

COMBUSTION KINETICS OF PETROLEUM COKE
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Abstract. The study examined the physico-chemical characteristics and combustion kinetics of petroleum coke or petcoke (PCK). The results revealed that PCK contains significantly high carbon, fixed carbon, and calorific value with low sulphur, and ash content. The combustion characteristics of PCK revealed the temperatures of ignition ranged from 764 to 795 K; peak decomposition from 808 to 875 K and burn-out from 857 to 933 K. The combustion performance and reactivity analyses were examined based on the ignition ratio, devolatilization ratio, burnout ratio, and combustion characteristic factor were performed. The activation energy and pre-exponential factor were determined also. The results revealed that PCK is highly reactive during combustion contrary to previous reports in the literature. Overall, the findings demonstrate that combustion is a practical approach for energy recovery from petcoke.

Keywords: combustion, petcoke, isoconversional model, kinetics.

1. Introduction

Over the years, the growing demand for energy has increased crude oil refining capacity worldwide [1]. As a result, it is estimated that crude oil refining capacity will surpass 118.1 million barrels per day by the year 2020. This demonstrates the petroleum refining industry will experience a significant growth due to a rising energy demand from India, China, and the United States. On the other hand, the refining of crude oil generates large quantities of liquid, sludge and solid waste products such as petroleum coke (petcoke). According to Bayram *et al.* [2], approximately 30 kg of petcoke is generated from refining one tonne of crude oil. Currently, India, China, and the United States account for 70 % of global petcoke

production estimated at 127 million tons per annum with a market value of \$7 billion. However, due to the planned expansion of coking plant capacity and plummeting supply of light sweet crude worldwide, petcoke production is expected to increase by 3–5 % by the year 2020 [3, 4].

Petcoke is the solid black residue generated from the thermal cracking or delayed coking of heavy grades of crude oil in petroleum refineries [5]. It typically consists of high carbon, fixed carbon and polycyclic aromatic hydrocarbons (PAH) depending on the origin of the crude oil [6]. In theory, petcoke can be categorised into four broad groups, namely sponge, honeycomb, shot or needle coke based on its physical structure [7]. Needle coke is considered a high-grade coke produced from processing the rich aromatic feedstock. It contains low sulphur (<1 wt %) ideal for the production of carbon graphite anodes, aluminium, and stainless steel. However, petcoke from heavy crude distillation contains high concentrations of sulphur and vanadium (approx. 500 ppm) which poses a significant risk to human health, safety, and the environment [8, 9]. Furthermore, the growing trend in heavy crude distillation will increase the global petcoke production exacerbating waste disposal challenges in refineries.

Consequently, researchers are exploring more economical and environmentally friendly technologies to valorise the growing global stock of petcoke. The long-term objective is to exploit the high heating value, carbon, fixed carbon in addition to the low moisture and ash content of petcoke for the production of energy, fuels, and chemicals. As a result, innovative energy technologies such as pyrolysis [10, 11], gasification [12, 13] and combustion [14, 15] have been explored to valorise petcoke. However, low reactivity and stability, along with its low volatile matter and high fixed carbon make it a poor fuel for gasification [16, 17]. Similarly, the pyrolysis of petcoke into bio-oils, char and fuel gases is considered inefficient due to the significant input requirements for downstream processing, storage logistics, and energy utilization based on existing power infrastructure worldwide. In addition, Yuan *et al.* [11] reported that the pyrolysis of petcoke is prone to the emission of nitrous pollutants.

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Therefore, the combustion or co-firing of petcoke with biomass or coal is the logical alternative for valorisation [17]. However, this will require comprehensive characterization and in-depth understanding of the physico-chemical, thermal and kinetic properties of petcoke for efficient utilisation as potential high-value fuel for combustion. Therefore, this study aims to examine the fuel characterization, thermal degradation behaviour and kinetic properties of petcoke for combustion. It is envisaged that findings will address the gap in knowledge regarding the fuel properties and combustion performance of petcoke for future energy recovery. Lastly, the study will provide data required for the efficient design, optimization, and scale-up of future energy conversion systems.

2. Experimental

2.1. Materials Preparation

Petcoke was supplied by Cemerlang Coke Industrial (Batu Pahat, Malaysia) as a waste by-product of the Petronas Refinery in Melaka, Malaysia. Next, the petcoke was sieved using the 60 mesh (250 μm) RetschTM analytical sieve and stored in airtight storage bags prior to characterization.

2.2. Fuel Characterization

The fuel characterization of petcoke was examined by ultimate, proximate, and bomb calorific analyses as reported in Table 1 in dry ash free (*daf*) and dry basis (*db*). The ultimate analysis was performed using an elemental analyser (vario MACRO Cube, Elementar Analysen Systeme GmbH, Germany). The proximate analysis was performed based on the ASTM standards D3173 for moisture, D3174 for ash and D3175 for volatile matter, whereas fixed carbon was determined by difference. The higher heating value (HHV) was determined by bomb calorimetry (IKA C2000, USA) according to ASTM Standard D2015. All tests were repeated three times to ensure the reliability of the results.

The combustion of petcoke was examined by non-isothermal thermogravimetric analysis (TGA) using the TG Analyser (Q500 TA Instruments, USA). The Q500 TGA is a hi-res, stepwise, modulated temperature controlled thermobalance periodically subjected to temperature and buoyancy calibration to ensure the reliability of the weight change data. In this study, the thermal analysis was performed by heating approximately 11 mg of petcoke in a platinum crucible within 303–1273 K under the dynamic heating rates of 10, 20 and 30 K/min under air atmosphere. The flow rate of the purge gas was 50 ml/min.

Next, the mass loss curves were analysed using the TA Instruments thermal analysis software (Universal version 4.5A). The burning profile of petcoke and its temperature profile characteristics (TPC); ignition (onset) (T_i); peak decomposition (T_p) and burn-out (T_o) temperatures were deduced. Based on the TPCs, the combustion performance parameters, such as ignition ratio (I_R), burn-out ratio (B_R), devolatilization ratio (D_R) and combustion characteristic factor (CCF) for petcoke were deduced [14, 18].

$$I_R = \frac{T_i}{T_o} \quad (1)$$

$$B_R = \frac{T_p}{T_o} \quad (2)$$

$$D_R = \frac{(T_o - T_i)}{T_p} \quad (3)$$

$$CCF = \frac{\left(\frac{dw}{dt}\right)_{\max} \cdot \left(\frac{dw}{dt}\right)_{\text{average}}}{T_i^2 T_o} \quad (4)$$

From the weight loss (TG , %) data, the kinetic analyses of petcoke combustion were performed based on the isoconversional models of Flynn-Wall-Ozawa (FWO) and Kissinger-Akahira-Sunose (KAS). Section 2.3 presents the theoretical assumptions and mathematical equations of the FWO and KAS models.

2.3. Model Theory

The kinetic analysis of petcoke combustion can be described by the one-step global model which presumes the degradation of carbonaceous materials in a single step reaction expressed as:



Therefore, the rate of degradation of the solid petcoke can be described as [19]:

$$\frac{da}{dt} = k(T)f(a) \quad (6)$$

The term $k(T)$ is the temperature dependent rate constant in the Arrhenius law (Eq. 7):

$$k(T) = A \exp\left(-\frac{E_a}{RT}\right) \quad (7)$$

Whereas $f(a)$ is the reaction model (Eq. (8)) that describes the thermal degradation of petcoke:

$$f(a) = (1-a)^n \quad (8)$$

The terms A , E_a , R , T , and n are the pre-exponential factor (min^{-1}), activation energy (kJ mol^{-1}), molar gas constant ($\text{J mol}^{-1} \cdot \text{K}^{-1}$), temperature (K) and reaction order, respectively. The solid-state degradation of petcoke is a first order reaction based on the degree of conversion (α):

$$a = \frac{m_i - m_a}{m_i - m_f} \quad (9)$$

where m_i , m_a and m_f describe the initial, actual and final mass of the sample after degradation. Based on Eqs. (7)-(9), the fundamental expression for the petcoke combustion kinetics can be deduced by substituting Eqs. (7)-(8) into Eq. (6), which yields the expression:

$$\frac{da}{dt} = A \exp\left(-\frac{E_a}{RT}\right) \cdot (1-a)^n \quad (10)$$

The effect of multi-heating rates during non-isothermal thermal degradation of petcoke can be defined by the heating rate expression:

$$b = \frac{dT}{dt} \quad (11)$$

After separation of variables Eq. (10) can be re-written as:

$$\frac{da}{(1-a)^n} = \frac{A}{b} \exp\left(-\frac{E_a}{RT}\right) dT \quad (12)$$

Consequently, Eq. (12) can be integrated to obtain the integral conversion function $g(\alpha)$ which describes the degradation kinetics of petcoke combustion as presented in Eq. (13):

$$g(\alpha) = \int_0^\alpha \frac{da}{(1-a)^n} = \frac{A}{b} \int_0^T \exp\left(-\frac{E_a}{RT}\right) dT \quad (13)$$

In this work, the isoconversional models of Flynn-Wall-Ozawa (FWO) and Kissinger-Akahira-Sunose (KAS) were employed to calculate the kinetic parameters for petcoke combustion. Based on Eq. (13), the expression for Flynn-Wall-Ozawa (FWO) is [19]:

$$\ln b = \ln\left(\frac{AE_a}{Rg(\alpha)}\right) - 5.331 - 1.052\left(\frac{E_a}{RT}\right) \quad (14)$$

The FWO model was used to determine the kinetic parameters; apparent activation E_a from the slope ($-1.052 E_a/RT$) of the plot of $\ln b$ against $1/T$ whereas the A pre-exponential factor was determined from the intercept $\ln[AE_a/Rg(\alpha)]$. Likewise, the petcoke combustion kinetics was examined by the Kissinger-Akahira-Sunose (KAS) model [20]:

$$\ln \frac{b}{T^2} = \ln\left(\frac{AR}{E_a g(\alpha)}\right) - \left(\frac{E_a}{RT}\right) \quad (15)$$

From the plot of $\ln(b/T^2)$ against $1/T$ the apparent activation energy E_a was calculated from the slope ($-E_a/RT$), whereas the A pre-exponential factor was calculated from the intercept $\ln[AR/E_a g(\alpha)]$.

3. Results and Discussion

3.1. Fuel Characteristics

The fuel characteristics of petcoke comprising the ultimate, proximate, and bomb calorific values are reported in dry ash free (*daf*) and dry basis (*db*) in Table 1.

As observed, petcoke contains significantly high carbon (*C*), fixed carbon (*FC*) and calorific values. The

heating value is significantly high indicating low quantities of the fuel are potentially required for energy utilization in thermal power plants [21]. Furthermore, the composition of pollutant precursor elements; nitrogen (*N*), sulphur (*S*), and ash (*A*) are significantly lower than the reported values in literature. This indicates that the petcoke sample is of fuel grade quality with a significantly low potential for emitting gaseous NO_x, and SO_x into the environment [22]. Lastly, the results indicate that petcoke is a potential feedstock for thermal conversion processes.

3.2. Thermal Decomposition

The thermogravimetric (TG) and derivative thermogravimetric (DTG) curves for petcoke combustion are presented in Figs. 1 and 2. As it is observed in Fig. 1, the TG curves exhibited the downward sloping trend typically observed for thermal decomposing materials [23]. This indicates the temperature variation during TG analysis which significantly influences degradation of petcoke, can be ascribed to the thermal cleavage of chemical bonds [24] in the petcoke structure.

Similarly, the increase in heating rate from 10 to 30 K/min influenced the degradation of petcoke. As observed, the TG curves shifted from the left to the right or higher temperatures during thermal analysis. This is largely due to the heat transfer limitation or thermal lag that shortens the time required for the decomposition of the sample during TG at higher heating rates [19, 25]. In addition, the results indicate that the thermal degradation of petcoke is more efficient, reactive or thermally less stable at lower heating rates.

The combustion of petcoke was also examined by DTG analysis as presented in Fig. 2. The DTG curves indicate that thermal degradation of petcoke resulted in a single prominent endothermic peak for each heating rate examined in the study. Conversely, the time of reaction range, size and symmetry size of the peaks were different, as observed by the increase in heating rate during thermal degradation. Hence, it can be surmised that petcoke decomposition or combustion during TGA occurs in a single step reaction with a significant weight loss occurring within the temperature range of 623–1023 K.

In addition, the results reveal that petcoke was completely decomposed below 1023 K which indicates it is highly reactive under oxidative conditions. The reactivity of petcoke is further examined and described in Subsection 3.3.

3.3. Thermal Reactivity

The thermal reactivity of petcoke was examined based on the temperature profile characteristics (TPC); ignition (T_i); maximum decomposition (T_p) and burn-out (T_o) temperatures presented in Table 2. As observed, the TPC increased with the increase in the heating rate during thermal degradation. In this study, T_i ranged from 764 to 795 K; T_p –

from 808 to 875 K and T_o – from 857 to 933 K. However, the average values of T_i , T_p , and T_o were 781, 844, and 898 K, respectively. This indicates that on average the multi-heating rate decomposition or combustion of petcoke occurs in the temperature range of 781–898 K.

The reactivity of petcoke was also examined from its residual mass after combustion. The values ranged from 0.29 to 1.69 % with an average of 0.85 % which indicates that petcoke is highly reactive based on 98.31–99.71 % decomposition during combustion. The near complete combustion of petcoke observed in this study is in good agreement with Yoon *et al.* [12] and Yuzbasi *et al.* [15].

The combustion performance of petcoke was also examined based on its ignition ratio (I_R), devolatilization ratio (D_R), burn-out ratio (B_R), and combustion characteristic factor (CCF) as presented in Table 3.

The results indicate the I_R value ranged from 79.17 to 84.10 %; D_R from 17.37 to 22.85 %; B_R from 91.17 to 91.66 % and CCF from $7.86 \cdot 10^{-8}$ to $3.81 \cdot 10^{-7}$ %/min. The

average values were 81.45, 20.29, 91.46 and $2.21 \cdot 10^{-7}$ for ignition ratio I_R , devolatilization ratio D_R , burn-out ratio B_R , and combustion characteristic factor CCF , respectively. As observed, the ignition ratio decreased gradually with the increase in the heating rate from 10 to 30 K/min during petcoke combustion. This may be due to increased resistance to heat and mass transfer by evolving flue gas species during thermal analysis. Similar observations have been reported for thermally degradation carbonaceous materials during TGA [24, 26]. In contrast, the burn-out ratio increased from 10 to 20 K/min but decreased at 30 K/min as observed in Table 3. Lastly, the D_R and CCF values increased during petcoke combustion due to the effect of higher heating rates.

3.4. Isoconversional Kinetic Plots

Figs. 3 and 4 present the kinetic plots for the combustion of petcoke based on Flynn-Wall-Ozawa (FWO) and Kissinger-Akahira-Sunose (KAS) isoconversional models.

Table 1

Comparative fuel characteristics of petcoke

Element/Fuel properties	Symbol	Units	Petcoke
Carbon	C^{daf}	wt %	92.61
Hydrogen	H^{daf}	wt %	4.49
Nitrogen	N^{daf}	wt %	1.46
Sulphur	S^{daf}	wt %	0.67
Oxygen	O^{daf}	wt %	0.78
Moisture	M	wt %	0.79
Volatiles	VM^{db}	wt %	14.65
Ash	A^{db}	wt %	0.57
Fixed carbon	FC^{db}	wt %	84.79
Heating value	HHV^{daf}	MJ/kg	36.81

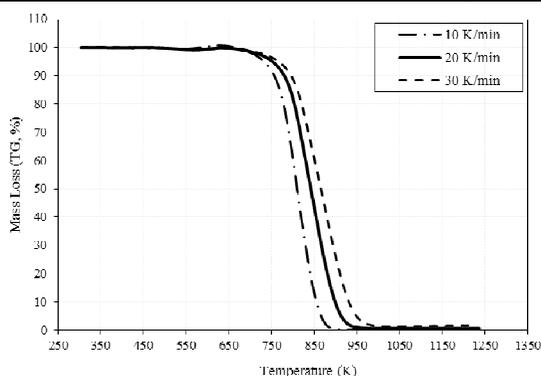


Fig. 1. TG curves for petcoke combustion

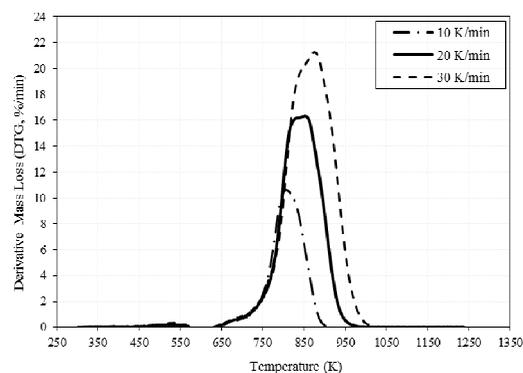


Fig. 2. DTG curves for petcoke combustion

Table 2

Temperature profile characteristics of petcoke

Heating rate, K/min	T_i , K	T_p , K	T_o , K	Residual weight, %
10	764.63	808.18	857.57	0.29
20	783.88	850.51	903.05	0.57
30	795.90	875.17	933.45	1.69

Table 3

Reactivity of petcoke during combustion

Heating rate, K/min	I_R , %	D_R , %	B_R , %	CCF, %/min
10	84.10	17.37	91.55	$7.86 \cdot 10^{-8}$
20	81.08	20.64	91.66	$2.04 \cdot 10^{-7}$
30	79.17	22.85	91.17	$3.81 \cdot 10^{-7}$

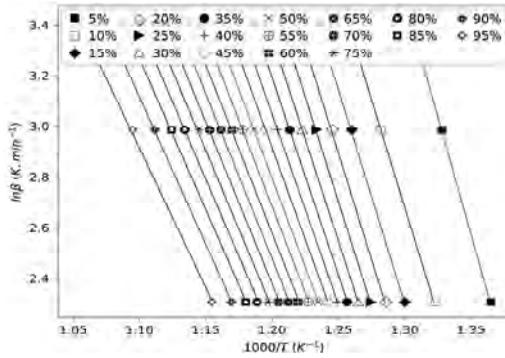


Fig. 3. FWO kinetic plots for petcoke combustion

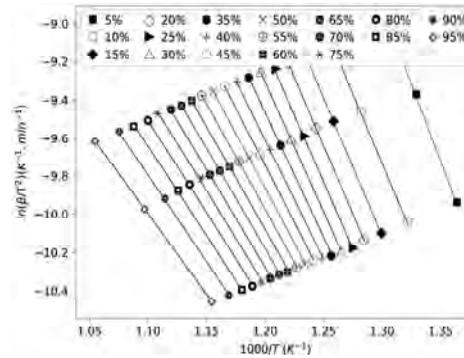


Fig. 4. KAS kinetic plots for petcoke combustion

Table 4

Kinetic parameters for petcoke combustion

α	Flynn-Wall-Ozawa (FWO)			Kissinger Akahira Sunose (KAS)		
	R^2	E_a , kJ/mol	A , min ⁻¹	R^2	E_a , kJ/mol	A , min ⁻¹
0.05	0.9979	146.82	$1.19 \cdot 10^{13}$	0.9972	134.82	$3.13 \cdot 10^0$
0.10	0.9997	134.29	$7.29 \cdot 10^{11}$	0.9996	121.34	$1.51 \cdot 10^3$
0.15	0.9999	133.33	$4.29 \cdot 10^{11}$	0.9998	120.17	$8.48 \cdot 10^4$
0.20	0.9998	132.52	$2.96 \cdot 10^{11}$	0.9997	119.21	$5.68 \cdot 10^4$
0.25	0.9996	130.41	$1.78 \cdot 10^{11}$	0.9994	116.90	$3.25 \cdot 10^4$
0.30	0.9993	127.10	$9.25 \cdot 10^{10}$	0.9989	113.33	$1.58 \cdot 10^4$
0.35	0.9989	122.89	$4.31 \cdot 10^{10}$	0.9984	108.83	$6.74 \cdot 10^3$
0.40	0.9987	118.47	$1.97 \cdot 10^{10}$	0.9980	104.10	$2.82 \cdot 10^3$
0.45	0.9985	114.15	$9.32 \cdot 10^9$	0.9977	99.48	$1.21 \cdot 10^3$
0.50	0.9985	110.34	$4.79 \cdot 10^9$	0.9977	95.40	$5.71 \cdot 10^2$
0.55	0.9986	107.28	$2.77 \cdot 10^9$	0.9978	92.11	$3.07 \cdot 10^2$
0.60	0.9988	104.87	$1.77 \cdot 10^9$	0.9980	89.51	$1.84 \cdot 10^2$
0.65	0.9988	102.87	$1.20 \cdot 10^9$	0.9981	87.32	$1.19 \cdot 10^2$
0.70	0.9987	100.79	$8.13 \cdot 10^8$	0.9979	85.05	$7.55 \cdot 10^1$
0.75	0.9986	98.51	$5.34 \cdot 10^8$	0.9977	82.58	$4.65 \cdot 10^1$
0.80	0.9987	96.23	$3.50 \cdot 10^8$	0.9977	80.08	$2.85 \cdot 10^1$
0.85	0.9989	94.14	$2.34 \cdot 10^8$	0.9981	77.78	$1.79 \cdot 10^1$
0.90	0.9988	91.78	$1.49 \cdot 10^8$	0.9979	75.17	$1.05 \cdot 10^1$
0.95	0.9981	86.89	$6.50 \cdot 10^7$	0.9964	69.84	3.94
Average	0.9989	113.35	$7.23 \cdot 10^{11}$	0.9982	98.58	$1.83 \cdot 10^5$

As observed, the plots for both models displayed parallel lines for conversions $\alpha = 0.05$ to 0.95 during PCK conversion within 303–1273 K. This indicates that the degradation mechanism for PCK combustion is governed by a complex set of the first-order reactions occurring concurrently [27].

3.5. Kinetic Parameters

The kinetic parameters (activation energy, E_a , and pre-exponential factor, A) were calculated from the slope and intercepts of the plots in Figs. 3 and 4. Table 4

presents the kinetic parameters for PCK combustion based on the FWO and KAS kinetic models. The results indicate that the activation energy, E_a , for PCK combustion ranged from 86.89 to 146.82 kJ/mol with an average value of 113.35 kJ/mol for FWO, whereas the values for KAS ranged from 69.84 to 134.82 kJ/mol with an average value of 98.58 kJ/mol. Conversely, the pre-exponential factor ranged from $6.50 \cdot 10^7$ to $1.19 \cdot 10^{13}$ min⁻¹ with an average value of $7.32 \cdot 10^{11}$ for FWO, and the values for KAS ranged from 3.94 to $3.13 \cdot 10^6$ min⁻¹ with an average value of $1.83 \cdot 10^5$ min⁻¹. The correlation coefficients for the PCK

combustion were in the range of 0.9982–0.9989 for both kinetic models examined.

Furthermore, the results of the kinetic analyses of PCK combustion indicate that the fuel thermal degradation occurs rapidly under the examined conditions. This is evidenced by the low values of activation energy and pre-exponential factors reported for the combustion of the fuel. In addition, the results demonstrate that the energy requirement for thermal conversion of petcoke (PCK) is minimal as characterised by the high potential for effective collision of its reactive species. Overall, the results highlight the potential of combustion as a practical approach for future energy recovery from petcoke due to its rapid kinetics and complete decomposition under the conditions investigated in this study.

4. Conclusions

The study examined the combustion kinetics of petcoke (PCK) based on the *isoconversional* kinetic models. The physico-chemical and fuel characteristics of PCK were also examined by ultimate, proximate and calorific analyses. The results indicate that PCK contains significantly high carbon, fixed carbon and calorific along with low nitrogen, sulphur, and ash content which highlights the environmentally friendly nature of the fuel. In addition, the isoconversional Flynn-Wall-Ozawa (FWO) and Kissinger-Akahira-Sunose (KAS) models successfully predicted the kinetics of PCK combustion. The average activation energy was 113.35 kJ/mol for FWO and 98.58 kJ/mol for the KAS model. The pre-exponential factors were $7.32 \cdot 10^{11} \text{ min}^{-1}$ for FWO and $1.83 \cdot 10^5 \text{ min}^{-1}$ for the KAS model. The kinetic parameters indicate that PCK combustion occurs rapidly based on the first order kinetics. Overall, the results demonstrate that petcoke may be a feedstock for future energy recovery.

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КІНЕТИКА ЗГОРЯННЯ НАФТОВОГО КОКСУ ЗГІДНО ІЗОКОНВЕРСІЙНОГО МОДЕЛЮВАННЯ

Анотація. Вивчено фізико-хімічні характеристики та кінетику згоряння нафтового коксу або пек-коксу (НК). Визначено, що НК має високий вміст карбону, зв'язаного карбону та високу теплотворну здатність, і низький вміст сульфурну та золи. Показано, що температура займання НК коливається в межах 764–795 K, пік розкладання 808–875 K та температура вигорання 857–933 K. За допомогою аналізу продуктивності згоряння та реакційної здатності визначено коефіцієнт запалювання, коефіцієнт перетворення, коефіцієнт вигорання та характеристичний коефіцієнт згоряння. Визначено також енергію активації та предекспонентний множник. Показано, що НК дуже реактивний під час згоряння всупереч даним літератури. На основі проведених досліджень встановлено, що спалювання є практичним підходом для видобутку енергії з НК.

Ключові слова: згоряння, нафтовий кокс, ізоконверсійне моделювання, кінетика.