

ORIGINAL RESEARCH ARTICLE

Influence of Na_2CO_3 on combustion performance of civil clean coke

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ABSTRACT

Civil clean coke is a low-pollutanted civil clean fuel made of coal and multi-functional composite additives by high-temperature carbonization, which is of positive significance to solve the problem of air pollution caused by civil bulk combustion. However, problems such as high ignition temperature and poor combustion performance exist in the use of civil clean coke in the early stage. In this paper, Lvliang fat coal is taken as the research object. Coke samples are prepared by adding different additives and high-temperature carbonization. The influence of Na_2CO_3 on the combustion performance of civil clean coke is analyzed by TG-DTG, BET, thermodynamics kinetics and other methods. The results show that the civil clean coke prepared by high temperature dry distillation with 1.0% Na_2CO_3 added, the ignition temperature by impregnation method is 53 K lower than that of coke without additives; after adding Na_2CO_3 , the TG and DTG curves of coke shifted significantly to the left, the average combustion rate increased from 0.38 mg/min to 0.75 mg/min, and the comprehensive combustion index increased from $2.01 \times 10^{-10} \text{ mg}^2/(\text{min}^2 \cdot \text{K}^3)$ to $11.14 \times 10^{-10} \text{ mg}^2/(\text{min}^2 \cdot \text{K}^3)$; after adding Na_2CO_3 , the specific surface area of coke is significantly increased and the pore structure is more developed, which promotes the oxygen transfer in the combustion process and significantly reduces the apparent activation energy of the combustion system. The apparent activation energy in the low temperature zone is reduced from 454.28 kJ/mol to 306.85 kJ/mol, and the apparent activation energy in the high temperature zone is reduced from 557.36 kJ/mol to 95.36 kJ/mol. The combustion performance of civil clean coke is improved.

Keywords: Na_2CO_3 ; Civil Clean Coke; Ignition Temperature; TG-DTG; Combustion Characteristics; Apparent Activation Energy; Pollution Control

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1. Introduction

According to statistics, China's annual coal consumption is about 4 billion tons, of which about 20% is used for domestic stoves. Domestic burning of coal has the characteristics of low combustion efficiency and high pollutant emission, which is one of the main reasons for smog^[1,2]. How to solve the problem of raw coal combustion pollution and actively promote the prevention and control of air pollution is imminent. Replacing bulk coal with a clean energy supply (such as electricity, gas, etc.) is one of the important ways to control bulk coal. However, China is geographically complex. In the vast rural areas, rural-urban fringe, and other remote areas, the progress of changing coal to gas and coal to electricity projects has not been completed or the gas supply is tight, and the cost of government subsidies is large. Therefore, using coal-based clean fuel instead of bulk coal has become one of the preferred ways^[3,4].

At present, the carbon-based clean fuels with feasible technology, low cost, and widely recognized by users mainly include civil briquette, civil blue carbon, and civil clean coke^[5]. Among them, civil clean coke is the product of high-temperature carbonization of coal, which has the characteristics of smoke-free, low dust, low sulfur oxide, and nitrogen oxide emissions, and is an effective measure to solve the pollution of bulk coal^[6,7]. Since 2015, more than 700,000 tons of civil clean coke have been promoted in Taiyuan, Yuncheng, Jinzhong and Dalian, and good environmental benefits have been achieved. However, in the process of promotion, it is found that civil clean coke has problems such as high ignition temperature and ignition delay. How to improve the combustion performance of civil clean coke is of great significance to the practical application and promotion of civil clean coke.

So far, most of the combustion performance-related studies have been carried out on the improvement of the combustion performance of coal and semi-coke. Additives that can improve the combustion performance of coal can be roughly divided into alkali metals, alkaline earth metals, transition metals, rare earth metals, waste additives, etc.^[8-10]. Zou *et al.*^[11] studied the effects of four iron-containing substances such as hematite, converter dust, converter mud, and blast furnace dust on the combustion performance of pulverized coal, and found that hematite, converter dust, and converter mud can significantly improve the combustion performance of pulverized coal, while blast furnace dust has an inhibitory effect on the combustion of pulverized coal. Gong *et al.*^[12] studied the catalytic effect of rare earth metal oxide CeO₂ and transition metal oxide Fe₂O₃ on coal combustion, and found that the higher the degree of coalification, the smaller the particle size of coal, and the more obvious the catalytic combustion supporting effect of the two additives. Liu *et al.*^[13] studied the effects of

MnO₂, CaCO₃, and other substances on the combustion performance of lean coal and found that MnO₂ can significantly promote the combustion of lean coal. Chen *et al.*^[14] studied the effect of calci-

um-based additives on the combustion performance of inferior coal with high ash content. The addition of CaCl₂ can improve the combustion efficiency of pulverized coal. Gong *et al.*^[15] added Fe₂O₃ and CeO₂ to the semi-coke, respectively, and the combustion performance of the semi-coke was significantly improved. Zhou *et al.*^[16] studied the effects of Cao, Fe₂O₃, and MnO₂ on the combustion performance of anthracite and semi-coke. The results showed that the improvement of combustion performance was related to the properties of additives, the properties of the fuel itself, and combustion conditions. So far, the research on the combustion performance of civil clean coke has not been reported.

Based on this, in this paper, the civil clean coke is prepared by high-temperature dry distillation after adding additives to coal. The effects of different additives, addition amount, and addition method on the combustion performance of civil clean coke are investigated. The mechanism of Na₂CO₃ improving the combustion performance of civil clean coke is discussed through TG-DTG, BET and thermal analysis kinetics, in order to provide theoretical guidance for the improvement of the combustion characteristics of civil clean coke.

2. Experimental method

The coal used in the experiment is Lvliang fat coal. Samples were prepared according to the national standard method and were grinded to less than 3 mm for standby. See **Table 1** for element and industrial analysis of experimental coal. FeCl₃, CuO, KOH, Na₂CO₃, and CaCO₃ used in the experiment are analytical pure reagents, and the manufacturer is Sinopharm Chemical Reagent Co., Ltd.

The coal samples were mixed by the mechanical mixing method and impregnation method. Mechanical mixing method: accurately weigh certain amounts of additives and mix them with 100 g of air-drying based pulverized coal below 3 mm, and then mix them evenly for standby.

Impregnation method: mix 100 g of air-drying-based pulverized coal below 3 mm with 10 mL of impregnation solution containing certain amounts of additives. After standing for 12 h, dry it

in a 378 K drying oven for 2 h. The impregnated

coal sample is placed in the dryer for use.

Table 1. The ultimate analysis and proximate analysis of testing coal

Sample name	Elemental analysis $w_{ad}/\%$					Industrial analysis $w_{ad}/\%$				$Q_{net,ad}/(\text{MJ}\cdot\text{kg}^{-1})$
	C	H	O	N	S	M	A	V	FC	
Lvliang fat coal	79.25	4.60	4.91	1.38	0.89	0.60	8.37	29.13	61.90	31.35

Add 10 mL of distilled water to the above-mixed coal sample and stir it evenly. Tamp the coal sample through a mold to obtain a cylindrical coal cake (with a bulk density of 1.1 g/cm^3). The coal cake is carbonized in a tubular furnace at high temperatures to obtain civil clean coke.

Temperature rise procedure of tubular furnace: the constant temperature for 10 minutes after rising from room temperature to 1,073 K at a temperature rise rate of 10 K/min, and constant temperature for 60 minutes after rising from 1,073 K to 1,323 K at a temperature rise rate of 5 K/min. Put the prepared coal cake into the high-temperature tubular furnace at 1,073 K, take out the sample after the program is completed, and prepare the sample to obtain the coke analysis sample with a particle size of less than 0.2 mm.

TG and DTG curves were obtained by using Setsys Evolution type high-temperature thermogravimetric analyzer produced by Setaram company. The sample mass is 7 mg, the temperature rise range is room temperature to 1,273 K, the temperature rise rate is 15 k/min, the carrier gas is air, and the carrier gas flow rate is 100 mL/min. In addition, the ignition temperature of coke was measured by Hebi Shenhua ignition point instrument^[17].

The pore structure characteristics of coke samples were tested by ASAP2020 type adsorption instrument produced by Micromeritics in the United States. The mass fractions of C, H, O, N and S of coke samples were determined by Elemental Vario MICRO cube element analyzer.

3. Experimental results and discussion

3.1 Effect of different additives on ignition temperature of civil clean coke

In the experimental coal, FeCl_3 , CuO , KOH , Na_2CO_3 and CaCO_3 (mass ratio of additives to coal samples) were added by the impregnation method,

respectively; then the civil clean coke was obtained by high-temperature carbonization. The experimental coal without additives was also obtained by high-temperature carbonization. The ignition temperature of different civil clean coke is investigated, and the results are shown in **Figure 1**.

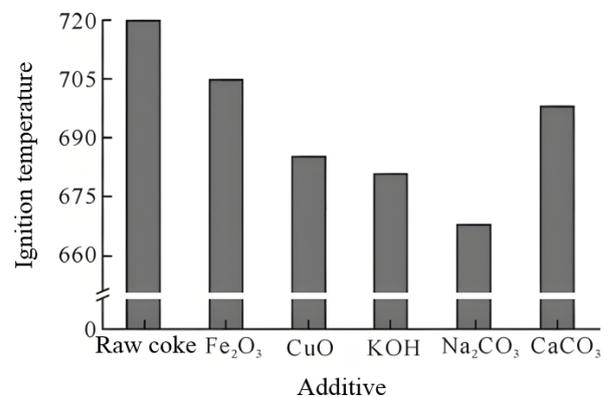


Figure 1. The ignition temperatures of civil clean coke with different additives.

It can be seen from **Figure 1** that after adding different additives, the ignition temperature of civil clean coke decreases to varying degrees; the ignition temperature of the original coke is 720 K. After adding FeCl_3 , CuO , KOH , Na_2CO_3 and CaCO_3 , the ignition temperature decreases by 15, 35, 40, 53 and 22 K, respectively, among which the effect of alkali metal Na_2CO_3 and KOH on the ignition temperature of civil clean coke is more significant. In order to further explore the combustion supporting effect of alkali metals in civil clean coke, Na_2CO_3 , the additive with the largest drop in ignition temperature, is taken as the research object for specific analysis.

3.2 Conditions for Na_2CO_3 to improve combustion performance of clean coke

3.2.1 Effect of Na_2CO_3 addition method on ignition temperature of coke

The additional methods of additives include the mechanical mixing method and the impregnation method. Civil clean coke is prepared by adding 1.0% Na_2CO_3 in two methods respectively. **Figure**

2 shows the effect of the Na_2CO_3 addition method on the ignition temperature of civil clean coke. It can be seen from **Figure 2** that the ignition temperature of the coke sample obtained by the mechanical mixing method is 688 K, which is 21 K higher than that obtained by the impregnation method. This is because Na_2CO_3 is loaded on the surface of coal samples through mechanical mixing, and the impregnation method can make Na_2CO_3 contact with coal particles more closely, and promote the change of carbon skeleton in the process of high-temperature carbonization of coal, which makes the ignition temperature of coke obtained by impregnation method significantly lower than that of mechanical mixing method.

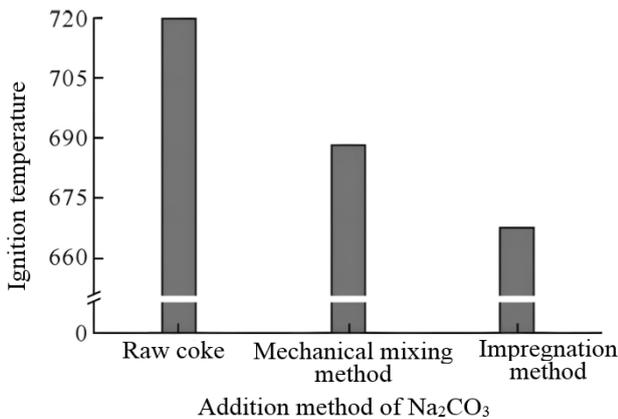


Figure 2. The effect of Na_2CO_3 Adding methods on the ignition temperature of civil clean coke.

3.2.2 Effect of Na_2CO_3 addition amount on ignition temperature of civil coke

Civil clean coke was prepared by adding 0.5%, 1.0%, 1.5%, 2.0% and 2.5% Na_2CO_3 by impregnation method. **Figure 3** shows the effect of Na_2CO_3 addition amount on the ignition temperature of civil clean coke.

It can be seen from **Figure 3** that the ignition temperature of civil clean coke decreases to varying degrees after adding Na_2CO_3 in the impregnation method; with the increase of Na_2CO_3 addition, the ignition temperature of civil clean coke decreases first and then increases; when the addition amount is 1.0%, the ignition temperature drops most significantly; when excessive Na_2CO_3 is added, part of Na covers part of the carbon surface, blocking part of the pores, and the oxygen transfer resistance increases, which hinders the combustion of coke and

makes the ignition temperature rise^[18].

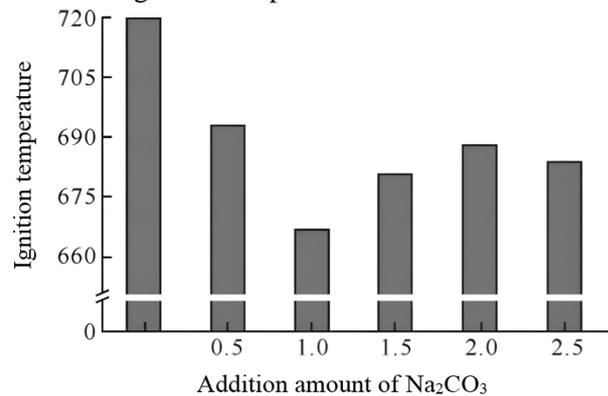


Figure 3. The effect of Na_2CO_3 addition amount on the ignition temperature of civil clean coke.

3.2.3 Effect of adding 1.0% Na_2CO_3 on coking characteristics of coal samples

When determining the volatile matter, the solid matter remaining in the crucible is called coke slag. Judging the characteristics of coke slag on the original coal sample and the coal sample obtained by adding 1.0% Na_2CO_3 by the impregnation method, the upper and lower surfaces of coke slag have silver-white metallic luster and obvious expansion, but the height is lower than 15 mm, which belongs to the seventh category of coke slag characteristics. It shows that the addition of Na_2CO_3 has no obvious effect on the coking characteristics of coal samples.

3.3 Influence of Na_2CO_3 on combustion performance of civil clean coke

3.3.1 Thermogravimetric curve analysis

Figures 4(a) and **4(b)** show the weight loss curve (TG) and weight loss differential curve (DTG) of the experimental coal when the coke and the original coke are burned by impregnation with 1.0% Na_2CO_3 .

It can be seen from **Figure 4(a)** that the curves of the two coke samples basically coincide in the temperature range of 300 ~ 763 K, and the weight loss phenomenon is not obvious, because the experimental samples are obtained by high-temperature distillation, and the water and volatile content are low. Compared with the original coke, the curve of coke added with Na_2CO_3 moves to the left obviously in the temperature range of 763 ~ 1,074 K, indicating that Na promotes the combus-

tion of fixed carbon; after adding Na_2CO_3 , the final temperature of coke combustion is advanced, and the percentage of combustion loss is reduced by 3.16%. This is because Na_2CO_3 promotes combus-

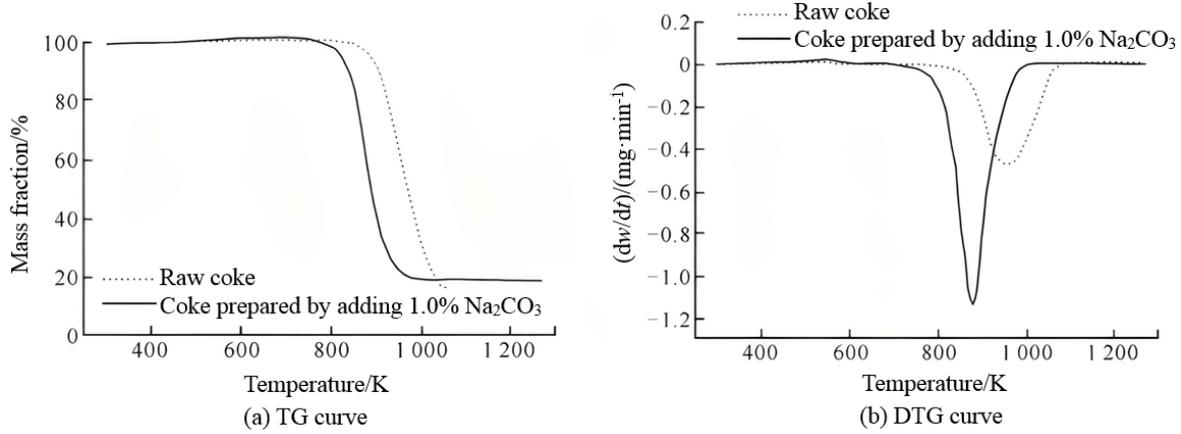


Figure 4. The TG-DTG curves of the raw coke and coke added with Na_2CO_3 .

Table 2. The combustion indexes of the raw coke and coke added with Na_2CO_3

Sample name	Initial combustion temperature T_i/k	Burnout temperature T_h/k	Average combustion rate $\bar{v}/(\text{mg}\cdot\text{min}^{-1})$	Maximum burning rate $v_{\max}/(\text{mg}\cdot\text{min}^{-1})$	Comprehensive combustion index $S/(\times 10^{-10}\cdot\text{mg}^{-2}\cdot\text{min}^{-2}\cdot\text{K}^{-3})$
Raw coke	918	1,074	0.38	0.48	2.01
Coke prepared by adding 1.0% Na_2CO_3	873	998	0.75	1.13	11.14

It can be seen from **Figure 4(b)** that the DTG curve of coke made by adding Na_2CO_3 is similar to the original coke, and the ignition mode has not changed, but the curve moves to the left, the peak width narrows, and the maximum combustion rate increases significantly, indicating that adding Na_2CO_3 can significantly improve the combustion performance of coke without changing the ignition mode.

3.3.2 Comprehensive combustion index analysis

In this paper, the comprehensive combustion index s is used to describe the combustion of the sample^[19].

$$S = \frac{(dw/dt)_{\max} (dw/dt)_{\text{mean}}}{T_i^2 T_h} \quad (1)$$

Where: $(dw/dt)_{\max}$ is the maximum combustion rate, mg/min ; $(dw/dt)_{\text{mean}}$ is the average combustion rate, mg/min ; T_i is the initial combustion temperature, K ; T_h is the burnout temperature, K . Here T_i is determined by TG-DTG method^[20].

tion, which makes the heat release rapidly in a shorter time, resulting in a small increase in the amount of carbon residue. At the same time, adding Na_2CO_3 increases the ash content of coke samples.

Table 2 shows the combustion indexes of raw coke and Na_2CO_3 added coke obtained from TG curve. It can be seen from **Table 2** that after adding 1.0% Na_2CO_3 , the coke combustion is advanced, the initial combustion temperature and burnout temperature are reduced, the combustion time is short, and the combustion is more concentrated; the average combustion rate, the maximum combustion rate and the comprehensive combustion index increased. It can be seen that the addition of Na_2CO_3 has a significant impact on the combustion characteristics of coke, which is mainly due to the increase of combustion rate and the decrease of initial combustion temperature and burnout temperature.

3.4 Improvement mechanism of Na_2CO_3 on combustion performance of civil clean coke

3.4.1 Pore structure analysis

Figure 5 shows the N_2 adsorption and desorption isotherm curve of raw coke and coke samples prepared by adding 1.0% Na_2CO_3 . It can be seen from **Figure 5** that the adsorption capacity of coke prepared by adding Na_2CO_3 at the maximum rela-

tive pressure is $60 \text{ cm}^3/\text{g}$, which is much larger than $15 \text{ cm}^3/\text{g}$ of the original coke; the N_2 isothermal adsorption and desorption curves of the two samples belong to the type IV isothermal curve in the classification of IUPAC isotherms. The coke made by adding Na_2CO_3 has H_3 hysteresis ring in the range of relative pressure $p/p_0 = 0.5 \sim 1.0$, and the original coke has H_3 hysteresis ring in the range of $p/p_0 = 0.7 \sim 1.0$. It is considered that both of them have slit holes formed by the accumulation of sheet particles, and the pore size distribution of coke made by adding Na_2CO_3 is more extensive; the curve of both rises in the high-pressure section ($p/p_0 = 0.9 \sim 1.0$), and the increase of adsorption capacity indicates the existence of macropores with particle accumulation^[20].

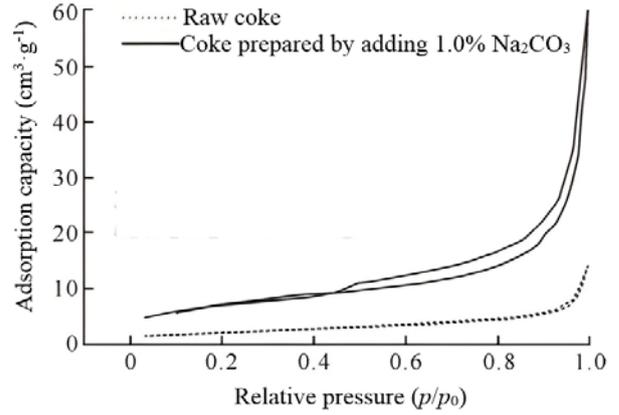


Figure 5. The N_2 adsorption-desorption isotherm of the raw coke and coke added 1.0% Na_2CO_3 .

Table 3 shows the pore structure parameters of raw coke and coke samples prepared by adding 1.0% Na_2CO_3 .

Table 3. The pore structure parameters of the raw coke and coke added 1% Na_2CO_3

Sample name	Specific surface area/($\text{m}^2 \cdot \text{g}^{-1}$)	Pore volume/($\text{cm}^3 \cdot \text{g}^{-1}$)	Aperture /nm
Raw coke	8.94	0.017	8.66
Coke prepared by adding 1.0% Na_2CO_3	21.30	0.079	11.03

It can be seen from **Table 3** that after adding Na_2CO_3 , the pore volume and pore diameter of coke are significantly changed. Compared with the original coke, the specific surface area is $21.30 \text{ m}^2/\text{g}$, which is increased by 1.4 times; the pore volume is $0.079 \text{ cm}^3/\text{g}$, which is increased by 3.6 times; the pore size is 11.03 nm , which increases by 2.4 nm . It further shows that the pore structure of the coke sample made of Na_2CO_3 is richer than that of the original coke, which provides sufficient gas-solid contact surface and gas diffusion channel for the combustion of coke, promotes oxygen transfer, accelerates the reaction rate of coke, and improves the combustion performance.

3.4.2 Thermokinetic analysis

Generally, the gas-solid reaction conforms to the following kinetic equation^[21,22]:

$$\frac{d\alpha}{dt} = Kf(\alpha) = Ae^{-E/(RT)} f(\alpha) \quad (2)$$

Where, α is the reaction conversion, t is the reaction time, K is the reaction rate constant, $f(\alpha)$ is a function related to solid unburned reactants and reaction rate, A is the frequency factor, E is the ac-

tivation energy of the reaction, R is the gas constant of $8.314 \text{ J}/(\text{mol} \cdot \text{K})$, and T is the reaction temperature.

Through thermogravimetric curve, reaction conversion α is

$$\alpha = \frac{(m_0 - m_t)}{(m_0 - m_w)} \quad (3)$$

Heating rate β is

$$\beta = \frac{dT}{dt} \quad (4)$$

Arrange formula (2) ~ (4) and arrange the Coats-Redfern integral to obtain the approximate solution^[23]:

When $n = 1$

$$\ln \left[\frac{-\ln(1-\alpha)}{T^2} \right] = \ln \left[\frac{AR}{\beta E} \left(1 - \frac{2RT}{E} \right) \right] - \frac{E}{RT} \quad (5)$$

When $n \neq 1$

$$\ln \left[\frac{1 - (1-\alpha)^{1-n}}{T^2(1-n)} \right] = \ln \left[\frac{AR}{\beta E} \left(1 - \frac{2RT}{E} \right) \right] - \frac{E}{RT} \quad (6)$$

Take points $\left(\ln \left[\frac{AR}{\beta E} \left(1 - \frac{2RT}{E} \right) \right], \frac{1}{T} \right)$ from the TG curve and regress the curve with the least square method to obtain the linear equation: $y = ax + b$.

According to the characteristics of thermogravimetric curve, the low temperature zone and high temperature zone are treated, respectively, with the maximum combustion rate point as the boundary. The fitting equation and correlation coefficient of reaction order n are 0, 1/2, 2/3, 1 and 2, respectively (**Table 4**). The activation energy E of the reaction is obtained from the fitting line, and the frequency factor A is obtained from the intercept. The calculated kinetic parameters of the coke sample are shown in **Table 5**.

It can be seen from **Table 5** that in the low temperature zone, the apparent activation energy of coke before and after Na_2CO_3 addition is 454.28 kJ/mol and 306.85 kJ/mol, respectively. The addi-

tion of Na_2CO_3 reduces the apparent activation energy in the low temperature zone, and Na_2CO_3 can improve the reaction rate in the low temperature zone to a certain extent; in the high temperature zone, the apparent activation energy of coke before and after adding Na_2CO_3 is 557.36 kJ/mol and 95.36 kJ/mol, respectively. The apparent activation energy of coke prepared by adding Na_2CO_3 is much lower than that of original coke. The reaction rate in the high temperature combustion zone is significantly increased and the combustion time is short, which is consistent with the previous analysis. It can be seen that the addition of Na_2CO_3 reduces the apparent activation energy of the system in the whole combustion reaction process, improves the reaction rate, and makes the TG and DTG curves significantly move towards the direction of low temperature.

Table 4. The fitting equations and related coefficients with different reaction orders

Sample name	Reaction order	Low temperature zone		High temperature zone	
		Fitting equation	Correlation coefficient R^2	Fitting equation	Correlation coefficient R^2
Raw coke	0	$y = -40\ 045x + 36.441$	0.9775	$y = -5\ 714.3x - 0.1375$	0.9149
	1/2	$y = -42\ 621x + 39.586$	0.9830	$y = -8\ 401.6x + 3.2123$	0.9395
	2/3	$y = -43\ 628x + 40.814$	0.9849	$y = -10\ 546x + 5.6792$	0.9552
	1	$y = -45\ 892x + 43.568$	0.9882	$y = -18\ 175x + 14.178$	0.9822
	2	$y = -54\ 640x + 54.187$	0.9910	$y = -67\ 039x + 67.41$	0.9981
Coke prepared by adding 1.0% Na_2CO_3	0	$y = -31\ 170x + 18.291$	0.9067	$y = -2\ 911.8x - 11.137$	0.8045
	1/2	$y = -32\ 464x + 19.781$	0.9199	$y = -8\ 444.1x - 5.2798$	0.9825
	2/3	$y = -32\ 916x + 20.301$	0.9241	$y = -11\ 470x - 2.1343$	0.9975
	1	$y = -33\ 851x + 21.378$	0.9322	$y = -20\ 146x + 6.7932$	0.9863
	2	$y = -36\ 908x + 24.892$	0.9530	$y = -66\ 694x + 54.207$	0.8928

Table 5. The kinetic parameters

Sample name	Temperate zone	Reaction order	Correlation coefficient R^2	Frequency factor A/s^{-1}	Activation energy $E/(kJ \cdot mol^{-1})$
Raw coke	Low-temperature zone	2	0.9910	3.53×10^{16}	454.28
	High-temperature zone	2	0.9981	7.93×10^4	557.36
Coke prepared by adding 1.0% Na_2CO_3	Low-temperature zone	2	0.9530	2.38×10^{16}	306.85
	High-temperature zone	2/3	0.9975	1.35×10^4	95.36

4. Conclusion

(1) Adding 1.0% Na_2CO_3 by impregnation method can significantly improve the combustion characteristics of coke, significantly shift the TG and DTG curves to the left, and significantly increase the comprehensive combustion index.

(2) The coke with 1.0% Na_2CO_3 has a richer

pore structure than the original coke, providing sufficient gas-solid contact surface and gas diffusion channel for coke combustion, and promoting oxygen transfer.

(3) After adding 1.0% Na_2CO_3 , the apparent activation energy of the whole combustion system decreases, the apparent activation energy in the

low-temperature zone decreases from 454.28 kJ/mol to 306.85 kJ/mol, and the apparent activation energy in the high-temperature zone decreases from 557.36 kJ/mol to 95.36 kJ/mol. The reaction rate increases, which significantly improves the combustion performance of coke.

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Conflict of interest

The authors declare that they have no conflict of interest.

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