions can also occur depending on the pyrolysis conditions.

Cylindrical tire particles with diameters between 7.5 and 22 mm have been devolatilized in a macro-TG reactor at temperatures between 490 and 840 °C in a spiritless environment by Larsen et al. [80]. The influence of particle size and surrounding temperature on the rate of devolatilization is noted to be significant. Thus, larger particle diameters and lower temperatures increase the pyrolysis period.

4.4. Feedstock Composition

The structures of tires vary depending on brand, type, and age [81,82]. Companies do not divulge the composition and the processing method of their tires, and they stay unrevealed [80]. Natural and synthetic rubber are found in different percentages in tires depending on the category (truck tire, passenger-car tire, and motorcycle tire). Today, tires are comprised of about 19% natural rubber and 24% synthetic rubber, which is a plastic polymer. The rest is made up of metal and other compound fillers [83]. The ratio of natural to synthetic rubber in TT is approximately 2:1 and nearly 4:3 in PCT [84].

Variations in the aromatic content of pyrolysis oil are reportedly associated with the original tire composition, especially the natural and synthetic rubber contents. For instance, Uçar et al. [60] observed that truck tires with a natural rubber content of 51 wt.% yields 15.4 vol.% aromatic content. Conversely, passenger-car tires with a lower natural rubber content of 35 wt.% produce a higher aromatic content of 41.5 vol.%. Additionally, Seidelt et al. [85], noticed that for natural rubber primarily made of xylene and isoprene dimmer, the SBR generates ethylbenzene, stirene, and cumene.

Therefore, passenger car tires with higher synthetic rubber content are likely to deliver oil with greater value for chemical production, owing to their higher aromatic content. Truck tires with superior natural rubber content tend to produce oil with larger potential as a vehicle fuel due to its lower sulfur content [86].

4.5. Pyrolysis Time

Corresponding to [57], the pyrolysis time is also referred to as the residence time of the tire inside the reactor, or simply reaction time; from previous literature reviews we can determine that pyrolysis time is related directly to particle size, meaning larger particles size require longer residence times while, inversely, lower heating rates require longer pyrolysis times. Numerous studies have shown a time–temperature link, i.e., higher temperatures require shorter tire residence times, and lower temperatures require longer residence times [87,88]. Recently, a derivative relationship has been obtained between two parameters (characteristic heating time and pyrolysis time) for olive stone particles with constant values of bed temperature [89].

4.6. Catalyst Role

Catalysts can be used to enhance pyrolysis rate, oil yield, oil quality, and yields of composites such as aromatics for chemical production to boost pyrolysis rate and oil quality. Some researchers have attempted to use (ITQ-21 and ITQ-24) as additives to industrial HMOR zeolite for the catalytic pyrolysis of waste tires; Dũng et al. [90] indicated that the catalyst affects the yield of kerosene, gasoline, and asphaltenes in pyrolytic oils. However, Kar [91] heated expanded perlite (volcanic-origin rock comprised mostly of silica and alumina) up to 850 °C to 1000 °C, and produced a porous catalyst with which metals were then combined to serve as catalysts.

Pyrolysis catalysts are very expensive, so some experimental studies have attempted to prepare low-cost catalysts similar to fly ash for the pyrolysis of HDPE and LDPE plastics. Tests were conducted at variable mass fractions (5%, 10%, and 15%) of catalyst. The maximum yield was 5%, and it decreases with further addition of catalyst. Conversely, any increase in mass portion of catalyst leads to reduced reaction temperature and retention periods [92]. Chen et al. [93] reported that applying high-alkalinity red mud as a catalyst for coal pyrolysis in a fixed bed and the CO_2 gasification of its resultant char improves the quality of tar and increases its weight percentage by 20.0% compared with coal pyrolysis alone.

Table 3 summarizes the effects of various parameters on fuel-additive production.

Table 3. Effects of pyrolysis processing parameters on fuel-additive production from rubber waste.

Processing Parameters Used	Made of (Natural/Synthetic Rubber Waste)	Reference
Temperature	Combination	[94]
Higher heating		[95]
Particle Size	Synthetic	[96]
Feedstock Composition	Combination	[53]
Residence Time	Synthetic	[97]
Atmospheric Pressure		[98]
Catalyst		[99]

5. Process Improvements for Pyrolysis to Be More Environmentally Acceptable

The use of liquid products from scrap rubber by combusting them in an internal combustion engine is one technique that may be less damaging to the natural environment [100]. It was also suggested that chemical recycling is undoubtedly energy consumption, especially during the pyrolysis process [101]. Besides a wide range of benefits to the community and economic sector, more research and development on the pyrolysis process needs to be explored, especially related to the environmental aspect. Qijing Wu et al. concluded that the pyrolysis-based high-value utilization of waste tires has more benefits compared to the conventional pyrolysis. Results showed reducing energy usage has a positive influence on reduction in major environmental effects; this resource saving is followed by environmental protection. Moreover, over 90% of the ecological impacts of ozone layer depletion and pollutants are decreased, and over 85% of the environmental loads of abiotic mineral and fossil fuel depletion are reduced [102].

Microwave pyrolysis is another possible technology to be explored. It has gained wide approval in the eyes of thermal chemistry scientists and presents more profits [103]. Electromagnetic rays of microwaves can be conveyed during the material directly. The processing has been claimed to provide many benefits, including environmental friendliness and process time reductions [104]. Chia Yang and Ani improved a controlled microwave heating technique by using activated carbon as a catalyst, due to its characteristic as a good microwave absorber, this study could contribute to carbon dioxide emission reduction and a move toward sustainable energy usage. Additionally, as the heating temperature rises quicker, and the heating duration is reduced, this will contribute in energy savings [105]. Recently, an experiment by Jin Hu indicated that applying ultraviolet (UV) will accelerate the aging of granules of plastics prior pyrolysis process. This will contribute to total energy saving [106]. According to Mohajerani et al. the life cycle assessment (LCA) analysis for recycling discarded rubber tires in building materials yielded certain advantages in terms of human health and environmental effect [107]. According to a review paper by

Sienkiewicz et al., rubber waste can be an environmentally friendly component after being converted as filler in polymer–rubber composites [108].

6. Environmental Assessment Outcomes

Table 4 shows the power consumption and gas emissions for different possible methods for rubber recycling. It was observed that pyrolysis could contribute higher amounts hazardous gases to environment. However, possible technology such as microwave pyrolysis would significantly reduce the power consumption. Additionally, the pyrolysis process would become a more environmentally friendly process.

Table 4. The amount of energy and gas emissions generated by various techniques of rubber recycling.

Recycling Type	Power KW	CO_2 Emission	Hazardous Gas Release (H_2S, CO, etc.)	Reference
Pyrolysis	15	2.9%	3.5%	[109,110]
Microwave Pyrolysis	3 (2.45 GHz)	<1%	<1%	[111]
Thermo-mechanical recycling (devulcanization)	9	-	-	[112]

7. Conclusions and Future Aspects of Rubber Waste as a Fuel Additive

Waste tires are perfect candidates for pyrolysis to recover energy owing to their origin (petroleum source). Moreover, temperature is the dominant factor influencing the physical and chemical properties of pyrolysis products, gas, liquid, and solid. Other prominent factors include reactor pressure, particle size, heating rate, feedstock composition, pyrolysis time/tire residence time, and catalyst role.

Additionally, 500 °C appears to be the premium temperature at atmospheric pressure after achieving total tire conversion. Processes at very high temperatures (over 1000 °C) usually lead to increased solid fraction, therefore made a boost to additional secondary reactions involving the solid pyrolytic Carbon Black particle with no liquid fraction production.

Future research related to waste rubber is needed to establish a new development model for increasing oil yield as a function of major parameters, including temperature, heating rate, type of reactor, and rubber particle size. This effort would certainly contribute to reduce the ongoing struggle against the climate crisis.

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