



TIRE WASTE AS A POTENTIAL MATERIAL FOR CARBON ELECTRODE FABRICATION: A REVIEW

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Abstract

Nowadays, tire waste is a big issue since the rapid growth of vehicle population worldwide. The disposal of tire waste should be done properly due to the environmental impact caused by stockpile the tire waste in landfill. Many kinds of research had been conducted to dispose of tire waste efficiently for getting valuable material from tire waste by using pyrolysis technology; one of them is carbon. In this review paper, production and application of carbon from tire waste presented. In several studies, carbon derived from tire waste was used as an adsorbent, electrode in energy storage application, like batteries and fuel cell.

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INTRODUCTION

The development of the automotive industry in Indonesia lead to the massive number of vehicles. Until 2017, there were more than 138 million vehicles in Indonesia, with 113 million was motorcycle. This huge volume of vehicle still increasing every years. In 2018, the association of Indonesia automotive industries (Gaikindo) recorded 1.1 million new cars were sold in Indonesia, and 6 million new motorcycles went to the road. It resulted high demand for tire as a vital component of vehicle.

Association of Tire Manufacturers Indonesia (APBI) stated that 69 million of motorcycles tires and 84 million of car tires including commercial truck tires were sold in 2018. It caused serious disposal problem once that tires are used up. Same phenomena also happen worldwide. Approximately, 2.7 billion tires were produced globally in 2017 and only 1 billion were disposed [1]. US every year spend 350 million passenger-car tires which are 85% go into landfill or stockpiled which need huge space and economic cost [2]. The disposal of tire waste become hot issue because landfilling of the wastes tire is no longer permitted in some country

because of its degradation resistance and bring impact to the environment. Legal regulation in South Africa and the European Union prohibit the disposal of tire waste in landfill sites [3] as listed in Table 1.

Table 1. Legislation of tire waste management [4][5]

Country	Legislation
South Africa	National Environmental Management Waste Act of 2008, Act No. 59 of 2008 Regulation 6(3) of the Tire waste Regulations requires all tires manufacturers registered under REDISA to comply with the approved REDISA Integrated Tire waste Management Plan (IWTMP)
European Union	Directive on the Landfill of Waste 1999/31/EC (1999) prohibited of disposal tire in the landfill since 2003 End of Life Vehicle Directive 2000/53/EC (2000) - recovered process in the end of vehicles life, all tires must be removed before vehicle being scrapped. Directive on Incineration of Waste-2000/76/EC -the directive prohibits combustion of end of life tires in older cement kilns
China	The Ministry of Industry and Information of China in July 2012 published an ordinance that the recovery rate that should be achieved at least 10,000 tons per years for recycling companies.

The disposal of waste tire should be done properly. The most common way to handle waste tire is piled up in landfill which is facilitate breeding space for mosquitos and spread of mosquito-borne disease later on. These piled up tire also represent fire danger and causing health and environmental problem.

MATERIAL AND METHOD

Tire and Its Disposal

Generally, Tires consist of four main components namely; tread which is directly contacted the road, plies which is a portion of the tire on that treads are vulcanized, sidewall that absorbs shock from the terrain and lastly bead heels which is fit to the rim [6]. Figure 1 shows these items.



Figure 1. Transversal cut of tire [7]

Tires mainly made from rubber and black carbon, carbon are the most main component. Other additives also added during the manufacturing process as accelerators and filler. Table 2 shows typical average of tire composition. Off course, those compositions maybe different for each tire, since the tire manufacturer keeps the composition as a secret recipe.

Tire Reconstruction

This involves re-treading tire for prolonging tire life. But not of all tire can be re-treading. It is only possible for specific tires, where the casing still good.

Table 2. Average composition of a new passenger and truck tire [8]

Contains	Passenger tire (%)	Truck tire (%)
Rubber	47.0	45.0
Carbon black	21.5	22.0
Metals (e.g. steel)	16.5	21.5
Textile (e.g. nylon)	5.5	-
Zinc oxide	1.0	2.0
Sulphur	1.0	1.0
Additives (e.g. clay, silica)	7.5	5.0

The well-managed tire can be re-treading at least two times [9]. This is the best way of recovery [6].

Recycling

This involves shredding or grinding of the tires for producing rubber chips. Many types of research show that rubber chips have been used for concrete [10][11], another common product produced from rubber chips such as noise barriers, floatation devices, mats, roofing materials, sports track, and so on [12].

Recovery of Energy

Many researchers conduct research in energy recovery from waste tire by thermal valorization using three technologies: pyrolysis, combustion, and gasification [13, 14, 15]. Figure 2 illustrates theses research classification.

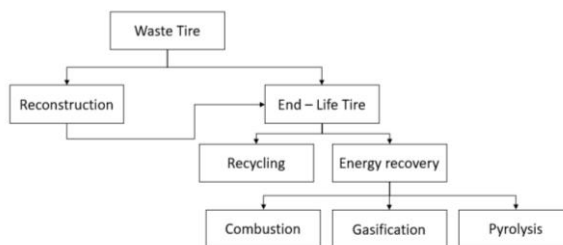


Figure 2. Tire Disposal Scheme

Combustion process used directly tire waste as fuels in incinerators due to high calorific value from tire waste. The advantage of combustion use tire waste is the maximum heat recovery that can be achieved, less production cost and environmentally acceptable process. It is make tire waste promising material for energy recovery. But, in other side, there is no material recovery from this process, high operating cost and emission of CO2 should also be considered [16]. Tire waste can be used as fuels in cement kilns. Although in European Union some of older cement kiln directive combustion of tire waste are prohibited, another attention is needed for ensure the environmental impacts of this process [17].

Gasification of tire waste is a process in which steam or air reacted with tires in an endothermic reaction and produce syngas (CO and H2) and other by-products such as CO2, char and hydrocarbon [18][19].

Pyrolysis is a thermal decomposition of tire waste in non-oxygen condition. Tire typically decomposed at 400°C temperature. Product derived from pyrolysis process are tire pyrolysis oil (TPO) 40-60 wt. %, pyro-gas 5-20 wt. % and pyro-char 30-40 wt. %. TPO can be used for diesel fuel. It also can be used in the synthesis of carbon nanotubes. Pyro-gas usually used for fuel in the

pyrolysis process. The gaseous fraction is composed of non-condensable gases like H_2 , CO , CO_2 , C_2H_4 , and C_3H_6 . Pyro-char can be used as a source of activated carbon since it contains very high carbon. Activated carbon can be used as an adsorbent for water treatment as good as for adsorb harmful gas. Recently many researchers studied carbon derived tire waste for energy storage applications including Na, K, and Li-ion batteries, supercapacitor, and also as a catalyst for biodiesel production [17][20]. Figure 3 shows the schematic diagram for the waste tire pyrolysis product and its application.



Figure 3. Schematic Diagram of Tire Waste Pyrolysis Product and Its Application [17]

Early Works in Tire Pyrolysis Process

Pyrolysis has been used to for a long time period to produce charcoal from biomass in the 1700s to early 1900s, pyrolysis used for production of fuel gas and smokeless solid fuel commercially. Generally, pyrolysis involves the heating of the raw material to temperatures higher than 400 C in non-oxygen environment, in an inert or a self-generated atmosphere, which lead to the volatilization and the decomposition of various structures that make up the raw material. When the raw material is heated under those conditions, many reactions take place including dehydrogenation, dehydration, aromatization cracking, isomerization, and condensation [21][22].

According to Dodds et al. [23] which quotes in Martinez et al. [21] a large number of tire waste pyrolysis projects has carried out since 1960 in several countries like United States, United Kingdom, Germany, and Japan. This pyrolysis process carried out in laboratory, pilot-plant and

semi commercial scale reactor. Deeper comprehensive description of the pyrolysis process and characteristics that implemented in the country that mention above can be seen elsewhere [23,24,25,26,27,28]. From an industrial point of view, several authors [26,27,29] pointed out that pyrolysis of tire waste was first attempted in the early 1970s by the US Bureau of Mines collaborating with the Firestone Company in the United State. 10 tires per day at laboratory unit and produced 3.8 l of liquid, 1.4 kg of gas, 3.2 kg of char and 1 kg of steel and char for every tire pyrolysis processed [26].

Types of Pyrolysis

Pyrolysis classified in many types depend on the operating conditions, such as temperature, heating rate, and volatiles residence time. As simple, pyrolysis can be classified as slow and fast pyrolysis. Fast pyrolysis sometimes also called as flash pyrolysis, although this term can also mean higher heating rates and short vapor residence times than the fast pyrolysis). Another classification of pyrolysis based on the environment used are steam-pyrolysis, oxidative pyrolysis, catalytic-pyrolysis, hydro-pyrolysis and vacuum pyrolysis. Depending on the heater system pyrolysis can be classified as the microwave or plasma pyrolysis.

Conventionally, fluidized, and entrained beds reactors are associated with fast pyrolysis whilst Fixed Bed Reactors (FBR) with slow pyrolysis (a batch or semi-batch process). However, it is possible to do fast pyrolysis using Fixed bed reactor by adjusting the heating rate and volatile residence time for research purposes. Other types of reactors that may be categorized for carry out fast pyrolysis are auger reactor and the rotating cone (usually used for liquid production since the heating rate is high and the vapor residence time relatively short).

Several different reactors, such as fixed bed reactor (batch or semi batch), screw kiln reactor, rotary kiln reactor, vacuum and fluidized-bed reactor have been used for pyrolysis of tire waste. Aydın et al. [30] investigated the pyrolysis in a 1.15 l capacity fixed bed reactor under nitrogen condition with temperature range from 400 to 700°C with steel and fabric removed. The result was the oil yield increased approximately 10 % with the increasing of 100°C temperatures from 31 wt.% at 400°C becoming 40 wt.% at 500°C . Table 3 shows the results.

Table 3. Product Derived of Waste Tire

Reactor	Temp. (°C)	Product yield (Approx. in %)			Additional information	Reference
		Oil	gas	Char		
Fixed, batch	700	38.5	17.8	43.7	Heating rate 15 C/min; 175 g tire	[31]
Fixed bed, batch	500	40.26	11.86	47.88	400 – 700 C temperature	[30]
Fixed bed, batch	425	60	10	30	Heating rate 10 C/min; 10 g tire	[32]
Rotary reactor	450	51.61	10.95	33.05	Heating rate 20 K/min. particle size 10 – 15 mm.	[34]
Rotary reactor	500	51.78	7.92	33.66	Heating rate 20 K/min. particle size 10 – 15 mm.	[34]
Rotary reactor	550	51.83	12.11	33.50	Heating rate 20 K/min. particle size 10 – 15 mm.	[34]
Rotary reactor	600	51.14	7.31	32.92	Heating rate 20 K/min. particle size 10 – 15 mm.	[34]
Auger	550	42.6	16.9	40.5	more than 500 kg of shredded tire waste were pyrolyzed in 100 h of operation	[21]
Closed batch	450	63	7	30	350–450 C Temperature; 30 K/min heating rate; 20 g tire	[21]
Fixed bed , batch	400	38.8	27.2	34	350–600 C Temperature; heating rate 5 and 35 C/min	[33]
Vacuum	500	56.5	33.4	10.1	Pilot scale semi continuous	[35]
Conical spotted bed	575	58	8	34	425 – 575 C temperature; Nitrogen flow rate 8 L/min	[44]
Fixed bed, Semi batch	750	63	11	26	Bicycle /motorcycle tire; 200 g; 20 C/min heating rate	[45]
Fixed bed, Semi batch	750	46	41	13	Jeep / car tire; 200 g; 20 C/min heating rate	[45]
Fixed bed, Semi batch	750	47	39	14	Truck / tractor tire; 200 g; 20 C/min heating rate	[45]
Fluidized bed	750	31.9	38	28.5	750–780 C Temperature; throughput 30 kg/h; tire piece	[42]
Ablative	495	54.2	11.5	34.4	Throughput 50 kg/h, scrap tire size 1.3 mm	[37]
Ablative	570	49.8	15.4	34.8	Throughput 50 kg/h, scrap tire size 3.4 mm	[37]

There was a consequent increase in gas yield. They studied the influence of nitrogen flow rate on product yield and found only negligible differences in yield. Islam et al. [31] investigated pyrolysis also in fixed bed reactor and pyrolyzed 750 g batches of tire waste they found a high oil yield of 55 wt.%, 36 wt.% of char and little gas yield approximately 9 wt.% at 475°C pyrolysis temperature. Kar [32] conducted pyrolysis process of 10 g tire waste using fixed bed reactor with 10°C/min heating rate, in nitrogen purged environment. They investigated the influence of pyrolysis temperature from 375 to 500°C. The result was the maximum oil yield of 60.0 wt.% oil obtained at 425°C. The oil yield decreased to 54.12 wt.%. The gas yield increased from 2.99 to 20.22 wt.% and char yield decreased from 50.67 to 26.41 wt.% as the temperature of pyrolysis was increased from 375 to 500°C.

However, for similar pyrolysis conditions, Banar et al. [33] reported pyrolysis of tire with steel removed produced only 38.8 wt.%, the maximum oil yield, with 34 wt.% char and 27.2 wt.% gas yield at 400°C pyrolysis temperature and heating rate of 5°C/min.

Antoniou et al. [34] conducted experimental quartz rotary kiln set-up for pyrolysis investigations of shredded waste tire with various of size 5-10 mm, 10-15 mm and 15-20 mm with steel removed. The samples was mixed car and truck tires and were supplied by a tire recycling company in Cork, Ireland, which has an output of 2500 t/year. The results show that temperature (450, 500, 550 and 600 °C), heating rates (60 and 20 K/min) and particle sizes influence the yields of oil, gas and char.

Vacuum pyrolysis technology was studied over a wide range of temperatures by Pakdel et al. [35] and Mirmiran et al. [36]. They used the PDU (pyrolysis development unit). The main component of this pilot plant is a semi continuous vertical cylindrical pyrolyzer with a diameter 0.7m and 2m height with six heating zones that stabilize the horizontal layers of granules at specific temperatures: $t = 226$ (highest layer), 295, 366, 404, 450 and 510°C. The maximum reactor temperature and total pressure were 510°C (at the bottom of the pyrolyzer) and 1 kPa, respectively. On average, product yielded from this process was 55 wt% oil, 25 wt% carbon black, 9 wt% steel, 5 wt% fiber residues, and 6 wt% gas. Black

Black [37] investigated pyrolysis of rubber from tires with particle size 1.3 mm and 3.4 mm in a continuous ablation reactor, which has a capacity of 5–20 kg/h. Feedstock loaded into the hopper manually. To prevent leaked gas, it was sealed after being. Material transported into the reactor by a discharge screw with variable speed. The reactor system was externally heated in six independently controlled zones of rotating vertical discs to produce continuous sliding contact between the metal surfaces and heated up to 550 °C and the particles of feedstock between them. After separation of the solid phase in the cyclone, the vapors of oil and gases from the reactor were condensed in a three-stage fractionation unit which utilizes two liquid quench columns, followed by a conventional water-cooled heat exchanger. The remaining non-condensable gases were directed to the filter to remove oil droplets and after measuring were analyzed. The yields of ablative pyrolysis products (liquids 49.6% wt., gases 16.9% w.t and carbon black 33.5% wt.)

Kaminsky and his co-workers from Hamburg were pioneers of polymer and rubber pyrolysis testing in fluidized bed reactors, they were the first investigator of the thermal decomposing method of polymers. Now, this approach known as the Hamburg process [38,39,40,41,42,43]. The study started from experiments based on a scale-up. It performed in three different laboratory plants with throughput 70 g/h, 300 g/h and 500 g/h of tire powder, then in a technical pilot plant with a 30 kg/h throughput of tire pieces, and ending at a semi-industrial pilot plant for whole tires with a 200 kg/h throughput [40,41,42,43]. Kaminsky et al. [43] reported a flow diagram of a fluidized bed reactor for the pyrolysis of two types of spent rubber materials. One type consists of truck tires with particle size 0.8–1.6 mm, and the other one was discarded rubber gloves. This laboratory scale equipment with a throughput of up to 3 kg/h with 154mm diameter cylindrical reactor, the fluidized bed medium consisted of 9 kg quartz sand, set in motion by preheated nitrogen or steam.

Product Derived from Waste Tire-Carbon

The production of tire carbons usually consists of physical activation using steam or carbon dioxide as oxidizing agents. The overall process typically consists of two steps: thermal pyrolysis to break down the cross-linkage between carbon atoms and usually conducted in relatively low temperature typically 400–700 °C temperature with the presence of nitrogen or helium, and the second step is activation with activating gas at higher temperature 800–1000 °C for develop of

the tire carbon porosity. Carbon characteristics are greatly influenced by activation temperature, the choice of the activating agent (carbon dioxide or steam) and process temperature [46].

Application of Carbon Derived from Tire Waste

Carbon derived from tire waste widely used as an adsorbent. It has been used to adsorb phenols, basic dyes and metals [47], phenols and p-chlorophenols [48], butane [49] and natural gas [50]. Shah et al. [51] compared the methylene blue adsorption using commercial activated carbon and acid-treated tire activated carbon. Shah et al. reported that acid-treated tire activated carbon has adsorption capacity higher than the commercial activated carbon. A comprehensive description about carbon derived from tire waste as an adsorbent can be found elsewhere [52].

There are not many reports which started the application of tire derived carbon as electrodes in energy storage application like supercapacitors and batteries, or fuel cells. Recently pyro char has been used to obtained electrode materials for Li, K, Na-ion battery, supercapacitor and electrocatalyst (ORR). Tire-derived activated carbon has several advantage such as a high surface area with porosity, high electrical conductivity and delocalized electrons. Paranthaman et al. reported tire-derived activated carbon for oxygen reduction reaction [53].

Rambau et al. [56] used activated carbon derived from tire waste for hydrogen storage application with best hydrogen storage capacity of 1.4 wt. % at 1 bar. Zhi et al. [60] reported pyro char treated chemical activation using H_3PO_4 and Zhao et al. [61] reported a steam-based activation procedure to modify tire pyrolytic char for its application as electrodes in double layer supercapacitors. Boota et al. [62] demonstrated a carbon-polymer composite electrode derived from tire waste for pseudo capacitor application. Naskar et al. [63] derived carbon from the pyrolysis of sulphonated tire rubber and demonstrated its application as Li-ion battery anode obtaining a reversible capacity of 390 mAh g^{-1} after 100 cycles. Sharma et al. [25] used carbon derived from tire waste which activated with KOH. The treated carbon is used for Li-ion battery application and shows a high specific capacity of ~880 mAh/g (at 50 mA/g) with nearly 80% capacity retention after 100 cycles. Li et al. [64] investigated the used of carbon from tire waste as an anode in sodium batteries. Carbon achieved from pyrolyzing acid-treated tire at temperature of 1100 °C, 1400 °C and 1600 °C show capacities getting higher as high as temperature which are 179, 185 and 203 mAh/g,

respectively, after 100 cycles at a current density of 20 mA/g in sodium-ion batteries with good electrochemical stability. This work opens a new path for tire waste recycling and its applicability in electrochemical applications including energy storage devices, as listed in Table 4.

Chen et al. [65] investigated the used of carbon derived tire waste as anode in microbial fuel cell. The electrode obtained from waste tires

carbonized at 800 °C gave a current density of 23.1±1.4 Am⁻², which is much higher than that achieved with traditional graphite felt anodes (5.5±0.1 Am⁻²). Passaponti et al. [54] used carbon derived from waste tire as highly efficient catalyst for alkaline fuel cell by using microwave assisted pyrolysis.

Table 4. Surface characteristics of tire carbon under different condition

References	Charring Conditions (carbonization) (°C, h)	Activation Temperature (°C)	Activation time (h)	BET surface area (m ² /g)	Additional information
[54]	-	150	1	50	Sample use a commercial tire (Michelin model
[54]	-	300	1	55	Agilis 81–195/65 R16C) and chopped into chip (8
[54]	-	450	1	296	cm ³), pyrolysis is done by microwave oven
[54]	-	600	1	83	(2.4kW, 10 min)
[55]	500, 0.5	From 20 to 900	-	44.72	12g of side wall part of tire used as sample. N2 flow rate = 100mL/min.
[55]	500, 0.5	From 20 to 900	-	121.47	12g of tread part of tire used as sample. N2 flow rate = 100mL/min.
[45]	750, 1	-	-	56.87	Bicycle / motorcycle (LVT) tire used as sample
[45]	750, 1	-	-	28.76	Jeep / car (MVT) tire used as sample
[45]	750, 1	-	-	41.76	Truck / tractor (HVT) tire used as sample
[45]	750, 1	-	-	109.01	Mixed of LVT, MVT and HVT (1:1:1) used as sample.
[52]	500, 5	900	5	220.48	Demineralization using 4M NaOH at 30oC for 3h.
[52]	500, 5	900	5	369.27	Demineralization using 4M NaOH at 60oC for 3h.
[52]	500, 5	900	5	268.11	Demineralization using 4M NaOH at 90oC for 3h.
[52]	500, 5	900	5	429.26	Demineralization using 4M HNO3 at 30oC for 3h.
[52]	500, 5	900	5	454.80	Demineralization using 4M HNO3 at 60oC for 3h.
[52]	500, 5	900	5	473.35	Demineralization using 4M HNO3 at 90oC for 3h.
[56]	-	800	1	775.41	Solid char mixed with KOH with ratio 1:4 (char:KOH) and compacted , resulting a pellet.
[57]	550, 100	700	-	76.5	Carbon black derived from continuous auger Reactor (more than 500 kg waste tire pyrolyzed). 2M HCl used as demineralized agent.
[57]	550, 100	700	-	76.3	Carbon black derived from continuous auger Reactor (more than 500 kg waste tire pyrolyzed). 4M HCl used as demineralized agent.
[57]	550, 100	700	-	75.4	Carbon black derived from continuous auger Reactor (more than 500 kg waste tire pyrolyzed). 6M HCl used as demineralized agent.
[57]	550, 100	700	-	79.1	Carbon black derived from continuous auger Reactor (more than 500 kg waste tire pyrolyzed). 8M HCl used as demineralized agent.
[44]	425, 0.5	-	-	46.5	The pyrolysis runs were carried out in continuous regime by feeding 1.3 g/min of tire rubber.
[44]	475, 0.5	-	-	63.3	The pyrolysis runs were carried out in continuous regime by feeding 1.3 g/min of tire rubber.
[44]	575, 0.5	-	-	80.5	The pyrolysis runs were carried out in continuous regime by feeding 1.3 g/min of tire rubber.
[58]	-	850	-	496	The activated carbons (ACs) were milled and sieved to <0.100 mm for the elemental and proximate analyses
[59]	1000, 4	-	-	30	After heating, a black carbon was recovered and grinded using an agate mortar, washed with water, filtered and dried under vacuum at room temperature. No further processing of the material was performed.

Another common application for carbon is as a sensor. Zhai et al. [66] applied black carbon for piezo resistive sensor. Li et al. [67] in his paper describe the current development of carbon used for nitrite sensor. Some researcher [68,69,70] modified the carbon for electrochemical sensor for acetaminophen detection. But author cannot find any literature that carbon derived from tire waste has been used for sensor application. It can be an interesting for every researcher to conducting a research based on this gap.

CONCLUSION

Tire waste always provided everywhere as long as the vehicle still exist in this world. Tire waste can be processed as a valuable material such as activated carbon. Active carbon derived from tire waste has a wide application exclude it was vulcanized to be a new tire. It can be used for adsorbent, energy storage application like battery and fuel cell. But author cannot find any literature yet that carbon activated derived from tire waste was used as a sensor. It will be an interesting for future research.

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