GENERATOR EXHAUSTS CONTROL IN NIGERIA USING ACTIVATED CARBON FROM DISCARDED RUBBER TYRES

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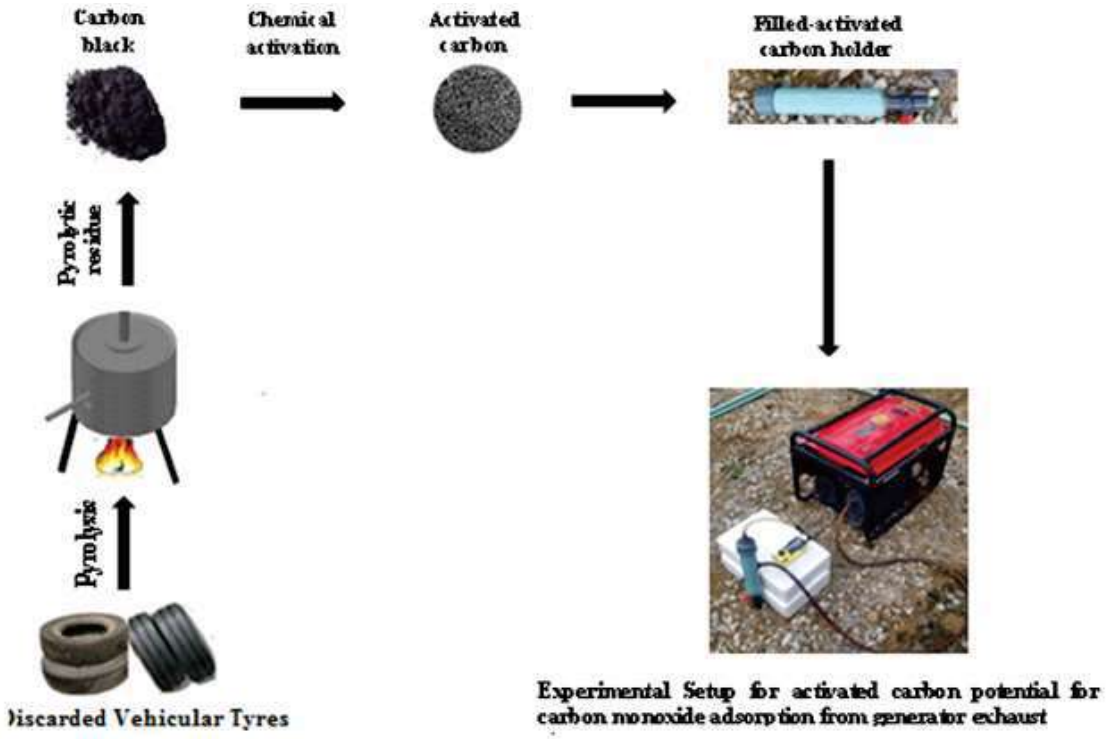

Experimental Setup for activated carbon potential for carbon monoxide adsorption from generator exhaust

Abstract

Many Nigerian homes,* offices, shops, and establishments *are powered by generators due to inaccessible electricity from the national grid. Consequently, many people suffer from life threatening cardiac complications due to their exposure to carbon monoxide poisoning from generator emissions. Besides, improper disposal of discarded rubber wastes which unfortunately has become a norm in Nigeria constitutes menace to human health. The purpose of this study was to develop effective system for pyrolytic conversion of discarded rubber tyres to activated carbons, which could be used as adsorbents for reducing generator emissions in Nigeria. Carbon black produced from the pyrolysis of the rubber tyres were chemically activated using potassium hydroxide, potassium carbonate and hydrochloric solution of mixture of copper (I) chloride and aluminium (III) chloride, with impregnation ratios ranging from 0.67 to 2.33. The range of values for surface areas, pore volumes and pore sizes of the resultant activated carbons included 285 – 846 m^2/g, 0.116 – 0.354 cm^3/g, and 2.92 -6.54 nm, respectively. The carbon monoxide reduction efficiencies of the activated carbons were very high ranging from 94.0 to 95.5% indicating the

feasibility and the effectiveness of this project in finding solutions to the twin problems of carbon monoxide poisoning from generator, as well as, improper disposal of discarded rubber tyres in Nigeria. Commercial application of this project will allow effective management of discarded automobile tyres and also make locally produced activated carbon to be affordable, thereby, reducing the dependency on imported activated carbon. Availability of locally produced activated carbon will make feasible the domestic application of the activated carbon as adsorbent for carbon monoxide from generator exhausts.

Keywords: Carbon monoxide, Emission poisoning, Rubber wastes, Pyrolysis, Efficiency, Activated carbon, Waste management, Generator exhausts, Carbon black, Emission reduction

1.0 Introduction

It is unfortunate that countless number of people in developing countries, like Nigeria, have either died or are suffering with life-threatening cardiac complications due to carbon monoxide (CO) poisonous fume inhalation from sources such as gasoline or diesel –powered engines (generators), wood cook stoves, car exhausts, and poorly ventilated rooms or chambers (Seleye-Fubara et al. 2011, Afolayan et al. 2014). In Nigeria, the major indoor source of carbon monoxide is petrol or diesel-fuelled generators since most homes, offices, shops, and establishments are run on generators because of the poor or inaccessible electricity supply from the national grid. Although other air pollutants such as nitrogen dioxides (NO_2), sulphur dioxide (SO_2) and particulates beside carbon monoxide are also emitted from the sources; carbon monoxide seems to be of major concern because of its deadly nature on human health. Meanwhile, improper disposal of non-biodegradable wastes, such as discarded vehicular tyres, is also a major challenge in Nigeria, since the improper waste disposal incurs environmental, health, economic, as well as, social costs. Vehicular rubber tyre is a material often made from petroleum rubber with weight composition of styrene-butadiene rubber (SBR) (62.1%), carbon black (31.0%), extender oil (1.9%), zinc oxide (1.9%), stearic acid (1.2%), sulphur (1.1%) (Macias-Garcia et al. 2003) and accelerator (0.7%) (Merchant and Petrich 1993, Zabaniotou and Stavropoulos 2003).

While most developed countries are working towards zero waste practice; which is referred to complete recycling and recovery of all resources from waste materials, most developing countries have not given much attention to the huge amount of waste generated daily in the countries. The wastes are usually dumped in unsanitary landfills, burnt in open fields or even thrown in water channels, thereby causing untold environmental pollution and adverse health effects. The global challenge for humanity is to design effective waste management system for climate change mitigation. Many researchers have explored the potentials of reprocessing waste tyres to valuable products such as artificial reefs, aggregates in asphaltic concrete, fuel for cement kilns, and activated carbon production (Teng et al. 1995; Topcu 1995, Eldin and Senouci 1999; Corinaldesi and Moriconi 2004, Corinaldesi et al. 2011, Trouzine et al. 2012). Besides, value added-products such as fuel gases; liquid oil and carbon black have also been recovered from waste materials through pyrolysis process (Zabaniotou and Stavropoulos 2003). It is a process that has since been employed as an environmentally friendly tool for treatment and adding value to waste. Pyrolysis is a process that involves heating a material to a temperature range of 250° - 500°C, in the absence of oxygen, so that the material can be volatilized and decomposed to oil, gas and char or carbon black (Martínez et al. 2013). Carbon black is a form of paracrystalline carbon with short- and medium-range ordering in their lattice but lacking crystal-like ordering (Voyles et al. 2001). It has a high surface-area-to-volume ratio, although lower than that of activated carbon.

Developing countries, like Nigeria, are faced with myriads of problems, among which are inefficient

waste disposal and frequent power outages. The unstable electricity supply has necessitated the use of diesel or gasoline powered generators as an alternative power supply in the countries. Oftentimes, the generators are located indoors or in enclosed spaces to prevent the generators from being stolen or damaged. The improper location of the generators exposes people to the damaging effects of the toxic exhaust emissions from the generators. The negative consequences of the inefficient disposal of discarded vehicular tyres and the toxic generator exhaust emissions in indoor environment cannot be overemphasized. One approach to finding solution to the challenges of improper disposal of discarded vehicular tyres, as well as, indoor carbon monoxide poisoning from generators, is to develop an efficient process for the production of activated carbon from locally available raw materials such as discarded vehicular tyres, for the purpose of using it for CO adsorption from generator exhausts. Activated carbon (AC) is a porous solid adsorbent that has been processed to develop its internal porosity, surface area, as well as surface reactivity, for the removal of pollutants such as insecticides, herbicides, chlorinated hydrocarbons, heavy metal ions, among others, from any medium. Adsorption process for pollutants removal is often favoured because the technology is simple, cost effective, and does not require high energy consumption. Activated carbon is often preferred to other adsorbents such as zeolites, porous clay ore, silica gel, activated alumina, molecular sieve adsorbents, among others, for removal of gaseous pollutants because of the high micro porous volume and adsorption kinetics (Bhave and Yeleswarapu 2020). Extensive research work had been carried out to investigate the effects of various activating chemical reagents on the quality of activated carbon produced during the activation process (Macias-Garcia et al. 2003). The various activating chemicals that had been used were zinc chloride ($ZnCl_2$), phosphoric acid (H_3PO_4), sulphuric acid (H_2SO_4), potassium hydroxide (KOH), sodium hydroxide (NaOH), and potassium carbonate (K_2CO_3) (Tsai et al. 1997, Hayashi et al. 2002, Fierro et al. 2006, Singh et al. 2008, Ashmore and Dimitroulopoulou 2009, Yorgun et al. 2009). Chemical activation process is often preferred to physical activation process because chemical activation process occurs at lower temperature and shorter time. The chemical activation process also produces activated carbon with better porous structure than the one produced during physical activation process (Yalçn and Sevinc 2000, Yorgun et al. 2009).

In Nigeria, there has not been so much done on the development of efficient process

for managing discarded automobile tyres in such a way as to convert the discarded tyres to value-added product like activated carbon with the aim of using it as adsorbent for reducing generator toxic emissions (Zabaniotou and Stavropoulos 2003, Teng et al. 2000, Caturla et al. 1991, Hari-Rudijanto et al. 2017, Thakre et al. 2017, Vishnu and Manikandan 2015, Tan et al. 2008, Setiawan and Al-Jauhari 2015, Li et al. 2000). The purpose of the study was, therefore, to minimize land, air and water pollution due to indiscriminate disposal of waste rubber tyres and generator exhaust emissions. The objective was achieved by developing a pyrolysis process for the conversion of discarded automobile tyres into valuable products including fuel gases, liquid oil and carbon black. The carbon black was converted to activated carbon which was used for CO adsorption from the generator exhausts. The coverage of the research work included the collection of discarded automobiles tyre wastes in communities around Lagos State University (Ojo Campus, Lagos, Nigeria), the crushing of the discarded tyres to granular particles, the setting up of pyrolysis equipment for converting the waste tyres to carbon black residue, the chemical activation of the carbon black to activated carbon, characterization of the carbon black and activated carbons produced, as well as, the testing of the activated carbon effectiveness in the removal of CO from generator exhausts. The Brunauer-Emmett-Teller (BET) analysis was used for the evaluation of specific surface area in m^2/g of the carbon black and activated carbon, yielding important information in studying the effects of surface porosity and particle size. The carbon content of the activated carbon and carbon black produced during the experiment was also determined.

The project is significant in the sense that it will help to create an effective management of discarded

automobile tyres through their conversion into valuable products including carbon black which can be subsequently used for making activated carbon. This will make locally produced activated carbon to be affordable, thereby reducing the dependency on imported activated carbon. Besides, the project also demonstrates the feasibility of using discarded tyre-based activated carbon for mitigation of indoor air pollution due to generator exhaust emissions.

2.0 Materials and Methods

The research methodology was divided into five sections which included, design and fabrication of equipment for pyrolysis of discarded rubber tyres, pyrolysis of the discarded rubber tyres, activation of pyrolytic carbon black residue, characterization of carbon black and activated carbon, as well as, assessment of carbon monoxide-removal efficiency of the activated carbon adsorbent.

2.1 Experimental Materials

Regarding the feedstock, waste rubber tyre was the only raw material used for the pyrolysis process during the research work. The waste tyre samples were collected from dumpsites around Lagos State University, Ojo, Lagos, Nigeria. Reinforced steel was removed from the waste tyres before they were shredded manually into chips, washed with clean water, and then spread under the sun for drying. Chemical balance, measuring cylinder, and stop clock were also used during the pyrolysis experimental process for measuring feedstock weight, cooling water volume, and progressing reaction time, respectively. Empty containers were prepared for the collection of condensed liquid oil and char during the pyrolysis process. The set-up for the pyrolysis process is shown in Figure 1. For the activation process, five different chemicals were purchased including potassium hydroxide (KOH), potassium nitrate (K_2NO_3), aluminium chloride ($AlCl_3$), hydrochloric acid (HCl) and copper (I) chloride (CuCl). Aluminium chloride, hydrochloric acid and copper (I) chloride were mixed together to form a hydrochloric solution of aluminium chloride and copper (I) chloride. To assess activated carbon removal efficiency of the toxic gaseous emissions from gasoline-powered generators; a 2 KVA Senwei gasoline generator and a gas analyzer were rented. Activated carbon holder equipment was also constructed using plastic material. The height and diameter of the activated carbon holder were 220mm and 50mm, respectively (Plate 1).

2.2 Equipment Design and Fabrication for Pyrolysis of Discarded Rubber Tyres

The pyrolysis system comprised of gas-fired furnace, pyrolysis reactor, heavy oil condenser (15 litre-capacity), two cyclones for light oil condensation (each of 40 litre-capacity), scrubber for gas cleaning (30 litre-capacity), and gas storage bag (Figure 1). The condensers and the scrubber have the same thickness of 5mm. The pyrolysis reactor was a cylindrical steel vessel with thickness, internal diameter and capacity of 12 mm, 250 mm, and 25 kg of shredded discarded rubber tyres, respectively. The reactor vessel was closed with two pairs of flanges, each with thickness of 10mm (top and bottom). There was a hole in the center of the reactor vessel which served as the gas exit. The gas-fired furnace was made in such a way that it would provide uniform heat to the pyrolysis reactor. Solar-power temperature sensor was inserted in the reactor vessel to measure the temperature in the pyrolysis reactor.

2.3 Pyrolysis of the Discarded Rubber Tyres

The main reactor containing 9 kg of the dried shredded tyres was placed inside liquefied petroleum (LPG) gas-fired furnace for batch pyrolysis process. Before starting the pyrolysis process, between 20 and 30 litres of clean water were measured into each of the two cyclones used for light oil condensation. The 9 kg of shredded tyres was pyrolyzed at a temperature range between 236°C and 488°C with an average temperature and heating rate of 382.44°C and 0.636°C, respectively. The

temperature was maintained for about 5 hours (residence time) at 488°C. At the end of the pyrolysis experiment, the non-condensable gas was collected in a gas storage bag and utilized to power 3.5 KVA gasoline generator with fuel gas adapter. The 60 watts compact fluorescent lamp connected to the generator lightened up throughout the running period of the generator. The condensable liquid oil was collected in two different containers; the first container was used for heavy oil collection, while the second was used for light oil collection. About 4 liters of heavy oil and 1 liter of light oil were collected. The furnace was left to cool down to room temperature (25°C) before the char or carbon black left in the reactor vessel was removed.

2.4 Activation of Pyrolytic Carbon Black Residue

Activated carbon; which is a form of carbon material used as adsorbent due to its large porous structure and specific surface area which enable adsorption of molecules on its surface, is commonly prepared by two basic processes which include physical or gas activation method and chemical activation. In the physical activation, the source material or carbonized material is developed into activated carbon by exposing it to oxidizing atmospheres (oxygen, carbon dioxide or steam) at temperatures above 250°C. In the chemical activation, the carbon material is impregnated with certain chemicals such as strong acid, strong base or salt. Chemical activation is often preferred to physical activation because of its lower temperature, shorter time needed for activating the material, as well as, better quality consistency (Ioannidou and Zabaniotou 2007). In this study, chemical activation method was used to prepare the activated carbon.

The carbon black produced during the pyrolysis process was subjected to chemical activation using three different chemicals including potassium hydroxide (KOH), potassium carbonate (K_2CO_3) and hydrochloric solution of mixture of copper (I) chloride (CuCl) and aluminium (III) chloride ($AlCl_3$). Different weight ratios or impregnation ratios (IR) of activating chemicals to carbon black were used. Impregnation ratio (IR) is the ratio of the amount of activating chemicals to the amount of char or carbon black used, as given in Equation 1 (Kairvehi et al. 2001, BaÅŸar 2006). Regarding potassium hydroxide as the activating chemical, weight or impregnation ratios of 50:50 (IR=1) and 40:60 (IR=0.67) were used while for both potassium carbonate and hydrochloric solution of copper (I) chloride (CuCl) and aluminium (III) chloride, weight or impregnation ratios of 50:50 (IR=1) and 70:30 (IR=2.33) were used. The carbon black was ground to pellet form before mixing it with various activating chemicals in 1000mL measuring cylinder at room temperature. The carbon black was impregnated with the chemicals for 24hrs after which the impregnated materials were packed into some plastic containers.

$$IR = \frac{Amount\ of\ Activating\ Chemical}{Amount\ of\ char\ on\ carbonblack}$$

2.5 Characterization of Carbon Black and Activated Carbon

The Brunauer-Emmett-Teller (BET) analysis was carried out using Quantachrome NovaWin-Data Acquisition and Reduction for NOVA instruments (1994-2013, version 11.03). BET analysis was used to determine the specific surface area and pore size distribution of the carbon black and the activated carbons produced during the experiment. The carbon content of the activated carbon and carbon black produced during the experiment was also investigated. The information from the analyses is useful in the prediction of the dissolution rate of the adsorbate on the adsorbent, as the rate is proportional to the specific surface area.

2.6 Assessment of Carbon Monoxide-Removal Efficiency of the Activated Carbon

The activated carbons produced were tested for their removal efficiency of toxic emissions, in

particular, carbon monoxide, from portable generator. The generator used for the study was a 2 KVA Senwei gasoline generator with generator number of EC5800CX while the analyzer was a multi 4 stage gas analyzer with model number of EXIBD1 Multi-GAS analyzer. The prepared activated carbon particles were packed into the activated carbon holder with one end of the packed holder attached to a hose and plugged to the exhaust pipe of the generator while the other end of the holder was connected to the gas analyser (Plate 1). The duration for the assessment of carbon monoxide-removal efficiency of the activated carbon adsorbent was 3 minutes.

Adsorption capacity or removal efficiency of activated carbon is influenced by factors such as concentration, pH, and ionic strength of the activating medium or solution; nature of the adsorbate materials, as well as, the activated carbon preparation conditions including activation temperature, activation time and chemical impregnation ratio (IR) which affect the pore development and surface characteristics of the activated carbon produced (Kairvehi et al. 2001, BaÅŸar 2006). The removal efficiency of pollutant is illustrated by the relation given in Equation 2 (BaÅŸar 2006).

$$E\,(\%) = \frac{C_i - C_f}{C_i}$$

where E= Efficiency of pollutant removal by the activated carbon, C_i = initial concentration of pollutant and C_f= final concentration of pollutant.

1.0 Results and Discussion

During the pyrolysis of the discarded rubber tyres, only 3.7 kg of carbon black or char

was obtained from the 9kg of the rubber tyres loaded into the pyrolysis reactor. The limitation of the study was the technical hitch of inefficient design of the air-fuel intake port of the pyrolysis furnace. The air-fuel intake port of the pilot scale pyrolysis reactor could not promote good air to fuel mixing for complete combustion as indicated by the yellow flame produced in the furnace throughout the period of the investigation. This might be responsible for the loss of valuable time and heat energy experienced during the experimental stage. It clearly showed that the reactor design was not efficient and needed to be improved upon in future related research to avoid loss of valuable fuel energy and environmental pollution.

The results of the BET and carbon content analyses on the carbon black and activated carbons produced during the experiment are given in Table 1 while the results of the experiments performed regarding the activated carbon - adsorption efficiency of the generator gaseous emissions are given in Table 2. The pore sizes of the resultant activated carbons (Table 1) are very big when compared to the kinetic diameter of CO, CO_2 and H_2S (generator gaseous emissions) which are 0.376, 0.33 and 0.36nm, respectively. This is important since adsorption increases when the pore diameter of the adsorbent is one to five times bigger than the adsorbate diameter. According to International Union of Pure and Applied Chemistry (IUPAC), the pore sizes of the activated carbons and the carbon black with range from 2.92 to 6.54nm were mesopores since their pore widths were between 2 and 50nm (Lowell et al. 2006). For the surface area, it can be assumed that the adsorption characteristics of adsorbent with higher surface area will be better because there will be more available spaces where the adsorbates could be retained; adsorption capacity of adsorbent is highly dependent on the surface area of the adsorbent (March et al. 1984, Reynolds 1982). The large surface area of activated carbon, among other influencing factors, is responsible for their adsorption properties

Regarding the adsorption capacity performance of the activated carbons produced, it was observed that the activated carbon efficiency for removing carbon monoxide (CO) from the generator exhaust emissions was relatively high regardless of the nature of the activating chemicals and the impregnating ratios (IR) used. The range values of CO removal efficiencies for all the chemically activated carbons were between 94.0 and 95.5%. When the activated carbons were impregnated

with activating chemicals including KOH, K_2CO_3 and hydrochloric solution of mixture of CuCl and $AlCl_3$ using the IR of 1, the adsorption capacity efficiencies were 95.0, 94.9 and 95.5%, respectively. When the impregnating ratios different from one were used, the efficiency values of the activated carbon materials for activating chemicals KOH, K_2CO_3 and hydrochloric solution of mixture of copper (I) chloride and aluminium (III) chloride were 94.9, 94.6 and 94.0%, respectively. Although activated carbon impregnated with hydrochloric solution of mixture of CuCl and $AlCl_3$ using IR of 1 had the least surface area, pore volume and pore size of 285 m^2/g, 0.116cm^3/g and 2.92nm, respectively; the activated carbon was observed to have the highest CO removal efficiency (Table 2). This might be attributed to his high carbon content, as well as, the strong π-complexation bonds formed between CO and CuCl.

The activated carbon efficiencies for the removal of CO_2 from the generator exhausts were very low regardless of the nature of the activating chemicals and impregnating ratios. This indicates that the impregnated activated carbon materials are not good adsorbents for carbon dioxide from the generator exhausts. When the activated carbons were impregnated with activating chemicals including KOH, K_2CO_3 and hydrochloric solution of mixture of CuCl and $AlCl_3$ using the IR of 1, the adsorption capacity efficiencies were 40.3, 40.6 and 40.3%, respectively. When the impregnating ratios different from one were used, the efficiency values of the activated carbon materials for activating chemicals KOH, K_2CO_3 and hydrochloric solution of mixture of copper (I) chloride and aluminium (III) chloride were 40.6, 39.7 and 40.9%, respectively. Similar to CO_2 removal, activated carbon removal efficiency for hydrogen sulphide removal was very low. The activated carbon efficiencies for removing hydrogen sulphide from the generator exhausts when IR of 1 was used for activating chemicals KOH, K_2CO_3 and hydrochloric solution of mixture of CuCl and $AlCl_3$, were 42.5, 39.7 and 42.2%, respectively. The activated carbon efficiencies for removing hydrogen sulphide from the generator exhausts when IR different from one was used for activating chemicals KOH, K_2CO_3 and hydrochloric solution of mixture of CuCl and $AlCl_3$, were 39.1, 36.9 and 41.6%, respectively.

4.0 Conclusion

This study has demonstrated that through pyrolysis process, discarded automobile tyre could be converted to a value-added product like activated carbon which could be used to reduce carbon monoxide concentration in the generator exhausts. This is significant as it will reduce the number of people that may die from carbon monoxide poisoning due to exposure to gasoline or diesel powered generator exhausts.. The aim of this study was achieved in the sense that the discarded rubber tyres were converted to activated carbon materials which were then verified to be effective in the removal of carbon monoxide from generator exhaust. Commercial application of this research project in Nigeria will allow effective management of discarded automobile tyres, as well as, make locally produced activated carbon to be affordable, thereby, reducing the dependency on imported activated carbon. Availability of locally produced activated carbon will make the domestic application of the activated carbon as adsorbent for carbon monoxide feasible.

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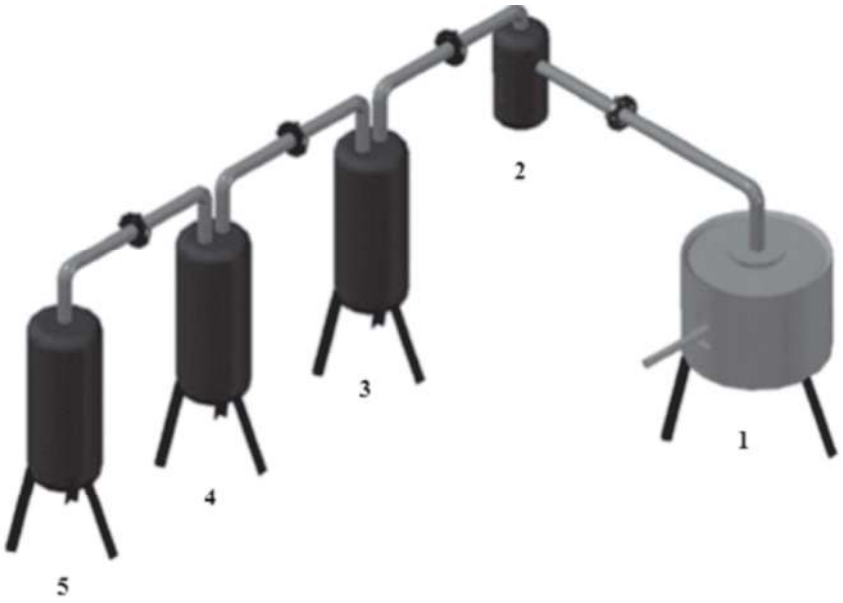

Figure 1: Pyrolysis system set-up

1: Pyrolysis reactor enclosed in gas-fired furnace
2: Heavy oil condenser
3: First cyclone for light oil condensation
4: Second cyclone for light oil condensation
5: Scrubber for gas cleaning

Plate 1: Experimental Setup for the Investigation of the activated carbon removal efficiency of the toxic gaseous emissions from gasoline-powered generator

Table 1: BET and Carbon Content Analyses of Carbon Black and Activated Carbons produced during the Experiment

S/N	Material	Activating Chemical	Brunauer Emmet Teller (BET) Analysis							Carbon content using SEM Compostional Analysis	
			Surface area (m^2/g)	Pore Volume (cm^3/g)	Pore size (nm)	BET Summary				Atomic concentration (%)	Weight Concentration (%)
						Slope	Intercept	Correlation Coefficient, r	C constant		
1	Activated Carbon -1a (AC_{1a})	Potassium hydroxide (KOH)	627.5	0.221	6.54	3.501	2.049	0.999392	2.708	28.30	12.13
2	Activated Carbon -2a (AC_{2a})	Potassium carbonate (K_2CO3)	846.0	0.354	5.75	3.319	0.797	0.990636	5.165	31.77	15.73
3	Activated Carbon -3a (AC_{3a})	Mixture of CUCl, $AlCl_3$ and HCl	285.0	0.116	2.92	9.618	2.601	0.989829	4.698	38.82	18.72
4	Carbon black	none	188.4	0.072	3.11	13.259	5.226	0.996314	3.537	57.53	32.73

Table 2: Activated carbon - adsorption efficiency for generator gaseous emissions reduction

S/N	Activated Carbon (AC)	Activating Chemical	Amount of Activating Chemical used (g)	Amount of carbon black used (g)	Impreg-nation ratio (IR)	Carbon monoxide (CO)			Carbon dioxide (CO_2)			Hydrogen Sulphide (H_2S)		
						CO Initial ppm	CO Final	CO Reduction Efficiency (%)	CO_2 initial	CO_2 Final	CO_2 Reduction Efficiency (%)	H_2S Initial	H_2S Final	H_2S Reduction Efficiency (%)
1	AC_{1a}	Potassium hydroxide (KOH)	200	200	1	3.34	0.14	95.0	3.40	2.03	40.3	3.2	1.84	42.5
2	AC_{1b}	Potassium hydroxide (KOH	160	240	0.67	3.34	0.17	94.9	3.40	2.02	40.6	3.2	1.95	39.1
3	AC_{2a}	Potassium carbonate (K_2CO3)	200	200	1	3.34	0.17	94.9	3.4	2.02	40.6	3.2	1.93	39.7
4	AC_{2b}	Potassium carbonate (K_2CO3)	280	120	2.33	3.34	0.18	94.6	3.4	2.05	39.7	3.2	2.02	36.9
5	AC_{3a}	Mixture of CUCl, $AlCl_3$ and HCl	305	305	1	3.34	0.15	95.5	3.4	2.03	40.3	3.2	1.85	42.2
6	AC_{3b}	Mixture of CUCl, $AlCl_3$ and HCl	427	183	2.33	3.34	0.20	94.0	3.4	2.01	40.9	3.2	1.87	41.6