

Application of Genetic Algorithm for Evaluation of Kinetic Parameters of Coal Pyrolysis

S. S. Dhumal and R. K. Saha

Chemical Engineering Department

Indian Institute of Technology, Kharagpur, India

Email: rks_iit@yahoo.co.uk

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Abstract

Complex reactions such as coal pyrolysis reaction are often analyzed using the distributed activation energy model, in which the reactions are assumed to consist of a set of irreversible first-order reactions that have different activation energies and a constant frequency factor. A Gaussian distribution has usually represented the difference in the activation energies. The parameters in the Gaussian distribution, mean activation energy, E_{v0} , and standard deviation, σ_v , are determined to fit experimental data. This problem is related to the multivariable optimization. The values of k_{r0} , E_{v0} & σ_v varied so widely such that available optimization techniques to predict the kinetic parameters of pyrolysis from experimental data are difficult. Genetic algorithm is used in searching the global optimum, based on the idea of natural selection in evolution, and is generally suitable, for cases where the wide variation in parameters to be optimized, such as kinetics of coal pyrolysis. The kinetic parameters of devolatilization evaluated from thermogravimetric data for a typical Indian coal show that they vary widely with the rate of heating of coal. The variation in k_{r0} is from $1\text{E}+5$ to $1\text{E}+16 \text{ s}^{-1}$, E_{v0} from 28,000 to 70,500 cal/mol and σ_v from 5,000 to 19,000 cal/mol. As compared to the method of Miura reported in 1995, the present method has an advantage that it can be applied for non-linear heating rate.

Nomenclature

$f(E_v)$	distribution function for devolatilisation activation energy (mol/cal)
k	first order devolatilisation reaction rate constant (s^{-1})
k_m, k'	devolatilisation reaction rate constant ((volatile conc.) $n-1 \text{ s}^{-1}$)
k_{r0n}	frequency factor for devolatilisation ((volatile conc.) $n-1 \text{ s}^{-1}$)
n	order of devolatilisation reaction
t	time (s)
E_v	activation energy of reaction for devolatilisation (cal/mole)
E_{v0}	mean activation energy for devolatilisation (cal/mole)
R	gas constant (cal/mole K)
T	temperature of the devolatilisation (K)
V	fraction of volatiles loss from coal up to time t

V^*	effective volatile content of coal
σ_v	standard deviation in activation energy distribution (cal / mol)
i	suffix for species number

Introduction

The mechanism involved in the rapid thermal decomposition of coal is exceedingly complex and the influence of many experimental variables is not well understood. Coal is an intermediate product of very slow decomposition of organic matter deposited in prehistoric times. The final product would be pure carbon. Measurable devolatilisation rate does not appear until temperatures of 350°C to 400°C are reached, and therefore its extent, apart from types of coal, is strong function of temperature and rate of heating. Coal is a complex organic polymer consisting of aromatic clusters of several fused rings linked together by assorted hydrocarbon and heteroatom (O, N, and S) linkages. Heating causes the structure to decompose, the weaker bonds rupturing at lower temperatures and the stronger ones at higher temperatures and the fragments that are volatile attempt to escape from the particle. Some of the fragments are highly reactive free radicals subject to a variety of secondary reactions such as cracking and re-polymerization. Generally, such secondary reactions are undesirable in practice, since they deposit a portion of the volatile matter as a solid or char and diminish gas and liquid yields. The extent of secondary reactions can be reduced by enhancing the transport of volatile fragments away from the reactive environment, such as by operating at reduced pressures with smaller and more widely dispersed particles.

The complexity of these phenomena has handicapped the development of models suitable for design and scale-up. Considerable research on coal devolatilisation has been conducted over the years. Several past reviews are available. The best source is probably Lowry's [1] compilation of review article. In that work, H. C. Howard [2] reviewed coal pyrolysis reactions (mostly information obtained by slow heating techniques), and C. G. V. Fredersdorff and M. A. Elliott [3] reviewed the state of the art in coal gasification technology. Devolatilisation research at low heating rates was summarized by S. Badzioch [4]. W. I. Jones [5] similarly reviewed a number of pyrolysis studies, most of which were done at low heating rates. Essenhigh and Howard [6] reviewed the devolatilisation literature in a monograph on combustion and explosion phenomena in coal dust.

Wiser et al. [7] treated the volatile loss data isothermally, integrating first and second order versions of kinetic equations, then inserting an appropriate value of V^* depending on the temperature. Similarly, Badzioch and Hawksley [8] correlated data with an expression derived by isothermal integration of first order kinetic rate expression and substitution of a temperature dependent function for V^* . A powerful but convenient technique available for modeling the numerous chemical reactions involved in the decomposition process is the use of a statistical distribution of activation energies. Such a model simulates the decomposition behavior over an extraordinarily broad range of conditions without creating a large number of adjustable constants. Secondary reactions are very difficult to model because they occur in competition with the transport and escape of volatiles. It was considered by Pitt's [9] by assuming the multiple parallel reaction models to represent the coal devolatilisation.

Miura [10] evolved a procedure which takes into account the dependence of k_{r0} on activation energy E_v and estimate both the distribution curve of the activation energy, $f(E_v)$, and the frequency factor, k_{r0} , from three sets of weight loss data at three different heating rates. With k_{r0} vs. E_v relationship established,

Miura and Maki [11] evolved another procedure by which the $f(E_v)$ curve can be generated with the help of a single weight loss curve. But the method described by Miura is applicable for linear heating rates only. It is not possible to apply for non-linear heating rates.

Katare et al. [12] evaluated the thermal decomposition kinetic parameters of borax pentahydrate by using genetic algorithm technique and made use of these variables for the comparison of theoretical calculation with the experimental results. Sachin and Bulutcu [13] found the application of genetic algorithm for efficient parameter estimation of large kinetic models. They applied this technique for estimating the parameters of a model for the propane aromatization on a zeolite catalyst.

This paper represents a genetic algorithm based technique for the evolution of kinetic parameters of devolatilization of coal. This technique is advantageous because it gives the kinetic parameters of devolatilisation for any case either linear or non-linear heating rates. Experimental data on coal pyrolysis at different temperatures and at different heating rates for different coals of Indian origin were obtained in a laboratory scale thermo-gravimetric set up fabricated locally. To obtain the kinetic parameters of coal pyrolysis (k_{r0} , E_{v0} and σ_v), the multiple reaction models with the assumption of Gaussian distribution of activation energy and genetic algorithm solution technique is used. It is very difficult to compare the results obtained by the methods mentioned above with that of Miura, because both methods are totally different from the solution point of view.

Experimental Details

To study kinetics of pyrolysis of different types of Indian coals, a simple thermo gravimetric set up [14] has been fabricated for this purpose in the lab. The experimental set-up as shown in Fig. 1 consist of an electrically heated tubular reactor of ID 60 mm, length 630 mm and is insulated with asbestos rope. A Chromel-Alumel thermocouple is placed inside the tube, very near the to the sampling pan and connected to a temperature indicator. A 300-mesh stainless steel sieve basket containing coal is suspended from the single pan balance by a fine nichrome wire in the centre of the tube. A pre-determined constant nitrogen flow is maintained during heating from the lower end of the pipe (near about 2 LPM). Temperature is varied from 25 °C to 800 °C. The rate of heating is adjusted manually by using the rheostat. The weight loss is noted continuously till the weight became constant. Using the devolatilisation models, which are to be described in the latter section, uses this weight loss data and time temperature history to determine the devolatilisation kinetic parameters of coal. The char at the end of each experiment was taken out and its volatile matter and ash content determined. The proximate and ultimate analysis of any type of coal was determined before thmo-gravimetric experimentation. Also some TG – DTA data were evaluated for different heating rates like 20, 30 and 40°C/min, which were used for evaluation of kinetic parameters.

Coal Pyrolysis Model

The modeling of coal pyrolysis [15] in terms of total volatiles yield is still important, even though the more powerful approach based on the individual compounds or classes of compounds that comprise the volatiles. Information on total volatiles, from weight loss measurements, has been more readily available than data on the evolution of individual species. Consequently much of the modeling work has been focused on total volatiles evolution or weight loss behavior.

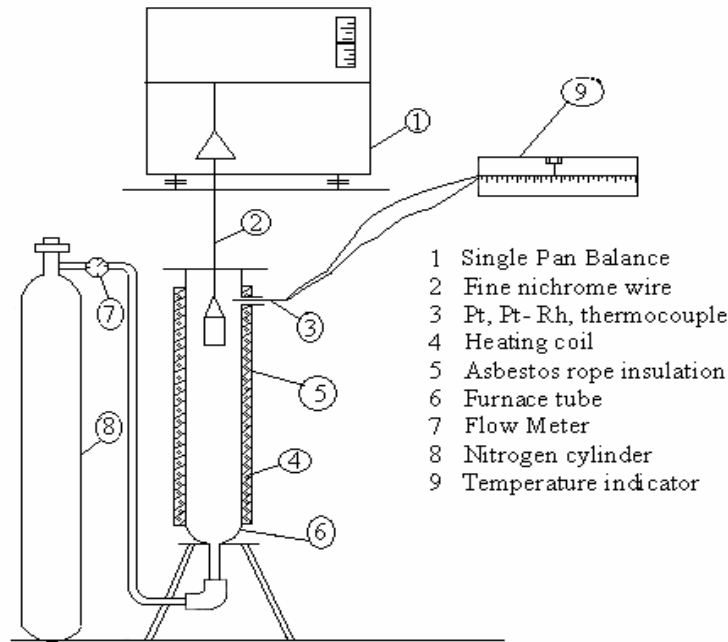


Fig. 1 Layout of the Thermogravimetric analysis setup

The complex decomposition and transport phenomena involved in coal devolatilisation are not amenable to exact description. Many authors [16] have approximated the overall process as a first order decomposition occurring uniformly throughout the particle. Thus, the rate of devolatilisation is expressed as:

$$\frac{dV}{dt} = k(V^* - V) \quad (1)$$

where, $V \rightarrow V^*$ as $t \rightarrow \infty$. The unknown parameters k and V^* have been the focus of kinetic studies. Badzioch and Hawksley [8] emphasize that V^* is to be carefully distinguished from volatile matter, V . The rate constant k in Equation (1) is typically correlated with the temperature by Arrhenius expression. Many authors have contended that a simple, first order model is inadequate. One attempt to improve the utility of Equation (1) has been to describe devolatilisation as a series of several first order processes occurring at different time intervals. Thus Shapatina et al. [17] depict three stages (1 to 0.06, 0.06 to 0.1, 0.1 to 180s), and Stone et al. [18] use time intervals typically much longer and dependent on coal type and temperature. Another approach has been to use a n^{th} order rate expression:

$$\frac{dV}{dt} = k'(V^* - V)^n \quad (2)$$

Thus, Wiser et al. [7] found that $n=2$ gave the best fit to their data over the first 3600s of weight loss, while $n=1$ was preferred for longer times.

Quantitative methods for modeling the temperature dependent behavior of coal devolatilisation are clearly not well established. Wiser et al. [7] treated their data isothermally, integrating first- and second- order versions of Equation (2) and then inserting an appropriate value of V^* depending on temperature. Similarly, Badzioch and Hawksley [8] correlated data with an expression derived by isothermal integration of Equation (1) and substitution of a temperature dependent function for V^* . Thus neither of these two groups of investigators considered the difficult integration involved in nonisothermal cases where a finite heating period exists.

Pitt's [9] recognition of this problem led him to adapt, as described Vand's [19] treatment of a large number of independent, parallel rate processes. The thermal decomposition of coal was assumed to consist of a large number of independent chemical reactions representing the rupture of various bonds within the coal molecule. Depending on the strengths, the rupture of different covalent bonds of coal molecules takes place at different temperatures with different rates. Since the thermal decomposition of a single organic species is typically described as an irreversible reaction that is first order with respect to the amount of unreacted material remaining, the rate of volatile evolution originating from a particular reaction within the coal structure was described by using the following equation, a subscript i is used to denote one particular reaction:

$$\frac{dV_i}{dt} = k_{ri}(V_i^* - V_i) \quad (3)$$

If k_{ri} is of the Arrhenius form, integration of the above equation for isothermal condition to find the amount of volatiles yet to be released gives:

$$V_i^* - V_i = V_i^* \exp(-k_{r0i} t \exp(-E_v / RT))_i \quad (4)$$

Values of k_{r0i} , E_{vi} and V_i^* cannot be predicted a priori and must be estimated from experimental data, a problem that increases with the number of reactions postulated. The problem is simplified if it is assumed that the k_{ri} 's differ only in activation energy, i.e., $k_{r0i} = k_{r0}$ for all i , and that the number of reactions is large enough to permit E_v to be expressed as a continuous distribution function $f(E_v)$, with $f(E_v) dE_v$ representing the fraction of the potential volatile loss V^* that has an activation energy between E_v and dE_v . Then V_i^* becomes a differential part of the total V^* and may be written:

$$dV^* = V^* f(E_v) dE_v \quad (5)$$

$$\int_0^\infty f(E_v) dE_v = 1 \quad (6)$$

The total amount of volatile yet to be released is obtained by summing up the contribution from each reaction or by integrating Equation (4) over all values of E_v and using equation (5). Thus

$$\frac{V^* - V}{V^*} = \int_0^\infty \exp[-k_{r0} t \exp(-E_v / RT)] f(E_v) dE_v \quad (7)$$

Pitt's results are not considered reliable at the initial stages of decomposition because he has considered first order kinetics. As described previously, only one kinetic expression is not helpful to represent the coal pyrolysis. Equation (4) and (7) are generalized as follows to allow for nonisothermal processes:

$$V_i^* - V_i = V_i^* \exp\left(-\int_0^t k_{ri} dt\right) \quad (8)$$

$$\frac{V^* - V}{V^*} = \int_0^\infty \exp\left[-\int_0^t k_r dt\right] f(E_v) dE_v \quad (9)$$

In addition, $f(E_v)$ can be approximated as a gaussian distribution with mean activation energy E_{v0} and standard deviation σ_v . Thus

$$f(E_v) = [\sigma_v (2\pi)^{\frac{1}{2}}]^{-1} \exp\left(-\frac{(E_v - E_{v0})^2}{2\sigma_v^2}\right) \quad (10)$$

Assuming that $k_{ri} = k_{r0i} \exp(-E_{vi} / RT)$ with k_{r0i} being a constant having the same value (k_{r0}) for all i , Equations (9) and (10) give

$$\frac{V^* - V}{V^*} = \frac{1}{\sigma_v (2\pi)^{1/2}} \int_0^\infty \exp[-k_{r0} \left(\int_0^t \exp\left(\frac{-E_v}{RT}\right) dt\right) - \frac{(E_v - E_{v0})^2}{2\sigma_v^2}] dE_v \quad (11)$$

Equations (10) and (11) permit the study of coal decomposition data under nonisothermal condition by evaluating its four kinetic parameters (V^* , E_{v0} , σ_v , k_{r0}). The equation (11) is solved by using the genetic algorithm technique in order to find the optimum values of the three parameters by using the experimental results from thermogravimetric analysis.

Genetic Algorithm

Genetic algorithm differs from classical optimization methods in that there is a non-zero 'probability' of attaining the global optimum. Genetic algorithms, on the other hand, are simple to implement and involve evaluations of only the objective function and the use of certain genetic operators to explore the design search space. The genetic algorithm is a probabilistic technique that uses a population of design variables rather than a single design variable at a time. It is analogous to natural selection in the evolution of living organisms in that the fittest members in the population have a better chance to survive, reproduce and thus transfer their genetic material to the successive generations. The initial population is formed by a set of randomly generated members. Each generation consists of members whose constituents are the individual variables that characterize a design variable and these are embedded in a binary string. Each member is evaluated using the objective function and is assigned a fitness value, which is an indication of the performance of the member relative to the other members in the population. A biased selection depending

on the fitness value, decides which members are to be used for producing the next generation. The selected strings are the parents for the next generation, which evolves from the use of two genetic operators namely crossover and mutation. These operators give a random displacement to the parent population and generate a new population of design variables. The crossover operator takes two parent strings, splits them at a random location and swaps the sub-strings so formed. A probability of crossover determines whether a crossover should be performed. The mutation operator inverts a bit in the string depending on the probability of mutation. The new strings formed are evaluated and the iteration continues until a maximum number of generations have been reached or until a user defined termination criterion has been met. The control parameters that have to be initially specified are the population size, the crossover and mutation probabilities, the maximum number of generations, upper and lower bound of design variables and the termination criterion.

There are many alterations that may have to be introduced into the basic genetic algorithm described above, depending on the problem [20]. For example, the whole population can be used for reproduction (generational replacement), only a part of the population can undergo reproduction (steady state replacement) or the best member in the population can be passed on to the next generation without any changes (elitist selection). The crossover operation can occur at a single point or at more than one point (multi point crossover). The fitness function may be based on the objective function value or on the position of the member in the population (linear normalization). The control parameters of the genetic algorithm may be fixed at a particular value or can be made to vary as the genetic algorithm progresses. Real numbers may be used for representing the design variables or they may be mapped to a binary string. There is no single variation that out-performs the others for all types of problems and therefore the designer has to decide as to which variations to implement.

The initial population for the genetic algorithm is randomly generated within a given search space. The design variables can be represented either as binary strings or as real numbers. The advantage of the binary representation is that conventional crossover and mutation operations become very simple. The disadvantage is that the binary numbers have to be converted to real numbers when the design variable is to be evaluated. Moreover, binary operations consume a lot of computer memory as well as computation time. On the other hand if crossover and mutation operators can be formulated to act on real numbers, the binary representation can be eliminated. At each of the randomly generated design variable the objective function is evaluated. The problem in hand is maximization problem; the objective function value cannot be used as a measure of fitness of the design variable. These have to be transformed so that a better design variable will have a higher fitness value. This can be done in two ways [20]. First, fitness can be determined by scaling the objective function value and assigning an inversely proportional number. This is a simple method but may lead to premature convergence when a single member dominates the population. The second method is to rank each design variable in the population in the ascending order of the objective function value and assign a fitness value proportional to the rank. This ensures that design variables, which are neither bad nor very good, remain in the competition to reproduce thus preventing the domination of the population by a single relatively exceptional design variable. In this research, binary number representation and ranking of the design variable has been used.

The selection of members for the next generation is done by the Roulette Wheel selection method [21] wherein members with high fitness value have a higher probability of getting replicated in the next generation. The selected members are then operated on by one of the two genetic operators, crossover or mutation, both of which have a user-defined probability of occurrence. The crossover operator has been formulated to use binary strings. It takes two design variables and compares their objective function values. The difference between the corresponding design variables is determined and each design variable is given an increment proportional to this difference. If this increment results in an infeasible design variable, the

value of the increment is reduced until a feasible design variable is generated. The amount of increment and the rate of decrease of the increment are parameters that have to be initially specified. This is analogous to the direct search procedure in numerical optimization. The conventional mutation operator acts on individual bits of the binary string associated with a design variable and inverts the bit if a probability test is satisfied. In effect, it generates a new design variable from the old design variable. The design variables resulting from the crossover or mutation operation are evaluated and subjected to the selection procedure. This iteration process continues until a maximum number of generations have been reached. In the present study, the fitness (objective) function used is:

Maximize:

$$F = -Abs\left\{\sum_{i=1}^n [(V/V^*)_{Exp.} - (V/V^*)_{GA}]\right\} \quad (12)$$

Subject to: (for non-isothermal case only)

$$LB \leq k_{r0}, E_{vo}, \sigma_v \leq UB \quad (13)$$

The GA generates the random number in between the LB and UB of the variables, which are to be optimized. By using the time temperature history and genetic algorithm, it is very easy to calculate the (V/V^*) by Equation (11).

Results and Discussion

A typical analysis of coal, used for experimentation, is given in Table 1. The thermogravimetric analysis is used to obtain the volatile loss data. The coal size used was -72 + 100. The results are taken at different linear heating rates like 20, 30, 40 °C/min. This is shown in Fig. 2. Also in Fig. 3 is shown some volatile loss data vs. temperature history at non-linear heating rates.

Table 1 Analysis of Indian coal

Proximate Analysis		Ultimate Analysis (d.m.m.f basis)	
Moisture	4 %	Carbon	71.0 %
Volatile Matter	25 %	Hydrogen	3.9 %
Ash	11 %	Oxygen	11.35 %
Fixed Carbon	60 %	Nitrogen	1.75 %
		Sulphure	1.0 %

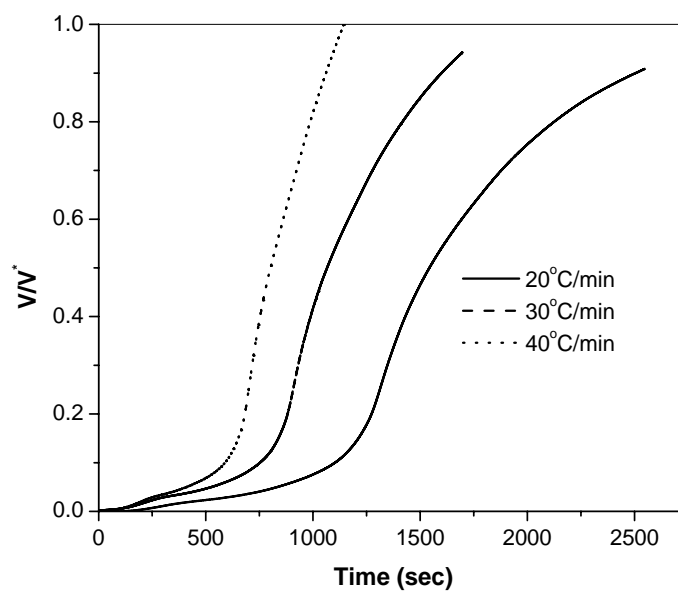


Fig. 2 Plot of V/V^* vs. time for linear heating rate

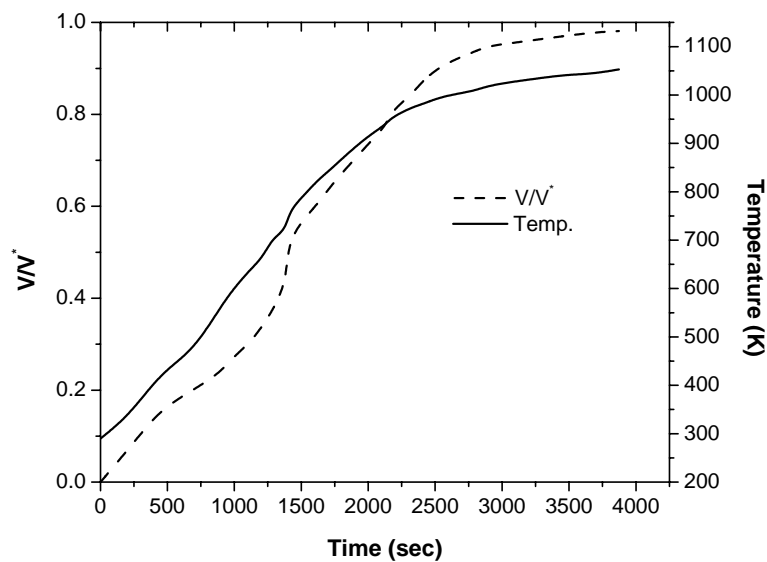


Fig. 3 Plot of volatile loss and temperature vs. time for non-linear case

In order to calculate the kinetic parameters of devolatilisation of coal, the genetic algorithm technique described above is used. A user must be familiar with the lower and upper bound of the variables, which are to be optimized by using the GA method. The user then can adjust the values of the genetic operators according to his/her own choice. In nature the crossover probability is much higher near about 0.8 to 0.9 as compared to the mutation probability which having the value of 0.001. The values of the kinetic parameters obtained from the genetic algorithm for different cases are given in the Table 2.

Table 2 Optimum values of kinetic parameters for different cases

Case	k_{r0} (s^{-1})	E_{v0} (cal/mol)	σ_v (cal/mol)
Non-Isothermal	5.6562E+16	70442.11	19303.859
20°C/min	1E+5	28897.4	6840.99
30 °C/min	1E+5	28560.4	5086.71
40 °C/min	1E+9	43065.9	6622.76

It is revealed from Table 2 that the kinetic parameters evaluated vary widely with the rate of heating. Fig.4 shows that how genetic algorithm improves the search as the number of generations increases. Finally the best fitness and the average fitness of the generation are nearly equal. The distributed activation energy function obtained by using the values of E_{v0} and σ_v for each case is shown in Figs. 5 and 6 show comparison between the theoretical (V/V^*) profiles obtained by using the Equation (11) with the kinetic parameters determined above and the experimental profiles. It is observed that the theoretically obtained profiles are reasonably in good agreement with the experimental profiles. It is not possible to compare the obtained results with the results obtained by the Miura because both methods are totally different in solution procedure.

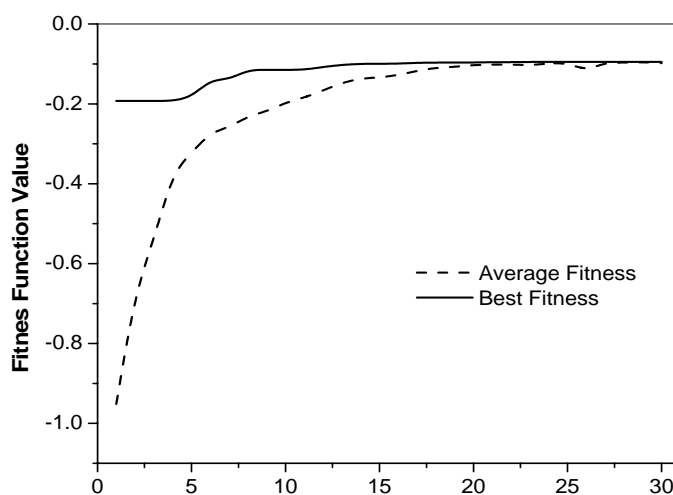


Fig. 4 Fitness function vs. generation number

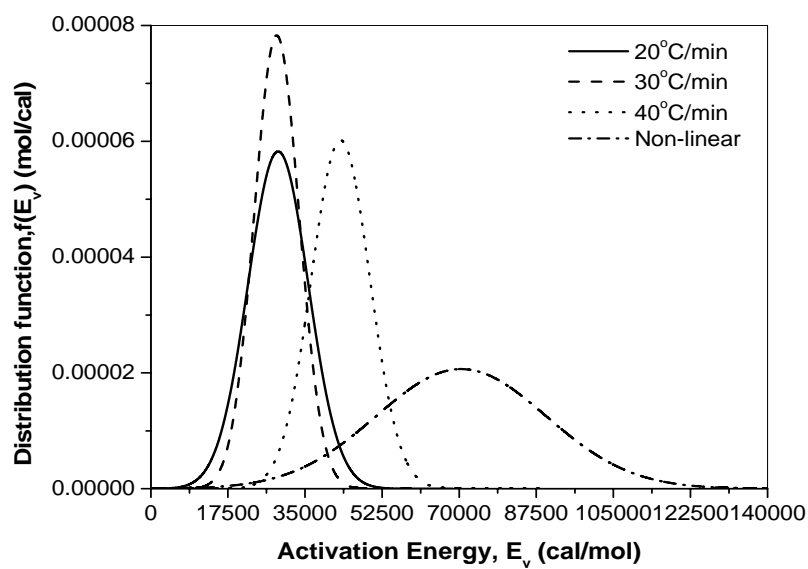


Fig. 5 Plot of activation energy distribution function for different cases

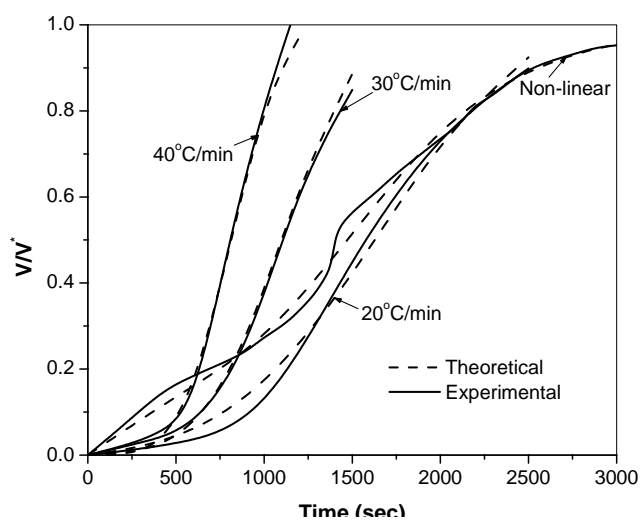


Fig. 6 Comparison of theoretical and experimental results

GA is useful for analyzing thermogravimetric data for evaluation of kinetic parameters either with linear or non-linear rate of heating of coal once we know the time – temperature history and volatile loss data. But the Miura method is applicable only for the linear heating rate while the present method is valid both for isothermal and non-isothermal heating rates.

Conclusion

Evaluation of kinetic parameters of coal is a multivariable optimization problem, which can be handled very well by the genetic algorithm. Genetic algorithm can be applied for all problems which having the very large search space for optimization. Genetic algorithm gives the better results, which are in good agreement with the experimental data obtained in the present case of a typical Indian coal. The kinetic parameters of devolatilisation vary widely with the rate of heating of the coal. This variation in k_{r0} is from $1\text{E}+5$ to $1\text{E}+16\text{ s}^{-1}$, E_{v0} from 28,000 to 70,500 cal/mol and σ_v from 5,000 to 19,000 cal/mol. As compared to the method presented by the Miura, in 1995, this method has an advantage that it can be applied for the non-linear heating rate as well. Comparison of the results obtained by GA methods with those obtained by Miura method seems inappropriate, as they are totally different from solution point of view.

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