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# Isoconversional Kinetic Analysis of Distributed Activation Energy Model Processes for Pyrolysis of Solid Fuels

Weixuan Wu,<sup>†</sup> Junmeng Cai,<sup>\*,†,‡</sup> and Ronghou Liu<sup>†</sup>

<sup>†</sup>Biomass Energy Engineering Research Center, Key Laboratory of Urban Agriculture (South) Ministry of Agriculture, School of Agriculture and Biology, Shanghai Jiao Tong University, 800 Dongchuan Road, Shanghai 200240, People's Republic of China

<sup>‡</sup>State Key Laboratory of Heavy Oil Processing, China University of Petroleum, 18 Fuxue Road, Beijing 102249, People's Republic of China

**ABSTRACT:** Seven different distributed activation energy model (DAEM) processes, which were used to describe the pyrolysis kinetics of wood, algae, lignin, corn stalk skin, kerogen, cellulose, and coal, were analyzed by means of the Friedman differential isoconversional method. It has been shown that the activation energies obtained from the Friedman method are independent of the heating rate. For all DAEM processes considered, the effect activation energies evaluated by the Friedman isoconversional method showed significant dependence on the conversion. The kinetic parameters obtained by means of the Friedman isoconversional method were used to reconstruct the conversions and rates data. The agreement between the reconstructed data and the data calculated by the DAEM is excellent at the presimulated and extrapolated conditions for all the DAEM processes. This indicates the DAEM process can be taken as an example to check the accuracy of some integral isoconversional methods in the determination of the activation energy.

## 1. INTRODUCTION

The pyrolysis of solid fuels has attracted much attention because it is the process to produce liquid fuels and some valuable chemicals. What's more, the pyrolysis process is the first step in the combustion process. The knowledge of the pyrolysis kinetics of solid fuels is fundamental in the modeling of pyrolysis processes and the design of the corresponding reactors. The isoconversional methods are the most commonly used for performing the kinetic analysis of the pyrolysis of solid fuels, such as coal and biomass. According to ISI Web of Science database, more than 1000 citations can be found in the literature for those isoconversional kinetic methods. Several papers on this topic have been recently published in this journal.<sup>1</sup>

The advantage of the isoconversional methods is that they can be used for describing the varying activation energy processes. Some papers stated that the isoconversional methods are undoubtedly the quickest way to derive kinetic parameters for complex reaction profiles involving multiple processes.<sup>2</sup> However, Criado et al.<sup>3</sup> employed some isoconversional methods to perform a critical study of the use of isoconversional methods for the kinetic analysis of competitive reactions and found that the activation energies obtained from the isoconversional methods are dependent on the heating rate. Cai et al.<sup>1b</sup> carried out a critical study of parallel and successive reactions by means of the iterative linear integral isoconversional method<sup>4</sup> and found that the activation energies obtained by means of the isoconversional methods are dependent on the range of heating rates for those complex processes. However, the basic assumption of the isoconversional methods is that the reaction model and the kinetic parameters at a certain extent of conversion are independent of the heating rate.<sup>2c</sup> Thus, considering the apparent activation energy obtained from the isoconversional methods as the real value of this parameter has

no physical meaning if the competitive, parallel, or successive reactions are concerned.

The distributed activation energy model (DAEM) process is a multiple parallel reaction process. The DAEM assumes the pyrolysis mechanism of solid fuels take a large number of independent, parallel, first or *n*th order reactions with different activation energies reflecting variations in the bond strengths of species composing solid fuels (such as coal, biomass, kerogen).<sup>5</sup> The systematic analysis of the DAEM process by means of isoconversional methods is still missing in the literature. This is the scope of the present work.

## 2. DISTRIBUTED ACTIVATION ENERGY MODEL

The DAEM generally holds a basic assumption that an unlimited number of parallel single step decomposition reactions take place and that the reactivity distribution can be represented by a continuous distribution of activation energies. The DAEM for the pyrolysis of solid fuels can be applied to either the total amount of volatiles released or to the amount of an individual volatile constituent.<sup>6</sup> The derivation of the DAEM can be found in the literature.<sup>7</sup>

$$\alpha = \begin{cases} 1 - \int_0^\infty \left[ 1 - (1-n) \int_0^T \frac{A}{\beta} \exp\left(-\frac{E}{RT}\right) dT \right]^{1/(1-n)} g(E) dE & n \neq 1 \\ 1 - \int_0^\infty \exp\left[- \int_0^T \frac{A}{\beta} \exp\left(-\frac{E}{RT}\right) dT\right] g(E) dE & n = 1 \end{cases} \quad (1)$$

where  $\alpha$  is the degree of conversion,  $n$  is the reaction order,  $A$  is the frequency factor,  $\beta$  is the heating rate,  $E$  is the activation energy,  $R$  is the universal gas constant,  $T$  is the temperature, and  $g(E)$  is the activation energy distribution.

Differentiating eq 1 with respect to  $T$ , the rate of reaction is then

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Table 1. Summaries of the Parameter Values of Seven Different DAEM Processes

process	activation energy distribution	frequency factor	reaction order	ref
1	Gaussian distribution $g(E) = \frac{1}{\sqrt{2\pi}\sigma} \exp\left[-\frac{(E - E_0)^2}{2\sigma^2}\right]$ $E_0 = 123.52 \text{ kJ mol}^{-1}, \sigma = 5.97 \text{ kJ mol}^{-1}$ ( $E$ is expressed in $\text{kJ mol}^{-1}$ )	$1.57 \times 10^8 \text{ s}^{-1}$	first-order	11
2	Gaussian distribution $g(E) = \frac{1}{\sqrt{2\pi}\sigma} \exp\left[-\frac{(E - E_0)^2}{2\sigma^2}\right]$ $E_0 = 190.02 \text{ kJ mol}^{-1}, \sigma = 14.73 \text{ kJ mol}^{-1}$ ( $E$ is expressed in $\text{kJ mol}^{-1}$ )	$2.6021 \times 10^{16} \text{ s}^{-1}$	6.63	12
3 <sup>a</sup>	Gaussian distribution $g(E) = \frac{1}{\sqrt{2\pi}\sigma} \exp\left[-\frac{(E - E_0)^2}{2\sigma^2}\right]$ $E_0 = 135 \text{ kJ mol}^{-1}, \sigma = 15 \text{ kJ mol}^{-1}$ ( $E$ is expressed in $\text{kJ mol}^{-1}$ )	$A = A_0 T^m$ $A_0 = 92670, m = 2.3$ ( $A$ is expressed in $\text{min}^{-1}$ )	first-order	13
4	Gaussian distribution $g(E) = \frac{1}{\sqrt{2\pi}\sigma} \exp\left[-\frac{(E - E_0)^2}{2\sigma^2}\right]$ $E_0 = 134 \text{ kJ mol}^{-1}, \sigma = 2.1 \text{ kJ mol}^{-1}$ ( $E$ is expressed in $\text{kJ mol}^{-1}$ )	$A = e^{a+bE}$ $a = 2.0$ $b = 0.146$ ( $A$ is expressed in $\text{s}^{-1}$ and $E$ is expressed in $\text{kJ mol}^{-1}$ )	first-order	14
5	Weibull distribution $g(E) = \frac{\lambda}{\eta} \left( \frac{E - \gamma}{\eta} \right)^{\lambda-1} \exp\left[-\left(\frac{E - \gamma}{\eta}\right)^\lambda\right]$ $\lambda = 15.8, \gamma = 34.1 \text{ kcal mol}^{-1}, \eta = 16.5 \text{ kcal mol}^{-1}$ ( $E$ is expressed in $\text{kcal mol}^{-1}$ )	$7.54 \times 10^{15} \text{ s}^{-1}$	first-order	15
6	logistic distribution $g(E) = \frac{\pi}{\sqrt{3}\sigma} \frac{\exp\left[-\frac{\pi(E-\mu)}{\sqrt{3}\sigma}\right]}{\left\{1 + \exp\left[-\frac{\pi(E-\mu)}{\sqrt{3}\sigma}\right]\right\}^2}$ $\mu = 258.5718 \text{ kJ mol}^{-1}, \sigma = 2.6601 \text{ kJ mol}^{-1}$ ( $E$ is expressed in $\text{kJ mol}^{-1}$ )	$1.6218 \times 10^{17} \text{ s}^{-1}$	1.1101	16
7	double-Gaussian distribution $g_1(E) = \frac{1}{\sqrt{2\pi}\sigma_1} \exp\left[-\frac{(E - E_{01})^2}{2\sigma_1^2}\right]$ $g_2(E) = \frac{1}{\sqrt{2\pi}\sigma_2} \exp\left[-\frac{(E - E_{02})^2}{2\sigma_2^2}\right]$ $g(E) = w g_1(E) + (1-w) g_2(E)$ $E_{01} = 202 \text{ kJ mol}^{-1}, \sigma_1 = 7.33 \text{ kJ mol}^{-1}$ $E_{02} = 237 \text{ kJ mol}^{-1}, \sigma_2 = 52 \text{ kJ mol}^{-1}$ $w = 0.442$ ( $E$ is expressed in $\text{kJ mol}^{-1}$ )	$8 \times 10^{12} \text{ s}^{-1}$	first-order	17

<sup>a</sup>The values of  $\sigma$  and  $m$  are different from those in the literature.

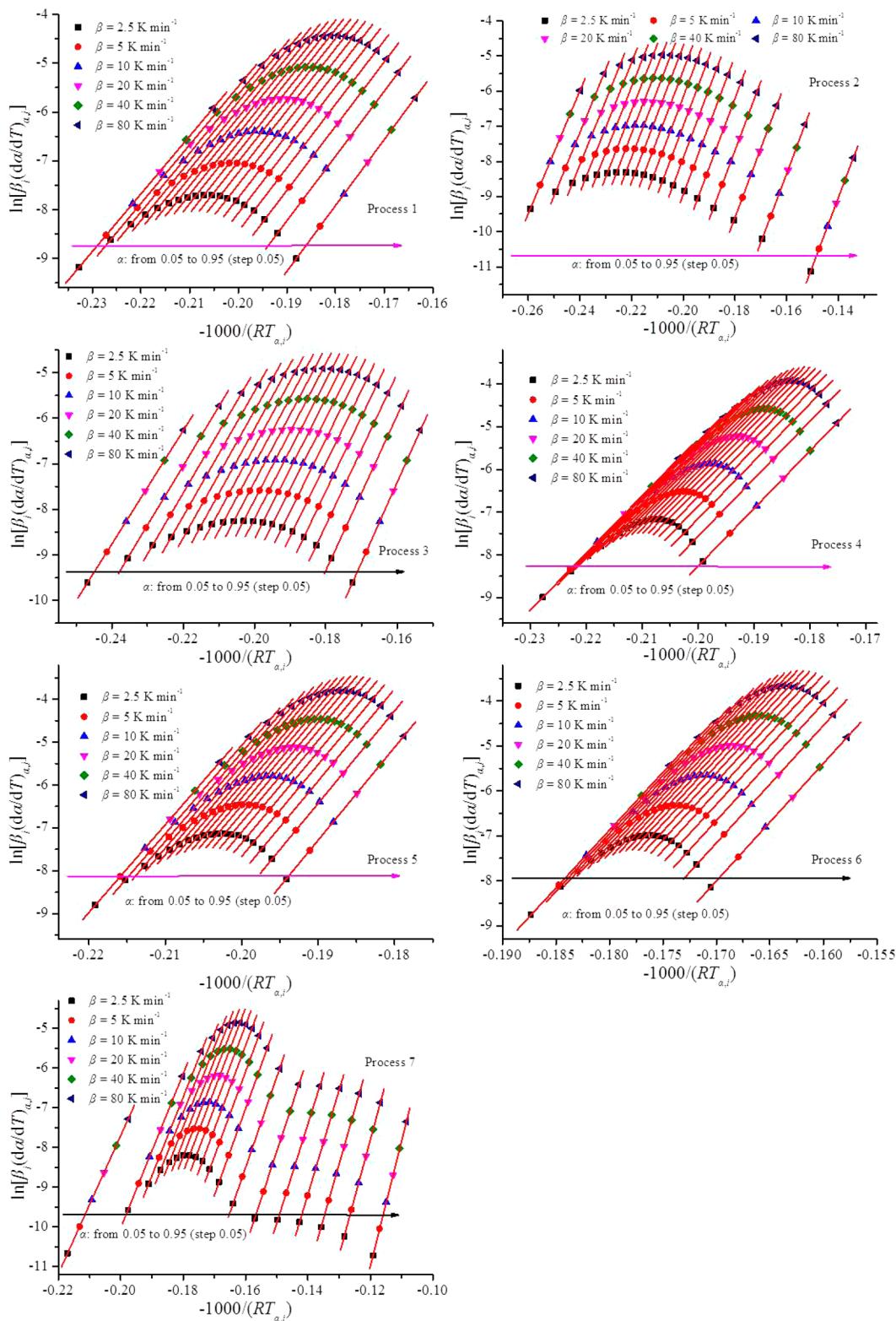
$$\frac{d\alpha}{dT} = \begin{cases} \int_0^\infty \frac{A}{\beta} \exp\left(-\frac{E}{RT}\right) \left[1 - (1-n) \int_0^T \frac{A}{\beta} \exp\left(-\frac{E}{RT}\right) dT\right]^{n/(1-n)} g(E) dE & n \neq 1 \\ \int_0^\infty \frac{A}{\beta} \exp\left(-\frac{E}{RT} - \int_0^T \frac{A}{\beta} \exp\left(-\frac{E}{RT}\right) dT\right) g(E) dE & n = 1 \end{cases} \quad (2)$$

Equations 1 and 2 have no closed form solutions, but they can be numerically solved. A computer program was written in the MATHEMATICA software system to perform the numerical calculations of eqs 1 and 2 where the inner  $dT$  integral and the outer  $dE$  integral were calculated by means of the 'NIntegrate' function of the MATHEMATICA software system.

### 3. FRIEDMAN ISOCONVERSIONAL METHOD

Since several years isoconversional methods have been widely used to study the kinetics of solid-state reactions by means of thermoanalytical methods.<sup>8</sup> For nonisothermal data, the

most popular methods are represented by the Friedman differential method and by the Ozawa–Flynn–Wall (OFW) and Kissinger–Akahira–Sunose (KAS) integral methods.<sup>9</sup> The OFW and KAS integral methods were derived assuming a constant activation energy.<sup>10</sup> This assumption obviously introduces some systematic error in estimating the activation energy values, if the latter varies with the degree of conversion.<sup>3</sup> This error does not appear in the Friedman method. For this reason, the Friedman method was used in this study.



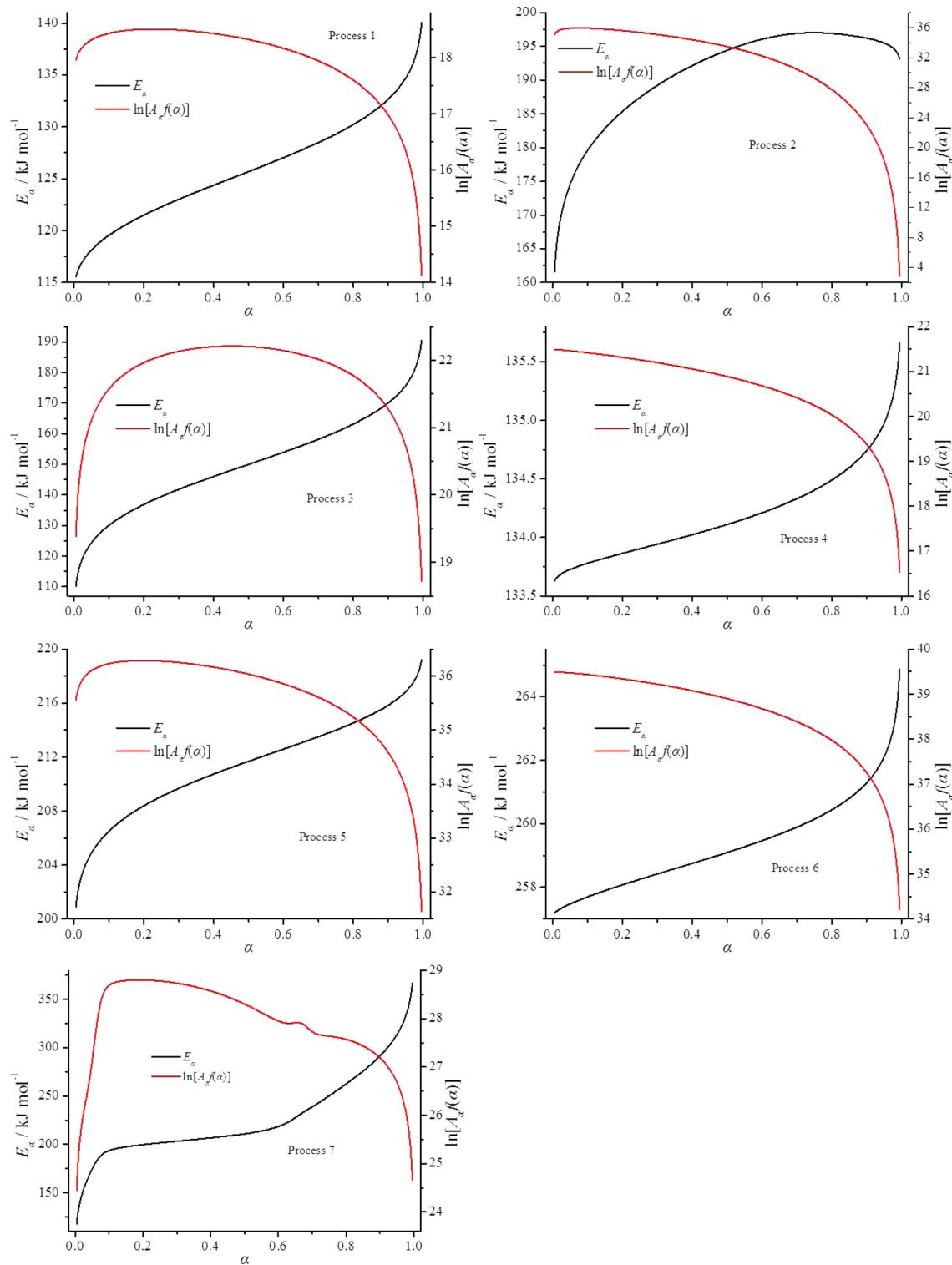
**Figure 1.** Dependence of  $\ln[\beta_i(d\alpha/dT)_{\alpha,i}]$  vs  $-1000/RT_{\alpha,i}$  at the different  $\alpha$  values for all the used heating rates (solid lines are linear fitting corresponding to different  $\alpha$  values).

The Friedman differential isoconversional method is based on the general equation in the following form:

$$\beta \frac{d\alpha}{dT} = A e^{-E/RT} f(\alpha) \quad (3)$$

where  $f(\alpha)$  is the reaction model. Using eq 3, the Friedman method that allows the activation energy to be obtained for each conversion degree, can be expressed as

$$\ln\left(\beta_i \frac{d\alpha}{dT}\Big|_{\alpha,i}\right) = \ln[A_f(\alpha)] - \frac{E_a}{RT_{\alpha,i}} \quad (4)$$



**Figure 2.** Obtained  $E_a$  and  $\ln[A_\alpha f(\alpha)]$  values as a function of  $\alpha$  for seven simulated DAEM processes.

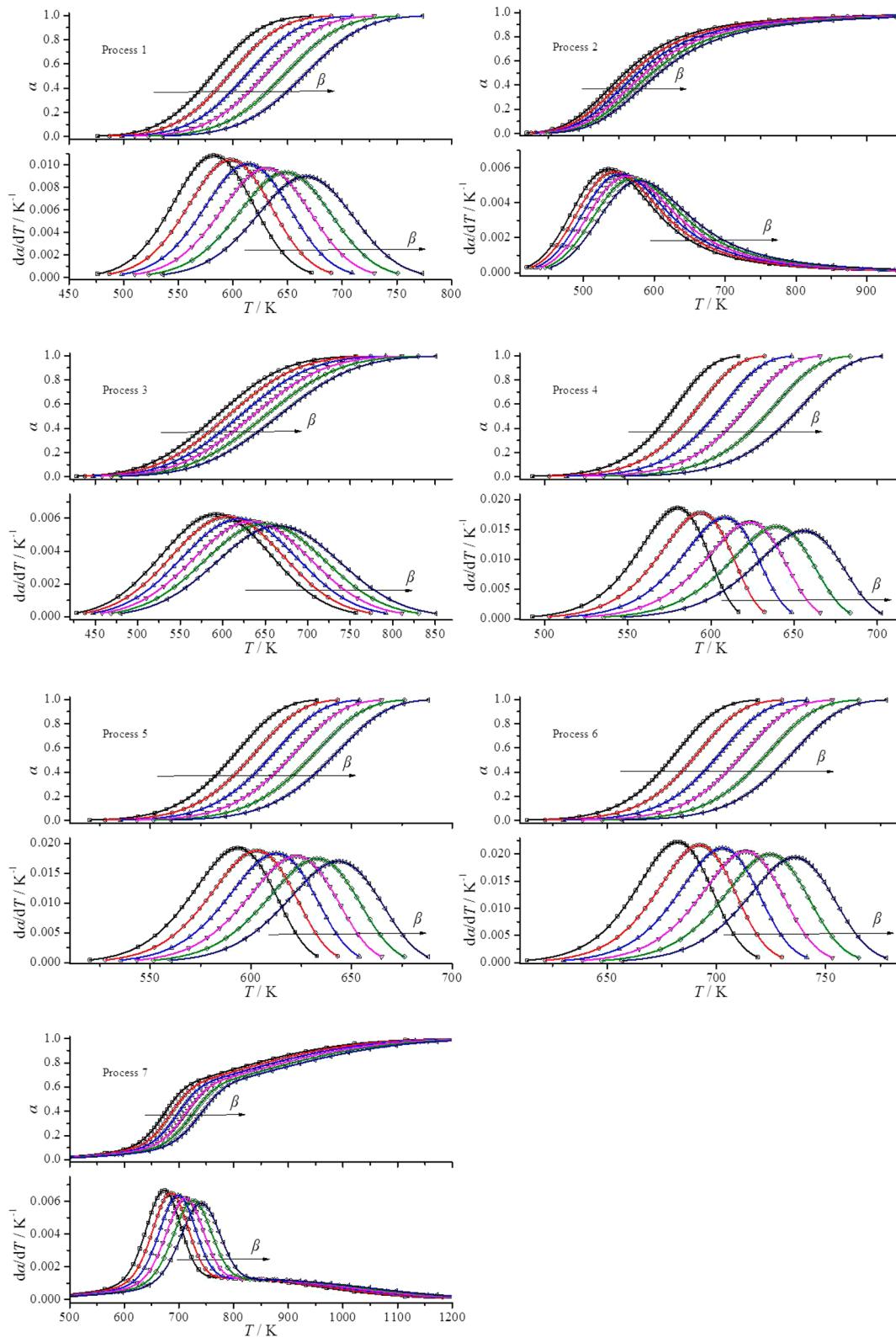
where the subscript  $i$  denotes the ordinal number of a non-isothermal experiment conducted at the heating rate  $\beta_i$  and the subscript  $a$  is the quantities evaluated at a specific conversion degree  $\alpha$ . For a given  $\alpha$ , the plot  $\ln[\beta_i(d\alpha/dT)_{\alpha,i}]$  versus  $(-1/RT_{\alpha,i})$  should be a straight line whose slope and intercept can be used to estimate  $E_a$  and  $\ln[A_\alpha f(\alpha)]$ , respectively.

The  $\alpha - T$  and  $d\alpha/dT - T$  data can be reconstructed based on the obtained  $E_a$  and  $\ln[A_\alpha f(\alpha)]$  values using eq 4. A program coded in MATLAB using the function ‘ode45’ was used to solve

the ordinary differential eq 4. The procedure for reconstruction can also be used to obtain conversions and rates at extrapolated conditions.

#### 4. RESULTS AND DISCUSSION

In order to carry out the systematic analysis of nonisothermal DAEM processes by means of isoconversional methods, some simulated nonisothermal DAEM processes were investigated. In those processes, different models were considered and the

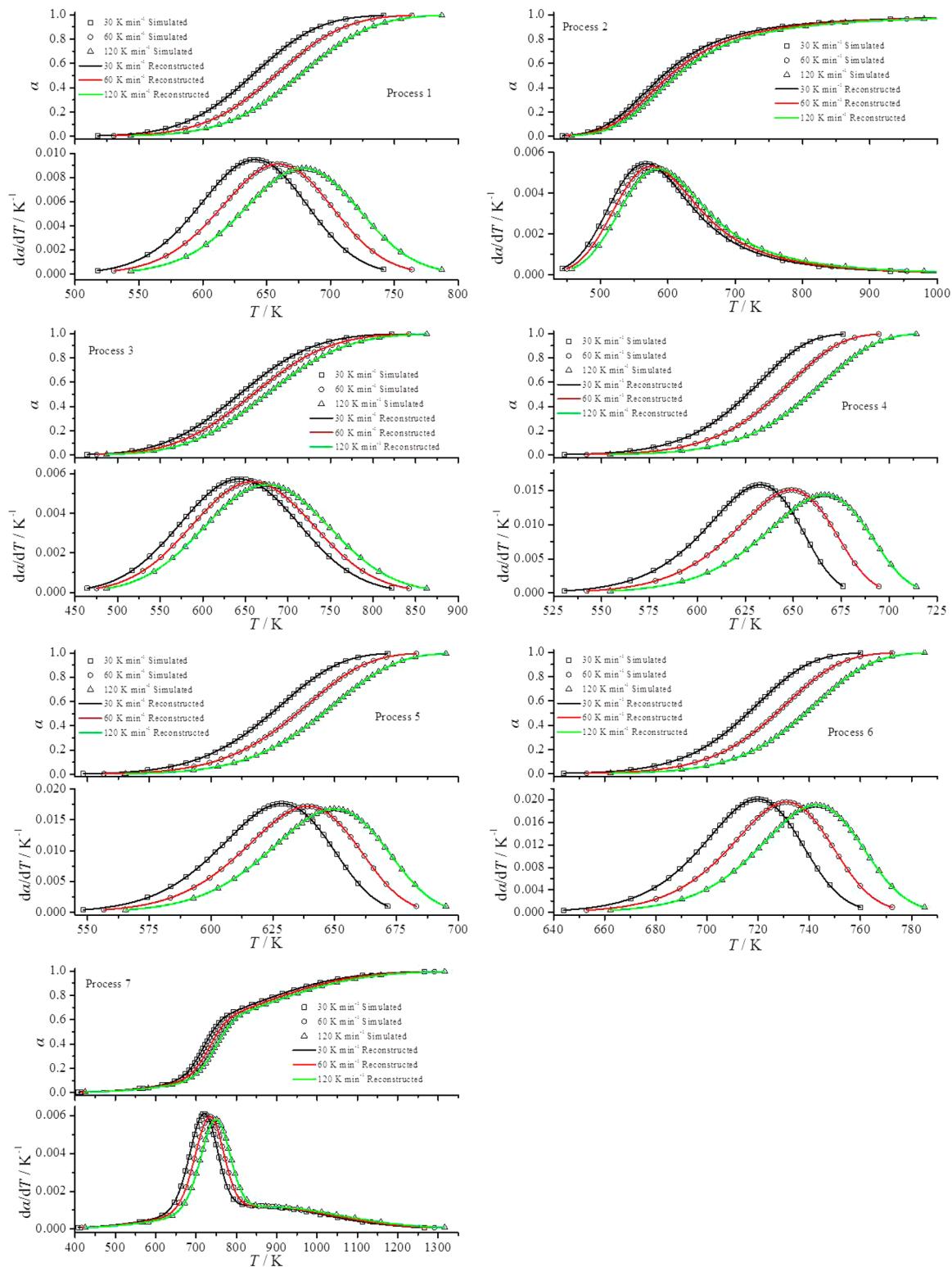


**Figure 3.** Comparison of the  $\alpha - T$  and  $d\alpha/dT - T$  data simulated from the DAEM and the curves obtained from the procedure for reconstruction at the heating rates of 2.5, 5, 10, 20, 40, and 80  $K \text{ min}^{-1}$ . The points present the data simulated from the DAEM and the solid lines represent the curves obtained from the reconstruction.

corresponding kinetic parameters were taken from the literature, see Table 1. They were used to describe the pyrolysis kinetics of wood,<sup>11</sup> algae,<sup>12</sup> lignin,<sup>13</sup> corn stalk skin,<sup>14</sup> kerogen,<sup>15</sup> cellulose,<sup>16</sup> and coal.<sup>17</sup> To investigate the effect of the heating rate on the

obtained activation energies, the simulated data at six heating rates (2.5, 5, 10, 20, 40, and 80  $K \text{ min}^{-1}$ ) for each process were analyzed.

By means of the MATHEMATICA program, the simulated  $\alpha - T$  and  $d\alpha/dT - T$  data for all processes are obtained.



**Figure 4.** Comparison of the  $\alpha - T$  and  $d\alpha/dT - T$  data simulated from the DAEM and the curves obtained from the procedure for reconstruction at the extrapolated heating rates of 30, 60, and 120 K min<sup>-1</sup>. The points present the data simulated from the DAEM and the solid lines represent the curves obtained from the reconstruction.

Based on the simulated  $\alpha - T$  and  $d\alpha/dT - T$  data, the Friedman plots ( $\ln[\beta_i(d\alpha/dT)_{\alpha_i}]$  versus  $(-1/RT_{\alpha_i})$ ) at different  $\alpha$  values ( $\alpha$  varies from 0.05 to 0.95 with a step 0.05) for all processes are shown in Figure 1. For each process, the perfect linear relationship was obtained in the Friedman plot ( $\ln[\beta_i(d\alpha/dT)_{\alpha_i}]$

versus  $(-1/RT_{\alpha_i})$ ) for the whole set of heating rates considered. This indicates that the activation energies obtained by the Friedman method are independent of the range of heating rates.

Based on the slope and intercept of the fitting straight line of the Friedman plot,  $E_\alpha$  and  $\ln[A_\alpha f(\alpha)]$  as a function of

conversion were obtained and shown in Figure 2, where it can be seen that the activation energies obtained from the Friedman method are strongly dependent upon the degree of conversion for each DAEM process.

The obtained  $E_\alpha$  and  $\ln[A_\alpha f(\alpha)]$  values obtained from the Friedman method were used to reproduce the data. The resulting data for all DAEM processes are compared in Figure 3 for the  $\alpha - T$  and  $d\alpha/dT - T$  curves at the heating rates of 2.5, 5, 10, 20, 40, and  $80 \text{ K min}^{-1}$ . In Figure 3, the points represent the data obtained from the numerical calculations of the DAEM, and the solid lines represent the data obtained from the procedure for reconstruction. From this figure, it can be seen that the  $\alpha - T$  and  $d\alpha/dT - T$  curves obtained from the Friedman reconstruction are in excellent agreement with those expected from the data numerically calculated from the DAEM. A comparison of the reconstructed data and the data obtained from the numerical calculation of the DAEM at the heating rates of 30, 60, and  $120 \text{ K min}^{-1}$  was done to verify whether the procedure for reconstruction could be used at extrapolated conditions as shown in Figure 4. It can be observed the reconstructed curves fit the simulated data very well.

According to the above analyses, the DAEM process is equivalent to a real varying activation energy process. In the DAEM process, a number of single processes overlap, and the varying activation energies have the contribution of those processes. Therefore, the activation energies obtained from the Friedman method can be taken as reference values for checking the accuracy of new integral isoconversional methods that take into account the variation of activation energy with conversion when the DAEM process is considered.

The activation energies,  $E_\omega$ , obtained from the Friedman method for the DAEM process can be used for making kinetic prediction. And in the literature,<sup>18</sup>  $d\alpha/dE_\alpha$  can be used to estimate the activation energy distribution,  $g(E)$ , in the DAEM.

## 5. CONCLUSIONS

- (1). Seven different DAEM processes (different activation energy distributions, different frequency factors and different reaction orders) for the pyrolysis of solid fuels were analyzed by means of the Friedman differential isoconversional method. The activation energies obtained from the Friedman method are independent of the range of heating rates for all DAEM processes.
- (2). The DAEM process is equivalent to a real varying activation energy process. The activation energies obtained from the Friedman method can be taken reference values for checking the accuracy of new integral isoconversional methods when the DAEM process is investigated.
- (3). For the DAEM process, the activation energies obtained from the Friedman method can be used for making kinetic predictions.

## ■ AUTHOR INFORMATION

### Corresponding Author

\*Tel.: +86-21-34206624. Fax: +86-21-34205744. E-mail: jmcai@sjtu.edu.cn.

### Notes

The authors declare no competing financial interest.

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