

Effect of Activated Carbon and ZSM-5 on Oil Yield from Microwave Assisted Pyrolysis of Automotive Paint Sludge (APS)

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Abstract

Automotive paint sludge (APS) contains valuable material which can be recovered by microwave-assisted pyrolysis as studied by previous researches. In this study, a combined application of microwave absorber and catalyst is used in order to study the effect of both components in microwave-assisted pyrolysis of APS. Activated carbon and ZSM-5 acted as microwave absorber and catalyst, respectively were used to investigate their effect on pyrolytic oil produced from APS. Parameters used were microwave power (W), radiation time (T), amount of microwave absorber and amount of catalyst. It was observed that catalyst of 1.5% yielded the highest amount of oil product, which is 1.84% when mass loading, power, time and MWA are fixed. Meanwhile, MWA of 6% produced the highest amount of oil product, which is 7.75% when mass loading, power, time and catalyst are constant. On the other hand, the calorific value (CV) for oil products is found to be in the range of 37 MJ/kg to 39 MJ/kg, which is within the range of commercialized liquid fuel of 36 MJ/kg to 48 MJ/kg. The outcome of this work is beneficial for the alternative utilization of APS particularly as renewable fuel.

Keywords: activated carbon; automotive paint sludge (APS); catalyst ZSM-5; catalytic microwave-assisted pyrolysis.

1. Introduction

1.1. Automotive Paint Sludge (APS)

Automotive paint sludge (APS) is mainly generated from painting process in car manufacturing plant. Car manufacturing process consists of four main stages which are stamping, welding, painting, and assembly. During the painting process in the spray booth, excess paint was accumulated in the water stream in the tank under the spray booth. The excess spray paint were injected with paint killer and then coagulates to form APS and collected at the sludge tank [1]. Based on data, demands of car are increasing steadily over the year which can be seen in Fig. 1, in which contributing to the production of APS [2]. There are in average 200 tonne of APS generated each year in Malaysia and the primary sources of hazardous waste in automotive manufacturing plant is mainly generated from automotive painting process, which comprises of 80% of environmental concerns in manufacturing plants [3], [4]. Table 1 showed chemical composition of APS. Xylene, a cyclic hydrocarbon, is the highest percentage of compound in APS. Short term exposure to xylene caused irritation to sensory organ while long term exposure lead to respiratory problem. According to Zorpas and Inglekazis, painting and coating account for the largest share of environmental impacts (62%) during the manufacturing of automobiles [5]. Treatment of APS before being disposed of at the landfill is quite tedious and time consuming. Hence, there is a need to search for alternative way of disposal of APS that requires short period of treatment at the same time is able to recover valuable products as shown in Table 2.

Table 1: Automotive paint sludge chemical composition [6]

Raw material composition	wt.%
Xylene	43.00
Solvesso 150	19.80
Toluene	15.00
Ethyl Acetate	5.75
Ester	4.88
Glycol Ether	2.69
Acrylic Resin	1.54
Cellosolve Acetate	1.46
Amino Resin	1.12
Acrylic copolymer	1.12

Table 2: Elemental results of APS from X-ray fluorescence spectroscopy (XRF) test [7], [8]

Element	Concentration
Aluminium oxide Al ₂ O ₃	18.01 wt%
Silicon dioxide, SiO ₂	2.15 wt%
Titanium dioxide, TiO ₂	54.21 wt%
Barium oxide, BaO	11.26 wt%
Bromine, Br	300 ppm

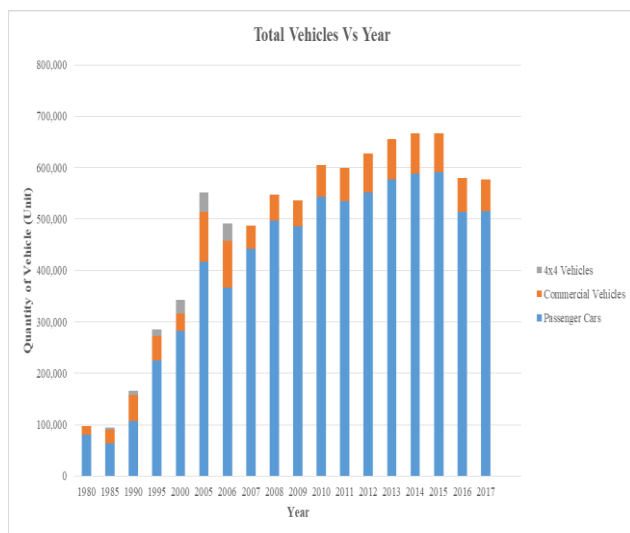


Fig. 1: Production of vehicles in Malaysia from 1980 to 2017 [2]

1.2. Microwave-assisted pyrolysis

Microwave-assisted pyrolysis is a process in which decomposition of hydrocarbon materials by heating them in the absence of oxygen or any other reagents using microwave radiation energy [9]. Products of the process are pyrolytic oil, char and syn gas. Microwave-assisted pyrolysis has many advantages as compared to conventional pyrolysis including uniform, rapid, and fast heating without pre-treatment process such as drying and grinding [10],[11]. Microwave-assisted pyrolysis application towards energy and valuable material recovery is vast. Various studies have been conducted to recover oil from waste material such as lignocellulosic biomass (wood, rice husk and rice straw) by Bo Zhang *et al.* [10],[12], Lam *et al.* [13] on waste palm oil, and J.H Ng *et al.* [14] used crude glycerol of waste palm oil to produce bio-oil. Other non-bio material such as oil waste from oil shipping and coal solid can also be pyrolyzed with microwave-assisted pyrolysis to recover oil from the material which has been reported by Wan Adibah *et al.* [15] and Faisal Mustaq *et al.* [16].

2. Methodology

2.1. Materials

Automotive paint sludge (APS) which is the main feedstock of this study was collected from an automotive manufacturer in Klang Valley, Malaysia. APS is in slurry form was collected from the water wash chamber under spray booth section. No pre-treatment of APS is needed prior to microwave assisted pyrolysis. Weight of the sample is fixed at 200 g which yielded the highest oil product in previous study [3]. Meanwhile, microwave absorber (MWA) used in this study is activated carbon and catalyst used is ZSM-5.

2.2. Microwave Assisted Pyrolysis of Automotive Paint Sludge

Domestic microwave (2.45 GHz) has been used to pyrolyze APS. To determine the effect of catalyst on pyrolytic oil, APS is placed in a quartz reactor with fixed loading amount of 200 g at 600 W, 50 min and fixed amount of activated carbon of 10 wt.%. Meanwhile, to investigate the effect of activated carbon on pyrolytic oil, APS is placed in a quartz reactor with fixed loading amount of 200 g at 1000 W, 30 min and fixed amount of catalyst of 1.5 wt.%. Amount of activated carbon and catalyst ZSM-5 used in this experiment are based on parameter setting as shown in Table 3 and Table 4.

Table 3: Parameter Setting 1

No.	Loading (g)	Power (W)	Time (min)	MWA %	Catalyst %
1	200	600	50	10	0.5
2					1.0
3					1.5

Table 4: Parameter Setting 2

No.	Loading (g)	Power (W)	Time (min)	Catalyst %	MWA %
1	200	1000	30	1.5	6
2					8
3					10

For both processes, APS was placed in a quartz reactor and mixed thoroughly with activated carbon (microwave absorber). Meanwhile, catalyst ZSM-5 was placed in a silica crucible without a lid. The silica crucible is then placed inside the quartz reactor in the microwave. 250 ml/min of nitrogen gas is purged into microwave system for 10 minutes prior to experiment commencement in order to ensure inert condition in the quartz reactor. After experiment starts, nitrogen gas inlet is set to 150 ml/min until experiment ends. Experiment is repeated as per setting in Table 3 and Table 4. Nitrogen gas is used as carrier gas for inert condition preparation in the quartz reactor in many studies such as used by Lam *et al.* [13],[17],[18].

Three condensers arranged in series to collect the condensable gas, whereby the oil product were collected. The arrangement of microwave-assisted pyrolysis of APS is shown in Fig. 2.

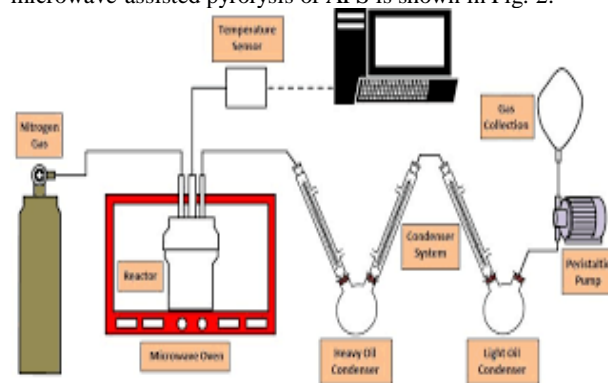


Fig. 2: The arrangement of equipment in microwave-assisted pyrolysis of APS [3]

The resulting liquid product was then separated by using liquid-liquid extraction method. Hexane was used as a solvent for the extraction as it is suitable for non-polar hydrocarbon separation [19]. Hundred milliliter of hexane was added to the liquid product obtained from condenser in each cycle of experiment. In this study, five parameters were used, in which three parameters (loading, power and time) were set constant and another two (MWA % and catalyst %) were varied in each setting. The resulting liquid product from the extraction process formed an oil fraction and an aqueous fraction. The amount of oil product which has been obtained was then collected and recorded for further analysis.

3. Results and Discussion

3.1. Product Distribution

From the result, it is shown that effect of combination of microwave absorber and catalyst affect the selectivity of product in microwave pyrolysis. As shown in Fig. 3 (variation of catalyst), the amount of liquid product was reduced when the catalyst was increased from 0.5% to 1.5%, which are 12.18%, 11.70%, and 10.20%. However, in terms of oil product, amount of oil was increased with increasing amount of catalyst which recorded as 0.64%, 1.42%, and 1.84%, respectively. Variation of the catalyst amount used in this study have an impact on the selectivity of product, which means the introduction of catalyst ZSM-5 would

further crack long hydrocarbon chain to produce more gases, as compared to liquid oil product in pyrolysis process as mentioned in a review by R. Miandad *et al.* [20]

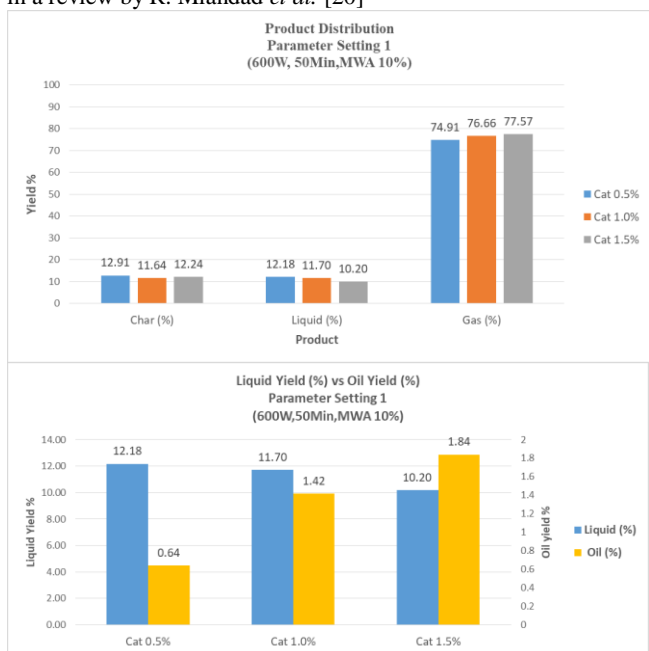


Fig. 3: Product Distribution of Parameter Setting 1 (600W, 50Min, MWA 10%) – Effect of Catalyst Weight Variation

In Fig. 4 (variation of activated carbon or microwave absorber (MWA)), it is shown that liquid product was reduced as MWA varied at 6%, 8%, and 10%. The amount of liquid product was reduced accordingly from 25.68%, 20.94%, and 20.43%, respectively. Meanwhile, oil fraction was also reduced with increasing amount of MWA, which were 7.75%, 4.84%, and 2.34%.

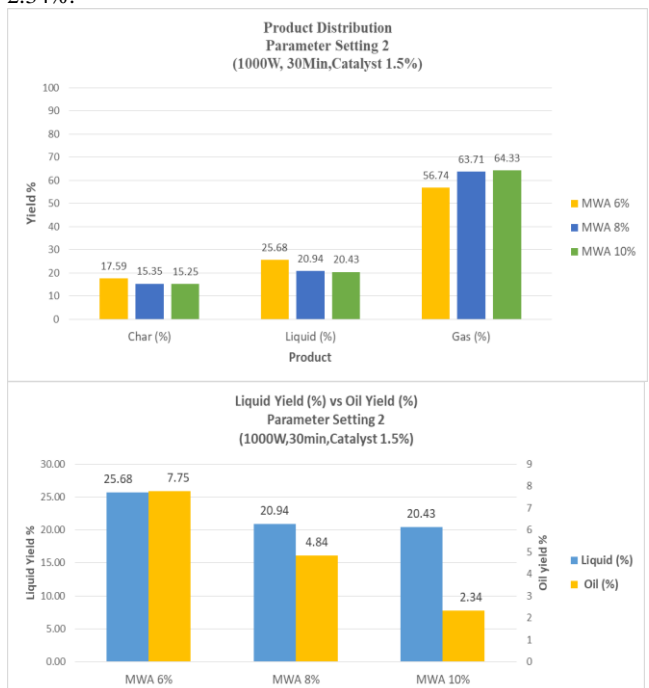


Fig. 4: Product Distribution of Parameter Setting 2 (1000W, 30Min, Catalyst 1.5%) – Effect of MWA Weight Variation

From visual observation, it can be seen that parameter setting 1 and parameter setting 2 produced yellowish liquid product. However, the former product has shown lighter yellowish liquid as compared to the former product, which was darker with appearance of blackish, insoluble and very viscous tar.

3.2. Calorific Value (CV)

In terms of calorific value (CV), there are no significant trends of calorific values among oil products as shown in Fig. 5 and Fig. 6. Calorific values obtained from oil products in this study was in the range of 37 MJ/kg to 39 MJ/kg.

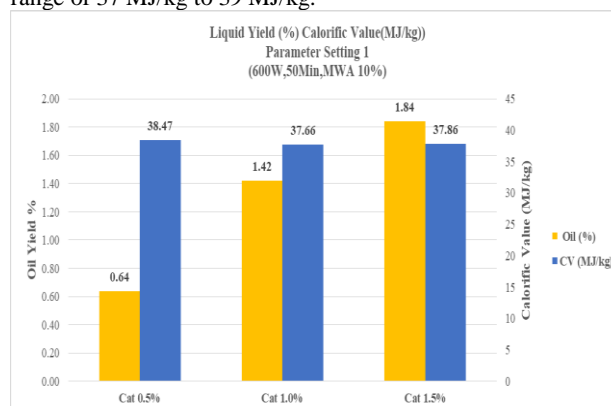


Fig. 5: Calorific Value of Parameter Setting 1 (600W, 50Min, MWA 10%) – Effect of Catalyst Weight Variation

From Fig. 5 and Fig. 6, it can be seen that the combine effect of the absorber and catalyst was insignificant with regard to calorific value (CV). In parameter setting 1 (Fig. 5), calorific value obtained from catalyst variation of 0.5%, 1.0%, and 1.5% were 38.47 MJ/kg, 37.66 MJ/kg, and 37.86 MJ/kg. Meanwhile, for parameter setting 2 (Fig. 6), MWA variation of 6%, 8%, and 10% resulted in calorific value of 37.28 MJ/kg, 37.44 MJ/kg, and 37.20 MJ/kg, respectively.

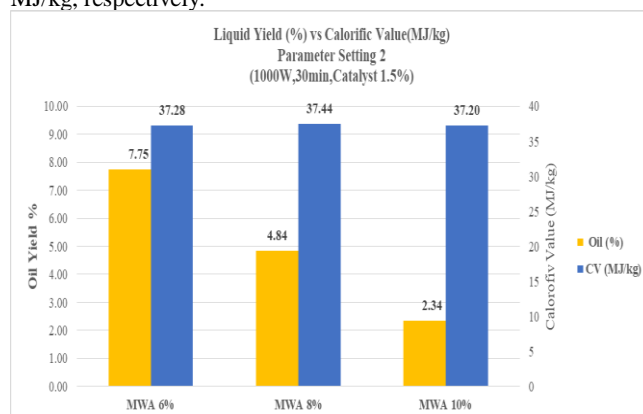


Fig. 6: Calorific Value of Parameter Setting 2 (100W, 30Min, Catalyst 1.5%) – Effect of MWA Weight Variation

4. Conclusion

Microwave assisted pyrolysis of automotive paint sludge (APS) with the presence of absorber and catalyst have an impact on the yield of pyrolytic liquid product. It can be concluded that by having constant amount of microwave absorber (MWA) of 10%, variation in catalyst weight showed increasing trends of amount of oil product as catalyst amount was increased. It can be explained that with the same temperature during pyrolysis of APS, increasing amount of catalyst would increase the selectivity of oil product. Hence, more oil would be produced when amount of catalyst increased. Meanwhile, with constant amount of catalyst (1.5%), variation of MWA showed increasing amount of oil product as amount of MWA reduced. This can be described that at the same catalytic rate, increasing amount of MWA would cause temperature in the quartz reactor during pyrolysis of APS to increase. Hence, at a certain point, secondary reaction occurred which is thermal cracking. Thermal cracking would act to further crack hydrocarbon chain to produce more non-condensable gas instead of oil. As for calorific value (CV), presence of absorber and catalyst do not affect CV significantly. It has been found that

calorific values obtained in this study were in the range of 37MJ/kg to 39MJ/kg. The CV obtained in this study are within commercial liquid fuel properties range which is in between 36 MJ/kg to 48 MJ/kg [21],[22],[23].

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