

# Logarithmic Differential Evolution (LDE) for Optimization of Kinetic Parameters in Pyrolysis of Biomass

Pratik N. Sheth

Lecturer, Department of Chemical Engineering  
Birla Institute of Technology and Science (BITS)  
PILANI – 333 031 (Rajasthan) India  
E-mail: pratik@bits-pilani.ac.in

B. V. Babu\*

Dean-Educational Hardware Division (EHD)  
& Professor of Chemical Engineering  
Birla Institute of Technology and Science (BITS)  
PILANI – 333 031 (Rajasthan) India  
Phone: +91-1596-245073 x 259 ;  
E-mail: bvbabu@bits-pilani.ac.in;

## ABSTRACT

Pyrolysis is the thermal decomposition of organic matter under inert atmospheric conditions, leading to the release of volatiles and formation of char. It is also a first step in the biomass gasification. Understanding of kinetic parameters is essential for the design of a suitable pyrolysis reactor. In the proposed kinetic model of this study, the kinetic scheme of biomass decomposition by two competing reactions giving gaseous volatiles and solid charcoal is used. Differential evolution is used to find the kinetic parameters by minimizing the square of the error between the reported experimental data of thermogravimetry of hazelnut shell and simulated model predicted values of residual weight fraction. Logarithmic DE, an improved version of simple DE, is proposed by incorporating logarithmic initialization and logarithmic mutation to take care of wide ranges of variable values. Logarithmic DE is found to yield better kinetic parameters in terms of objective function and gave better fit with experimental data.

## Categories and Subject Descriptors

G.1.6 OPTIMIZATION: Global optimization; J.2 PHYSICAL SCIENCE AND ENGINEERING: Engineering

**General Terms:** Algorithms

## Keywords

Biomass; Pyrolysis; Kinetics; Parameter Estimation; Modeling; Simulation; Optimization; Evolutionary Algorithm; Initialization; Mutation; Differential Evolution; Logarithmic

## 1. INTRODUCTION

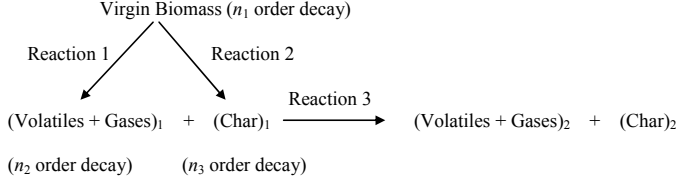
Decomposition of a compound, in the absence of oxygen, by the action of heat alone to produce various organic gaseous products, charcoal and tar, is known as pyrolysis [1]. Pyrolysis is not only an independent process, but also a first step in the gasification or the combustion process. Hazelnut shell is an abundantly available agriculture residue. Dogru et al. [2] used a pilot plant scale downdraft gasifier to investigate gasification potential of hazelnut shells. It is necessary to understand the kinetics of pyrolysis in order to design a suitable pyrolysis reactor. Balci et al. [3] proposed several kinetic models for hazelnut pyrolysis and validated by thermo-gravimetric

experiments. In these kinetic models, the rate expression based on first-order decomposition of the reactive solid is defined in terms of fractional conversion. Demirbas [4] performed thermo-gravimetric experimental runs and presented the weight loss data for different particle sizes of ground hazelnut shell and for various heating rates. Kinetic analysis has also been carried out but the expression for the kinetic constants with respect to temperature is not developed, and the experimental data validation with theoretical models for these experiments is not reported in the literature. Experimental and modeling studies have been conducted on pyrolysis by many researchers [1-10].

In the present study, kinetic model based on apparent decomposition rate [3] is modified by incorporating the kinetic scheme proposed by Koufopoulos et al. [8, 9]. To find kinetic parameters of proposed model, an objective function based on least square error between experimental data and simulated results has to be minimized. Model simulation results are validated with the data reported in literature [4]. A population based search algorithm, Differential Evolution (DE), which is simple and robust and has proven successful record, is employed for optimization in the present case [12, 13]. However, for some problems simple DE gives poor population distribution for cases where the range of limits is very wide (more than three orders of magnitude). In the present case of kinetic parameter estimation, lower and upper limits of frequency factors are  $10^{10}$  and  $10^{18}$  respectively. Hence simple DE is modified first by including the logarithmic initialization (LIDE). The algorithm is further improved by incorporating the logarithmic mutation also and named as logarithmic DE (LDE). Optimum kinetic parameters are found by minimizing the objective function using simple DE, LIDE and LDE and obtained results are compared.

## 2. KINETIC MODELING & SIMULATION

The pyrolysis reactions can be described by means of the following scheme proposed by Koufopoulos et al. [8, 9]. This model indicates that the biomass decomposes to volatiles, gases and char. The volatiles and gases may further react with char to produce different types of volatiles, gases and char where the compositions are different. Therefore, the primary pyrolysis products participate in secondary interactions (Reaction 3), resulting in a modified final product distribution.



The kinetic equations for the mechanism, shown above are represented by Eq. (1) through Eq. (3).

$$r_1 = k_1 B^{n_1} \quad (1)$$

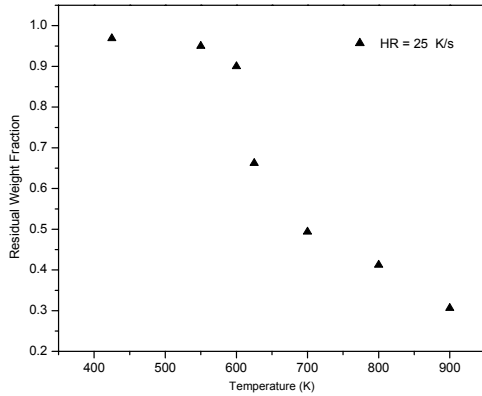
$$r_2 = k_2 B^{n_2} \quad (2)$$

$$r_3 = k_3 G_1^{n_3} C_1^{n_3} \quad (3)$$

To find the kinetic parameters of the above-mentioned reactions, the square of the error between the reported experimental data of thermogravimetry of hazelnut shell and theoretical values of residual weight fraction is minimized. Thermogravimetry data of hazelnut shell is reported as % weight loss versus temperature [4]. The data has been recalculated in terms of residual weight fraction, where, residual weight fraction is defined as given by Eq. (4)

$$\text{Residual Weight Fraction } (W) = \frac{(\text{Residual Weight})}{(\text{Initial Weight})} \quad (4)$$

The recalculated experimental data of 0.180 mm size and for heating rate of 25.0 K/s are plotted and shown in Fig. 1.



**Fig. 1 Residual Weight Fraction versus Temperature (K)**

To find the residual weight fraction theoretically, net rate of production of different species by reaction 1 & 2 in terms of rate of reactions are found. Due to small size (0.180 mm) of the hazelnut shell sample taken in the experiments, the secondary reaction [reaction 3] is neglected. The residual weight fraction is calculated using Eq. (5).

$$W = B + C_1 \quad (5)$$

For simplicity, the order of reactions 1 & 2 are taken as 1.0. Then Eq. (1) & (2) reduce to Eq. (6) and Eq. (7) respectively.

$$\frac{dB}{dt} = -(k_1 + k_2)B \quad (6)$$

$$\frac{dC_1}{dt} = k_1 B \quad (7)$$

So the change of residual weight fraction with time [Eq. (8)] obtained by addition of Eq. (6) and Eq. (7).

$$\frac{dW}{dt} = -k_1 B \quad (8)$$

To find temperature ( $T$ ) at a particular time ( $t$ ), following equation [Eq. (9)] is used [3, 4, 7-9],

$$T = (HR)t + T_0 \quad (9)$$

Differentiating Eq. (9) would result in

$$dT = (HR)dt \quad (10)$$

Using Eq. (8) & (10), relations of change of residual weight fraction with temperature is found, which is given by Eq. (11).

$$\frac{dW}{dT} = -k_1 B \frac{1}{HR} \quad (11)$$

Using Eq. (6) & (10), relation of change of biomass weight fraction with temperature can be found & given by Eq. (12).

$$\frac{dB}{dT} = -(k_1 + k_2)B \frac{1}{HR} \quad (12)$$

Arrhenius relation of kinetic constants is given by Eq. (13) in which  $i = 1$  and 2 for reaction 1 and reaction 2 respectively.

$$k_i = A_i \exp\left(\frac{-E_i}{RT}\right) \quad (13)$$

Values of frequency factor and activation energy of both reactions are found by minimizing the objective function (Eq. 14).

$$F(A_1, E_1, A_2, E_2) = \sum_{j=1}^n (W_{\text{exp},j} - W_{\text{cal},j})^2 \quad (14)$$

Sheth and Babu [10] found the kinetic parameters for isothermal pyrolysis of beech wood saw dust using a MATLAB subroutine. When the same inbuilt MATLAB optimization function is used to find the kinetic parameters by minimizing the Eq. (14) for non-isothermal pyrolysis of hazelnut shell in the present study, it yielded different converged values of frequency factor and activation energies as optimum with different initial guesses. It indicates that the present objective function is highly nonlinear and complex in nature, having local optima. Most of the traditional optimization algorithms based on gradient methods have the possibility of getting trapped at local optimum depending upon the degree of non-linearity and initial guess. Differential Evolution (DE) is applied by taking Eq. (14) as an objective function to be minimized to find the global optimum set of kinetic parameters. The key parameters of control in DE are: NP- population size, CR- cross over constant, and F weight applied to random differential (scaling factor). The details of DE algorithm and pseudo code are available in the literature [12, 13-17]. The key parameters of DE are problem dependent. However, certain guidelines and heuristics are available for the choice of these parameters [15]. Based on these heuristics, the values of DE key parameters for the present problem are set as follows:

$$\text{NP} = 40, 80, 120, 160, 200; \text{CR} = 0.9; \text{F} = 0.5$$

The limits of the frequency factors and activation energies are given below, which is chosen based on the available literature for kinetic modeling of different biomasses.

Limits of frequency factors ( $A_1$  &  $A_2$ ) = (1.0e+10, 1.0e+18)

Limits of activation energies ( $E_1$  &  $E_2$ ) = (1.0e+04, 3.0e+05)

To find the theoretical value of the residual weight fraction (W), forward finite difference technique [21, 22] is applied to Eq. (11) to Eq. (13) with the following initial conditions.

At time  $t=0$ ;  $T_0 = 325$  K;  $B = 1.0$ ;  $C_1 = 0.0$ ;  $G_1 = 0.0$

### 3. RESULTS AND DISCUSSION

Table-1 shows the kinetic parameters of reaction 1 ( $A_1$  and  $E_1$ ) and reaction 2 ( $A_2$  and  $E_2$ ) for the heating rate of 25.0 K/s for different size of population (NP). NP value is varied from 10 to 50 times of the dimension of the problem. With increase in NP value, objective function value is decreased and different set of kinetic parameters are found as optimum values. It clearly indicates that kinetic parameter estimation is a multimodal problem and having a number of local minima. In addition it also yielded the frequency factors of the order of  $10^{17}$ , whereas the limits cover a wide range from  $10^{10}$  to  $10^{18}$  (i.e. forcibly getting trapped towards the upper limit only).

Table-1. Kinetic Parameters of reaction 1 and reaction 2 found using simple DE for heating rate of 25 K/s

Number of Population (NP)	Kinetic Parameters				Objective Function Value [Eq. (14)]
	$A_1$ (1/s)	$E_1$ (J/mol)	$A_2$ (1/s)	$E_2$ (J/mol)	
40	4.566886671e+017	2.113759061e+005	5.720718806e+017	2.126114848e+005	9.130447823e-003
80	4.558503512e+017	2.113970396e+005	4.765050853e+017	2.117037535e+005	9.128729091e-003
120	3.709092314e+017	2.102901874e+005	3.507645032e+017	2.100792813e+005	9.109103152e-003
160	1.416632742e+017	2.055108395e+005	9.999999843e+017	2.155763135e+005	9.048729762e-003
200	1.337902880e+017	2.052421797e+005	9.999999843e+017	2.156096494e+005	9.041855517e-003

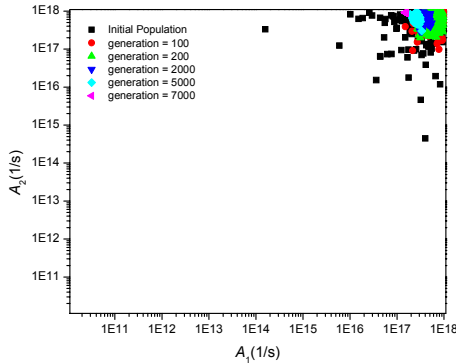


Fig. 2 Population Distribution of Frequency Factors for NP = 200 using simple DE

Fig. 2 and Fig. 3 show the variation of population distribution of frequency factors ( $A_1$  and  $A_2$ ) and activation energies ( $E_1$  and  $E_2$ ) with the number of generations for an NP value of 200. For frequency factors (Fig. 2) the initial population covers only a part ( $10^{16}$  to  $10^{18}$ ) of the entire range ( $10^{10}$  to  $10^{18}$ ). Initial population of activation energies almost covers the entire range from lower to upper limit (Fig. 3), but majority of the points lie in the range from  $10^5$  to  $3 \times 10^5$  and very few points lie near lower limit i.e.  $10^4$ . This is due to linear mapping rule used in DE for initialization of normalized population. According to the mapping rule in the initialization of normalized population

$$\text{New variable} = \text{Minimum value of the variable} + \text{Random no.} \times (\text{Maximum value of the variable} - \text{Minimum value of the variable}) \quad (15)$$

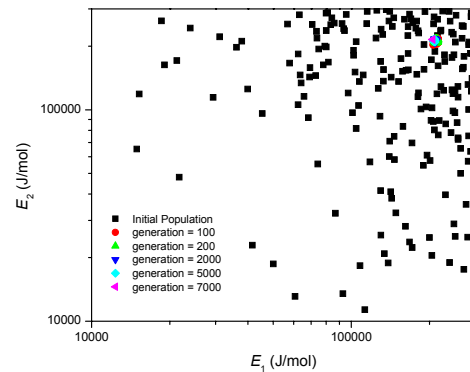


Fig. 3 Population Distribution of Activation Energies for NP = 200 using simple DE

Table -2 shows the variable value with different random numbers ranging from 0.1 to 1.0 for a minimum value of  $10^{10}$  and a maximum value of  $10^{18}$ . For a change in value of random number from 0.1 to 1.0, the change in variable value is only one order of magnitude ( $1.0 \times 10^{17}$  to  $1.0 \times 10^{18}$ ), which is not good enough change taking into account of the possible range of variable value ( $10^{10}$  to  $10^{18}$ ). By this mathematical operation, the new variable value found in the initialization of DE would have the order of magnitude equal to that of the maximum value. So the optimum value found using simple DE may be a local minimum or maximum and not the global one.

#### 3.1 Logarithmic Mapping Rule

To overcome the problem of population distribution, logarithmic mapping rule is proposed for initialization of normalized population and given by Eq. (16)

$$\text{Variable value} = \text{AntiLog} \{ \text{Log}(\text{minimum value}) + (\text{random number}) [\text{Log}(\text{maximum value}) - \text{Log}(\text{minimum value})] \} \quad (16)$$

New variables are found using Eq. (16) with a minimum value of  $10^{10}$  and maximum value of  $10^{18}$  for different values of random numbers ranging from 0.1 to 1.0. Wide distribution of new variable values, which is essential to cover wide range of the said variable, are obtained and given in Table - 2.

**Table -2 Variable value for different random numbers (Min =  $10^{10}$  and Max =  $10^{18}$ )**

Random Number	Variable Value using simple initialization (Eq. (15))	Variable Value using logarithmic initialization (Eq. (16))
0.1	$1.000 \times 10^{17}$	$6.309 \times 10^{10}$
0.2	$2.000 \times 10^{17}$	$3.981 \times 10^{11}$
0.3	$3.000 \times 10^{17}$	$2.512 \times 10^{12}$
0.4	$4.000 \times 10^{17}$	$1.585 \times 10^{13}$
0.5	$5.000 \times 10^{17}$	$1.000 \times 10^{14}$
0.6	$6.000 \times 10^{17}$	$6.309 \times 10^{14}$
0.7	$7.000 \times 10^{17}$	$3.981 \times 10^{15}$
0.8	$8.000 \times 10^{17}$	$2.512 \times 10^{16}$
0.9	$9.000 \times 10^{17}$	$1.585 \times 10^{17}$
1.0	$1.000 \times 10^{18}$	$1.000 \times 10^{18}$

For the present problem of kinetic parameter estimation for a heating rate of 25 K/s, logarithmic mapping is used to initialize the normalized population vectors and simple differential evolution is applied to find the global optimum value of kinetic parameters (We call the LIDE). Fig. 4 and Fig. 5 shows the population distribution of frequency factors and activation energies respectively, for NP = 200 with respect to number of generations in case of LIDE. In comparison with simple DE (Fig. 2 and Fig. 3), LIDE gives better population distribution for  $A_1$ ,  $A_2$ ,  $E_1$ , and  $E_2$ . Table - 3 shows the optimum kinetic parameters and the value of objective function found by using the LIDE for different value of NP. Comparison for NP = 40, 120 and 160 from Table - 1 (simple DE) and Table - 3 (LIDE) shows that the objective function value is less for simple DE and for NP = 80 and 200, LIDE gave better results in terms of optimum value of objective function. It may be noted that the optimum values of frequency factors are of the order of  $10^{17}$ . Also in Fig. 4 and Fig. 5, the population distribution just after 20<sup>th</sup> generation narrows down and all points lie very close to the upper limit value of the variable. This kind of population distribution change with respect to no. of generations in DE is due to the mutation operator. In the mutation operation of the DE, weighted difference vector is calculated by difference of two randomly chosen vectors. Noisy random vector is calculated by adding weighted difference vector and the randomly chosen Target vector. Mathematically it is written as given by Eq. (17).

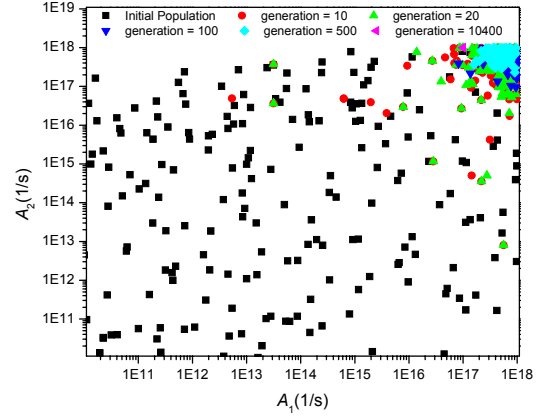
$$\text{Noisy Random Vector} = \text{Target Vector} + \text{Scaling Factor} (\text{difference of two randomly chosen vectors}) \quad (17)$$

For a Scaling factor of 0.5, and the order of three randomly chosen vector of  $10^{10}$ ,  $10^{12}$  and  $10^{17}$  respectively

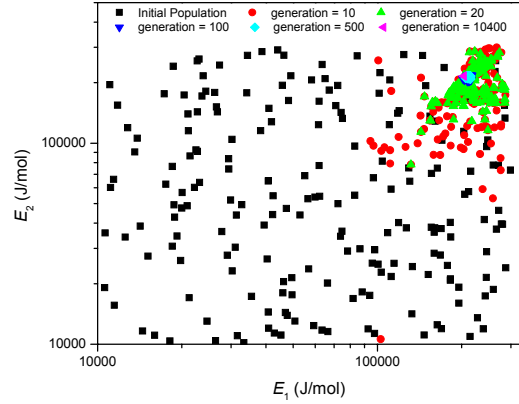
$$\text{Noisy Random Vector} = 10^{10} + 0.5 (10^{17} - 10^{15}) = 4.95 \times 10^{16}$$

Because of this linear operation, all the members of the mutant population would be of highest order among the three

randomly chosen vectors (irrespective of their numerical values) and after few generations, the entire population of points lie near the upper limit of the variable.



**Fig. 4 Population Distribution of Frequency Factors for NP =200 using LIDE**



**Fig. 5 Population Distribution of Activation Energies for NP =200 with LIDE**

### 3.2 Logarithmic Mutation

To overcome the problem of population distribution generation after generation, logarithmic mutation is proposed and given by Eq. (18).

$$\text{Noisy Random Vector} = \text{AntiLog} \{ \log(\text{variable}[c]) + F (\log(\text{variable}[a]) - \log(\text{variable}[b])) \} \quad (18)$$

Where a, b and c are randomly chosen number from the population size. For a Scaling factor of 0.5, and the order of three randomly chosen vector of  $10^{10}$ ,  $10^{12}$  and  $10^{17}$  respectively

Noisy Random Vector

$$= \text{AntiLog} \{ \log(10^{10}) + F (\log(10^{17}) - \log(10^{15})) \} = 1.0 \times 10^{11}$$

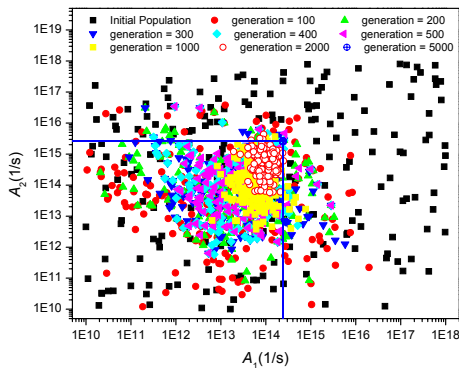
By implementation of logarithmic mutation in DE, better mutant population in terms of wide population distribution is expected and so better chances to get the global optimum values. We call this logarithmic initialization and logarithmic mutation of DE as Logarithmic DE (LDE).

**Table-3. Kinetic Parameters of reaction 1 and reaction 2 found using LIDE for heating rate of 25 K/s**

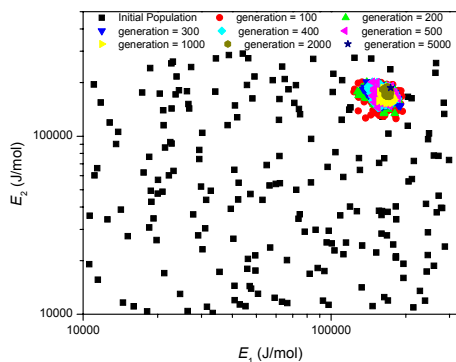
Number of Population (NP)	Kinetic Parameters				Objective Function Value [Eq. (15)]
	$A_1$ (1/s)	$E_1$ (J/mol)	$A_2$ (1/s)	$E_2$ (J/mol)	
40	3.625545542e+017	2.101262519e+005	2.405592550e+017	2.080204938e+005	9.135202071e-003
80	1.684361501e+017	2.064137617e+005	6.975696814e+017	2.137319632e+005	9.071062357e-003
120	3.732242417e+017	2.104686034e+005	4.505817354e+017	2.115734744e+005	9.125753720e-003
160	2.426530209e+017	2.082492439e+005	9.999999843e+017	2.155842635e+005	9.085364436e-003
200	9.959812379e+016	2.037569391e+005	9.999999843e+017	2.156303008e+005	9.021881334e-003

**Table-4. Kinetic Parameters of reaction 1 and reaction 2 found using Logarithmic DE (LDE) for heating rate of 25 K/s**

Number of Population (NP)	Kinetic Parameters				Objective Function Value [Eq. (15)]
	$A_1$ (1/s)	$E_1$ (J/mol)	$A_2$ (1/s)	$E_2$ (J/mol)	
40	1.540784554e+014	1.713920488e+005	2.005103335e+013	1.619462097e+005	6.442233304e-003
80	3.885485326e+014	1.758750841e+005	4.551904297e+012	1.541520486e+005	6.441844469e-003
120	2.284807833e+014	1.734125580e+005	1.961720679e+014	1.735975231e+005	6.425304514e-003
160	2.119764183e+014	1.731095826e+005	6.750124424e+014	1.800125476e+005	6.421839787e-003
200	2.106331972e+014	1.730996444e+005	3.014892950e+015	1.876928166e+005	6.294910790e-003



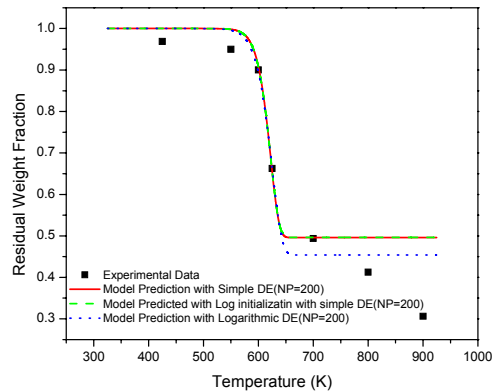
**Fig. 6 Population Distribution of Frequency Factors for NP =200 with logarithmic DE**



**Fig. 7 Population Distribution of Activation Energies for NP =200 with logarithmic DE**

To compare the performance of LDE with simple DE and LIDE, LDE is applied to the present problem of kinetic parameter estimation for a heating rate of 25 K/s. Table - 4

shows the optimum kinetic parameters and the value of objective function found by using the LDE for different values of NP. The objective function value is the least using LDE in comparison with simple DE (Table -1) or LIDE (Table - 3) for any NP Value. The optimum kinetic parameter values found using LDE are also quite different from that of the values found using simple DE and LIDE. Fig. 6 shows the population distribution of the frequency factors with number of generations using LDE. Initial population is widely spread over the entire range and population distribution after subsequent generations are also spread over a wide range.



**Fig. 8 Experimental and Theoretical Residual Weight Fraction for heating rate of 25 K/s**

Comparison of Fig 6 with Fig.4 and Fig. 2 shows that LDE gives wide spread of population distribution and so could give better value of optimum variables. The lines shown in Fig. 6, i.e. Drop lines drawn on to x-axis and y-axis, show the population after 5000 generations, where all the points merge to give optimum value of frequency factors. Fig. 7 shows the variation of population distribution with number of generations

for activation energies. Using the optimum kinetic parameters for NP = 200 and found by simple DE (Table - 1), LIDE (Table - 3) and LDE (Table - 4) are used to find the residual weight fraction and the results are compared with experimental data as shown in Fig. 8. It shows that model predictions for LDE fits better amongst model predictions found using simple DE and LIDE.

#### 4. CONCLUSIONS

Based on the results obtained and discussions in the earlier sections, the following conclusions are drawn.

- Kinetic parameters (frequency factors and activation energies) for the two competing reactions, found using simple differential evolution (DE) are not global.
- Optimum value of frequency factors are biased towards the upper limit of the range chosen using simple DE.
- Logarithmic initialization with simple DE (LIDE) yields very good initial population distribution but failed to give better value of kinetic parameters in comparison with simple DE.
- Logarithmic DE (LDE) which is a combination of logarithmic initialization and logarithmic mutation with simple DE yields kinetic parameters which give better value of objective function.
- Kinetic parameters found using LDE gives global optimum set when compared with experimental data.

#### NOTATION

$B$	Concentration of Biomass
$C_1$	Concentration of Charcoal 1
$CR$	Crossover constant
$F$	Scaling Factor
$G_1$	Concentration of volatile component 1
$HR$	Heating Rate
$j$	Data point variable
$k_i$	Kinetic constant of reaction $i$
$n$	Total no. of data points
$NP$	Population size
$r_i$	Rate of reaction $i$
$t$	Time
$T_0$	Initial Temperature
$W$	Residual Weight Fraction
$W_{exp}$	Experimental value of the residual weight fraction
$W_{cal}$	Calculated value of the residual weight fraction
$z$	Normalized Conversion

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