

Retraction

Retracted: Experiment and Algorithm Research of Coal Direct Liquefaction Residual Oil Pyrolysis and Coking Technology Based on Lumped Kinetic Engineering

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This article has been retracted by Hindawi following an investigation undertaken by the publisher [1]. This investigation has uncovered evidence of one or more of the following indicators of systematic manipulation of the publication process:

- (1) Discrepancies in scope
- (2) Discrepancies in the description of the research reported
- (3) Discrepancies between the availability of data and the research described
- (4) Inappropriate citations
- (5) Incoherent, meaningless and/or irrelevant content included in the article
- (6) Manipulated or compromised peer review

The presence of these indicators undermines our confidence in the integrity of the article's content and we cannot, therefore, vouch for its reliability. Please note that this notice is intended solely to alert readers that the content of this article is unreliable. We have not investigated whether authors were aware of or involved in the systematic manipulation of the publication process.

Wiley and Hindawi regrets that the usual quality checks did not identify these issues before publication and have since put additional measures in place to safeguard research integrity.

We wish to credit our own Research Integrity and Research Publishing teams and anonymous and named external researchers and research integrity experts for contributing to this investigation. The corresponding author, as the representative of all authors, has been given the opportunity to register their agreement or disagreement to this retraction. We have kept a record of any response received.

References

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Research Article

Experiment and Algorithm Research of Coal Direct Liquefaction Residual Oil Pyrolysis and Coking Technology Based on Lumped Kinetic Engineering

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With the development of computer operation technology and algorithms, the lumped dynamic model is more practical, and the development and application are more comprehensive. Among them, the direct coal liquefaction residual oil pyrolysis and coking technology, as a coal-to-liquid process, can increase the oil yield of the coal liquefaction process and reduce environmental pollution. The purpose of this paper is to study the experiment and algorithm of coal direct liquefaction residual oil pyrolysis and coking technology based on lumped kinetic engineering. Starting from the lumped kinetic engineering, this paper takes the direct coal liquefaction residual oil pyrolysis and coking technology as the research object. Based on the experiment of small- and medium-sized equipment, the residual oil pyrolysis and coking experiment is carried out. This paper further analyzes the components of the experimental products and explores the factors that affect the yield of residual pyrolysis oil based on the five lumped kinetic model of coking. Experimental data shows that when the pyrolysis temperature is 450°C, the content of liquefied heavy oil HS in the pyrolysis oil is 47.87%, the content of asphaltene A is 44.28%, and the content of preasphaltene PA is 7.85%; the pyrolysis temperature is 500 °C. At this time, the content of liquefied heavy oil HS in the pyrolysis oil is 54.97%, the content of liquefied heavy oil HS in the pyrolysis temperature, the content of preasphaltene PA is 4.8%. It can be seen that, with the increase of pyrolysis temperature, the content of liquefied heavy oil HS increases, and the content of asphaltene A and preasphaltene PA decreases.

1. Introduction

Direct coal liquefaction residual oil pyrolysis and coking technology, as a coal-to-liquid process, effectively provide important technical support for ensuring our country's energy supply and safety, alleviating our country's shortage of petroleum resources and other prominent and sensitive issues [1,2]. However, the existing coal rapid pyrolysis and coking process still faces problems such as low coal utilization, poor tar quality, and reprocessing pyrolysis of the liquefied residue generated at the end of the hydrolysis process [3,4]. The research on the experiment and algorithm of coal direct liquefaction residual oil pyrolysis and coking technology based on lumped kinetic engineering will help to provide a new solution to this type of problem [5,6]. Regarding the research of lumped kinetic engineering, residual oil pyrolysis, and coking technology, many scholars at home and abroad have conducted multidirectional and indepth research on it. For example, Colantonio [7] conducted a pyrolysis experiment on a synthetic mixture of packaging plastic residues; Wang [8] conducted pyrolysis experiments on wood and forest residues and studied the carbon dioxide gas kinetics of pyrolysis coke; Wang [9] launched a continuous pyrolysis experiment of biomass coal residues and analyzed the results. Chen [10] studied the copyrolysis characteristics and kinetics of furfural residue and oil shale semicoke. It can be seen that since the development of this technology, its innovation and application research have received the attention of the majority of researchers. Therefore, research on the experiment and algorithm of coal direct liquefaction residual oil pyrolysis and coking technology based on lumped kinetic engineering has important theoretical and practical significance.

The purpose of this paper is to study the experiment and algorithm of coal direct liquefaction residual oil pyrolysis and coking technology based on lumped kinetic engineering. In this paper, starting from the lumped kinetic engineering, the direct coal liquefaction residual oil pyrolysis and coking technology is the research object, and the factors affecting the coal pyrolysis are analyzed. Then, the residual oil pyrolysis is carried out on the basis of the experiment of smalland medium-sized equipment. Finally, the components of the experimental products were analyzed, and the factors affecting the yield of residual pyrolysis oil were explored based on the five lumped kinetic model of coking. In the rest of the paper, the main contents are arranged as follows. Section 2 presents the experiment and the algorithm of coal direct liquefaction residual oil pyrolysis and coking technology has been proposed which is based on lumped kinetic engineering. In Section 3, further experimental research is carried out using the proposed methods. Section 4 presents the analysis of experimental results and experimental data. Lastly, the paper is concluded in Section 5.

2. Experiment and Algorithm of Coal Direct Liquefaction Residual Oil Pyrolysis and Coking Technology Based on Lumped Kinetic Engineering

In this section, the experiments are performed and the algorithm of coal direct liquefaction residual oil pyrolysis and coking technology has been proposed which is based on lumped kinetic engineering. To this end, the factors affecting coal pyrolysis are presented. In addition, the calculation of the rate constant of the coking reaction is carried out.

2.1. Factors Affecting Coal Pyrolysis. The thermogravimetric method is a common method to study the thermal reaction behavior of heavy oil. In a specific reaction device, by using inert gas to isolate the air, you can directly measure the quality of the oil sample and the reaction temperature data and then explore the relationship between the two based on the data. And according to the experimental data, through parameter estimation and optimization methods, the reaction kinetic parameters are obtained.

There are many factors that affect the thermogravimetric method. First of all, the density of the gas is different at different temperatures, which will affect the buoyancy of the gas to be measured in the experiment. In terms of experimental conditions, the heating rate of the reaction will also affect the thermogravimetric curve. According to the related experiments, it is found that an increase in the heating rate will aggravate the difficulty in the production of intermediate products [11]. In addition, the experimental environment and the amount of experimental materials will also affect the measurement results of this research method. 2.1.1. The Influence of Coal Structure. The structure of coal is complex, and there are phenomena of gas, liquid, and solid phases in pyrolysis, leading to many factors that affect coal pyrolysis, for example, coal maturity, high volatile content, and the coalification degree. Among them, the factor that has a greater impact on coal pyrolysis is the coalification degree, that is, the carbon content in coal. The carbon content affects the starting temperature. The higher the carbon content of coal, the higher the incidence of coal pyrolysis [12].

Secondly, the degree of coalification also affects the pyrolysis product and yield. The degree of coalification directly affects the quality of the product, including the size of the coke block, the yield of tar gas, and the strength of the cohesiveness.

The degree of coalification also affects the activity of the coal pyrolysis reaction. The higher the degree of coalification, the lower the reaction activity. Studies have found that, in the process of pyrolysis, different coal ranks have different crosslinking and different maturity. The temperature corresponding to the maximum production of tar and methane will shift. The higher the maturity, the higher the temperature.

2.1.2. The Influence of External Conditions

(1) Temperature. In external conditions, the temperature has a great influence on coal pyrolysis. The specific performance is as follows: as the temperature increases, the degree of coal cracking increases, the volatility increases, the content of coke or semicoke decreases, the pyrolysis temperature is lower, and the gas yield is lower. Finally, as the temperature increases, the gas yield increases, the calorific value decreases, and the coke and tar yield decrease. Therefore, according to the difference in temperature, it is divided into low-temperature pyrolysis, medium-temperature pyrolysis, and high-temperature pyrolysis so as to achieve the required different products.

(2) Heating Rate. The heating rate also has an effect on coal pyrolysis. Specifically, if the heating rate deviates from the gas precipitation temperature and the maximum precipitation temperature, the decomposition of some structures in the coal is too slow, and some of the coal has not been decomposed, which affects the efficiency of coal pyrolysis. Therefore, the rapid heating rate will cause the temperature to shift to the high-temperature direction, so that the volatile components that are decomposed will not be precipitated in time, causing delays.

(3) Change in Heating Rate. The change in heating rate is also an important factor affecting coal pyrolysis. Changing the heating rate will affect the deviation between the starting softening temperature and the starting curing temperature. As the rate changes, the index temperature will move to a higher temperature. In this case, the temperature range of the colloid will be affected and the adhesiveness will change.

(4) Constant Temperature Time. Constant temperature time is also a factor that affects coal pyrolysis. The constant

temperature time will affect the volatilization of the gas; regardless of the temperature, the longer the time, the more thoroughly the volatiles will be removed. But when the temperature is high, the removal of volatiles can be completely removed in a relatively short time at a constant temperature. As the pressure increases during coal pyrolysis, the volatilization of pyrolysis products is limited, and the yield of liquid products increases.

2.2. Calculation of Rate Constant of Coking Reaction. The number of reactions in the coking process is huge, and the calculation methods for different reaction rates are shown as follows:

$$k(T) = \frac{k_B T}{h} \exp\left(\frac{T\Delta S_m - \Delta E}{RT}\right),\tag{1}$$

where k_B is the Boltzmann constant, *h* is the Planck constant, *T* is the temperature, *R* is the ideal gas constant, ΔS_m is the entropy change before and after the reaction, and ΔE is the reaction energy barrier. Therefore, knowing ΔS_m and ΔE , the reaction rate constant at the specified temperature can be calculated.

3. Experimental Research

3.1. Experimental Materials. The original coal liquefaction residue (CHR) sample is a mixture of powdered particles and massive particles. The original mixed particle size sample is crushed and used for analysis and testing to ensure its uniformity. Among them, the CHR material is mixed with quartz sand in different proportions and fed into the pyrolyzer at a feed rate of 50–100 g/h, and the cooling water consumption is about 120 g/h.

3.2. Analysis of Experimental Products

3.2.1. Liquid Product Analysis. The tar product was analyzed by solvent extraction in a Soxhlet extractor using n-hexane and toluene solvents, and n-hexane soluble matter HS, toluene soluble matter—n-hexane insoluble matter A, and tetrahydrofuran soluble—toluene insoluble matter were obtained. The substances PA are, respectively, called liquefied heavy oil HS, asphaltene A, and preasphaltene PA.

3.2.2. Analysis of Solid Products. Morphology characterization of the bonding mixture: FEIQ scanning electron microscopy was used to characterize the morphology of the bond.

3.2.3. Semifocus Analysis. The measurement of semicoke products is directly weighed and measured after the pyrolysis experiment is completed and the temperature is naturally cooled.

3.2.4. Elemental Analysis. The element analysis of the semicoke is determined by an element analyzer to determine the content of C, H, O, N, and S in the semicoke sample.

3.2.5. Tar Separation. There are many methods for separating tar and water, including electrical separation commonly used in the oil refining industry, adsorption separation using lipophilic materials, and gravity separation and centrifugal separation using different oil-water densities.

In this experiment, the tar in the pyrolysis process will produce carbon dioxide. After the protective gas that the carbon dioxide passes into the reactor is diluted, it is discharged through the gas outlet at the bottom of the reactor and is gradually cooled by a three-stage water bath, so the generated pyrolysis water and part of the light oil are condensed in the third stage.

Due to the small scale of this experiment and the small amount of product produced, the mass yields of the experimental liquids are all weighed and measured with a highprecision balance. After each set of experiments is completed, the light oil and pyrolysis water in the third-stage collection bottle are allowed to stand and then separated. It can be seen from the stages divided by the pyrolysis process of coal that most of the pyrolysis water is produced before 350°C. Therefore, under the same experimental environment of the same group of experiments, the final pyrolysis has little effect on the change of temperature on the pyrolysis water output, which reduces the influence of the final temperature on the experiment.

3.3. Pyrolysis Test. A control group is set up, namely, the raw coal and its vitrinite group, and the main weight loss temperature range and maximum weight loss temperature of the two groups are analyzed in the pyrolysis process.

The molecular weight of the raw coal group is 2814, and the element content is C (80.6%), H (6.9%), N (1.00%), and O (11.4%).

The molecular weight of the vitrinite group is 3138, and the element content is C (79.18%), H (4.91%), N (1.00%), and O (14.90%).

3.4. Delayed Coking Experiment. After preheating, the residual oil in the raw material tank is pumped out by the raw material pump. It is first mixed with the high-temperature steam from the steam generator, and after reaching a certain temperature, it is mixed with the circulating oil from the bottom of the fractionation tower and enters the heavy oil heating furnace. After the furnace is heated to the outlet temperature, it quickly enters the coking tower through the oil pipeline for a coking reaction.

The produced coke product stays in the coke tower, and the produced high-temperature oil and gas enter the fractionation tower for fractionation from the top of the tower. Part of the heavy oil is pumped out from the circulating pump at the bottom of the fractionation tower for cyclic reaction, while the light components are cooled from the top of the tower and then enter the distillate oil receiving tank for collection, and the gas products are cooled and then calculated.

After the experiment, the device was cut off from water and power. After the coke tower was cooled, the coke tower was first weighed, the quality of the coke product was measured, and then the coke removal was started, and the amount of gas and oil produced was calculated.

3.5. All Lumped Codes of the Coking Reaction Process. The lumped code table of the coking reaction process is shown in Table 1.

Among them, the reaction network of five lumped reaction kinetics is shown in Figure 1.

According to Figure 1 and the residue reaction mechanism, the differential form of the five-set total reaction kinetic equation is as follows:

$$\frac{dOC}{dt} = -(k_1 + k_2 + k_3 + k_4)OC,$$

$$\frac{DG}{dt} = k_1OC,$$

$$\frac{dGL}{dt} = k_2OC,$$

$$\frac{dDF}{dt} = k_3OC,$$

$$\frac{dCGO}{dt} = k_4OC.$$
(2)

4. Experimental Data Analysis

This section presents the experimental research and analyzes the experimental data. First of all, the extraction analysis of pyrolysis oil is carried out. Then the comparison of solvent extraction composition of CHR pyrolysis oil and residual at different pyrolysis temperatures is proposed. Thirdly, the effects of liquefaction time on liquefaction performance are deliberated. Lastly, the discussion about the total specific surface area and pore volume of copyrolysis semicoke is given.

4.1. Extraction Analysis of Pyrolysis Oil. The results of solvent extraction of pyrolysis oil at different pyrolysis temperatures are shown in Table 2. When the pyrolysis temperature is 450°C, the content of liquefied heavy oil HS in the pyrolysis oil is 47.87% and the content of asphaltene A is 44.28%. The content of asphaltene PA is 7.85%; when the pyrolysis temperature is 500°C, the content of liquefied heavy oil HS in the pyrolysis oil is 54.97%, the content of asphaltene A is 40.23%, and the content of preasphaltene PA is 4.8%; when the pyrolysis temperature is 550°C, the content of liquefied heavy oil HS in the pyrolysis temperature is 550°C, the content of liquefied heavy oil HS in the pyrolysis oil is 58.14%, the content of asphaltene PA is 4.67%.

Observing Figure 2, it can be found that as the pyrolysis temperature increases, the content of liquefied heavy oil HS in the oil solution increases and the content of asphaltene A and preasphaltene PA decreases.

TABLE 1: Delayed coking reaction heavy oil lumped code.

Name	Code	Temperature
Oil and coke	OC	_
Gas	G	-
Gasoline	GL	0~180°C
Diesel fuel	DF	180~350°C
Coker gas oil	CGO	Above 350°C



FIGURE 1: Reaction network diagram of five lumped reaction kinetics.

TABLE 2: Solvent extraction results of residual pyrolysis oil at different pyrolysis temperatures.

Sample	HS	А	PA
Tar-450°C	47.87	44.28	7.85
Tar-500°C	54.97	40.23	4.80
Tar-550°C	58.14	35.44	6.42
Tar-600°C	65.26	30.07	4.67

4.2. Comparison of Solvent Extraction Composition of CHR Pyrolysis Oil and Residue at Different Pyrolysis Temperatures. The results of the solvent extraction composition of the pyrolysis oil and residue calculated on the basis of CHR are shown in Table 3. The content of the liquefied heavy oil HS in the pyrolysis oil is similar to that of the liquefied heavy oil HS in the residue, but the content of asphaltene A and preasphaltene PA is significantly different.

Observing Figure 3, it can be concluded that most of the liquefied heavy oil HS in the residue during the pyrolysis process is recovered and transferred into the pyrolysis oil, while most of the asphaltene A and preasphaltene PA undergone during the pyrolysis process. Reactions such as polycondensation formed THFIS, an insoluble tetrahydrofuran. In particular, preasphaltene PA undergoes polycondensation at a relatively low pyrolysis temperature, such as 450°C, to form products that are insoluble in tetrahydrofuran solvents, such as pyrolysis semitar and other heavy products. It can be seen that the increase in pyrolysis temperature is conducive to the conversion of asphaltene A in the residue to liquefied heavy oil HS, which may be caused by the enhanced pyrolysis of asphaltene.

Journal of Mathematics



FIGURE 2: Solvent extraction results of residual pyrolysis oil at different pyrolysis temperatures.

TABLE 3: Comparison of residue pyrolysis oil and residue solvent extraction composition at different pyrolysis temperatures.

Sample		HS	А		PA
CHR		10.51	37.12		8.69
Tar-450°C		7.03	6.37		0.89
Tar-500°C		10.84	7.62		1.37
Tar-550°C		11.78	7.54		1.26
Tar-600°C		12.51	5.70		1.23
	50 45 40 35 30 5 25 10.51 8.69 10 5 0 CHR	10.84 7.03 6.37	11.78 7. 5 4 1.26 <u>1</u> Tar-550°C	12.51 57 1.23 Tar-600°C	
	HS				
	A				
	PA				

FIGURE 3: Comparison of residue pyrolysis oil and residue solvent extraction composition at different pyrolysis temperatures.

4.3. Influence of Liquefaction Time on Liquefaction Performance. Coal liquefaction involves the fragmentation of the macromolecular structure of coal, the cracking of

chemical bonds, and hydrogenation. It is a complex process. Since these reactions take a certain amount of time to complete, it is necessary to study the effect of reaction time

TABLE 4: Liquefaction conversion rate and oil yield of raw coal and its vitrinite.

Time (min)	Conversion rate of raw coal	Raw kerosene yield	Vitrinite conversion rate	Vitrinite oil yield
0	74.13	37.16	88.16	28.70
10	78.32	42.41	91.24	62.47
20	80.47	50.07	92.35	67.05
30	82.48	53.47	93.16	69.46
40	83.46	54.79	94.62	70.18
50	84.28	55.16	95.41	73.24
60	85.14	57.24	94.19	74.16



FIGURE 4: Liquefaction conversion rate and oil yield of raw coal and its vitrinite.

on the result of liquefaction. Table 4 shows the results of liquefaction conversion rate and oil yield of raw coal and its vitrinite at the optimal liquefaction temperature.

It can be seen from Figure 4 that the liquefaction time has an impact on the conversion rate of liquefied coal and the oil yield, but its impact is not as large as that of temperature. With the increase of liquefaction time, the liquefaction conversion rate and oil yield of raw coal and its vitrinite first increase and then become stable. Considering the cost and other issues, the optimal liquefaction temperature (450°C) is determined, and the optimal reaction time is 60 minutes. Under the optimal liquefaction temperature and time, the raw coal liquefaction conversion rate was 85.14%, and the oil yield was 57.24%; the vitrinite liquefaction conversion rate was 94.19%, and the oil yield was 74.16%. It can be seen that the vitrinite group shows a higher liquefaction conversion rate and oil yield. 4.4. Total Specific Surface Area and Pore Volume of Copyrolysis Semicoke. The results of the change of the total specific surface area and pore volume of copyrolysis semicoke are shown in Table 5. When the temperature is 500° C, the total specific surface area of semicoke is 4.1 m^2 g and the total pore volume is 5.4 mg; when the temperature is 600° C, the total specific surface area of the semicoke is $4.2 \text{ m}^2 g$ and the total pore volume is 4.5 mg; when the temperature is 700° C, the total specific surface area of semicoke is 4.5 m^2 g and the total pore volume is 6.4 mg; when the temperature is 800° C, the total specific surface area of the semicoke is 7.5 m^2 g and the total specific surface area of the semicoke is 7.5 m^2 g and the total specific surface area of the semicoke is 7.5 m^2 g and the total specific surface area of the semicoke is 7.5 m^2 g and the

It can be seen from Figure 5 that as the degree of pyrolysis reaction deepens, the total specific surface area and total pore volume of the obtained semicoke will first decrease and then increase with the increase of pyrolysis temperature, reaching the minimum value at 600°C. And the total specific

Journal of Mathematics

Temperature (°C)	Total surface area	Total pore volume
HLSHR-char-500°C	4.1	5.4
HLSHR-char-600°C	4.2	4.5
HLSHR-char-700°C	4.5	6.4
HLSHR-char-800°C	7.5	8.5
number		
	temperature	
	Total surface area	
	Total pore Volume	

TABLE 5: Total specific surface area and pore volume of copyrolysis semicoke.

FIGURE 5: Total specific surface area and pore volume of copyrolysis semicoke.

surface area of copyrolysis semicoke changes slowly when it is between 500 and 600°C, and then as the pyrolysis temperature increases, the total specific surface area increases rapidly.

5. Conclusion

Since our country's national economic development and environmental protection are put under tremendous pressure by the mode of using coal as the main energy consumption. Therefore, the efficient and clean use of coal is a huge test faced by various coal utilization technologies, and it is of great practical significance to develop an efficient and environmentally friendly modern coal chemical industry. This paper carried out the research to complete the following tasks: based on the experiment of small and medium-sized equipment, carried out the residual oil pyrolysis and coking experiment, further carried out the composition analysis of the experimental products, and explored the influence of the residual pyrolysis based on the five-set kinetic model of coking.

Data Availability

The data underlying the results presented in the study are available within the manuscript.

Conflicts of Interest

The authors declare that there are no conflicts of interest.

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