



The Catalytic Impact of MnO Additive on the Selected Municipal Solid Waste Combustion Behavior Determined by Thermogravimetric Analysis

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ABSTRACT

The thermogravimetric (TG) and kinetic analysis have been performed to evaluate the catalytic impact of MnO additive on selected municipal solid waste (SMSW) combustion behavior. Nineteen kinds of the SMSW have been collected, cleaned, dried, and grounded to the size of less than 1 mm and then mixed with the same mass ratio. The addition of an additive to the SMSW was completed by virtue of blending the 10 mg SMSW with 0.5 mg the MnO powder and then was ground thoroughly in the mortar to obtain the homogeneous mixture of SMSW/5%MnO. Subsequently, the sample was brought into the crucible uniformly. The sample combustion in the chamber was carried out under non-isothermal conditions at the controlled heating program of 10 and 30 °C/min, a temperature range of 25-1000 °C, and an air atmosphere flowrate of 100 ml/min. The sample mass loss during temperature and time escalation was recorded by a computer automatically to be a TG curve. The 1st derivative of the TG curve to the temperature or time represented the rate of sample mass loss and denoted as the derivative thermogravimetric (DTG) curve. In accordance with both TG and DTG curves, then the characteristic temperature parameters were determined. The Coats-Redfern method has been adopted to evaluate kinetic parameters in this study. To understand the catalytic effect of the MnO, the blending SMSW/5%MnO results were compared with the pure SMSW ones. The results indicated that the MnO additive has promoted the initial temperature decomposition toward the lower value and provoked the increasing mass loss rate. Moreover, the presence of MnO also decreased the activation energy in the main combustion region. The overall results revealed that the MnO additive has significantly impacted the dynamic of thermal decomposition during the combustion processes.

Keywords:

Selected Municipal Solid Waste;
Combustion; Thermogravimetric;
Kinetics; MnO; Catalytic

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1. Introduction

The parallel growth of the people and their living standards have forced human mobilities more dynamics in urban society, which accompanied by lifestyle changes. These situations have pushed the increasing waste, which is the matter discarded as no longer useful or required after the completion of their daily necessities [1]. Tyagi *et al.*, [2] reported that worldwide municipal solid waste (MSW) generation reaches the levels of 1.3 billion tons per year and is estimated growth continuously to around 2.2 billion tons per year by 2025. Part of the huge amount of the MSW is plastic, that its production projected about 300 million tons annually [3].

In Indonesia, the rate of MSW generation from 2010 to 2025 is around 97,356 tons/day, and part of them is a synthetic waste (SW) especially plastic that has grown in the rate of 10,660 tons/day [4]. From the amount of plastic waste, at least 20% is disposed of in rivers and coastal waters. Considering the environmental impact of the enormous amount of waste, proper management must be being a critical concern currently.

Utilization of waste as an energy feedstock is believed as an attractive choice to decrease the disposal problem and mitigate pressure on the environment as well as supplying clean energy [5]. It would significantly reduce the volume by 95% and decrease mass by 90% [6-7]. Moreover, proper burning techniques of waste yielded reducing landfilling space, significant energy recovery, and reducing the environmental effect [8].

In recent years, studies on waste combustion for the purpose of energy generation have found significant progress. Hu *et al.*, [9] studied the co-combustion characteristic of MSW and paper mill sludge, and the results indicated that the presence of 80% paper mill sludge in blend led to the activation energy value achieved the minimum. Liu *et al.*, [10] had investigated the thermal behavior of MSW during pyrolysis and combustion by comparing their characteristics with and without additives (ZnO, Fe₂O₃, CuO, and Al₂O₃). It was reported that the additives had a catalytic effect during MSW ignition and combustion except for combustion with CuO. Boumanchar *et al.*, [11] examined the kinetics of the co-combustion of biomass, coal, and MSW and found that there were synergistic interactions between biochar, coal, and MSW with biomass. However, among the various published works, the impact of MnO additive on selected municipal solid waste (SMSW) combustion behavior was not found in the kinds of literature.

Manganese oxides are recognized for demonstrating high performance during the hydrocarbon oxidation [12]. The oxidation completion of ethyl acetate due to the catalytic activities of manganese oxides has reported by Santos *et al.*, [13]. The reactivity of MnO, MnO₂, and Mn₂O₃ for the CO oxidation has investigated by Ramesh *et al.*, [14]. The MnO played an excellent catalytic activity during the styrene oxidation has reported by Tseng and Chu [15]. These aforementioned overall literatures have strongly indicated the potential of manganese oxide as the catalyst during the oxidation process; however, the catalytic reactivities of MnO for the SMSW combustion have not discussed in the previous paper.

This paper presented the evaluation of the MnO additive on the SMSW combustion to investigate its catalytic impact. The experiment was performed under the apparatus of the thermogravimetric analyzer. The kinetic parameters during the combustion were examined by using a fitting method of Coats-Redfern.

2. Methodology

2.1 Materials Preparation

Detail of the SMSW material preparation had presented in our previous literature [16]. These SMSW materials consist of 19 kinds of synthetic waste that commonly encountered in the temporary shelter in Malang City, Indonesia. They were composed of various thick plastic, shampoo bottles, mineral bottles, banners, styrofoam, detergent packs, mattresses, plastic bags, flip-flops, jeans, water pipes, motorcycle body, rice wrapper plastic, oil bottles, sponges, rice bags, bottles of floor cleaner liquid, synthetic leather, and jerry cans of water. To warrant the SMSW were free from the impurities material, they were washed and then occupied two days sun-drying process during the dry season. The dry samples then were shredded by a rotating grater to attained the size less than 1 mm. The final SMSW powder was composed of 19 kinds of synthetic waste with the same mass ratio, in which every type of synthetic waste that weighs 0.5 g was decanted into a mortar to mix them entirely, and subsequently, they were ground.

The completion of SMSW with MnO additive was done by means of mixture the 10 mg SMSW with 0.5 mg MnO powder. A similar additive mass ratio has been found in the previous literature [10,17]. To ensure the homogeneous mixture, the samples were put into the mortar and subsequently were ground and mixed under the hand pressure.

2.2 Thermogravimetric Experiment

The main parameters that commonly used for featured the thermal behavior of the solid samples during the combustion process were thermogravimetric (TG) and derivative thermogravimetric (DTG) curves. These either TG and DTG data were obtained by conducting the experiment under the apparatus of the thermal analyzer (METTLER TOLEDO TGA/DSC1). A very small sample amount of 10 mg was inserted in the crucible and then subjected to the furnace that had been set with non-isothermal heating with a rate of 10 and 30 °C/min for each experiment. The combustion was performed in the temperature escalation started from ambient temperature to 1000 °C. The sufficiency oxidizer was given by means of flowing the air atmosphere continuously at a rate of 100 ml/min during the combustion process. The TG and DTG data that were generated during the experiment recorded by a computer automatically. From these TG and DTG data, subsequently, the characteristic temperatures were determined and then used for examining the kinetic parameters.

3. Results and Discussion

3.1 Effects of MnO Addition on The Thermal Behavior of the Samples

Figure 1 presented the TG and DTG of each sample tested at a heating rate of 10 °C/min (Figure 1(a) and 1(b)) and heating rate 30 °C/min (Figure 1(c) and 1(d)). Based on these TG and DTG curves, the parameters of characteristic temperature and the maximum mass loss rate in the main combustion region were specified as depicted in Table 1. In accordance with Figure 1 and Table 1, the combustion process of SMSW with and without additive could be divided into two regions.

The Region I for the SMSW at a heating rate 10 °C/min stretched in the temperature range of 235.92-365.91 °C. It was clearly observed that addition 5%MnO provoked the onset degradation temperature (T_o) shifted to lower temperature as well as the burnout temperature (T_b), where the Region I for the SMSW/5%MnO occurred at temperature range 207.35-313.30 °C. This region was correlated with degradation and combustion of organic volatile compound originated from polyvinyl chloride (PVC) [18]. This region also indicated by decreasing the value of the maximum mass loss rate

($-DTG_{max-1}$) due to the presence of 5% MnO. The temperature of the maximum rate (T_{max-1}) slightly shifted toward the higher value because of the addition of 5%MnO from 277.56 to 279.86 °C.

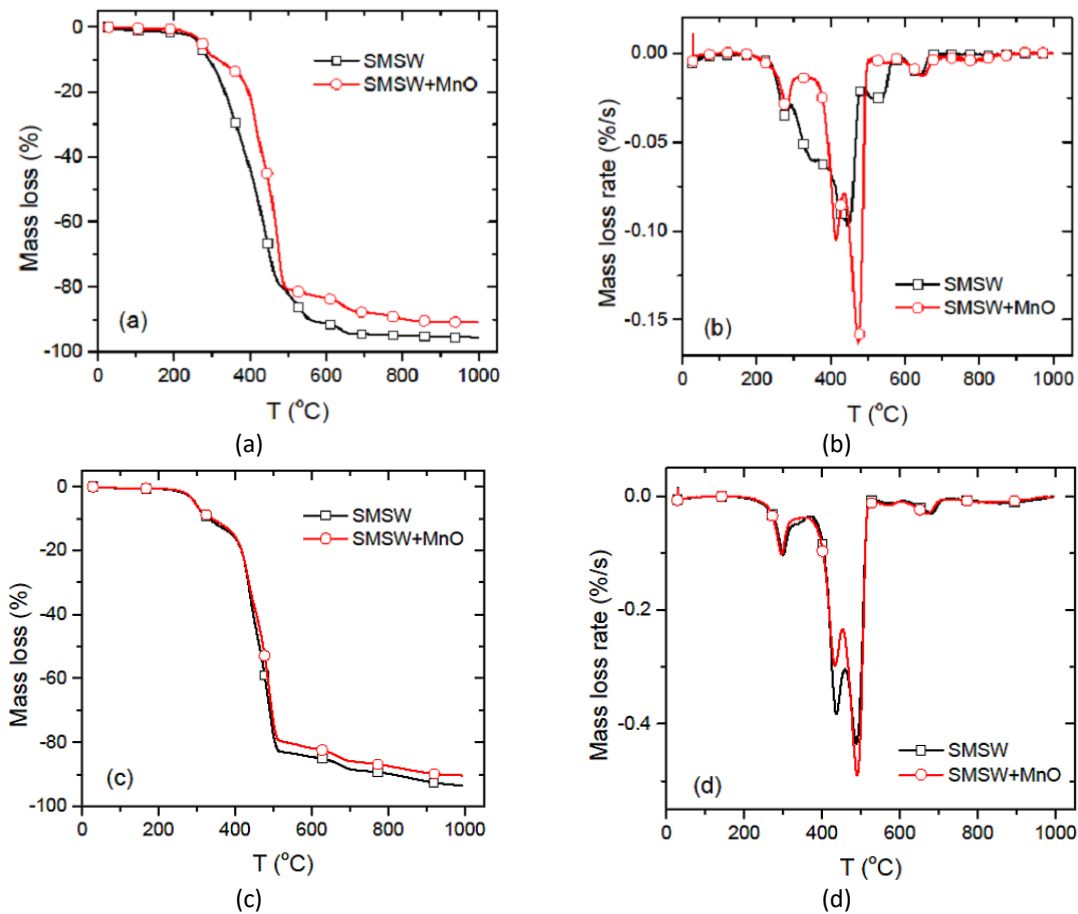


Fig. 1. (a) TG and (b) DTG of the SMSW and SMSW/5%MnO at a heating rate 10 °C/min; (c) TG and (d) DTG of the SMSW and SMSW/5%MnO at a heating rate 30 °C/min

Table 1

Characteristic parameters of the SMSW and SMSW/5%MnO at a different heating rate

β (°C/min)	Samples	Region I				Region II			
		T_{o-1} (°C)	T_{max-1} (°C)	T_{b-1} (°C)	$-DTG_{max-1}$ (%/s)	T_{o-2} (°C)	T_{max-2} (°C)	T_{b-2} (°C)	$-DTG_{max-2}$ (%/s)
10	SMSW	235.92	277.56	365.91	0.0352	365.91	446.78	482.23	0.0969
	SMSW/5%MnO	207.35	279.86	313.30	0.0307	313.30	473.05	509.70	0.1632
30	SMSW	261.52	299.68	349.95	0.1037	349.95	488.07	551.30	0.4348
	SMSW/5%MnO	243.20	295.07	341.29	0.1028	341.29	488.78	548.20	0.4903

T_o and T_b represented the onset degradation temperature and burnout temperature, respectively

T_{max} represented the temperature of the maximum rate

DTG_{max} represented maximum mass loss rate

Subscribe 1 and 2 specified the Region I and Region II, respectively

The Region II for the heating rate 10 °C/min, addition 5%MnO moved the onset combustion temperature to the lower value from 365.91 to 313.30 °C, but conversely for the burnout temperature that shifted to the higher value from 482.23 to 509.70 °C. The temperature of the maximum rate (T_{max-2}) shifted in the direction of the higher value due to the addition of 5%MnO from 446.78 to 473.05 °C. The maximum mass loss rate ($-DTG_{max-2}$) significantly increased by 68.42% from 0.0969 to 0.1632 %/s. This region was associated with the enormous degradation and

combustion of various constituents which composed the SMSW such as polypropylene (PP), polycarbonate (PC), polyethylene (PE), polystyrene (PS), polyethylene terephthalate (PET), polyamide (PA), and the second step PVC decomposition [19].

For the heating rate 30 °C/min, the similar characteristics with heating rate 10 °C/min were observed in the Region I and Region II for either SMSW and SMSW/5%MnO. However, the discrepancy between SMSW and SMSW/5%MnO in the 30 °C/min heating rate was not obviously apparent, on the contrarily with the 10 °C/min one. This phenomenon confirmed that the catalytic effect of the MnO during the combustion process would be significant in the low heating rate, regardless of whether its effect accelerative or inhibitive.

For an in-depth understanding of whether the MnO additive has significantly impacted the SMSW during combustion, the parameter Δm was presented as the following form [20-21]:

$$\Delta m = m_{SMSW/5\%MnO} - m_{SMSW} \quad (1)$$

where Δm signified the increasing mass loss of the sample after additives addition; $m_{SW/5\%MnO}$ represented the mass loss of the SMSW/5%MnO; and m_{SW} specified the mass loss of the pure SMSW at the same temperature. Figure 2 featured the incremental mass loss of the SMSW after MnO addition.

Figure 2 indicated that at a heating rate of 10 °C/min, the incremental mass loss significantly rising started from 245.76 °C and ended at 450.49 °C. This temperature range was associated with the main combustion region, in which a huge amount of the mass was combusted. It pointed out that the presence of 5%MnO into the SMSW had significantly impacted the thermal degradation of the SMSW at such as temperature range. This effect was decreased started from 450.49 °C up to 494.28 °C and then obviously apparent from 494.28 °C until to 566.26 °C.

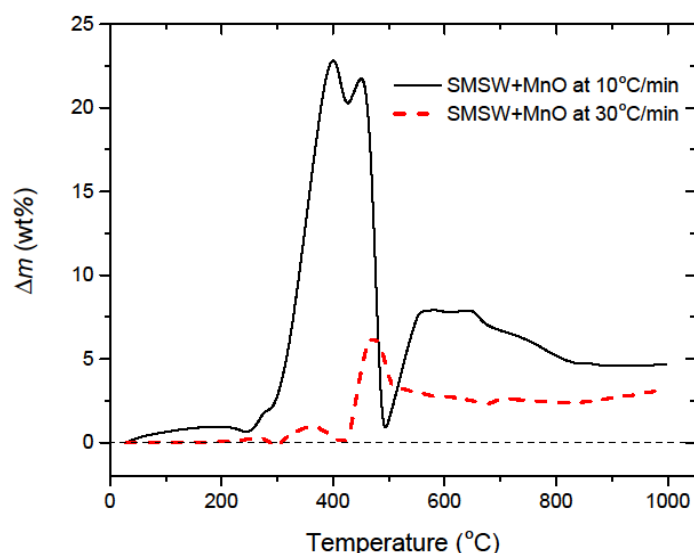


Fig. 2. The incremental mass loss of the SMSW after MnO addition

At a heating rate of 30 °C/min, the catalytic effectiveness of the MnO was lower compared to the 10 °C/min. It confirmed that the MnO additive had a more powerful effect at a lower heating rate. At heating rate 30 °C/min, the MnO noticeably more evident work at a very narrow range temperature, started from 421.29 °C and then declined at 473.03 °C. However, almost overall incremental mass

loss lines placed over the zero lines, which indicated that MnO had not inhibited the combustion process at whole temperature range, even though it was more effective for enhancing the combustion of the SMSW at only a certain temperature range. From this discussion, it could be deduced that the addition of MnO had an active effect on improving SMSW combustion.

3.2 Effects of MnO Addition on Kinetic Parameters

The adopted Coats-Redfern method for evaluating the kinetic parameters had been presented in the author's previous paper [22-23]. The experiment was performed under non-isothermal setting, where the expression of the conversion rate basically stated as follow:

$$\frac{d\alpha}{dt} = k(T)f(\alpha) \quad (2)$$

and from the Arrhenius known that:

$$k(T) = Ae^{\frac{-E}{RT}} \quad (3)$$

and function $f(\alpha)$ is:

$$f(\alpha) = (1 - \alpha)^n \quad (4)$$

where A is the frequency factor, which is a measure of the frequency of the reaction at which occurrence collisions of all molecules are taken place regardless of their energy level [24-25]. The exponential expression in Eq. (2) represents the portion of collisions that has kinetic energy adequately to bring about a reaction. Therefore, $k(T)$ is the rate constant which is representing the frequency of successful collisions [25]. E is the activation energy, considered as the energy threshold that is an obstacle that must be overcome before molecules reaction can be occurred to form products. R is the universal gas constant, and T is the absolute reaction temperature. $f(\alpha)$ signify the supposed model of the reaction mechanism and n is a specific exponent that known as the reaction order. α is lost mass fractions of the materials during thermal processes, expressed as:

$$\alpha = \frac{m_i - m_t}{m_i - m_f} \quad (5)$$

where m_i is the initial mass of the sample; m_t is the actual mass of the sample at time t ; m_f is the final mass that refers to the end of the mass event of interest.

Combining Eq. (2)-(4) and by substituting the parameter of time changes during non-isothermal processes that expressed as $dt = dT/\beta$, resulted in the basic equation for determining kinetics parameters as:

$$\frac{d\alpha}{(1-\alpha)^n} = \frac{A}{\beta} e^{\frac{-E}{RT}} dT \quad (6)$$

The Coats-Redfern is solving Eq. (6) by integrating to produce an exponential integral, and following the completion of this integral by using the Taylor series approach [26], and then linearization is performed to achieve the following expression:

$$\ln g(\alpha) = -\frac{E}{RT} + \ln \frac{AR}{\beta E} \quad (7)$$

where

$$\text{if } n = 1 \text{ then } g(\alpha) = -(\ln(1 - \alpha))/T^2$$

$$\text{if } n \neq 1 \text{ then } g(\alpha) = (1 - (1 - \alpha)^{(1-n)})/((1 - n)T^2)$$

Based on Eq. (7), the kinetic parameters were evaluated correspondingly to the characteristic temperature parameters that were listed in Table 1. The evaluation was conducted in the α interval of 0.05-0.95 at each separate temperature region. Initiated by determining any n values, the $\ln g(\alpha)$ was calculated in this α interval. Plotted the $\ln g(\alpha)$ against $1/T$ at numerous n values was performed and followed by the linear fitting processes resulted in straight lines with slopes and correlation coefficients of R^2 . The most appropriate n value means the best linear fitting is. The best linear fitting was specified by the nearby R^2 to the 1,0 value. Therefore, $R^2 - n$ were plotted to determine the most appropriate n , as shown in Figure 3(a) and 3(c). From this final n value, the final $\ln g(\alpha)$ was calculated at a given α interval and corresponding temperature and followed by plotting $\ln g(\alpha)$ against $1/T$ for gaining the final slope and intercept, as shown in Figure 3(b) and 3(d). From this final slope and intercept then the respective activation energy and frequency factor were determined, and these results were presented in Table 2.

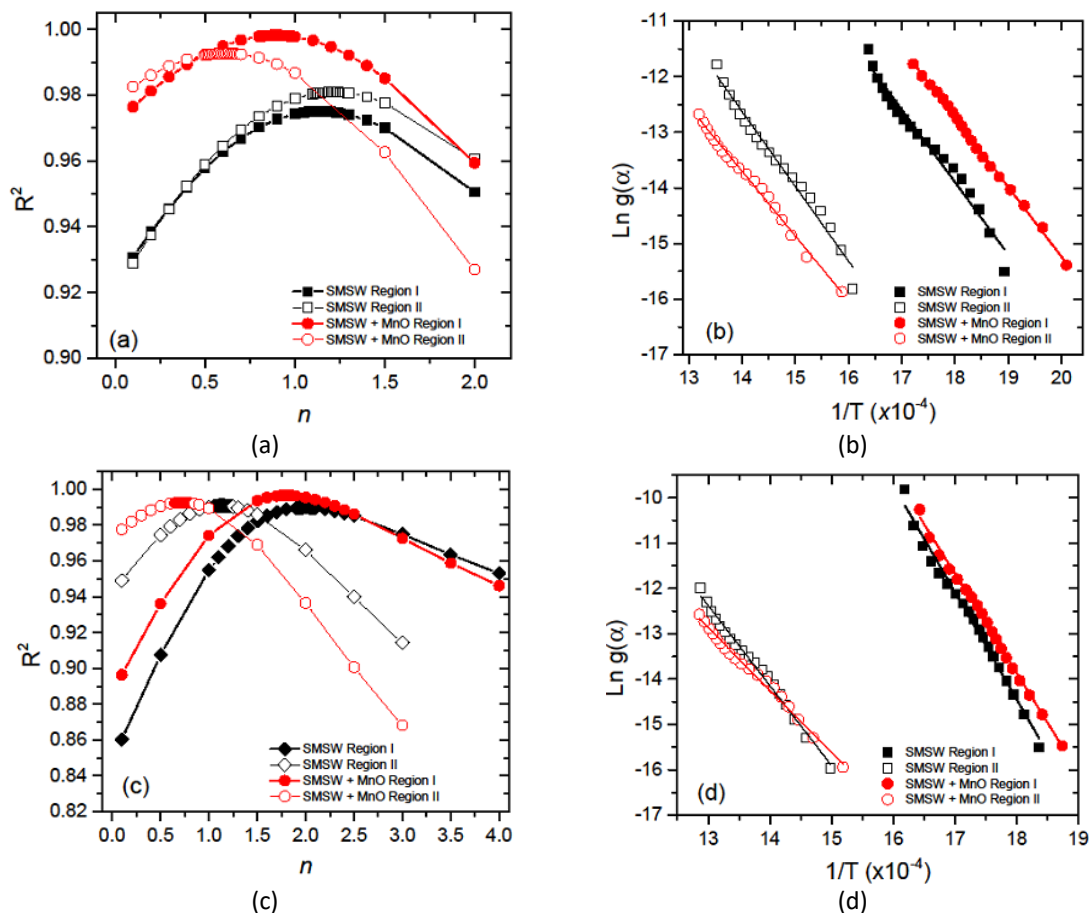


Fig. 1. (a) The $R^2 - n$ and (b) linear regression at a heating rate 10 °C/min; (c) The $R^2 - n$ and (d) linear regression at a heating rate 30 °C/min

Table 2 showed the overall correlation coefficients of activation energy were between the minimum value of 0.975 and the maximum one of 0.998, which confirmed that these kinetic evaluations had a high accurateness and therefore the results were acceptable. It could be observed from Table 2 that the presence of 5%MnO on the SMSW had decreased the activation on the whole

process. In the heating rate 10 °C/min, Region I decreased in the activation energy by 3.9% and Region II by 13.6%. Similar trends were encountered in the heating rate of 30 °C/min, where Region I declined in the activation energy by 7.3% and Region II had the most significant decrease in activation energy that was 22.3%. These results corroborated the hypothesis that the addition 5%MnO was highly promoted the combustion process toward an easier process as indicated by other aforementioned parameters stated previously.

Table 1

The kinetics parameters of the SMSW and SMSW/5% MnO combustion

β (°C/min)	Samples	Regions	Trendline equation	R^2	Kinetic Parameters		
					E (kJ/mol)	$\log A$ (min ⁻¹)	n
10	SMSW	I	$y = -13037x + 9.5715$	0.975	108.38	9.27	1.14
		II	$y = -13474x + 6.2407$	0.981	112.02	7.84	1.21
	SMSW/5%MnO	I	$y = -12527x + 9.8158$	0.998	104.15	9.36	0.90
		II	$y = -11640x + 2.5979$	0.993	96.77	6.19	0.62
30	SMSW	I	$y = -23595x + 28.028$	0.990	196.17	18.02	2.00
		II	$y = -17644x + 10.557$	0.991	146.69	10.30	1.13
	SMSW/5%MnO	I	$y = -21873x + 25.508$	0.997	181.84	16.89	1.80
		II	$y = -13709x + 4.9503$	0.993	113.97	7.76	0.74

4. Conclusions

The catalytic impact of the MnO additives during the combustion of SMSW under the two heating rates of 10 and 30 °C/min had been evaluated by using the thermogravimetric analyzer. From the TG-DTG curves could be deduced that presence 5%MnO promoted the SMSW combustion process toward the lesser temperature. DTG curves also indicated that by adding 5%MnO provoked the maximum mass loss rate toward the higher value, either in heating rate 10 or 30 °C/min. The incremental mass loss also significantly increased because of the 5%MnO addition, and more significant in the low heating rate. The activation energies in the whole region of combustion have obviously decreased due to the 5%MnO addition. These overall parameters were strengthening thought that the presence of 5%MnO has a positive impact on the SMSW combustion.

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