

Kinetic Model Development for Decomposition of Mixed Waste Plastic

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Abstract : High and low density polyethylene, HDPE and LDPE; polypropylene, PP; polystyrene, PS; and polyethyleneterephthalate, PET) make up the majority of municipal plastic waste (MPW). The goal of this study was to create a kinetic model to apply thermogravimetric analysis to uncover the real reaction mechanism of mixed waste plastic (TGA). To determine the mechanism of MPW pyrolysis, a variety of techniques including Kissinger, Akahira-Sunose, KAS, Malek, and linear model fitting were used. The results were evaluated using experimental data from TGA. Five types of waste plastic waste decomposition on different setpoint (10°C/min & 20 °C/min), temperature of 30°C to 530°C and the sample size 1-4 mm. in the model fitting method we use the Criado method. As a result of the fluctuation in apparent activation energy with conversion and kinetic model with heating rate, the results demonstrated that a complicated process, rather than a simple 1st order, occurs during the breakdown.

Key words: Mixed waste plastic, Thermogravimetric analysis, Kinetic parameters, Decomposition model etc.

1. Introduction

General Background:

Plastic trash production has risen as a result of the significant increase in plastic consumption. The majority (up to 60%) of India's overall plastics consumption is made up of polyolefins. With 52% of overall consumption, the packaging business leads all other industries in the use of plastics, followed by consumer goods and the construction sector. In the municipal solid waste (MSW) stream, waste plastics mostly consist of polystyrene (PS), polypropylene (PP), high density polyethylene (HDPE), low density polyethylene (LDPE), and polyethylene terephthalate (PET). Researchers are looking at alternative techniques including tertiary or chemical recycling, which includes pyrolyzing plastics into petroleum fuel, in response to the growing environmental concern brought on by the high rate of pollution from incinerators and decreasing disposal sites.

Kinetic modelling is a crucial tool for the objective design of recycling processes, but it is impossible to explicitly describe the majority of fundamental step reactions as a result of quantitative rate constants, and manual creation of models with the required complexity takes a long time. Due to its ease of use and the informative data provided by a thermogram, thermo-gravimetric analysis (TGA) is a commonly used technique to examine the reaction kinetics for pyrolysis & kinetic parameters. When it comes to reactor design and scaling up of industrially feasible processes, the behaviour of plastics during thermal degradation as well as its kinetics is crucial in determining the decomposition operation.

2018 saw the production of about 360 million tonnes of plastic worldwide. Everyday living and industrial operations are made more convenient by the widespread use of plastic items. But some single-use plastic goods, especially those used in the shipping and e-commerce sectors, decompose poorly when they are recycled, posing a major threat to both human health and the existence of marine life unless a method of cleaning them is devised. The various polymers during breakdown are important since the majority of plastic wastes are naturally made up of a mixture of several plastic types. The location and type of repeating units, the types of intramolecular forces, the degree of symmetry and homogeneity in molecular structure, and many other aspects all affect the enormous range of physico- chemical properties of polymers.

Kruse et al. combined the different component models to create a mechanistic model of the binary degradation of polystyrene (PS) and polypropylene (PP). Researchers looked at how well polystyrene (PS) and polypropylene (PP) mixed in the melt and discovered that if the mixing scale was low, each polymer's breakdown acted differently from the other. Conversely, a copyrolysis with partial contacts occurs when the mixing reaches the molecular size. Once more, Westerhout and co. The goal of the current study is the development of a kinetic model for the breakdown of mixed waste plastic, which includes combinations of PS, PP, HDPE, LDPE, and PET waste plastics. On the basis of kinetic model easily desing the reactor in industry.

Origin of problem:

Till now, Kinetic model not delvelop of mixed waste plastic (PP, PS, PET, HDPE & LDPE).

Objective of present work:

The study of kinetic model development for degradation of mixed waste plastic is the project's main goal. High density polyethylene, low density polyethylene, polypropylene, polyethylene terephthalate, and polystyrene are among the five types of plastics that make up mixed waste plastics.

2. Literature Review

Statistics of waste generation of plastics:

Due to the inability of all types of plastic trash to be recycled, it collected in open drains, low-lying areas, riverbanks, coastal areas, seashores, etc. Thus, most of the waste plastics produced in this manner. In India's main cities, 3501 TPD of plastic waste (PW) is produced.

State	Waste generation in tons/annum
Andhra Pradesh	243820
Chhattisgarh	5840
Goa	164250
Gujarat	251796.65
Haryana	55480
Himachal Pradesh	106.72
J&K	11748
Jharkhand	81030
Karnataka	77247

Kerala	50370
Maharashtra	10950
Madhya Pradesh	23400
Manipur	4380
Meghalaya	4599
Mizoram	1682.50
Tamil Nadu	205724.95
Sikkim	1668.05
Tripura	10950
Delhi	251850
Chandigarh	4964

Table 2.1: State-wise waste generation in India








Table 2.1 above displays the amount of waste plastic produced in each Indian state. According to this information, the state of Delhi in India produces the most waste plastic. Gujarat produces the second-highest amount of plastic waste after Delhi.

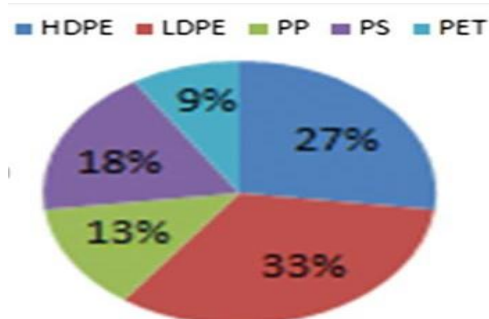
Table 2.2: Gujarat's largest cities' production of plastic waste

City	Total municipal plastic waste	Plastic Waste	Plastic Waste (Tones per day)
Dwarka	18	8.05	1.45
Gandhinagar	97	4.80	4.66
Rajkot	230	6.92	15.93
Vadodara	600	4.56	27.41
Surat	1200	12.46	149.62
Ahemdabad	2300	10.5	241.50

Table 2.2 above displays Gujarat's city-by-city generation of plastic waste. Ahmedabad and Surat are the Gujarati cities with the highest waste plastic generation rates among these big cities and metro areas.

Table 2.3: Types of waste plastics

Mark	Type	Recyclable	Abbreviation	Description & Common uses
	1	Yes	PET	Polyethylene Terephthalate Beverages.
	2	Yes	HDPE	Milk, detergent & oil bottles, toys, containers used outside, parts and plastic bags.
	3	Yes, But not common	PVC	Food wrap, vegetable oil bottles, blister packages or automotive parts.
	4	Yes	LDPE	Many plastic bags, shrink-wraps, garment bags or containers.
	5	Yes	PP	Refrigerated containers, some bags, most bottle tops, some carpets, and some food wrap.
	6	Yes, But not common	PS	Through away utensils, protective packing.
	7	Some	OTHER	Usually layered or mixed plastic.



In this figure-2.1, it's shows the composition of five types waste plastic. the composition of high density polyethelene 27%, low density polyethelene 33%, polypropylene 13%, polystyrene 18% & polyethelene terephthalate.

Table 2.3: India's city-level plastic waste composition in kg/MT

City	PET	HDPE/LDPE	PVC	PP	PS	OTHER	Total
Indor	4.92	40.52	3.91	27.86	6.44	4.39	88.05
Kolkata	1.79	102.98	2.52	2.01	3.97	2.8	116.09
Pune	0.53	74.33	0.35	1.40	1.32	0.02	77.96
Hyderabad	5.22	29.93	3.41	3.64	0	5.24	47.46
Bengaluru	1.52	79.3	0.87	0.77	0.76	1.61	84.83
Chennai	3.90	80.12	2.62	5.67	2.42	0.67	95.42
Delhi	3.17	77.24	1.44	6.21	9.65	3.71	101.44
Kochi	11.92	43.03	1.53	2.59	2.53	1.26	62.88
Patna	1.46	37.33	1.56	7.32	4.15	5.41	57.25
Mumbai	3.51	46.65	2.22	3.66	2.095	4.64	62.81

Table 2.4: Gujarat's city-level plastic waste composition in kg/MT

City	PET	HDPE/LDPE	PVC	PP	PS	OTHER	Total
Ahmedabad	0.67	92.41	0.82	6.75	4.40	0	105.07
Vadodara	0.42	35.70	1.76	0.64	3.88	3.28	45.69
Rajkot	7.64	42.91	3.55	1.17	2.14	11.85	69.28
Surat	1.25	117.8	0.90	0.68	1.39	2.64	124.68
Gandhinagar	0.77	38.35	0.944	1.52	6.01	0.46	48.06
Dwarka	12.40	45.81	3.66	3.39	8.38	8.52	80.79

According to this data, polypropylene and polystyrene are the next most prevalent types of waste plastic after high-density polyethylene (HDPE) and low-density polyethylene (LDPE).

Table 2.5: Vadodara's plastic waste day by day in kg/MT

Day	PET	HDPE,LDPE	PP	PS	Total
Day-1	0	48.9	0.565	2.97	52.435
Day-2	0.27	30.11	0.64	4.78	35.8
Day-3	0.988	28.096	0.704	3.90	33.688
Day-4	0.42	35.70	0.64	3.883	40.643

Theoretical considerations:

The waste plastic reaction's global kinetics model can be expressed as follows:

$$\frac{d\alpha}{dt} = k(T)f(\alpha) \quad (1)$$

Where,

T, the process's temperature (K) = the reaction model

α , The degree of conversion is demonstrated by ,

$$\alpha = \frac{m_0 - m_t}{m_0 - m_f} \quad (2)$$

m_0 , m_t & m_f are, respectively, the sample's initial mass, mass at time 't,' and ultimate mass. K, the reaction rate constant, was greatly influenced by temperature.

Many theorems can be used to predict how the k depends on temperature. The Arrhenius equation, which is typically utilised, is written as

$$k(T) = A \exp \left(-\frac{E}{RT} \right) \quad (3)$$

From equation (1) & (3)

The standard rate expression will take the form of

$$\frac{d\alpha}{dt} = A \exp \left(-\frac{E\alpha}{RT} \right) f(\alpha) \quad (4)$$

The degree of conversion varies with the procedure's time and temperature because the pyrolysis process is not isothermal. A parameter that related to the process's heating rate is introduced to $\beta = \frac{dT}{dt}$ lessen the complexity of the preceding mathematical equation.

$$\frac{d\alpha}{dT} = \frac{A}{\beta} \exp \left(-\frac{E\alpha}{RT} \right) f(\alpha) \quad (5)$$

Friedman Method (FR):

By taking the logarithm on both sides of the equation, the integral iso-conversional approach is obtained.

$$\ln\left(\frac{d\alpha}{dt}\right) = \ln(Af(\alpha)) - \frac{E_a}{RT} \quad (6)$$

The value of activation energy can be obtained by plotting $\ln\left(\frac{d\alpha}{dt}\right)$ against $\left(\frac{1}{T}\right)$ over the complete range of conversion.

Kissinger Method:

Without being aware of the reaction mechanism, this method enables determination of the kinetic parameters of solid-state reactions. Kissinger created a non-isothermal model-free method that eliminates the requirement to compute E_a for each conversion value when evaluating kinetic parameters. Through the use of a series of trials with various heating rates (β), where T_m is the temperature peak of the DTG curve, this method enables determining

the value of activation energy from a plot of $\ln\left(\frac{\beta}{T_m^2}\right)$ against $\frac{1}{T_m}$. The formula is as follow,

$$\ln\left(\frac{\beta}{T_m^2}\right) = \ln\left(\frac{AR}{E}\right) - \frac{E}{RT_m} \quad (7)$$

The plot's slope, which equals $-\frac{E}{R}$, can be used to determine the activation energy E_a .

Kissinger Akahira Sunise Method (KAS):

An integral isoconversional method is the Kissinger Akahira Sunose (KAS) method. The integral form of equation (5) is as follows:

$$g(\alpha) = \frac{\alpha}{f(\alpha)} = A \int_{T_0}^T \exp\left(-\frac{E_a}{RT}\right) dT \quad (8)$$

The integral in equation is evaluated using the Coats Redfern approximation (8),

$$\ln\left(\frac{E}{RT^2}\right) = \ln\left(\frac{A}{f(\alpha)}\right) - \frac{E_a}{RT} \quad (9)$$

This approximation yields the KAS method's representative form, which is

$$\ln\left(\frac{\beta}{T_m^2}\right) = \ln\left(\frac{A}{f(\alpha)}\right) - \frac{E_a}{RT_m} \quad (10)$$

β it is possible to computer the activation energy (E_a) from a plot of $\ln(\frac{1}{T^2})$ against $(\frac{1}{T})$.

Flynn Wall Ozawa method (FWO):

In this method, the temperature integral is evaluated using Doyle's approximation, as shownbelow.

$$p(\frac{E_a}{RT}) \cong -5.331 - 1.052(-\frac{E_a}{RT}) \quad (11)$$

As a result, the final FWO method expression is as follows:

$$\ln(\beta) = \ln(\frac{AR}{g(\alpha)}) - 5.331 - 1.052(\frac{E_a}{RT}) \quad (12)$$

A straight line can be drawn from a plot of $\ln(\beta)$ vs $(\frac{1}{T})$ to get the activation energy (E_a).

Prediction of decomposition model

Criado Method:

By creating master plots $Z(\alpha)$, this technique can be utilised to identify the kinetics of how a solid-state decomposition happens during the course of the process. To clarify a kinetic model for the reaction, a comparison is done between a theoretical estimate of master plots and its experimental counterpart.

$$Z(\alpha) = f(\alpha)g(\alpha) \quad (13)$$

$$Z(\alpha) = \frac{d\alpha}{dT} (\frac{E_a}{RT}) \exp \frac{E_a}{RT} p(x) \quad (14)$$

The theoretical estimate of the master plot is represented by the symbols $f(\alpha)$ and $g(\alpha)$, which stand for the reaction model and integral form of the reaction model in equation (13). Table 3 shows the reaction model for solid-state kinetics and the appropriate integral form.

Equation (14)'s factor $p(x)$ defines the temperature integral determined by Senum- approximation Yang's to the fourth degree.

$$p(x) = \frac{e^{-x}}{(x^3 + 18x^2 + 86x + 96)} \quad (15)$$

Where, $x = \frac{E_a}{RT}$

Coats-Redfern Method:

A crucial method is the Coats-Redfern method, which uses the thermal degradation mechanism. Its model equation is shown as follows.

$$\ln \frac{g(\alpha)}{T^2} = \ln \frac{AR}{\beta E} - \frac{E}{RT} \quad (16)$$

Table 2.7: The model information

model	differential form $f(\alpha) = 1/k \, d\alpha/dt$	integral form $g(\alpha) = kt$
nucleation models		
power law (P2)	$2\alpha^{1/2}$	$\alpha^{1/2}$
power law (P3)	$3\alpha^{2/3}$	$\alpha^{1/3}$
power law (P4)	$4\alpha^{3/4}$	$\alpha^{1/4}$
Avrami–Erofeev (A2)	$2(1-\alpha)[- \ln(1-\alpha)]^{1/2}$	$[- \ln(1-\alpha)]^{1/2}$
Avrami–Erofeev (A3)	$3(1-\alpha)[- \ln(1-\alpha)]^{2/3}$	$[- \ln(1-\alpha)]^{1/3}$
Avrami–Erofeev (A4)	$4(1-\alpha)[- \ln(1-\alpha)]^{3/4}$	$[- \ln(1-\alpha)]^{1/4}$
Prout–Tompkins (B1)	$\alpha(1-\alpha)$	$\ln[\alpha/(1-\alpha)] + c$
geometrical contraction models		
contracting area (R2)	$2(1-\alpha)^{1/2}$	$1 - (1-\alpha)^{1/2}$
contracting volume (R3)	$3(1-\alpha)^{2/3}$	$1 - (1-\alpha)^{1/3}$
diffusion models		
1-D diffusion (D1)	$1/(2\alpha)$	α^2
2-D diffusion (D2)	$-[1/\ln(1-\alpha)]$	$((1-\alpha)\ln(1-\alpha)) + \alpha$
3-D diffusion–Jander (D3)	$[3(1-\alpha)^{2/3}]/[2(1-(1-\alpha)^{1/3})]$	$(1-(1-\alpha)^{1/3})^2$
Ginstling–Brounshtein (D4)	$3/[2((1-\alpha)^{-1/3}-1)]$	$1 - (2/3)\alpha - (1-\alpha)^{2/3}$
reaction-order models		
zero-order (F0/R1)	1	α
first-order (F1)	$(1-\alpha)$	$-\ln(1-\alpha)$
second-order (F2)	$(1-\alpha)^2$	$[1/(1-\alpha)] - 1$
third-order (F3)	$(1-\alpha)^3$	$(1/2)[(1-\alpha)^{-2} - 1]$

3. Experimental

Thermogravimetric Analysis

To forecast the kinetics of solid state processes, thermogravimetric analysis is frequently used. The sample (of mg size) is immediately heated to the specified temperature in TGA, and its weight is tracked while the sample undergoes thermal decomposition. However, because of the inter-particle heat transfer and the time lag in arriving, this isothermal technique is the one that frequently occurs. Calculate temperature using the average temperature (room temperature). In order to characterize the thermal behaviour of particles, a different method known as non-isothermal precise with constant heating rate is used. The basic plots Temperature vs. weight loss (%) and Rate vs. Temperature are produced by the TGA investigation, and various processing techniques are designed based on their kinetics.

The TGA method is consequently beneficial for figuring out the order of reactions, kinetic parameters like activation energy and frequency factor, rate of decomposition, kinetic model, and thermal parameters like maximum decomposition temperature and characteristic temperature.

Kinetic characteristics like activation energy and the pre-exponential factor are computed using TGA curves. Since the values of the activation energy and pre-exponential factor vary when other process elements, such as the heating rate, inert flow, starting weight of the sample, and molecular weight of the sample, change, the choice of these parameters has also changed the kinetics of the reaction.

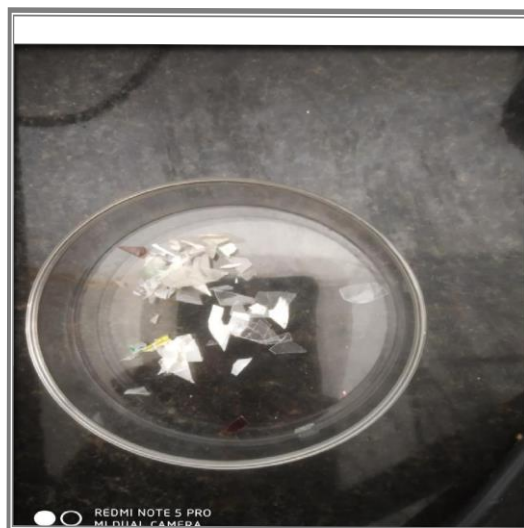
Sample preparation

The materials were crushed, then dried and sieved. For the analyses, the proportions of particles less than 2 mm were chosen. 100 mg is the weight of each sample. Figure 2.1 provides the content (in mg) of all five plastics, which is comparable to the composition of the waste plastic generated in Vadodara City.

Table-3.1: Mixed plastic composition (in mg) for TGA analysis

HDPE	LDPE	PP	PS	PET
27	33	13	18	9

Figure-3.1 100mg sample



Result and discussion

Thermogravimetric analysis

Five distinct heating speeds (10 and 20 °C/min) and a temperature range of 30 to 530 °C were used in the TGA studies. At SICART-Anand Vadodara, experiments were conducted on the thermogravimetric analyzer PERKIN ELEMER Thermal Analysis. The two curves were created from the TGA analysis using the weight loss (%) and temperature data at various heating rates (10, 20 °C). The TGA curves are displayed in Figures 1 and 2.

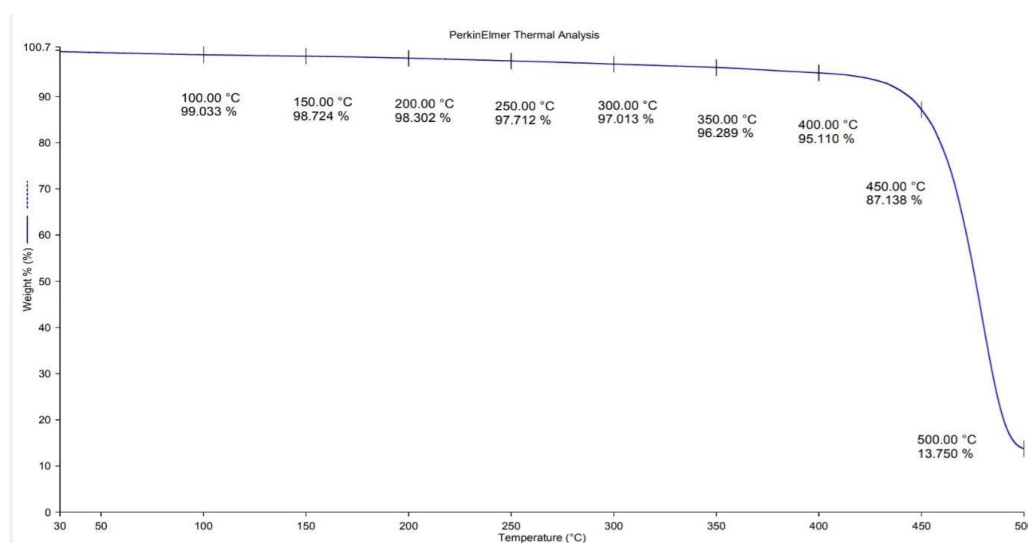


Figure 4.1 Temp vs weight (10°C/min)

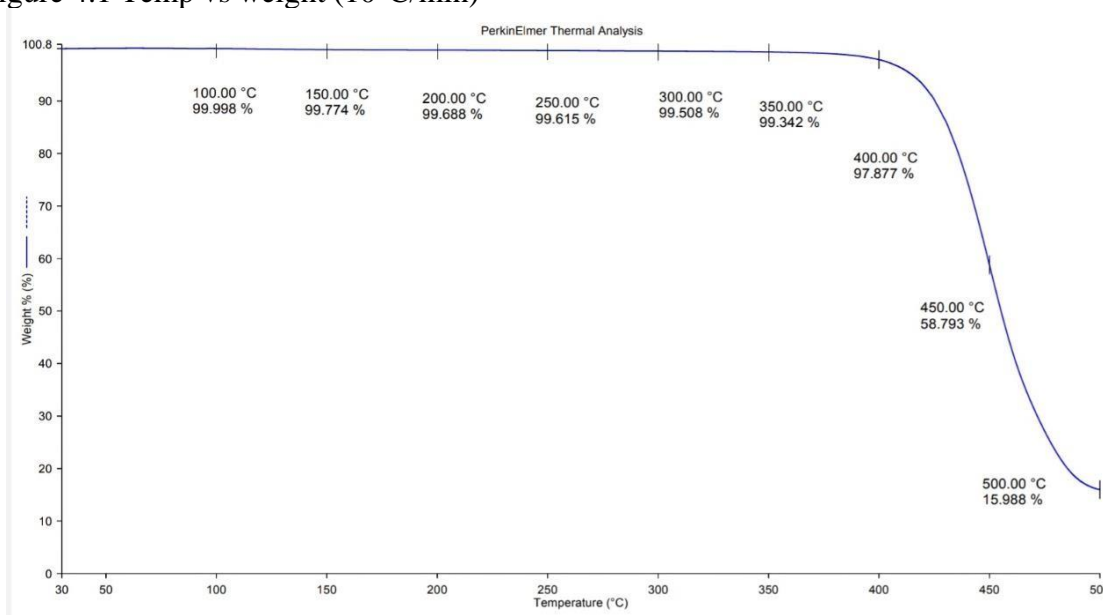


Figure 4.2- Temp vs weight (20°C/min)

We need to know the real temperature range for plastic degradation from the TGA analysis. The varying temperatures during the decomposition are causing weight loss. Looking at Fig.1 the temperature range demonstrated for the maximum weight losses at various heating rates for the mixed waste plastic is in Table 4.1.

Table 4.1 Maximum weight losses at various heating rates

Heating Rates	Maximum decomposition in a temperature range
10	422 °C to 502 °C
20	375 °C to 510 °C

The obtained data from the TGA, such as weight loss vs. temperature, is adequately modified for interpretation into kinetic triplets (E_a , n and A). These kinetic data allow us to determine which model best describes this breakdown reaction.

Evaluation of activation energy from iso-conversional methods

The activation energies were computed using the Kissinger, Flynn-Wall-Ozawa (FWO), Kissinger-Akahira-Sunose (KAS), and Friedman (FR) methodologies, taking into account all heating rates. First, using the iso-conversional Kissinger technique, the activation energy was

determined by plotting $\ln\left(\frac{\beta}{T_m^2}\right)$ versus $\frac{1}{T_m}$ in Fig. 4.3 and determining that the slope of the

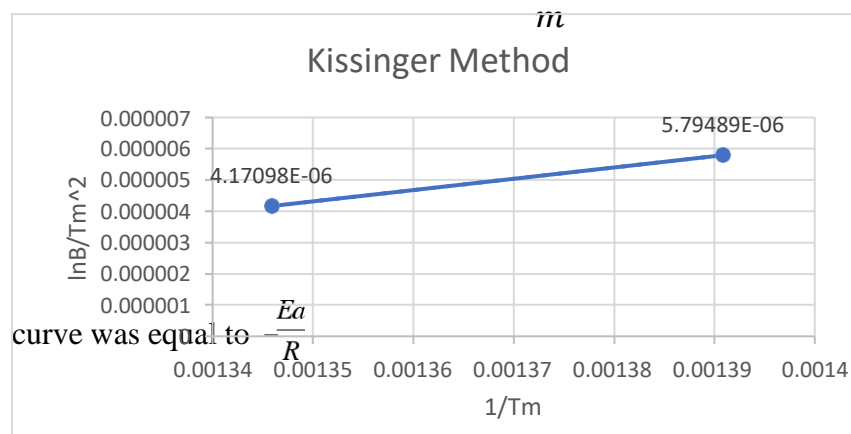
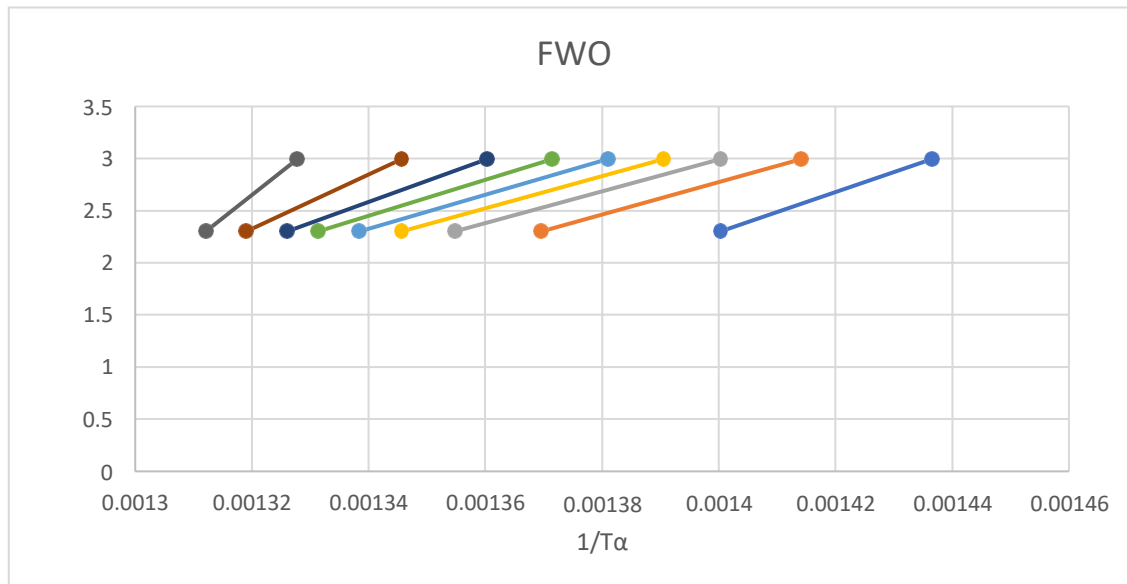


Fig. 4.3 Plot between $\ln\left(\frac{\beta}{T_m^2}\right)$ against $\frac{1}{T_m}$

Slope	R, KJ/mol.k	Ea, KJ/mol
2.3428	0.008314	1.9478

The activation energy was then calculated using the iso conversional Flynn-Wall-Ozawa (FWO), Kissinger-Akahira-Sunose (KAS), and Friedman (FR) techniques using the plots of $\ln(\beta)$ against $\frac{1}{T}$ (Fig. 4.4), $\ln(\frac{\beta}{T^2})$ against $\frac{1}{T}$ (Fig. 4.5), and $\ln(\beta \frac{d\alpha}{dt})$ against $\frac{1}{T}$ (Fig. 4.6).



The calculated activation energies for iso-conversional Flynn-Wall-Ozawa (FWO), Kissinger-Akahira-Sunose (KAS), and Friedman (FR) techniques are illustrated in Fig. 4.4, 4.5, and 4.6 for different conversion levels.

Table 4.2 - For iso-conversional Flynn-Wall-Ozawa (FWO), Kissinger-Akahira-Sunose (KAS), and Friedman (FR) techniques, activation energies (kJ/mol) at various conversions

	FWO	KAS	FR
0.1	159.1678	171.8695	296.84
0.2	129.3693	143.0691	159.085
0.3	126.578	140.2766	122.311
0.4	128.3276	140.1931	97.4788
0.5	135.5639	146.1102	78.0906
0.6	143.4684	156.4277	53.4805
0.7	168.1572	181.9741	8.39094
0.8	216.4591	225.7448	32.66681
0.9	367.5487	372.5218	28.31899

4.4 Prediction of decomposition model

Different solid-state kinetic breakdown models, which are categorised as nucleation, geometrical contraction, diffusion, and reaction order, are available to help us understand the mechanism by which the reaction is occurring in the reactor.

A model fitting method is used to predict the decomposition model for a recent study. Criado and Coats-Redfern approaches are employed in this model fitting to forecast the solid state decomposition model that will be used in a pathway of degradation of mixed waste plastics.

4.4.1 Criado Method:

This method should evaluate the theoretical estimate of master plots of various models versus developed experimental data, as mentioned in the literature section.

This approach allows us to determine $Z(\alpha)$, a function of $f(\alpha)$ and $g(\alpha)$.

$$Z(\alpha) = \frac{f(\alpha)g(\alpha)}{\frac{d\alpha}{dT} \left(\frac{E\alpha}{R} \right) \exp \left(\frac{E\alpha}{RT} \right) p(x)}$$

$$\text{Where, } p(x) = \frac{e^{-x}}{x} \quad (x^3 + 18x^2 + 86x + 96) \quad (x^4 + 20x^3 + 120x^2 + 240x + 120)$$

We have been using experimental data from isoconversional techniques, such as activation energy (E) and conversion rate (dα/dT). Our Eα/RT is represented by the term x in the

equation p(x). We can specifically mention activation energy, which comes from the FWO approach.

Figure 4.7 displays the graphic behaviour of all sixteen models mentioned in Table 4.2 as well as experimental data plotted between $Z(\alpha)$ vs α .

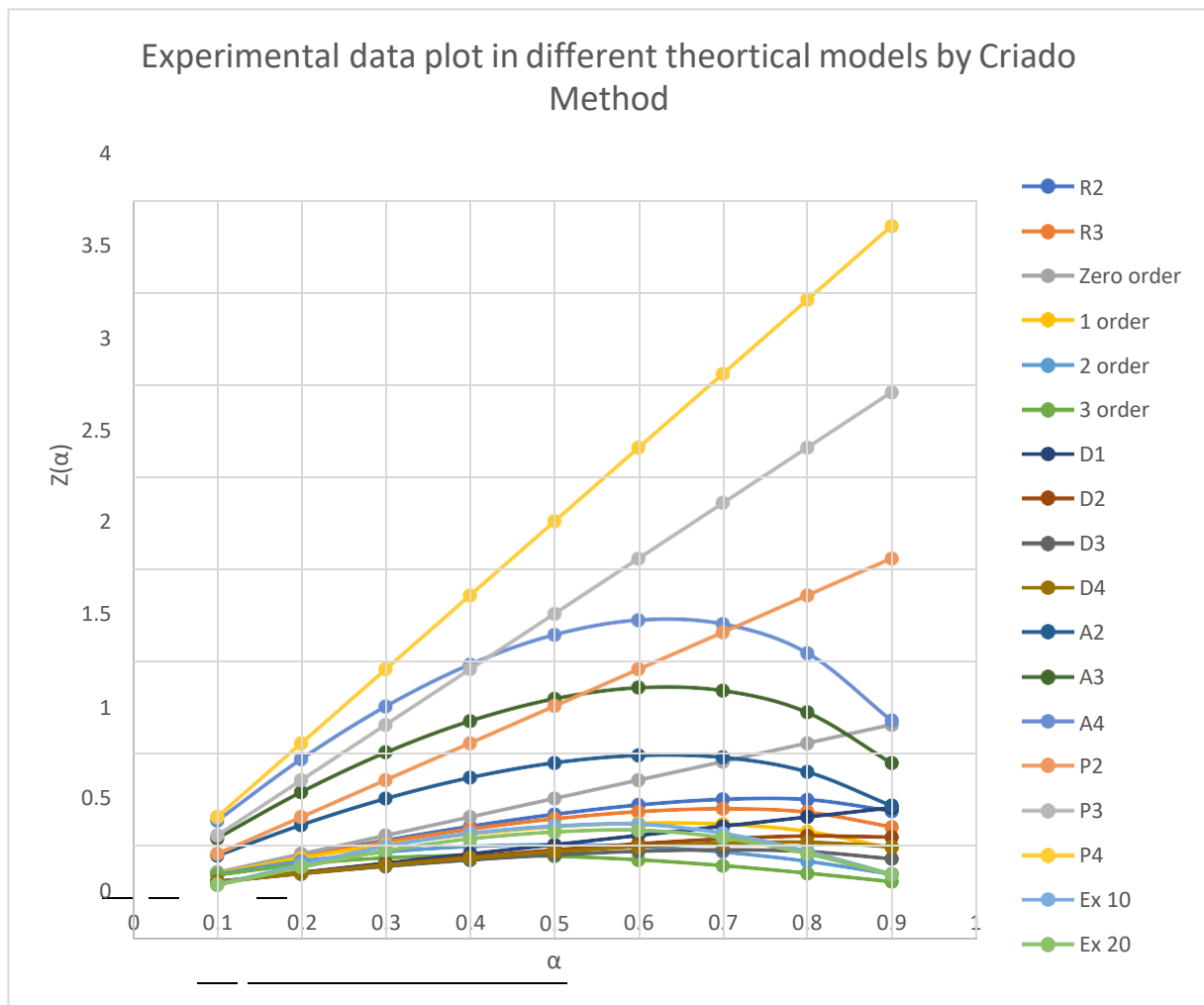


Fig. 4.7 illustrates this by highlighting the model curve line that best fits the experimental curve while ignoring other model curves.

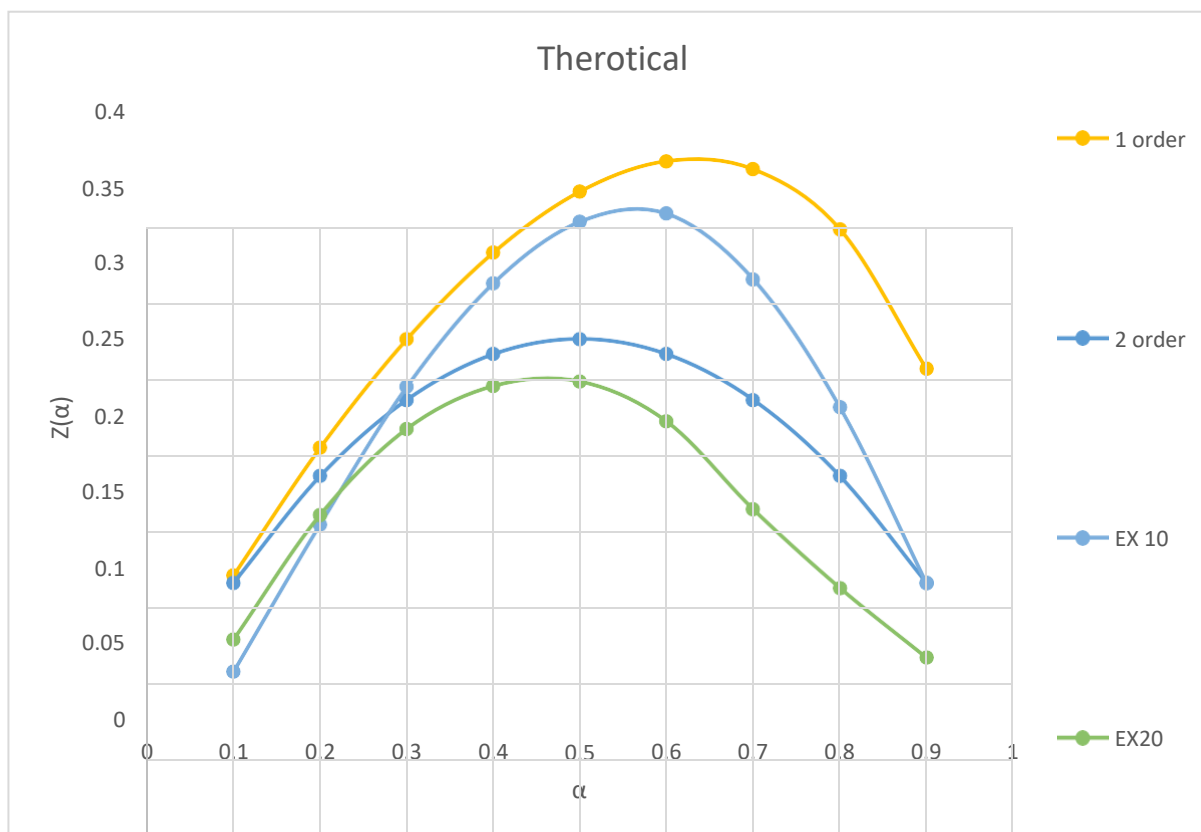


Fig.4.8 Model fitting between therotical & practical

The experimental curve, namely the heating rate of 10°C, is fitted on a first order basis, as seen in Fig. 4.8. Additionally, a 20°C heating rate experimental curve was fitted on a second order.

In first order, An arrangement of chemical reactions where the rate of the reaction is proportional to the quantity of the reactant and is dependent on the concentration of just one reactant & in second order, A concept used to describe a chemical reaction's reaction rate when it is proportional to the product of the concentrations of two of the reactants

In this result we can concluded, the 10°C/min decomposition fitting in first order because the reaction rate is controlled by only one reactant concentration. The behaviour of 20°C/min decomposition here the result of second order because the reaction rate is controlled by probably square root of component concentration or other waste component present in the feed mixture.

Conclusion:

To solve the problem of disposing of plastic trash, the municipal plastic waste stream, which primarily consists of high-density polyethylene, low-density polyethylene, polypropylene, polyethylene terephthalate, and polystyrene after separation, can be treated. Utilizing thermogravimetric analysis (TGA), mixed plastic (a simulated mixture of pure plastics) was thermally decomposed. Using many iso-conversional techniques, assess the activation energy and frequency component based on this TGA study (Friedman method, Kissinger method, FWO method and KAS method). With the aid of model fitting techniques like the Criado method, the value of a chosen activation energy was used to define the model fitting methodology employed on the decomposition data. It was applied to the plastics mixture simulation to forecast degradation. The outcome shows that it fits the reaction model for this breakdown process at first order in 10°C and also at second order in 20°C.

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