CO₂ Gasification Kinetics of Shenhua Bituminous Coal by Isothermal Thermo-Gravimetric Analysis

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ABSTRACT

This research performed the gasification kinetics of three Shenhua coal under CO₂ atmosphere using isothermal thermogravimetry. Results showed that isothermal gasification curves for three different coal samples revealed different gasification behaviour. Among the eleven kinetic models, A2 was the most suitable one to describe the gasification kinetics of three coal samples, because it can reproduce the experimental data very well with reasonable correlation coefficients. The activation energy for sample A, B and C were 95.9, 79.1, and 69.4 kJ mol⁻¹, respectively. The activation energy decreased with the increase of the particle size. The compensation relationship was observed between activation energy and frequency factor, and the mathematical expression was $\ln A = 0.1041 E + 0.54028$ with the correlation coefficients of 0.999.

Keywords: coal; CO₂ gasification; kinetics

1. Introduction

Coal gasification is a process in which carbon can be transformed into combustible gas such as CO, H₂ and CH₄ through incomplete combustion^[1, 2]. Coal gasification consists of two processes. Firstly, coal pyrolysis occurs, and volatiles and coal char release. Secondly, reactions between coal char and gasifying agent occur, as well as reactions between volatiles and gasifying agent^[3-5]. Coal gasification is typical gas-solid reaction. Due to the fact that the reaction rate of char gasification is much slower than that of coal pyrolysis, the char gasification rate determines the whole carbon conversion rate^[6-8]. The reaction between coal char and CO₂ is one of the most important reactions in gasification process^[9-11]. Therefore, it is of important significance to study

the gasification kinetics between coal char and CO₂^[10, 12, 13]. Many scholars focused on isothermal kinetics at different gasification temperatures by using a series of kinetic models^[3, 14-16]. Many factors such as coal rank^[17-19], temperature^[20], pressure^[21] have effect on coal char gasification rate under CO₂ atmosphere. The coal particle size also presents significant effects on coal pyrolysis^[22, 23] and gasification^[24] properties.

In this work, the gasification kinetic analysis between coal char and CO₂ has been investigated through isothermal thermogravimetry at three different temperatures (900, 950, and 1000°C). Eleven different gas-solid models were used to fit the experimental data and to predict the kinetic parameters. Three coal samples with different particles were used to investigate the effect of particle size on gasification kinetics.

2. Materials and methods

2.1. Materials

Coal used in this work were obtained from Shenhua mine. It was first ground by ball mill and then was screened. Three coal samples with different particle size ranges labelled as A, B and C were used to investigate the effect of particle size on gasification kinetics. Proximate and ultimate analyses of three coal samples were shown in Table 1. Information about particle size distribution was obtained using Malvern Mastersizer 3000. The average particle size of sample C was the highest, followed by sample B, and sample A had the lowest average particle size. The characteristic particle sizes were shown in Table 2.

Table 1. Proximate and ultimate analysis of coal samples.

Sample	Proximate Analysis (ad, %)				Ultimate analysis (daf, %)				
	M	A	VM	FC	C	H	N	S	O*
A	6.80	9.91	27.86	55.43	84.26	7.53	1.07	1.03	6.10
B	7.24	6.83	31.99	53.94	81.98	8.38	0.82	0.37	8.45
C	7.86	7.01	30.84	54.29	82.63	8.77	0.83	0.29	7.48

ad, air dried; daf, dry and ash free basis; *, by difference

Table 2. The average particle size for coal samples used in this work.

Average particle size	A	B	C
$D_v(50)/\mu\text{m}$	43.4	168	293

2.2. Thermogravimetric analysis

The CO₂ gasification characteristics were performed at different temperatures (1000, 950, and

900 °C) by means of a thermogravimetric analyser (LABSYS EVO). In each thermogravimetric experiment, about 8 mg coal sample was used. It was heated under inert atmosphere (N₂) from ambient temperature to the target temperature with the heating rate of 10 °C/min, and then kept at the target temperature for 30 min to ensure the complete of the pyrolysis. At last, the atmosphere of N₂ was changed to CO₂ and the target temperature was kept for further 60 min.

2.3. Isothermal method for kinetic analysis

The the degree of conversion (α) of coal char can be calculated according to the following equation:

$$\alpha = \frac{m_0 - m_i}{m_0 - m_\infty}$$

where m_0 is the initial mass of the coal char, m_i is the real-time mass of the coal char, m_∞ is the final mass at the end of the gasification of the coal char.

The reaction rate equation of CO₂ gasification of coal char can be described by the following equation:

$$\frac{da}{dt} = kf(a) \quad (1)$$

Where t is the time; k represents the gasification rate constant according to the Arrhenius equation, $k=A\exp(-E/RT)$; $f(\alpha)$ is the function which describes the relationship between the reaction function. A group of isotherms have been recorded at different temperatures, which can be fitted to different kinetic models according to the following equation, which have been obtained by integrating Eq.1:

$$\int \frac{da}{f(a)} = \int kdt \quad (2)$$

$$g(a) = kt \quad (3)$$

Where $g(\alpha)$ representing the integral form of the kinetic model. The plot $g(\alpha)$ versus the reaction time leads to a straight line, whose slope is the reaction constant. In consideration of the Arrhenius dependence of the rate constant with the temperature, the activation energy and the frequency factor can be obtained according to the linear regression analysis of rate constants at different temperatures.

$$k = A\exp\left(-\frac{E}{RT}\right) \quad (4)$$

$$\ln k = \ln A - \frac{E}{RT} \quad (5)$$

Table 2 Expressions for $f(\alpha)$ and $g(\alpha)$ based on various reaction mechanisms

Mechanism	$f(\alpha)$	$g(\alpha)$	Abbreviation
Power Law	$2\alpha^{1/2}$	$\alpha^{1/2}$	P2
Power Law	$3\alpha^{2/3}$	$\alpha^{1/3}$	P3
Power Law	$4\alpha^{3/4}$	$\alpha^{1/4}$	P4
Avrami-Eroféve	$2(1-\alpha)[- \ln(1-\alpha)]^{1/2}$	$[- \ln(1-\alpha)]^{1/2}$	A2
Avrami-Eroféve	$3(1-\alpha)[- \ln(1-\alpha)]^{2/3}$	$[- \ln(1-\alpha)]^{1/3}$	A3
Avrami-Eroféve	$4(1-\alpha)[- \ln(1-\alpha)]^{3/4}$	$[- \ln(1-\alpha)]^{1/4}$	A4
Contracting Sphere	$2(1-\alpha)^{1/2}$	$1-(1-\alpha)^{1/2}$	R2
Contracting Cylinder	$3(1-\alpha)^{2/3}$	$1-(1-\alpha)^{1/3}$	R3
One Dimensional Diffusion	$1/2\alpha$	α^2	D1
Three Dimensional Diffusion-Jander	$\frac{3(1-\alpha)^{2/3}}{2[1-(1-\alpha)^{1/3}]}$	$[1-(1-\alpha)^{1/3}]^2$	D3
First Order	$1-\alpha$	$-\ln(1-\alpha)$	F1

3. Results and discussion

3.1. TG curves of coal samples

TG curves of three coal samples at different gasification temperatures were shown in **Figure 1**. Results showed that the TG curve consisted of two stage, the first was the pyrolysis stage, which occurred in the inert atmosphere; the second was the gasification stage which required the participation of CO_2 . The steep TG curve during raising temperature (20-95 min) indicated that the weight of coal sample decreased fast, while during the isothermal process (95-130 min) in inert atmosphere the weight was reduced very slowly suggesting the complete of the pyrolysis. The gasification occurred during the isothermal process in CO_2 atmosphere, the steep TG curve indicated that the gasification rate of coal char was rather large.

Figure 1 TG curves of three coal samples at different gasification temperatures: (a), 1000 °C; (b), 950 °C; (c), 900 °C

Figure 2 displayed three isothermal curves for coal samples, corresponding to the gasification of coal char under CO₂ atmosphere. It can be found that three coal samples revealed different gasification behaviour.

Figure 2 Fit of the gasification isothermal curves to thirteen different kinetic models

3.2. Gasification kinetic analysis of coal samples

The plots of $g(\alpha)$ versus reaction time for each gasification isothermal curve using eleven different

kinetic models were presented as in **Figure 3**. It can be seen that the curves fitted to kinetic model of A, A3, and A4 were more similar to straight lines, indicating that these three models were more suitable for kinetic analysis of Shenhua coal samples.

Figure 3 Fit of the 1000 °C isothermal curves to a set of different kinetic models

The rate constants for sample A, B, and C, which were determined from the slope of the fitted straight line by linear fit, were shown in Table 2, Table 3, and Table 4, respectively, together with the activation energy calculated using each set of reaction constants. Among the eleven kinetic models, A2 was the most suitable kinetic model to describe the gasification kinetics of coal samples, whose coefficient correlation (R^2) for three samples were all equal to 0.99.

Table 2 rate constants and coefficient correlation (R^2) obtained from fitting the experimental curves for sample A in Figure 2 to a set of theoretical kinetic models.

Models	900 °C		950 °C		1000 °C		Result of isothermal analysis		
	R^2	$k(\text{min}^{-1})$	R^2	$k(\text{min}^{-1})$	R^2	$k(\text{min}^{-1})$	$E(\text{kJ mol}^{-1})$	$A(\text{s}^{-1})$	R^2
P2	0.95	0.022	0.92	0.023	0.88	0.024	12.7	0.08	1.00
P3	0.92	0.023	0.89	0.024	0.86	0.024	8.3	0.05	0.99
P4	0.89	0.023	0.87	0.024	0.85	0.025	5.9	0.04	0.99
A2	0.99	0.035	0.99	0.045	0.99	0.075	95.9	615.9	0.90
A3	0.97	0.031	0.98	0.036	0.99	0.061	84.3	164.1	0.81
A4	0.95	0.029	0.95	0.033	0.97	0.055	78.7	86.8	0.74
R2	0.99	0.015	0.99	0.018	0.99	0.030	89.6	135.7	0.88
R3	0.98	0.012	0.99	0.015	0.98	0.026	99.0	284.7	0.93
D1	0.98	0.016	0.98	0.020	0.94	0.023	43.7	1.4	0.99
D3	0.83	0.006	0.92	0.011	0.89	0.019	133.5	5529.9	0.99
D4	0.92	0.004	0.97	0.005	0.95	0.009	107.33	220.3	0.97
F1	0.89	0.053	0.92	0.089	0.89	0.154	131.9	39116.1	0.99

Table 3 rate constants and correlation factors obtained from fitting the experimental curves for sample B in Figure 2 to a set of theoretical kinetic models.

Models	900 °C		950 °C		1000 °C		Result of isothermal analysis		
	R^2	$k(\text{min}^{-1})$	R^2	$k(\text{min}^{-1})$	R^2	$k(\text{min}^{-1})$	$E(\text{kJ mol}^{-1})$	$A(\text{s}^{-1})$	R^2
P2	0.95	0.0221	0.93	0.02269	0.89	0.02416	11.0	0.08	0.87
P3	0.915	0.02306	0.90	0.02338	0.87	0.02442	7.1	0.05	0.81
P4	0.89	0.02359	0.88	0.02376	0.85	0.02457	5.0	0.04	0.72
A2	0.99	0.03546	0.99	0.04023	0.99	0.06743	79.1	110.4	0.75
A3	0.97	0.03127	0.97	0.03387	0.98	0.05353	66.1	25.7	0.68
A4	0.94	0.02952	0.95	0.03123	0.96	0.04797	59.6	12.5	0.64
R2	0.99	0.01499	0.99	0.01691	0.99	0.02687	71.9	22.4	0.76
R3	0.98	0.01191	0.99	0.0139	0.99	0.0232	82.2	50.9	0.80
D1	0.98	0.01668	0.986	0.01872	0.94	0.02278	38.5	0.9	0.94
D3	0.84	0.00669	0.90	0.00898	0.93	0.01744	118.3	1159.2	0.89
D4	0.93	0.00393	0.96	0.00481	0.97	0.00819	90.6	40.0	0.85
F1	0.89	0.05546	0.92	0.072	0.93	0.1443	117.9	9177.4	0.85

Table 4 rate constants and correlation factors obtained from fitting the experimental curves for sample C in Figure 2 to a set of theoretical kinetic models.

	900 °C		950 °C		1000 °C		Result of isothermal analysis		
Models	R^2	$k(\text{min}^{-1})$	R^2	$k(\text{min}^{-1})$	R^2	$k(\text{min}^{-1})$	$E(\text{kJ mol}^{-1})$	$A(\text{s}^{-1})$	R^2
P2	0.96	0.02	0.94	0.02	0.89	0.024	15.1	0.10	0.93
P3	0.92	0.02	0.91	0.02	0.86	0.024	9.9	0.06	0.90
P4	0.90	0.02	0.88	0.02	0.85	0.025	7.2	0.05	0.85
A2	0.99	0.03	0.99	0.041	0.99	0.058	69.4	38.5	0.83
A3	0.98	0.03	0.98	0.03	0.97	0.043	46.8	3.5	0.82
A4	0.95	0.03	0.95	0.03	0.94	0.038	35.2	1.0	0.80
R2	0.99	0.01	0.99	0.02	0.97	0.022	55.9	4.1	0.92
R3	0.97	0.01	0.99	0.01	0.986	0.019	72.6	17.7	0.90
D1	0.97	0.02	0.98	0.02	0.94	0.023	49.6	2.4	0.97
D3	0.79	0.006	0.88	0.008	0.98	0.016	128.1	2698	0.91
D4	0.89	0.003	0.95	0.004	0.96	0.007	85.9	22.3	0.94
F1	0.88	0.05	0.91	0.06	0.96	0.14	134.0	41253	0.84

Further, the plots of $\ln k$ versus the reverse of the temperature (as shown in Eq.5), from which the activation energies were calculated, were presented in Figure 4, Table 2, Table 3, and Table 4.

Figure 4 Plot of $\ln k$ versus the reverse of their corresponding temperature, constructed for every set of rate constants in Table 2, Table 3 and Table 4. The activation energy is extracted from the slope of the plots, being identical in each case.

Plots of α versus the reaction time for each 1000 °C isothermal curve, which were constructed using different kinetic models, were presented in **Figure 5**. It can be seen that the models of A2 could reproduce the conversion curve very well, with the correlation coefficient R^2 of 0.99. All results indicate that the kinetic model of A2 can describe the experimental data very well with reasonable correlation coefficients. The activation energy for sample A, B and C were 95.9, 79.1, and 69.4 kJ mol⁻¹, respectively.

Figure 5 Comparison of experimental data and calculated data using eleven different kinetic models

As for the effect of the particle size on the kinetic parameters, it can be seen from Figure 6 that, the larger the particle size was, the higher the activation energy was. The pre-exponential factor had the similar trend. A semilogarithmic plot of preexponential factor versus the activation energy resulted in a straight line (shown in Figure 7), which was known as the compensation effect^[9]. The mathematical expression was $\ln A = 0.1041 E + 0.54028$ with the correlation coefficients of 0.999.

Figure 6 The effect of the particle size on the kinetic parameters.

Figure 7 Compensation relationships between activation energy and frequency factor

4. Conclusions

Based on isothermal gasification kinetic analysis of three coal samples with different particle sizes, the following conclusions can be drawn:

- (1) The TG curves in this work consisted of two stages, the first was the pyrolysis stage, which occurred in the inert atmosphere; the second was the gasification stage, which required the participation of CO_2 . Three isothermal curves for different coal samples revealed different gasification behaviour.
- (2) Among the eleven kinetic models, A2 was the most suitable kinetic model to describe the gasification kinetics of coal sample, which can describe the experimental data very well with reasonable correlation coefficients. The activation energy for sample A, B and C were 95.9, 79.1, and 69.4 kJ mol^{-1} , respectively. The activation energy decreased with the increase of the

particle size. The Compensation relationship was shown between activation energy and frequency factor, and the mathematical expression was $\ln A = 0.1041 E + 0.54028$ with the correlation coefficients of 0.999.

Competing interests: The authors declare no conflict of interest.

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Figures

Figure 1

TG curves of three coal samples at different gasification temperatures: (a), 1000 oC; (b), 950 oC; (c), 900 oC

Figure 2

Fit of the gasification isothermal curves to thirteen different kinetic models

Figure 3

Fit of the 1000 °C isothermal curves to a set of different kinetic models

Figure 4

Plot of $\ln k$ versus the reverse of their corresponding temperature, constructed for every set of rate constants in Table 2, Table 3 and Table 4. The activation energy is extracted from the slope of the plots, being identical in each case.

Figure 5

Comparison of experimental data and calculated data using eleven different kinetic models

Figure 6

The effect of the particle size on the kinetic parameters.

Figure 7

Compensation relationships between activation energy and frequency factor