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ORIGINAL RESEARCH ARTICLE

Pyrolysis of municipal solid waste: Kinetics and thermodynamic parameters via Coats-Redfern method

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ABSTRACT

Renewable and sustainable energy resources are the dire need of time for environmental sustainability and to minimize the effects of global warming and climate change. The objective of this study was to investigate the thermos-kinetic parameters of municipal solid waste through pyrolysis. The sample was collected and prepared according to the American standards for test materials. Thermogravimetric analysis showed the three distinct regions, while the maximum degradation occurs in the second region within the temperature range of 230–400 ℃. A model-fitting approach using the Coats Redfern model was applied in this region to perform thermo-kinetic analysis. Based on the kinetic analysis, the D3 diffusion model showed the highest regression coefficient with an activation energy of 16–18 kJ/mole among all three diffusion models. Thermodynamic analysis showed that the pyrolysis process is endothermic, the product has more energy and a well-ordered arrangement of molecules confirmed by the positive change in enthalpy values and negative entropy values. The results demonstrate the usefulness of municipal solid waste in the creation of productive methods for converting to energy.

Keywords: waste; pyrolysis; Coats Redfern method; thermos-kinetic parameters

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1. Introduction

The exploration of cheap and easily available renewable energy resources has received greater attention due to exponentially increasing environmental problems. The excessive use of fossil fuels, especially the burning of low-quality coal[1,2] has increased the CO_2 level up to 421 ppm in the environment[3]. Climate change and global warming create negative impacts in almost every sector of human development[4,5]. To mitigate the increasing climate change effects, it is mandatory to stop the use of fossil fuels and replace them with renewable energy resources immediately[6] such as biofuel[7], green hydrogen[8] and solar energy. Several approaches have been proposed but there is a dire need to use a suitable energy mix that should be sustainable over long periods[9]. Solid fuels have been utilized since ancient times for steam generation and subsequent energy generation from the produced steam[10]. Replacing coal with biomass as a solid fuel can provide dual benefits such as solid waste management and renewable energy production. According to current practice, the solid waste is usually burned in the open atmosphere in most of the developing countries[11], which produces huge amounts of particulate matter, unburnt hydrocarbons, CO, and CO_2[12]. During winter season, in most Asian countries, there is a serious issue of photochemical smog[13] which is mainly produced by particulate matter and smoke produced by open burning and other industrial combustion processes. A proper management of solid waste and its utilization for energy production could be the better strategy to overcome the issue of carbon emissions and solid waste management[14]. There are several methods of biomass utilization such as direct combustion and pyrolysis to obtain different product like gaseous and liquid products along with the carbon-rich material that has many broad applications as a catalyst and adsorbent in many industrial applications[15]. The thermochemical biomass conversion process is also utilized for biomass conversion into different products, along with the application of different catalysts to increase the yield of selective products[16].

Pyrolysis is a process of heating the biomass at a high temperature in the absence of an oxidative environment. Usually, it takes place in three stages such as mild-, slow- and fast pyrolysis. The moisture removal starts during the mild pyrolysis at a temperature range of 200–300 ℃ depending on the volatile degradation of biomass, and it usually ends at 400–500 ℃ (called slow pyrolysis) depending on the nature of biomass and particle size. At the last stage or fast pyrolysis, the degradation of heavy molecular weight hydrocarbon at a temperature range of 700–900 ℃ leaving behind biochar which is carbon-rich, highly condensed aromatic structure with different porosity and high surface area which make it applicable as catalyst or adsorbent[17].

In our previous study, it was reported that municipal solid waste has an ideal proximate and elemental composition for being utilized as a renewable energy source[18]. The gravimetric analysis showed that it has been well degraded in the temperature range of 230–400 ℃. In this study, detailed kinetic and thermodynamic parameters for the same waste material have been calculated using model model-fitting approach to gain insight into the pyrolysis process. Thermo-kinetic parameters are very useful and provide crucial information for optimization of chemical reaction conditions and design of reactors for effective utilization of biomass towards energy conversion. In this regard, this study aims to provide basics for the effective utilization of municipal solid waste for energy production through pyrolysis or thermochemical conversion processes. Section 2 of this manuscript presents a detailed analysis of kinetics and thermodynamic parameters, and section 3 provides the understanding and significance of these parameters in the energy conversion process.

2. Materials and methodology

2.1. Sample collection and preparation

Biomass samples were collected from fifteen locations within the Chattogram City Corporation (CCC) (**Figure 1**). The specific locations of all sampling points together with sample code are presented in **Table 1**. A representative sample was collected from 15–20 different points in a quantity of 1 kg from each point, and mixed as a gross sample, which was crushed into very small pieces and dried in the open air. The dried sample was mixed thoroughly and divided into two parts several times until the final sample weight remained about 1 kg. The collected samples were dried in an oven at 40 ℃ for evaporation of surface moisture. Samples were kept in an inert oven for 16–18 h until the weight loss became constant. At the same time, the samples were crushed by using a biomass crusher. These crushed samples were sieved by using laboratory Tyler sieve shaker (model no. RX-29-10, made in the USA) to obtain a fine sample size of 250 µm. The homogenous grain size samples were then subjected to several analyses together with pyrolysis. To determine the composition of the biomass including moisture content, the volatile matter, fixed carbon content, and ash content were measured by proximate analysis according to ASTM standards. To check the elemental composition of biomass sample, elemental analysis was performed to obtain the weight percentages of carbon, hydrogen, oxygen, sulfur, and nitrogen. This data is helpful to investigate its heating value and pollutant emissions during its commercial scale application. Details about the proximate analysis, elemental analysis and thermogravimetric analysis have been provided in our previous study[18].

Table 1. Geographical locations of the selected study areas.

Sample code	Study area	Geographical location	
		Latitude	Longitude
SW-1	EPZ	22.29293	91.77662
SW-2	Agrabad	22.32445	91.81172
SW-3	Dewanhat	22.33893	91.81452
SW-4	GEC	22.35907	91.82154
SW-5	Nasirabad	22.36082	91.8233
SW-6	Muradpur	22.36597	91.82878
SW-7	Bahaddarhat	22.37027	91.84341
SW-8	Chandgaon	22.38875	91.85622
SW-9	ChawkBazar	22.35177	91.83305
SW-10	Didarmarket	22.34812	91.83842
SW-11	Anderkilla	22.34107	91.83667
SW-12	Jamalkhan	22.34581	91.83305
SW-13	New market	22.3344	91.83315
SW-14	Riazuddin Bazar	22.33653	91.8303
SW-15	Sadarghat	22.32402	91.82937

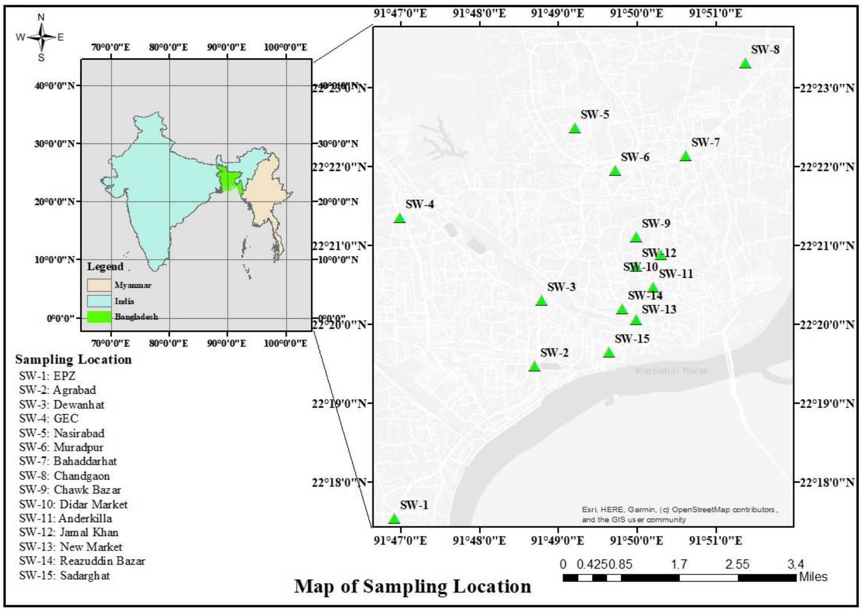

Figure 1. Sample collection spots within the study area of Chattogram City Corporation.

2.2. Kinetic analysis using Coats Redfern models

A non-isothermal model fitting technique called Coats-Redfern is used to evaluate the viability of the model-based approach and to obtain the best possible reaction model for decomposition of biomass. This Coats Redfern model is adopted here for the determination of kinetic parameters such as pre-exponential factor and activation energy of the sample. The basic equation of Coats Redfern models for the calculation of kinetic parameters is given below[19].

$$\ln\left[\frac{g(\alpha)}{T^2}\right] = \ln\frac{AR}{\beta Ea}\left(1-\frac{2RT}{Ea}\right)-\frac{Ea}{RT} \tag{1}$$

where $g(\alpha)$ is a function of different statistical models for the determination of kinetics of biomass pyrolysis, T is the absolute temperature, A is the collision frequency (pre-exponential factor), R is the universal gas constant, β is the heating rate and Ea is the activation energy.

The plot of left-hand side of Equation (1) versus $1/T$ for each reaction mechanism listed in **Table 2** yields approximately straight lines. The most suitable model for the explanation of biomass pyrolysis was selected based on the highest regression coefficient R^2 value.

Table 2. Reaction mechanism, model names with their $f(\alpha)$ and $g(\alpha)$.

Reaction mechanism	Model name	$f(\alpha)$	$g(\alpha)$
Chemical reaction order	Chemical reaction order 1 (F_1)	$1 - \alpha$	$-\ln(1 - \alpha)$
	Chemical reaction order 1.5 ($F_{1.5}$)	$(1 - \alpha)]^{3/2}$	$2[(1 - \alpha)^{-3/2} - 1]$
Diffusion	Parabolic law (D_1)	$1/2\alpha$	α^2
	Valensi Eq. (D_2)	$-[\ln(1 - \alpha)]^{-1}$	$\alpha + (1 - \alpha)\ln(1 - \alpha)$
	Ginstling-Brousshtein Eq. (D_3)	$3/2[(1 - \alpha)^{1/3} - 1]^{-1}$	$(1 - 2/3\alpha) - (1 - \alpha)^{2/3}$
Nucleation and growth	Avrami-Erofeev Eq. nucleation and growth ($N_{1.5}$)	$3(1 - \alpha)[-\ln(1 - \alpha)]^{2/3}$	$[-\ln(1 - \alpha)]^{2/3}$
	Avrami-Erofeev Eq. nucleation and growth (N_2)	$2(1 - \alpha)[-\ln(1 - \alpha)]^{1/2}$	$[-\ln(1 - \alpha)]^{1/2}$
Phase interfacial reaction	Shrinkage geometrical column (S_1)	$2(1 - \alpha)^{1/2}$	$1 - (1 - \alpha)^{1/2}$
	Shrinkage geometrical spherical (S_2)	$3(1 - \alpha)^{2/3}$	$1 - (1 - \alpha)^{1/3}$
Power law	Power law (P)	1	A

The activation energy was estimated from the slope of the line having a best fit to the experimental data. This model has been used extensively for the estimation of the kinetics of solid fuel's reaction.

2.3. Calculation of thermodynamic parameters

Thermo-gravimetric analysis also helps us in determining the thermodynamic parameters such as Gibbs free energy and entropy of the sample which was subjected to pyrolysis. Thermo-dynamic parameters can be determined based on kinetic data by using the following Equations (2)–(4).

$$\Delta H = Ea - RT \tag{2}$$

$$\Delta G = Ea + RT_{\mathrm{m}} \ln\left(\frac{K_{\mathrm{B}}T}{hA}\right) \tag{3}$$

$$\Delta S = \frac{\Delta \mathrm{H} - \Delta \mathrm{G}}{T_{\mathrm{m}}} \tag{4}$$

where, K_{B} is the Boltzmann constant which is equal to 1.381×10^{-23} m^2 kg/s^{-2} K^{-1}, T_{m} is the maximum temperature at which maximum decomposition occurs, h is the planks constant which is equal to 6.626×10^{-34} m^2 kg/s and R is a universal gas constant which is equal to 0.008314 kJ/mol K.

3. Results and discussion

Thermogravimetric analysis was performed at 5 ℃/min from room temperature to 900 ℃ in an inert atmosphere with a constant supply of nitrogen gas. For the proper evaporation of the moisture 10 min temperature pause was given at 110 ℃ and 30 min temperature pause was given at 900 ℃ for complete de-volatilization. The degradation pattern was divided into three distinct stages, the first one was the loss of moisture, second one corresponds to the loss of volatile fraction, which is an actual degradation of the biomass corresponding to a huge fraction of biomass degradation in this region. The third region corresponded to the degradation and breakdown of heavy molecular weight condensed structured hydrocarbons into small units and their subsequent degradation. The actual biomass pyrolysis started in the second degradation region, and it gives us important information about the self-ignition point of biomass for its effective storage and transportation. The amount of volatile matter present in the biomass sample is inversely proportional to the degradation starting point. So, it's very important to investigate the thermal behavior of the biomass in this temperature range for its effective storage, transportation, thermal conversion, and gasification process. A model-fitting approach using the Coats Redfern method was applied in the second degradation region to investigate the appropriate degradation model for MSW degradation. It has been proven from the published literature that most of solid fuel pyrolysis degradation reaction falls into 5 different reaction models listed in **Table 2**. A suitable degradation reaction model was selected based on the highest regression coefficient. Pre-exponential factor (collision frequency) plays an important role in chemical reactions, it was also calculated from the intercept of the line drawn by the Coats-Redfern method. All the graphs drawn for each model using the Coats Redfern method have been presented in **Figures 2** and **Figure 3** along with their respective equations of line and linear regression coefficient R^2.

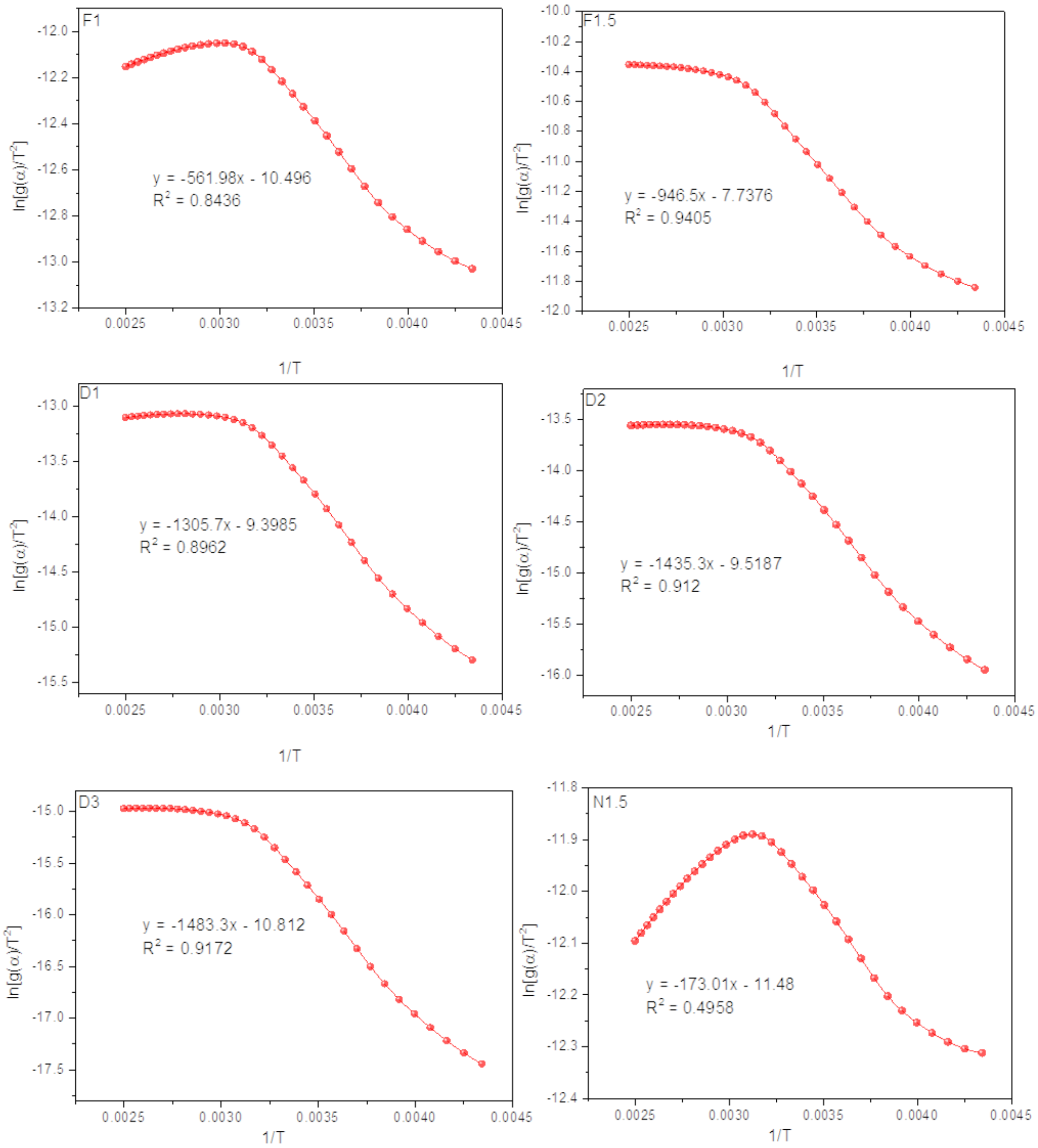

Figure 2. Linear plots of different models fitted in Coats Redfern method chemical reaction order 1 (F1), chemical reaction order 1.5 (F1.5), parabolic law (D1), Valensi equation (D2), Ginstling-Brousshtein equation (D3) and Avrami-Erofeev equation nucleation and growth (N1.5).

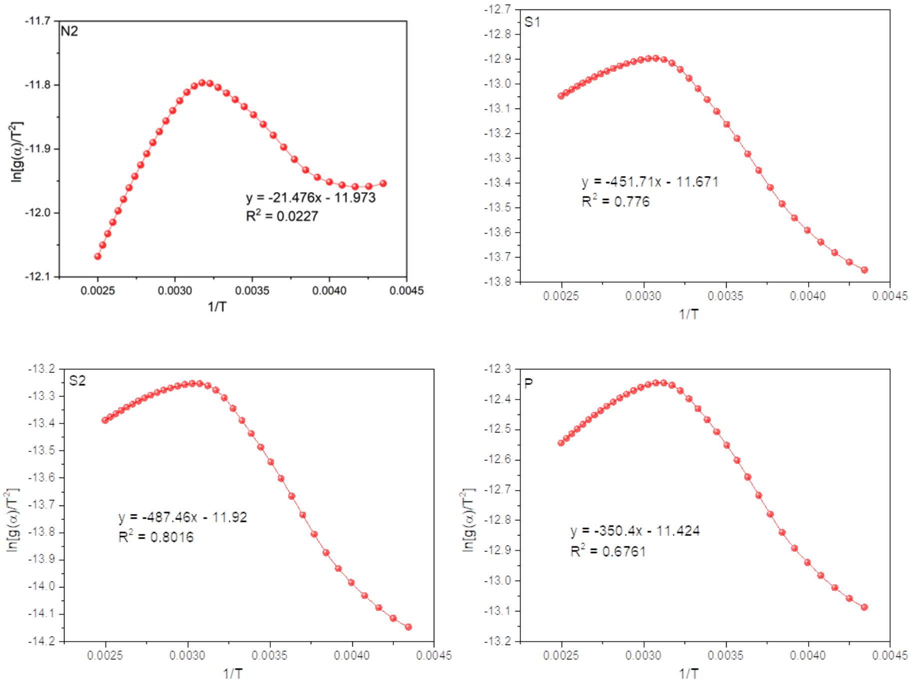

Figure 3. Linear plots of different models fitted in Coats Redfern method Avrami-Erofeev equation nucleation and growth (N2), shrinkage geometrical column (S1), shrinkage geometrical spherical (S2) and power law (P).

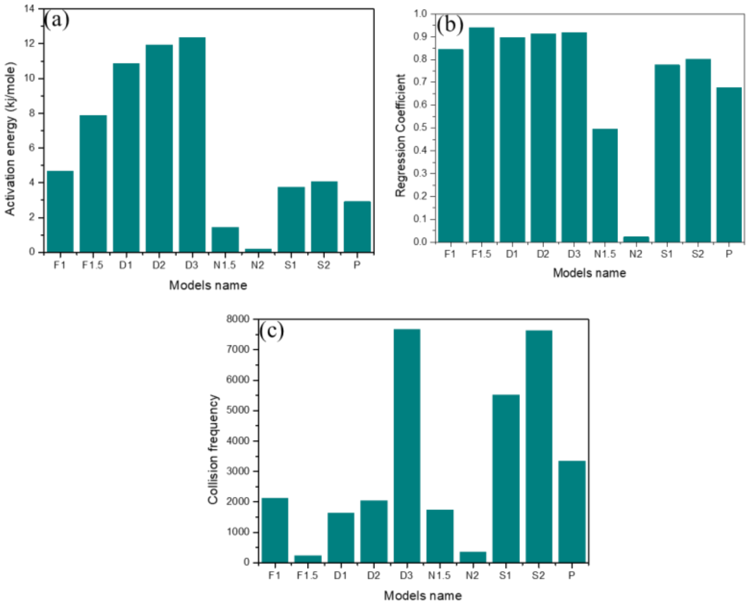

Figure 4. Best fitted model for the thermal degradation of municipal solid waste and their respective kinetics parameter, **(a)** activation energy (kJ/mole); **(b)** regression coefficient; **(c)** collision frequency.

3.1. Kinetic analysis

The kinetic analysis consists of the activation energy calculated from the slop of the plot ln(*g*(*a*)/*T*2 vs. 1/*T*, the regression coefficient of the linear plot of coats Redfern method fitted with different models and pre-exponential factor (collision frequency) estimated from the intercept of the linear plot as presented in **Figure 4**. Ten different models were applied and among all of them chemical reaction order (F1) and diffusion model (D1, D2 and D3) showed the highest regression coefficient indicating towards the suitable reaction model for the MSW pyrolysis. Chemical reaction order (1.5) and nucleation model (N2) showed poor linear fitting (regression coefficient) indicating towards an unsuitable model for MSW pyrolysis. Comparative observation on the values of activation energies and collision frequencies showed a perfect theoretical trend, with increasing collision frequencies the activation energies decrease. The maximum activation energy values were shown by diffusion models in the range between 16–18 kJ/mole with the highest regression coefficient. Nucleation models and spherical models including power law showed minimum magnitude of activation energy with higher collision frequencies and better regression coefficients of spherical models and power law but nucleation model (N2) showed a poor regression coefficient. Collision frequency values change between 50–600 per minute except chemical reaction order (F1.5) which showed almost negligible frequency with a poor linear regression coefficient. Based on the observation from kinetic analysis, it can be concluded that chemical reaction order (F1.5) is a highly inappropriate model for MSW degradation. The diffusion models, especially D3 could be the most suitable model for the explanation of the MSW pyrolysis process mechanism and other specifications for its commercial-scale application. The activation energy of MSW pyrolysis is very low as compared to other biomasses[20,21] indicating that it has a good amount of volatile matter suggesting towards its huge potential for practical applications.

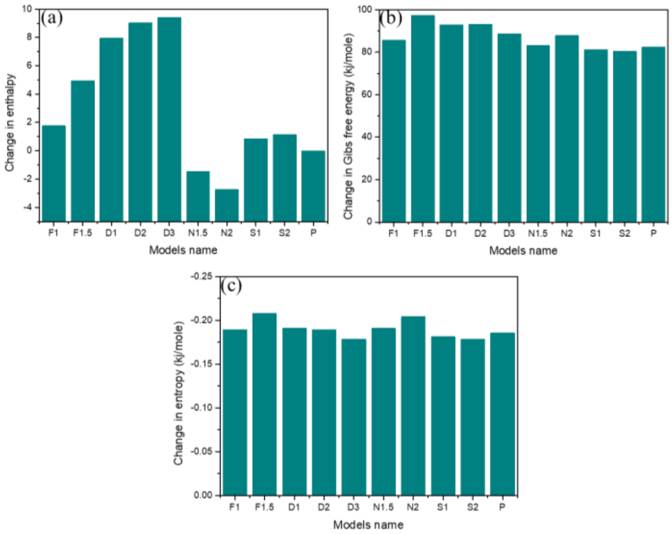

Figure 5. Best fitted model for the thermal degradation of municipal solid waste and their respective thermodynamic parameters, **(a)** change in enthalpy (kJ/mole); **(b)** change in Gibbs free energy; **(c)** change in entropy.

3.2. Thermodynamic analysis

The thermodynamic analysis consists of the measurement of the change in enthalpy of reaction and product, change in Gibbs free energy and change of entropy. Enthalpy in the measurement of the heat changes during the chemical reaction and provide some information about the exothermic and endothermic nature of the chemical reaction. In case of MSW pyrolysis, the change in enthalpy varies between 10–12 kJ/mole for high linear regression coefficient models. While the poor regression, coefficient models showed the magnitude of change in enthalpy between −2 to +2 kJ/mole as depicted by **Figure 5.** The negative value of the enthalpy showed the reaction is exothermic, and the product has less energy as compared to the reaction which is not true in biomass pyrolysis. The primary objective of pyrolysis is to achieve a high-energy-density alternative fuel. So, the negative value is highly inappropriate and positive values showed that the reaction is endothermic, and the product has higher energy as compared to the reactant which could be true, and it is also well correlating with our kinetic analysis. Hence, the enthalpy of the MSW pyrolysis is endothermic in nature and the product has more energy as compared to the reactant. Gibbs free energy is a measurement of available energy for chemical reactions at given temperature and pressure conditions. It indicates whether the reaction will be spontaneous or non-spontaneous in nature. The magnitude of the Gibbs free energy varied between 100–120 kJ/mole for all reaction models suggesting that the pyrolysis process is non-spontaneous, and heat is required from an external source to proceed with the chemical reaction. Entropy is the degree of the disorder of chemical species at given temperature and pressure conditions. In the case of MSW pyrolysis, the entropy showed negative values for all the models applied for the investigation of the reaction mechanism of MSW pyrolysis. A negative value of entropy showed the product is in a more ordered form as compared to the reactant. During the pyrolysis process at high temperatures there are a lot of free radicals available and due to fast kinetics, they can recombine with their desired one to minimize their energy and to gain stability.

3.3. Comparison with existing literature worldwide

A review of literature reveals that a considerable number of studies were performed to analyze the kinetic and thermodynamic properties of different biomass to understand their thermal behavior. As for instance, Naqvi et al.[19] investigated the kinetic and thermal properties of sewage sludge pyrolysis. Hernández et al.[22] employed numerous first-order reaction models and TGA-FTIR to examine the degradation of sewage sludge in different gases. The kinetics of undigested sewage sludge pyrolysis were investigated using the Friedman technique. They calculated a reaction order that was remarkably high, and employed the Coats-Redfern method to determine a suitable arbitrary reaction order in order to examine the sludge's pyrolytic activity[23]. The kinetic (activation energy and frequency factor) and thermodynamic (ΔG, ΔH, and ΔS) parameters of date seed pyrolysis using TGA at various heating rates (10, 20, 30, and 40 ℃/min) and using diverse reaction processes were investigated by employing the Coats-Redfern technique. The devolatilization region's average activation energy value was calculated to be 186 kJ/mol[24]. Based on the previous data reported in the literature, we use a similar methodology to investigate the thermal behavior of another organic waste sample which has never been done before, and it shows a very promising result for commercial scale applications.

3.4. Limitations

This study has limitation of experimental validation of calculated parameters through different temperature programmed and control detecting species like thermogravimetric analysis and mass spectrometer analysis (TGA-MS) and other in-situ analysis to investigate the real degradation mechanism of biomass pyrolysis with its product distribution. Diffuse reflectance infrared spectroscopy (DRIFTS) analysis is another promising analytical technique to investigate the different intermediates of biomass pyrolysis. In current research work, the above-mentioned analysis could be performed in future which may help in both parameters and product selectivity towards the commercial scale production of waste to energy.

4. Conclusion

This study performed the kinetic and thermodynamic analysis of MSW pyrolysis using the Coats Redfern method. The maximum degradation of MSW occurred in the temperature range of 230–400 ℃. A model-fitting approach was applied in this temperature range using the Coats Redfern method. A suitable model for the explanation of the approximate mechanism of MSW pyrolysis was selected based on the highest regression coefficient. Diffusion models showed higher regression coefficients among all models, particularly D3 showed the highest regression coefficient. The magnitude of activation energy was estimated between 18–20 kJ/mole and thermodynamic parameters indicated that the product is in a higher energy state as compared to the reactant with well-ordered structure indicated by the negative value of entropy. Further, Gibbs' free energy values confirmed the non-spontaneous nature of the pyrolysis process. Thermos-kinetic analysis of the MSW pyrolysis showed that it has a huge scope for low-temperature pyrolysis or gasification process for its effective utilization as an alternative renewable energy source.

Author contributions

Conceptualization, AMMH, MUK and FY; methodology, AMMH, MUK and FY; software, MA and MRU; validation, MA, TH and RS; formal analysis, AMMH, TH, MA and MRU; investigation, MA, RS and

MRU; data curation, AMMH; writing—original draft preparation, AMMH; writing—review and editing, MAA, MUK and FY; visualization; supervision, FY. All authors have read and agreed to the published version of the manuscript.

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Conflict of interest

The authors declare no conflict of interest.

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