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RECOVERY OF OIL FROM WASTE TYRES USING PYROLYSIS METHOD: A REVIEW

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ABSTRACT

Owing to the difficulty in disposal of tyres and environmental pollution due to accumulation of waste tyres, pyrolysis is carried out as an alternative, efficient and environmental valorisation process, rather than combustion-inceneration which primarily is characterized as a destructive one. Pyrolysis can be considered as a non-conventional method for tyre recycling, which seems to be very appropriate for complex materials, such as tyres. This technology could not only reasonably and effectively dispose waste tyre and tube without pollution, but also is effective in producing fuel that can reduce energy crisis. Thermogravimetry analysis reveals that the pyrolysis of tire rubber at atmospheric pressure starts at a temperature around 250^{o}C and finishes at a temperature of about 550^{o}C. In general, by pyrolysing waste tire three fractions are obtained: solid residue (around 40 wt.%), liquid fraction (around 50 wt.%) and gas fraction (around 10 wt.%). The general trend is an increase in yields of liquid and gas fractions as the temperature increases. From the works devoted to tire pyrolysis, which are investigated on the generation of liquid fuel results that derived liquids are a complex mixture of organic compounds containing a lot of aromatics.

KEYWORDS: Pyrolysis, Proximate Analysis, Elemental Analysis, Thermogravimetry Analysis, Hydrogenative Pyrolysis

INTRODUCTION

The disposal of waste tyre has become a major environmental concern globally and this can be attributed to the increase in automobile usage as well as population especially in areas of large population and highly industrialized nations. (Mazloom G. et al.,2009, Gao N. et al.,2009, Cheung et al.,2011, Senneca et al.,1999, Leung DYC., 1998). The problems caused by the waste tyres is majorly because they are not biodegradable and can last for several decades if no proper handling is carried out. A number of studies related to tyre pyrolysis had been reported in the literature for its conversion into valuable compounds. Developed countries have been paying great attention to the effective utilization of discarded tyres to achieve the goals of protecting environment, recycling resources and preserving energy. For many reasons, recycling of waste rubber has received much attention in recent years all over the world, due to the present rate of economic growth in utilization of the fossil energy fuels like, crude oil, natural gas or coal without saving for the future.

Waste tyres have a high content of volatile matters as well as fixed carbon that makes them an interesting solid as a fuel for energy production or hydrogenation processes and in pyrolysis processes to obtain different fractions of soild, liquid and gaseous products. Also tyre rubber has a lower ash content (≈3% vs. ≈7% for coal) and a lower carbon content (≈25% vs. ≈45% for coal) when compared to other activated carbon precursors for example coal and wood. The energy content or fixed carbon content of waste tyres can be exploited by thermochemical processes via pyrolysis into a more valuable fuel and useful chemicals.

Pyrolysis process was studied thoroughly by many authors. Pyrolysis is a process of decomposing the organic substances to lower molecular weight products, liquids or tars, chars and few amounts of gases(volatiles), by subjecting a material to heat in a reduced or no oxygen environment, which can be useful as fuels or chemical sources. Plastics pyrolysis usually proceeds at low (<400 ^{o}C), medium (400–600 ^{o}C) or high temperature (>600 ^{o}C). Despite the fact that pyrolysis is considered a major alternative to exploit the useful chemicals and resources from waste tyre, the process is still not in high usage and this is largely due to the high amount of energy required for the process, various attempts have been made by researchers to make tyre pyrolysis an economic viable process.(Mazloom G. et al.,2009, Cheung et al.,2011, Senneca et al.,1999, Ferrero et al.,1989).

CHARACTERISTIC ANALYSIS OF TYRES

Tyres are produced by more than 100 different species. The composition of different tyre parts like the tyre sidewall or the tyre tread varies due to the different desired characteristics of product. Tyres are composed of rubber compounds and textile or steel cords. Rubber compounds generally consist of elastomers (natural or synthetic rubber), carbon black, hydrocarbon oils, zinc oxide, sulphur and sulphur compounds and other chemicals such as stabilizers, anti-oxidants, anti-ozonants, etc. The rubbers mostly consist of blends of two or three rubbers together with tyre additives. Because of these complex mixtures, the pyrolysis of tyres seems to be a complicated process involving a large number of chemical reactions and complex interactions of the single components. In order to determine feedstock characteristics, analyses suitable for solid materials characterization are used such as proximate and elemental analyses.

Proximate Analysis

Proximate analysis separates the products into four groups: (1) moisture, (2) volatile matter, consisting of gases and vapors driven off during pyrolysis, (3) fixed carbon, the nonvolatile fraction of coal, and (4) ash, the inorganic residue remaining after combustion.

Elemental Analysis

A complementary method to proximate is the ultimate analysis. Its scope is the determination of the carbon and hydrogen in material, as found in gaseous products of feedstock’s complete combustion, determining also the content of sulphur, nitrogen, and ash in the material as a whole, and oxygen content by difference.

Table 1 shows the proximate analysis of scrap tires declared by various authors. The elemental analysis of waste tires found in the literature is presented in Table 2.

Table 1: Proximate Analysis of Scrap Tyre Rubber

Author	Volatile (wt %)	Fixed Carbon (wt %)	Moisture (wt %)	Ash (wt %)	Steel (wt %)
M. Juma et al.	61.61	22.66	1.72	14.01	-
Rodrigues et al. *	58.8	27.7	-	3.9	9.6
Jong Min Lee et al.	67.3	28.5	0.5	3.7	-
Yu Min Chang et al.	62.32	26.26	1.31	10.29	-
Gonzales et al.	61.9	29.2	0.7	8.0	-
Chen et al.	93.73**	-	0.54	5.3	-
Loresgoiti et al. *	59.3	27.6	-	3.5	9.6
Orr et al.	68.7	23.3	0.4	7.6	-
Williams and Bottrill	66.5	30.3	0.8	2.4	-
Atal and Levendis.	58.7	33.6	-	7.7	-

**- Based on reinforced tyre with steel cords   **- Including fixed carbon*

Table 2: Elemental Analysis of Scrap Tyre Rubber

Author	C (wt %)	H (wt %)	N (wt %)	S (wt %)	O (wt %)	Ashes (Inorganic)
M. Juma et al.**	81.24	7.36	0.49	1.99	8.92	-
Rodrigues et al. *	74.2	5.8	0.3	1.5	4.7	13.5
Jong Min Lee et al.	83.8	7.6	0.4	1.4	3.1	3.7
Yu Min Chang et al.	74.4	6.96	0.21	1.6	5.02	10.21

Table 2: Contd.,

Gonzales et al.	86.7	8.1	0.4	1.4	1.3	2.9
Chen et al.	81.16	7.22	0.47	1.64	2.07	7.44
Berrueco et al.	88.5	6.6	0.4	1.6	3.0	-
Arion et al.	73.8	5.3	0.44	1.71	0.11	17.8
Loresgoiti et al.*	74.2	5.8	0.3	1.5	5.1	13.1
Orr et al.	81.3	7.3	0.3	1.5	-	1.4
Williams and Bottrill	85.8	8.0	0.4	1.0	2.3	2.4
Lanoir et al.	82.63	7.5	0.36	1.69	-	-
Senneca et al.	86.7	6.9	0.3	1.9	1.0	3.3
Roy et al.	86.6	8.1	0.5	0.8	2.2	-
Cunliffe and Williams	86.4	8.0	0.5	1.7	3.4	2.4

- Based on reinforced tyre with steel cords* *- Based on free of ash*

EXPERIMENTAL PROCEDURE

The waste tyres are collected and segregated. Then the tyres are cut into small pieces and steel wires, fabric fibers were removed. Thick outer edges of the tyres were divided into small slices. These tyre slices were washed, dried and then filled in the pyrolysis reactor. Conventional pyrolyis was performed under N_2 environment and ambient pressure. Most tyre pyrolysis processes operate within a temperature range of 250–500^oC, although some processes are reported to operate at upto 900^oC. At temperatures above approximately 250^oC shredded tyres release higher amounts of liquid oil products and gases, while above 400 ^oC , the yield of oil and solid tyre-derived char may decrease relatively to gas production. The procedure is simply depicted below in figure 1.

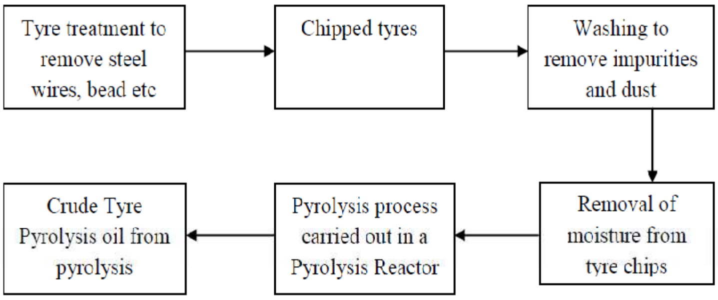

Figure 1: Pyrolysis Process of Waste Automobile Tyre

Pyrolysis can be classified as atmospheric, vacuum, catalytic, fast or slow according to the operation parameters applied. Alternatively hydrogenative pyrolysis was studied by Murena et al. used hydrogen as a medium for tyre pyrolysis. The pyrolysis process is generally carried out by many types of reactor. A number of studies and researches has been done to investigate the pyrolysis of waste tires in both laboratory and industrial scale. Subsequently, several laboratory, pilot pyrolyis reactor procedures and different conditions using end of life tyres (ELTs) as feedstock are presented below in a table 3. Many attempts have progressed, creating a wide range of industrial appliances aiming at fuel and material recovery. The manufacturers located around the globe give certain solutions to energy valorisation of ELTs. Some of those are mentioned below in table 4.

Table 3: Laboratory and Pilot Scale Pyrolysis Reactors by Various Authors

Author	Type of Pyrolysis Reactor	Temperature Conditions (^oC)
Williams et al., Berrueco et al., Cunliffe and Williams, Barbooti et al., Suuberg et al., Laresgoti et al., Jitkarnka et al., Arabiourrutia et al., Paradela et al., Diez et al., Zabaniotou et al., Dai et al.	Fixed – bed pyrolysis reactor (Nitrogen atmosphere)	300 to 720
Rodriguez et al., Murena et al., Laresgoti et al.	Autoclaves	
Kaminsky and Mennerich	Fluidized – bed reactor	500 to 700
Dai et al., Williams et al., Roy et al.	Vacuum pyrolysis reactor	
Tang and Huang et al.	Plasma pyrolysis	1500

Table 3: Contd.,

Diez et al.	Rotary Kiln pyrolysis	550
Helleur et al., Stanciulescu et al.	Ablative reactor pyrolysis	550
Aguado et al., Fabbri et al.	Flash pyrolysis	450 to 600
Qu et al., Boxiong et al., San Miguel et al., Olazar et al.	Catalytic pyrolysis	300 to 700
	Molten – salt pyrolysis	
	Microwave pyrolysis	
Diez et al., Lopez et al.	Pilot scale pyrolysis (Rotary kiln reactor type)	550

Table 4: Industrial Scale Pyrolysis Reactors in Various Companies

Reactor Type	Company	Temperature(^{o}C)
Indirectly fired kiln, grinding circuit, oil condensing and gas cleaning system.	Metso–Minerals Pyro Systems, USA	450
Pyrolysis liquefaction includes rotary kiln, rotary hearth unit, and the fluidized bed	Klean Industries, TPP, Kouei international.	430
Depolymerisation with catalysts	Jiangyin Xinda Machinery Co.,Ltd	normal temperature and pressure
Batch pyrolysis reactor	Jingcheng India	>400
Rotary pyrolysis reactor	Xinxiang Huayin Co.LTD	$350 < T < 450$
Batch pyrolysis reactor	Pyrocrat	$350 < T < 450$
Indirect Rotary Kiln	Pyreco	-
Modular pyrolysis Steam Cycle (MPSC) system in a rotary kiln using an indirect, external source of heat	Splainex	$400 < T < 600$
Batch Horizontal reactor	Hanocorp–Pyrogen	400

PROCESS CONDITIONS OF PYROLYSIS PRODUCTS

Pyrolysis of waste tires leads to the production of a solid carbon residue (char), a condensable fraction (pyro-oil) and gases. The percentage of each phase is influenced by process conditions, such as temperature, pressure, heating rate, particle sizes, heat exchange system, catalysis etc. Various authors have briefed about the operating parameters of pyrolysis in table 5.

Temperature

Gas yield increases with temperature as a result of more powerful thermal cracking in high temperatures and liquid's yield is almost stable on 500^{o}C and decreasing by raising temperatures. Higher temperatures raise lighter products' yield, as benzene and kerosene content raise with temperature. Tar yield, does not show an obvious trend in the studied temperature range and more specifically it has its maximum value at 600 ^{o}C, with similar yields in lower temperatures of 425^{o}C and 500^{o}C. Char yield obviously reduces with temperature. The removal of pyrolysis products from the hot zones, reduces the range of secondary reactions taking place that are known to raise char yield unlikely to oil yield. The specific surface of the produced char showed a significant raise by raising temperature and heating rate.

Heating Rate

For a given temperature, the heating rate (^{o}C/min) has a minor effect on products yield. In general, the faster the feed stock is heated to a given temperature, the less tyre-derived char and the more oil and gas are produced. The surface area of the solid product increases as heating rate or temperature increases. Higher heating rates in small residence times along with the immediate cooling, favour liquid yield as pyrolysis gas and vapours are condensed, before reaction cracks larger molecular weight molecules to gaseous products. At higher temperatures the main product is gas. Tyre pyrolysis at a low heating rate produces high amounts of char and gas. A high heating rate decreases liquid yield, while this does not happen on moderate heating rates.

Particle Size

The particle size of tyres was found to influence not to a great extent pyrolysis products. However, the larger the particle size is, the greater will be the amount of oils at high temperature range, while the yield of carbon black is almost constant under the same conditions.

Table 5: Influence of Some Process Conditions on Char, Liquid and Gas Yields Presented by Different Authors

Author	Temperature (^oC)	Heating Rate (^oC/min)	Pressure kPa	Sample Sizes	Solid (wt %)	Liquid (wt %)	Gas (wt %)
Williams et al.	300-720	5-50	101	-	35	55	10
Laresgoiti et al.	400-700	15	101	20-30 mm	43-53	28-40	7-9
Berrueco et al.	400-700	15	101	20mm	47-63	30-43	2.4-4.4
Gonzalez et al.	350-700	5-20	101	0.2-1.6 mm	37-40	55	4-11
Pakdel et al.	440-570	-	1.3-28	$3.8cm^3$	30.6-53.4	50-60	3.2-11.9
Barbooti et al.	400-460	-	101	2-20mm	32.5	51.0	16.6
Chang	200-600	-	101	20 mg	14-28	28-42	30-53
Cunliffe andWilliams	450-60	-	101	-	37-38	53-58	5-9
Roy et al.	25-500	15	0.8 - 2.8	-	35-36	62	1-3

THERMOGRAVIMETRY ANALYSIS

Both thermogravimetry (TG) and derivative thermogravimetry (DTG) are used as standard methods for studying the thermal degradation of waste rubber samples. From the thermogravimetry analysis provided by various authors (for example: Leung and Wang,1998, Yang et al,1995, Berrueco et al. 2005) results that more than one degradation temperature region during rubber pyrolysis is recorded.

Measurements provided in laboratory (M. Juma et al.,2006) sustain this fact (see Figure 4), however, it depends upon the composition of rubber compounds. The measured TG curves show two different mass loss regions over a temperature range of 250-550^{o}C. A typical behaviour of sample mass loss during pyrolysis measured by TG (M. Juma et al.,2006) is shown in figure 3.

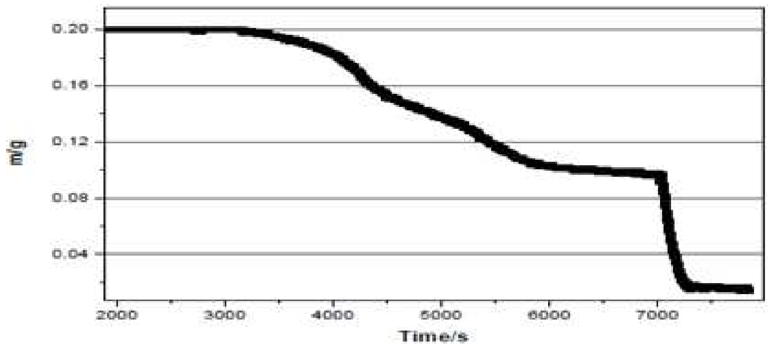

Figure 3: A Typical Behaviour of Mass Loss of a Scrap Tyre Sample during Pyrolysis

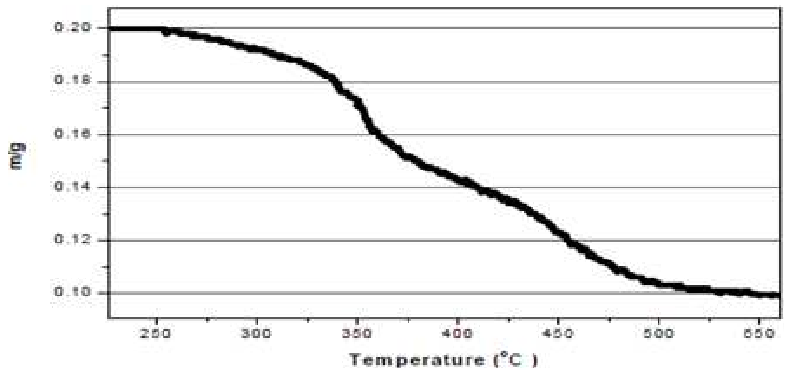

Figure 4 : Degradation Temperature Regions of Scrap Tyre Pyrolysis

The start and end temperatures of the pyrolysis process reported by various authors are compared in table 6.

Table 6: Start and End Temperatures of Pyrolysis of Scrap Tyres

Author	Start Temperature of Pyrolysis (^{o}C)	End Temperatures of Pyrolysis (^{o}C)	Heating Rate (^{o}C/min)
M. Juma et al.,2006	250	550	5
Berrueco et al.,1995	200	500	15
Leung and Wang,1998	200	550	10
Chen et al.	250	500	5
Senneca et al.	200	450	5
Chen and Qian.	200	550	10
Conesa et al.	250	500	5

DISCUSSIONS

From the published works results that scrap tire rubber consists of about 60 wt.% volatile organics, 30 wt.% fixed carbon and 10 wt. % ash. Elemental analysis shows that the tire rubber contains approximately 80 wt.% C, 7 wt. H, 0.4 wt.% N, 1.5 wt.% S, 3 wt.% O and 8 wt.% ash. From thermogravimetry analysis provided by various authors results that pyrolysis of tire rubber at atmospheric pressure starts at a temperature of about 250 ^{o}C and finishes at a temperature of around 550 ^{o}C. The products obtained by pyrolysis techniques are useful in many ways and can be a suitable alternative to fossil fuels and natural gas. A significant demand rises for the optimization of pyrolysis process conditions and various factors that influences the pyrolysis products yields and characteristics. The optimization of process parameters like temperature, heating rate and particle size is important for the product yield.

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