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Catalytic Pyrolysis of Waste Toner Powder using Low-Cost Local Clay

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Abstract— The global energy demand has gradually increased because of the raising of the global population and rapid urbanization, increasing the dependence on traditional fossil fuels. In response, renewable energy alternative sources are required to minimize the environmental effects and improve the resource recovery. The present work aims to study the catalytic pyrolysis of waste toner powder. Waste toner powder is a hazardous byproduct from the printing industry, which produces dangerous health and environmental issues due to inappropriate recycling techniques. To address this issue, this study presents innovative recycling techniques for converting Waste Toner Powder (WTP) into valuable bio-oil using locally sourced Tafla clay as a low-cost catalyst. The pyrolysis process was performed in a pilot-scale fixed-bed reactor. The catalyst pyrolysis of waste toner powder was carried out at a pyrolysis temperature of 550°C at different mass ratios of 10%, 20%, and 30%. The X-ray Fluorescence (XRF) analysis was selected to characterize Tafla clay, indicating the presence of some important metallic oxides such as SiO_2, CaO, Al_2O_3, and Fe_2O_3 that can be separately used as a catalyst. The Thermogravimetric Analysis (TGA) analysis was also conducted, illustrating that using Tefla clay can increase the degradation of waste toner powder. The bio-oil yield of waste toner powder thermal pyrolysis was 30%. While that of catalyst pyrolysis was 31%, 36%, and 17% at mass ratios of 10%, 20%, and 30%, respectively. The study findings emphasize the possibility of catalytic pyrolysis is a helpful solution for converting hazardous WTP into useful energy source using local and low-cost material.

Keywords: Pyrolysis; catalyst; Tafla clay; Pyrolysis; Waste toner powder.

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1 Introduction

The rapid rise of the world's population and the expanding urbanization resulted in an increase in global energy requests and hence an increase in the dependence on fossil fuels. This is because fossil fuels including petroleum, coal, and natural gas, save about 90% of the world's energy requirement [1,2]. Therefore, there is an urgent need to decrease the dependence on fossil fuels by finding renewable, sustainable, and low-polluting energy sources. The Waste-to-energy approach is one of the methods used to generate sustainable bioenergy. Converting waste to energy is an environmental and economic conversion process [3-5]. Some traditional methods are used to treat the different types of waste such as direct composting, landfilling, burning, and incineration. However, these methods have many disadvantages, like pollution of soil and water, health problems, and climate change. [6,7]. Therefore, there are other methods used to generate biofuel from wastes, these methods can be sorted into three major pathways: biochemical, physicochemical, and thermochemical. Thermochemical techniques include three primary methods: gasification, pyrolysis, and combustion. Thermochemical conversion techniques are preferable than biochemical methods because of their higher efficiency. Among thermochemical routes, the pyrolysis process is given more attention as a waste-to-energy conversion method due to its advantages, mainly the selective control of products [8].

Pyrolysis of waste is an endothermic decomposition process achieved in an inert condition at high temperature, converting waste into useful products of bioenergy, namely bio-oil, bio-gas, and biochar [9]. Nevertheless, the pyrolysis process has been limited by some substandard properties of its products, such as low bio-oil selectivity, lower content of hydrogen in the produced gas, poor stability, and porosity due to the bio-char amorphous structure [10,11]. Consequently, catalytic pyrolysis is used to enhance the quality of pyrolysis products and increase the use of pyrolysis in the waste-energy field. The catalytic pyrolysis has various benefits compared to the non-catalytic method. The foremost advantage is decreasing the temperature of pyrolysis by reducing the activation energy of the reaction [12]. Furthermore, using catalysts has given higher selectivity for desired products of pyrolysis [13-15]. Notwithstanding, catalytic pyrolysis has some limitations. The most important one is the reduction of catalyst performance with time, so the regeneration and durability of catalyst should be considered to enhance the economic value [16]. Therefore, using low-cost catalysts becomes a critical parameter in waste catalytic processes.

Different types of catalysts are investigated in the pyrolysis of numerous types of solid waste. The studies of catalytic pyrolysis focus on using commercial catalysts and low-cost catalysts. A core-shell hierarchical ZSM-5 (zeolite) was used in the corn stalk catalytic pyrolysis. Using catalysts resulted in enhancing the aromatic yield and facilitating the oxygen removal [17]. Using iron nitrite in the biomass catalytic pyrolysis resulted in many benefits such as accelerating aromatic hydrocarbon formation, promoting mesopores formation, and promoting hydrogen release [18]. The calcium oxide catalyst was used in the pyrolysis of eucalyptus residues using a rotary kiln reactor. The higher interaction between catalysts and volatiles in the reactor improved the deoxygenation. Using fractional condensation and improved deoxygenation produced an organic-rich fraction with 12.6 wt% oxygen content [19]. The catalyst co-pyrolysis of different food waste including plastic, eggshells, chopsticks, and bones was achieved using activated biochar and treated eggshells as catalysts. The production of aliphatic and aromatic hydrocarbon increased with higher heating values ranging from 36 MJ/kg to 44.44 MJ/kg. The C-O and C-H ratio of the produced bio-oil demonstrated the possibility of using it as conventional liquid fuel [20]. Two types of low-cost clay, attapulgite and montmorillonite were used as a catalyst in the apple pomace pyrolysis. The two types of clay generate the same bio-oil when they are used at similar process point. The points at which clay was added affected the bio-oil. The addition of clay to hydro char before hydrothermal carbonization produced an average hydrocarbon content of bio-oil twice that when it added after hydrothermal carbonization [21]. The catalytic co-pyrolysis of dealkaline lignin and low-density polystyrene was conducted using red clay catalyst. To estimate effect of pyrolysis temperature on co-pyrolysis, the experiments are performed at different temperatures of 600, 700 and 800°C. the study results gave a novel technique for improving the lignin depolymerization used with plastic wastes via red clay as a catalyst [22].

Waste toner powder is the toner that does not agglutinate the paper during the printing, and it is a byproduct from the printing industry. This type of waste is considered hazardous waste, so it should not be thrown in the trash because it can spread as fine particles, causing health and environmental problems. Therefore, the first objective of this study is converting waste toner powder into biofuel using the pyrolysis process. Catalysts are important to enhance pyrolysis efficiency. However, the high cost of catalysts and difficulty separating them from solid products result in increasing the cost of waste catalytic processes, which leads to the urgent need for developing low-cost catalysts. Hence, the second objective of this study is to develop low-cost new catalysts from local materials. Tafla clay is a very economically and abundant material that was used as a catalyst in this study.

2 Characterization and Experimental Set-Up

2.1 Characterization of feedstock

The ultimate and proximate analyses of Waste Toner Powder (WTP) are given in **Table 1**. The elemental composition of WTP and produced bio-oil and bio-char was conducted using an automatic analyzer (CHNS Vario EL III, an elementary German) at the microanalytical center, Faculty of Science, Cairo University, Egypt. The ultimate analysis of WTP indicated high carbon and hydrogen contents of 60.12% and 4.32%, respectively, which demonstrates that the WTP can be used as an energy-rated precursor. Also, the proximate analysis shows a high content of volatile matter with little moisture content. Also, the higher heating value of 25 MJ/kg and the 0.86 H/C are attributes of WTP's good fuel characteristics that are comparable to other pyrolysis feedstocks.

Table 1 The ultimate and proximate analysis of waste toner powder

Ultimate analysis					
C	**H**	**O_2**	**N_2**	**S**	**H/C**
60.12	4.32	16.425	17.65	1.51	0.86
Proximate analysis					
VM	**FC**	**MS**	**A**	**HHV (MJ/kg)**	
50	14	6	30	25	

VM: volatile matter, FC: fixed carbon, MC: Moisture content, A: ash

2.2 Characterization of catalyst

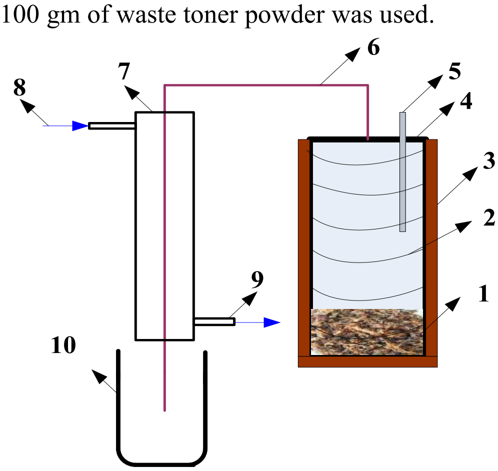
In this study, a new material called Tafla clay was used as a low-cost, available catalyst. Tafla clay is formed because of the decomposition and disintegration of rocks containing felspars. These clays are mostly used in the pottery industry due to their extreme softness and ease of shaping. The chemical composition of Tafla clay was determined using XRF analysis. **Table 2** indicates the chemical composition of Tafla clay as well as red mud and bentonite clay, which are common catalysts. As indicated in the table, Tafla clay has a chemical composition like that of bentonite clay. The chemical composition of clay indicated the presence of CaO, SiO_2, Al_2O_3, and Fe_2O_3 with a good percentage. These metallic oxides are used as catalysts in the pyrolysis process in many studies, such as SiO_2 [23,24], Al_2O_3 [23,25], CaO [19,26], Fe_2O_3 [25], and MgO [23]. The Tafla clay was used as received without any activation process and was mixed with a sample of WTP in a proportion of 20 wt% (20 g of catalyst/100 g of WTP).

Table 2 Chemical composition of Tafla, bentonite, and red mud clays

Raw materials	Constituents (wt.%)						
	CaO	SiO_2	Al_2O_3	Fe_2O_3	MgO	TiO_2	Other
Tefla clay	40.14	34.59	12.23	6.65	1.48	1.09	3.28
Bentonite clay [27]	2.5	46	17	6	2.2	0.2	26.1
Red mud [28]	5.3	8.5	23.8	36.5	-	13.5	25.9

2.3 Pyrolysis experiments and method

The pyrolysis experiments was carried out using a lab-scale fixed-bed reactor manufactured in the heat transfer lab at the mechanical engineering department, faculty of engineering, south valley university. The reactor was constructed from steel with a thickness of 2 mm, a diameter of 10 cm, and a total height of 20 cm. The reactor was insulated using fiberglass with a 2 cm wall thickness of. The condensation of gases is conducted using a shell and tube heat exchanger composed of two concentric tubes. The diameters of the inner and outer tubes are 1.5 and 5 cm, respectively, while their lengths are 50 and 70 cm, respectively. The reactor is heated using an electric heater with a total power of 1 kW. The temperature inside the reactor is measured using a thermocouple type K connected to a temperature controller unit to maintain the temperature at the required value. **Figure 1** illustrates the experimental test rig used in this study. In this experiment, about 100 gm of waste toner powder was used.



(1) Wastes (2) Electrical heater (3) Insulation (4) Reactor (5) Thermocouple (6) Bio-gas pipe (7) Condenser (8) Water inlet (9) Water outlet (10) Bio-oil collector

Fig. 1 The experimental set-up

3. Results and Discussions

3.1 Thermogravimetric analysis

The Thermogravimetric Analysis (TGA) and Derivative Thermogravimetric Analysis (DTG) of uncatalyzed and catalyzed waste toner powder are illustrated in **Fig. 2** and **Fig. 3**, respectively. Both TGA and DTG were carried out at a heating rate of 20 °C/min and a mass ratio of 20%. The figures indicate that the thermal degradation of waste toner powder takes place through three main stages. The first stage occurred in the temperature range from 30°C to 300°C. In this stage the moisture of the material is removed, and some volatile matters are released. The weight loss of this stage is the lowest. The second stage is called active pyrolysis, where the thermal degradation of material is observed in the temperature range of 300°C to 500°C, with the highest weight loss. The third stage starts at temperatures above 500°C. This stage includes the formation of char, which represents the solid portion of material that remains without pyrolysis.

Since the clay itself does not undergo thermal degradation, the content of degradable and volatile components in the mixture is decreased compared to pure WTP. Therefore, the early-stage mass loss appears lower, even though the absolute mass loss remains consistent with the WTP content in the mixture [25]. Without a catalyst, the weight loss starts at 350°C, and the maximum weight loss is observed between 400°C and 500°C. While, with catalyst, the degradation begins at a lower temperature of 300°C, with rapid weight loss taking place in the temperature range of 350°C –450°C. This showed that the utilization of Tafla clay accelerates the decomposition of waste toner powder and decreases the decomposition temperature, as indicated by the earlier start of weight loss and the more rapid reduction in weight. This shift toward lower temperatures demonstrates that the addition of a clay catalyst effectively decreases the activation energy required for the breakdown in WTP components, mainly carbon black pigments and polyester resins. After a temperature of 500°C, the sample weight stabilizes, showing the breaking down of the major decomposable components. The final weight loss of the catalyzed sample is lower than that of the non-catalyzed sample, demonstrating a higher degree of decomposition and lower solid residue remaining. **Figure 3** indicates the DTG curves of WTP without and with the addition of catalyst, illustrating the rate of mass loss (derivative weight (%/min)) as a function of temperature. The DTG curve indicates that the pure WTP has a maximum decomposition rate of –0.38 %/min at a major degradation peak around 425°C. This peak is related to the active stage of WTP pyrolysis, where most volatile components are released. On the other hand, a significant shift in the degradation behavior was observed for WTP and tefla clay mixture. The main decomposition peak occurs at a lower temperature of 395°C, with an increased peak decomposition rate of –0.45 %/min. This illustrates improved thermal degradation kinetics due to catalytic activity. The addition of the catalyst resulted in earlier and sharper peaks, suggesting that Tefla clay effectively decreased the activation energy needed for the decomposition, thereby increasing the breakdown of polymeric components in WTP.

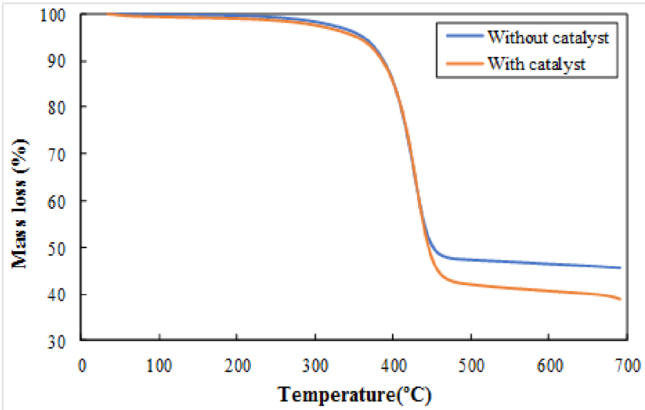


Fig. 2 The TGA for waste toner powder without and with Tafla clay catalyst

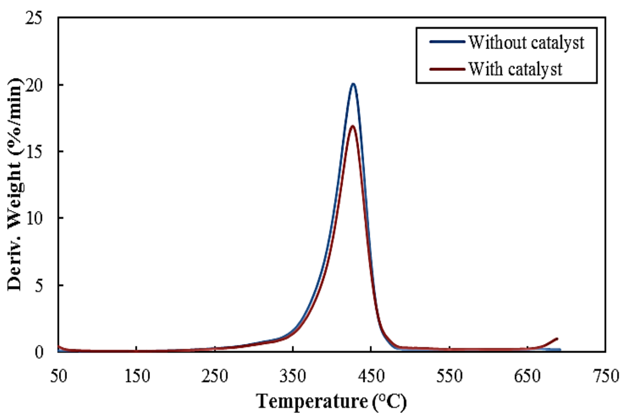


Fig. 3 The DTG for waste toner powder without and with Tafla clay catalyst

3.2 Effect of catalyst on the pyrolysis products

The effect of Tafla clay on the pyrolysis of waste toner powder was investigated at different mass ratios of 10%, 20%, and 30% at a pyrolysis temperature of 550 °C. **Figure 4** compares pyrolysis product yields for thermal pyrolysis (without catalyst) and catalyst pyrolysis (with Tafla clay) of waste toner powder. As shown in the figure, using Tafla clay in the pyrolysis of waste toner powder increased the yield of bio-oil to a certain value, then decreased with further increasing of the percentage of catalyst. As indicated by Figs. 2 and 3, the addition of catalyst accelerated the degradation of waste toner powder and enhanced the pyrolysis of it, which increased the yield of bio-oil. However, with further increase of catalyst, the bio-oil yield decreased. This is because increasing the catalyst can increase the secondary cracking reactions that decrease the content of volatile compounds, which condense to bio-oil, converting them into non-condensable gases. It is worth noting that, a further increase in catalyst content can reduce the bio-oil yield, due to excessive secondary cracking of volatile products, the 20% catalyst content represents an optimal balance between activity catalytic of clay and product yields under the studied conditions.

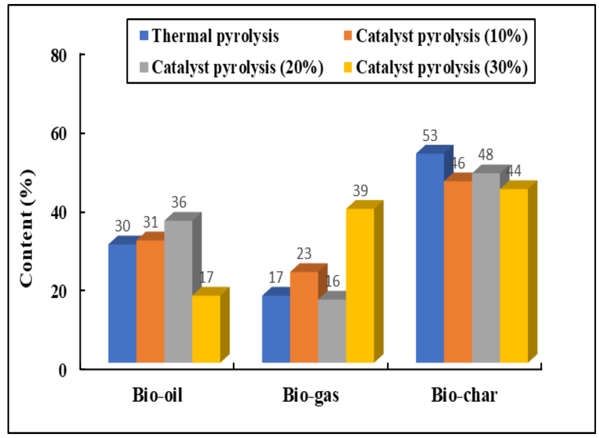


Fig. 4 The yield of pyrolysis products for thermal and catalyst pyrolysis of waste toner powder

4 Comparative study

For better understanding the potential of Tafla clay as a low-cost catalyst in the WTP pyrolysis, it is essential to compare its composition, catalytic behavior, and product selectivity with those of other commonly used low-cost catalysts such as red mud, and modified biochar. As given in **Table 2**, tafla clay contains significant amounts of CaO (40.14%), SiO_2 (34.59%), Al_2O_3 (12.23%), and Fe_2O_3 (6.65%). All these metallic oxides are known for their catalytic activity in pyrolysis reactions, contributing to various reaction mechanisms. Compared to red mud and bentonite clay, Tefla clay has a higher content of CaO, indicating its ability to promote deoxygenation reaction that decreases the content in bio-oil. Based on its chemical composition, Tafla clay can provide a balanced composition, integrating both acidic and basic functionalities, which is useful for conducting a broader range of catalytic impacts during pyrolysis. **Table 3** shows a comparison between Tafla clay and other types of low-cost catalysts. The table indicated that Tafla clay has competitive catalytic performance without needing any modification or pre-treatment process, which is considered a key advantage in terms of process simplicity and cost-effectiveness.

Table 3 Che comparison between Tefla clay and other catalysts

Feedstock	Catalyst	Key findings	Ref.
Waste toner powder	Tafela clay	- The bio-oil yield increased up to 36%. - The degradation was improved	Current study
Eucalyptus residues	Calcium oxide	- Deoxygenation was improved with 12.6 wt% oxygen content	[19]
Food waste and plastic wastes	Activated biochar and treated eggshell	- The higher heating values ranged from 36 MJ/kg to 44.44 MJ/kg.	[20]
Apple pomace	Attapulgite and montmorillonite clays	The yield of bio-oil increased twice	[21]
Lignin and plastics	Red mud	The depolymerization and hydrocarbon yield were improved	[22]

5 Conclusion

Waste toner powder is an excess powder left in the printer cartridge without use. It is treated as garbage and can't be recycled. Therefore, this study aims to use waste toner powder as a source of bio-oil. To enhance quality and increase the yield of bio-oil, catalysts are used in the pyrolysis process. In this study locally available, low-cost clay called Tafla clay was used as a catalyst in the pyrolysis of waste toner. The XRF analysis was applied to characterize the chemical composition of clay. Both TGA and DTG were selected to address the effect of Tafla clay on the pyrolysis of waste toner powder. The catalyst pyrolysis of waste toner powder was carried out at a pyrolysis temperature of 550 °C at different mass ratios (catalyst to feedstock) of 10%, 20%, and 30%. The main findings of this work are summarized as following:

- Tefal clay contains some important metallic oxides that can be separately used as catalysts, mainly SiO_2, CaO, Al_2O_3, and Fe_2O_3.
- The TGA and DTG analyses indicated that Tafla clay can accelerate the pyrolysis of waste toner powder and decrease the degradation temperature, and the activation energy required to carry out the pyrolysis process.
- The bio-oil yield of catalyst pyrolysis of waste toner powder was 31%, 36%, and 17% at mass ratios of 10%, 20%, and 30%.
- Further increase of catalyst decreases the bio-oil yield.

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