



Statistical Analysis of Catalytic Removal of Soot Particles Based on Big Data

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Abstract. Different temperature, power, flow rate and other factors have different effects on the removal of soot particles in the tail gas of simulated diesel vehicles, and the removal effect of each kind of soot particle catalytic removal method is also different. In order to further improve the effect of soot particle catalytic removal, a statistical analysis method of soot particle catalytic removal method based on big data is designed. Using large data technology to extract catalytic removal methods of soot particles, detailed analysis of each method was carried out, and the soot combustion performance of soot particles catalytic removal method was compared. The results showed that the removal of soot particles based on perovskite catalyst was more effective than that of soot particle removal method based on sol-gel preparation method, and that soot particles were catalyzed by low temperature plasma. The combustion performance of the removal method is better, and the catalytic removal performance is more superior.

Keywords: Big data · Soot particles · Catalytic removal · Sol-gel preparation · Low temperature · Plasma · Perovskite type catalyst

1 Introduction

With the rapid development of industrialization and urbanization, the situation of regional air pollution is increasingly serious. At present, a large number of emissions caused by the substantial increase of vehicle ownership, destroy the ozone layer of the atmosphere, cause the global greenhouse effect, make the whole earth temperature warm and climate abnormal [1]. With the deepening and recognition of the concept of “low carbon life”, energy saving and emission reduction of motor vehicles has been put on the agenda. Therefore, driven by the policy of energy conservation and emission reduction, diesel engine has been more and more widely used for its advantages of large power, high efficiency, good fuel economy, strong adaptability and wide power range. At present, almost all heavy vehicles have used diesel engines, and light vehicles have gradually used diesel engines. As the diesel vehicle is not pollution-free and zero emission, the exhaust emissions of diesel vehicles have caused serious environmental problems. In order to control the rapid development of diesel vehicles and continue to cause serious air pollution, countries around the world have developed relevant vehicle exhaust emission regulations, so as to urge vehicle manufacturers to improve their

products from the source to reduce the serious environmental pollution caused by the emissions of CO, HC, NO_x and soot particles [2].

At present, there are many catalytic removal methods for soot particles, each method has different catalytic removal effect. In order to further improve the catalytic removal effect of soot particles, a method of catalytic removal of soot particles based on big data is proposed. The method of catalytic removal of soot particles is extracted using big data technology. Each method is analyzed in detail and the catalytic removal of soot particles is compared. The flue gas combustion performance of the method provides a basis for the catalytic removal of soot particles. Big data refers to the data collection that can not be grabbed, managed and processed by conventional software tools in a certain period of time. Big data has five characteristics, that is, large amount, high speed, diversity, low value density and authenticity. It has no statistical sampling method, but only observes and tracks what happened. The use of big data tends to predict analysis, user behavior analysis or the use of some other advanced data analysis methods, so it is applied to the catalytic removal of soot particles.

2 Statistical Analysis Framework of Catalytic Removal of Soot Particles Based on Big Data

Big data is a large-scale data collection, and data analysts cannot extract, process, analyze, and manage it with general software within a certain period of time. The key technologies for processing big data include parallel processing technology for large-scale data sets, distributed databases, distributed file storage and processing systems, data mining, and cloud computing. Big data processing tasks cannot be processed in a single-machine serial computing mode. You must use a distributed architecture for calculation. Read the original data from HDFS and send it to the Spark cluster for efficient distributed parallel calculation. After analysis, write the result set to persistent Into the HBase cluster. In spark cluster, it is also divided into Worker nodes that are used to directly participate in calculation, interact with input and output interfaces, and Master nodes that are used for scheduling, task allocation and management of Worker nodes. The statistical process of catalytic removal of soot particles based on big data is shown in Fig. 1:

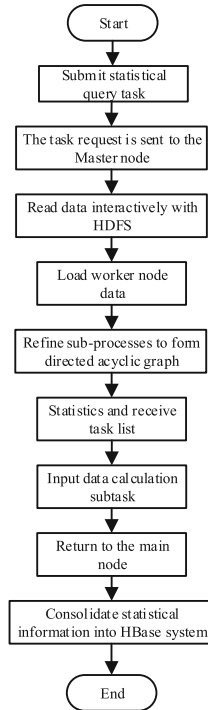


Fig. 1. Statistical flowchart of catalytic removal of soot particles based on big data

Step1: When the statistical query task of the soot particle catalytic removal method is submitted from the user client, the task request is sent to the Master node by the client, The Master node analyzes the size and distribution of the data collection. Because the data comes from HDFS, and HDFS has natural data segmentation (64 MB size per block area), the Master only needs to interact with the HDFS Master node to get the location of each block of data, then assign appropriate reading strategies according to the data location. These strategies include which data of each HDFS Slave node that each Worker node reads from;

Step2: In the process of loading the data of the Worker node, the Master node further refines the task into smaller sub-processes according to the query statement. These sub-processes have data dependencies, forming a directed acyclic graph, and The sub-processes are descriptions that the Worker node can directly execute within it;

Step3: After loading the statistics of the soot particle catalytic removal method and receiving the task list, the Worker node can start calculation based on the input data of the task and the task. After each subtask is calculated, it will be returned to the Master node A completed message is convenient for the Master node to track the progress of task processing in real time;

Step4: After each Worker node completes the calculation separately, according to the user's options when submitting the task, you can choose to schedule and merge the statistics of the soot particle catalytic removal method through the Master's scheduling,

and then write it together into the HBase system, or Each Worker directly writes its own output data to HBase.

The statistical analysis framework for the catalytic removal method of soot particles based on big data is shown in Fig. 2:

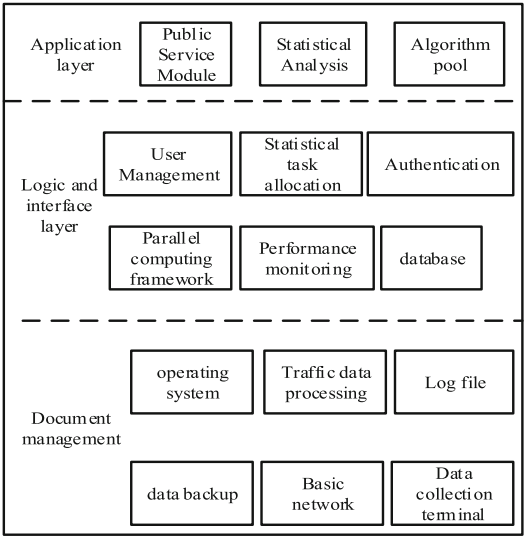


Fig. 2. Statistical framework of catalytic removal of soot particles based on big data

Through the above process, the relevant data about the catalytic removal method of soot particles are extracted, and then the data mining technology in big data technology is used to rank the relevant technologies, and the most commonly used removal methods are calculated. The expression is as follows:

$$f = \frac{g}{j} + \frac{x}{b} \tag{1}$$

In formula (1), g represents statistical data, j represents the definition of statistical indicators, b represents behavioral influencing factors, x represents the dimension of statistical indicators.

Through analysis, the most commonly used removal methods are the following three, the soot particle catalytic removal method based on the sol-gel preparation method, the soot particle catalytic removal method based on low temperature plasma, and the perovskite type catalyst Soot particle removal method.

3 Analysis of Catalytic Removal of Soot Particles

3.1 Catalytic Removal of Soot Particles Based on Sol-Gel Process

The soot particle catalytic removal method based on the sol-gel preparation method is mainly through alumina produced by Wenzhou Alumina Plant, zirconia produced by Sinopharm Group Chemical Reagent Co., Ltd., zirconia produced by Qingdao Ocean Chemical Group Company, Qingdao Ocean Chemical Silicon oxide produced by the group company, manganese acetate produced by Aladdin Reagent Co., Ltd., ammonia water produced by Sinopharm Group Chemical Reagent Co., Ltd., and ethanol produced by Sinopharm Group Chemical Reagent Co., Ltd. as the main experimental reagents [3]. Adopt gas chromatograph produced by Zhejiang Kexiao Instrument Co., Ltd., plasma power supply produced by Nanjing Suman Electronics Co., Ltd., dual dielectric discharge reaction tube, electronic balance produced by Shanghai Mingqiao Precision Scientific Instrument Co., Ltd. The temperature controllers produced by Beijing Huibolong Instrument Co., Ltd., mass flow meters produced by Shanghai Yifeng Electric Furnace Co., Ltd., muffle furnaces produced by Shanghai Yifeng Electric Furnace Co., Ltd. and Shangpu Compact Equipment are the main catalyst making instruments. Mainly used to prepare potassium titanate series catalysts with different K/Ti mass ratios, the precursors of the catalysts $\text{Ti}(\text{OC}_2\text{H}_5)_4$ and CH_3COOH , and the precipitant is H_2O [4]. Dissolve a certain amount of $\text{Ti}(\text{OC}_2\text{H}_5)_4$ in absolute ethanol, and add a certain amount of ethylene glycol to it as a plasticizer, and continue to stir at room temperature to obtain solution A; Then weigh a certain amount with an electronic balance Potassium acetate was placed on a magnetic stirrer and stirred well until potassium acetate was completely dissolved and dissolved in absolute ethanol. A certain amount of glacial acetic acid and a small amount of deionized water were added and stirred to obtain solution B. The solution at this time was a clear solution. Under constant stirring conditions, the solution B was added dropwise to the solution A, and the PH value of the solution was controlled to about 6 during the preparation process; stirring was continued for a while to obtain a sol-like substance, and the gel was obtained by standing. Then it is aged at room temperature for a period of time, placed in an electric blast drying oven at 100 °C for drying, then ground, and placed in a muffle furnace at 800 °C for roasting, and finally obtained $\text{K}_2\text{Ti}_2\text{O}_5$, $\text{K}_2\text{Ti}_4\text{O}_9$, $\text{K}_2\text{Ti}_6\text{O}_{13}$, $\text{K}_3\text{Ti}_8\text{O}_{16}$ and $\text{K}_4\text{Ti}_{10}\text{O}_{18}$ five potassium titanate catalysts [5]. Potassium titanate is a new type of high-performance material enhancer. It exists in the form of whiskers. This whisker is a new type of inorganic polymer compound with micro needle-shaped short fibers. Its chemical formula is $\text{K}_2\text{O} \cdot n\text{TiO}_2$ or $\text{K}_2\text{Ti}_2\text{O}_{2n+1}$. When $n = 2, 4, 6, 8$, it is called potassium dititanate, potassium tetratitanate, potassium hexatitanate and potassium octatitanate respectively [6].

Potassium titanate belongs to monoclinic system and C2/m point group. Ti the crystal structure of potassium dititanate with Ti coordination number of 5 is a chain-like layered structure formed by connecting the triangular double cones of TiO_6 through a common vertex. The layers are separated by 6.5 angstroms. Parallel, and K^+ is located between the layers of the layer structure, so it has chemical activity. Similarly, in the crystal structure of potassium tetra titanate whose coordination number of Ti is 6, it is a lamellar structure formed by the connection of the TiO_6 octahedron

through common edges and common apical angles. The layers are also parallel to the crystal axis, with a spacing of 8.5 angstrom. K^+ is located between layers, so it also has chemical activity. The K^+ ions in the middle layer of the structure of potassium diti-tanate and potassium tetra titanate crystals can be exchanged with other cations to replace the heavy metal ions in the wastewater, so they can be applied to the treatment of metal ions. In the crystal structure of potassium octatitanate different from the first two, the coordination number of Ti is 6, which is a tunnel structure formed by octa-hedron through common edges and common angles, rather than a layered structure. Among them, K^+ is in the middle of the tunnel, separated from the environment, and the K^+ in potassium octotitanate crystal has almost no chemical activity. Figure 3 is a schematic diagram of the cell structure of $K_2Ti_2O_5$ established by molecular simulation. The combination of the red and white spheres represents the triangularis of TiO_6 , and the purple spheres represent potassium ions. It can be seen that they are connected by common vertices. TiO structure is uniformly distributed at different levels, with large space gaps and obvious regularity. However, potassium ions are distributed between the levels of TiO structure, which provides excellent conditions for the good chemical activity of potassium ions.

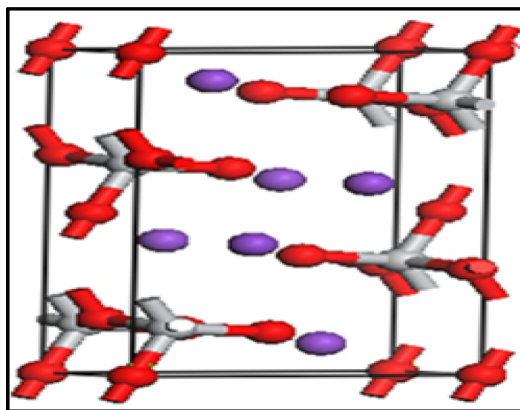


Fig. 3. Crystal cell structure of $K_2Ti_2O_5$

In addition, the following means of characterization were used for the removal effect of characterization:

First, XRD test was performed to determine the composition of the catalytic active agent by Rigaku D-max-rA Xray diffractometer. Experimental conditions: at room temperature, Cu target $K\alpha$ beam, graphite monochromator, X-ray wave-length = 0.1548 nm, step length: 0.02, tube pressure: 40 kV, tube flow: 100 mA, scanning range: $10^\circ \sim 90^\circ$, scanning rate: $10^\circ \cdot \text{min}^{-1}$.

Second: BET test was conducted on Micromeritics ASAP2000. The sample was pretreated at 623 K and adsorbed in a liquid nitrogen environment. The cross-sectional

area of nitrogen molecule was set at 0.162 m^2 . The specific surface area was obtained by the BET equation and the nitrogen adsorption isotherm.

Thirdly, SEM test was performed to observe the morphology and size of potassium titanate catalyst particles prepared by scanning the electroscope of JSM-6700 from Japan Jeol co., LTD.

Fourth: thermogravimetric (TG) determination. The combustion temperature of the catalyst for the catalytic oxidation of carbon smoke particles is determined on the thermogravimetric analyzer. The tga is a type TAC/DX thermal analyzer manufactured by the United States company. First, a certain amount of catalyst samples (about 10 mg) were taken and pretreated at 300°C for 60 min in N_2 atmosphere. Then, after the temperature dropped to 100°C , the gas passing through the catalyst sample was switched to a mixture of $20\%\text{O}_2/\text{N}_2$ and heated up to 800°C at a heating rate of $10^\circ\text{C}/\text{min}$.

Fifth, the morphology of the catalyst before and after the reaction was observed by transmission electron microscopy (TEM). The instrument was HT-7700 high-resolution electron microscopy (ht-7700) of Hitachi high-tech co., LTD. Before the test, the sample powder was dispersed evenly by ultrasound in anhydrous ethanol, and then a small amount of suspension was taken and transferred to the carbon film made of copper mesh. After being irradiated to complete drying by infrared lamp, the sample powder could be put into the instrument for testing. The soot combustion curve after catalytic removal of soot particles prepared based on sol-gel is shown in Fig. 4:

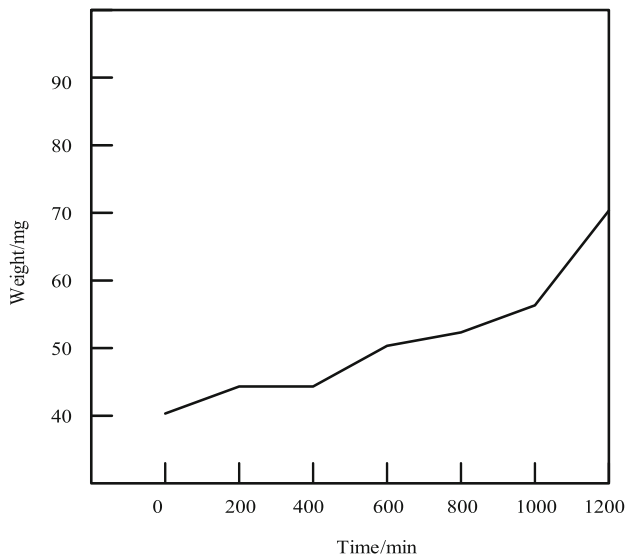


Fig. 4. Combustion curve of soot after catalytic removal of soot particles prepared by sol-gel process

The starting temperature and maximum oxidation rate temperature of different catalyst contents are shown in Table 1:

Table 1. Starting temperature and maximum oxidation rate temperature of catalyst

Catalyst content/%	The light-off temperature/°C	Maximum combustion temperature/°C
20	276	413
40	279	420
60	295	450
80	270	368
100	480	580

3.2 Catalytic Removal of Soot Particles Based on Low Temperature Plasma

The plasma-enhanced catalysis process is also known as the “two-stage process”, in which the plasma and the catalyst are placed separately and the catalytic reactor is located behind the plasma reactor. In the reaction system, the reaction process is mainly divided into two steps: first, the reaction gas is activated in the plasma reactor, which can produce relatively more reactive intermediate products and or produce odor; Then these intermediates leave the plasma reactor and enter the catalytic reactor to produce the final target product under the action of the catalyst. The reaction atmosphere is the air purified by the air generator. A mass flow controller is used to control the gas flow rate of 30 mL.min⁻¹. In the coordinated catalytic reaction between the plasma and the catalyst, the catalyst is filled in the discharge area of the plasma. The filling mass of the catalyst is 0.5 g, and the reaction temperature is within the range of 20 °C ~ 200 °C. The reaction products were detected online by gas chromatograph and then detected by thermal conductivity detector TCD.

In the catalytic oxidation of carbon smoke particles, the perovskite catalyst with ideal structure is not very active, while the perovskite catalyst with defect structure after distortion shows good catalytic activity. Therefore, the preparation of perovskite catalyst with defect structure has become a hot research topic. Generally speaking, the preparation methods of perovskite catalysts with defective structures mainly include A and B site ion doping and non-stoichiometric ratio synthesis [7], as follows:

a. Conduct ion doping at the a level and replace the high-priced A level ions (rare earth metals) with low-priced A level ions (alkali earth metals or alkali metals) doped into the lattice of perovskite.

b. Replace the low-priced A-bit ions with the high-priced A-bit ions at the a-bit ions, thereby reducing the valence state of the A-bit ions and producing the low-priced b-bit ions;

c. Carry out ion doping at the A-bit, and replace the high-priced A-bit ions with low-priced b-bit ions.

The following device was used to evaluate the performance of the catalyst. The first part is a catalytic reaction unit with a quartz tube reactor [8, 9] and a heating furnace. The second part is the exhaust gas detection device, namely gas chromatograph. The third part is the computer data output device.

The co-catalytic removal of soot particles by plasma was performed on the following plasma-driven catalytic devices.

The dielectric barrier plasma discharge was adopted, and the quartz tube with an internal diameter of 8.0 mm was used as the reactor and the barrier medium. A 3.0 mm diameter unembroidered steel rod is used as the high voltage electrode. At the same time, the stainless steel wire mesh wound on the surface of the quartz tube is used as the grounding electrode. The reaction device in the experiment is controlled by a programmed temperature rise controller in an open-type tubular resistance furnace. The high voltage power supply device is used to control the discharge voltage of the plasma.

The low temperature plasma discharge is dielectric barrier discharge. The catalyst is placed within the low-temperature plasma discharge region, that is, the combination mode of plasma and catalyst is plasma-driven catalytic reaction mode. Low-temperature plasma discharge power supply is high frequency and high voltage power supply (CTP-2000 K, nanjing suman electronics co., LTD.), which can provide 0–30 kV, 9–16 kHz sinusoidal ac voltage. The discharge power of dielectric barrier discharge plasma is an important parameter affecting plasma discharge. Due to the phase imbalance between the current and voltage of dielectric barrier discharge plasma, its power calculation and measurement are quite complicated. The power of dielectric barrier discharge can be determined accurately by analyzing the power of dielectric barrier discharge by using the pattern of discharge voltage and charge li sayuan in plasma.

The discharge voltage and current waveform and discharge frequency in the process of low-temperature plasma discharge were measured by oscilloscope [10, 11]. The discharge power of the plasma was measured by the lisaru method:

$$p(w) = \frac{s(c)}{j} \quad (2)$$

In formula (2), w represents the total charge on the capacitor, which is connected to the grounding electrode; s Represents the voltage at both ends of the capacitor; c Represents the discharge voltage, j represents the discharge frequency.

After removal of soot particles by catalytic removal method based on low-temperature plasma, the soot combustion curve is shown in Fig. 5:

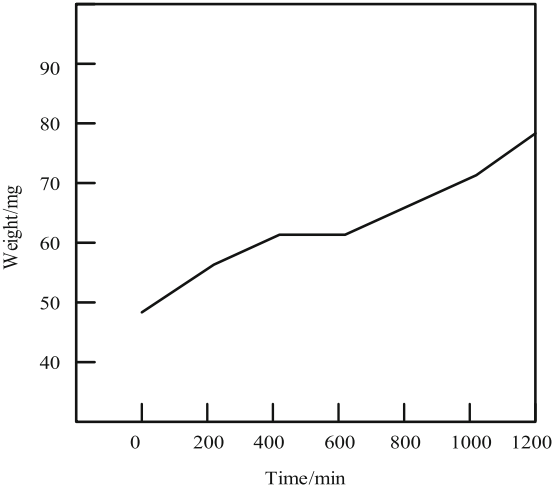


Fig. 5. Combustion curve of soot particles after catalytic removal based on low temperature plasma

The starting temperature and maximum oxidation rate temperature of different catalyst contents are shown in Table 2:

Table 2. Starting temperature and maximum oxidation rate temperature of catalyst

Catalyst content/%	The light-off temperature/°C	Maximum combustion temperature/°C
20	125	300
40	145	320
60	295	200
80	100	250
100	120	320

3.3 Removal of Soot Particles Based on Perovskite Catalyst

The catalytic activity of the catalyst was studied in a TPO unit, and the reaction product was analyzed online by gas chromatograph. The reaction device mainly consists of three parts: the catalytic reactor of the programmed temperature control device, the gas chromatograph (SP-3400, Beijing beifenruili co., LTD.) and the computer for recording data [12, 13]. The schematic diagram of the reaction device is shown in Fig. 6:

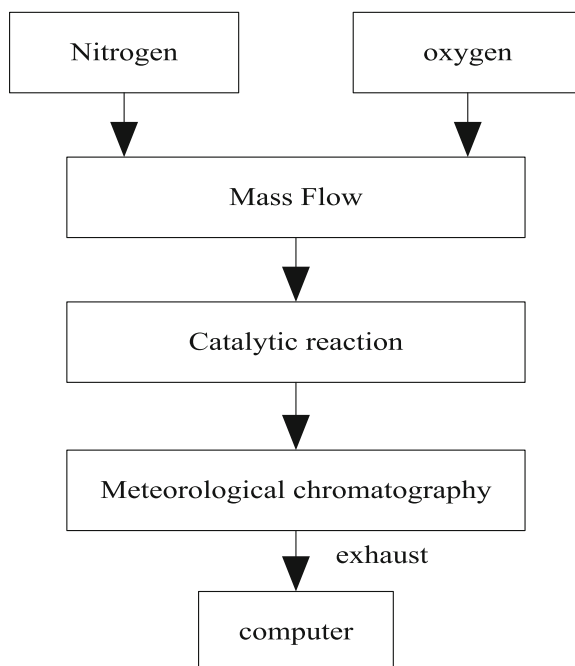


Fig. 6. Schematic diagram of temperature programmed reaction device

The performance of perovskite catalyst was tested by the simulated tail gas evaluation method. The specific process is as follows:

First of all, the contact mode between the soot and the catalyst is an important factor affecting the catalytic performance, and the catalyst always exhibits better activity under tight contact conditions than under loose contact conditions. But in practice, the contact between the carbon smoke and the catalyst in the tail gas device is loose. In order to comply with the actual conditions, the carbon smoke and the catalyst were directly mixed and ground in this experiment, so that the carbon smoke could be attached to the catalyst and fully contacted.

Second, in the process of the catalytic reaction, at a rate of about 1 L/min to device zhongtong into the air to ensure there is plenty of oxygen, the heating rate of $5\text{ }^{\circ}\text{C}/\text{min}^{-1}$, using the mass flowmeter control O_2 volume fraction (10%) and N_2 (gas) as the balance of flow rate of 200 ml/min, using the mixed gas to simulate the rich oxygen condition of diesel exhaust, the purpose is to test whether potassium titanate catalysts system has low temperature catalytic oxidation of carbon smoke particles combustion of diesel exhaust good activity. After the reaction, the gas was detected online by SP-3400 gas chromatograph, and the catalytic activity of the catalyst was evaluated according to the data.

Chemical kinetics is a branch of science that studies the reaction rate and reaction mechanism of the transformation of one chemical substance into another. However, the study of catalytic kinetics has become one of the most important components in the

field of catalysis. In the study of catalyst science and engineering, one of the important objectives of the study of catalytic kinetics is to establish an appropriate kinetic mathematical model for the reaction studied and to help explore the mechanism of its catalytic reaction. The study of reaction dynamics can help to provide an appropriate mathematical model. The model can accurately and reliably reflect the influence of reaction conditions such as airspeed, temperature and pressure on the selectivity of reactants, reaction speed and conversion rate, and provide a suitable mathematical model for the study of catalyst design and catalytic kinetics. Moreover, in a large range, this model has been able to reflect more accurately and confidently the influence of space speed, temperature, pressure and other reaction conditions on the selectivity, reaction rate and conversion of reactants. As the combustion of gasoline is more complex, the composition of the combustion product soot is also different under different circumstances. Since the combustion temperature of soluble organic matter is much lower than that of dry carbon smoke, the combustion of carbon smoke can be regarded as the combustion of dry carbon smoke. Oxygen is first adsorbed on the surface of the catalyst, and then combined with the active substances in the catalyst to form $[-MO]$. The active oxygen reinteracts with the soot particles, and some of it generates CO_2 , while some of it generates CO . The resulting gas molecules are then released through desorption of the desulfurization catalyst, resulting in the reduction of the catalyst.

In the study of reaction kinetics, reaction series is a very important factor. Each reaction level corresponds to its reaction characteristics. According to the reaction series, the reaction rate equation can be known. Based on the first order, second order and third order differential diagrams of carbon smoke oxidation, the reaction progression of carbon smoke combustion is determined in this paper.

Figure 7 shows the soot combustion curve after removal of carbonaceous smoke particles by the removal method based on perovskite catalyst:

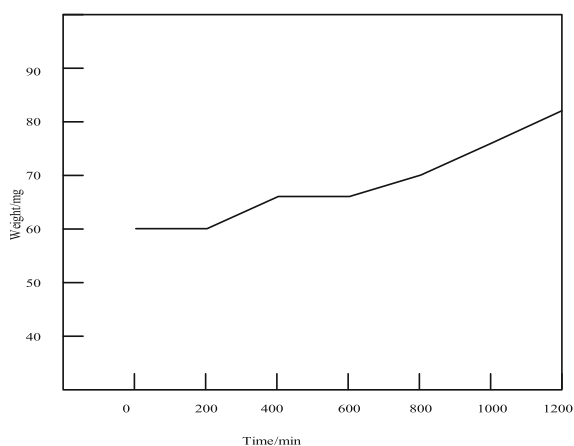


Fig. 7. Combustion curve of soot after removal of soot particles based on perovskite catalyst

The starting temperature and maximum oxidation rate temperature of different perovskite catalysts are shown in Table 3:

Table 3. Starting temperature and maximum oxidation rate temperature of catalyst

Catalyst content/%	The light-off temperature/°C	Maximum combustion temperature/°C
20	300	600
40	350	650
60	400	700
80	450	650
100	500	600

4 Discussion of Results

Using the programmed heating and oxidation device as the potassium titanate catalytic soot reaction device and the meteorological chromatograph, the post-reaction gas was detected online, and the experimental results were systematically analyzed and studied. Satisfactory results were obtained (Table 4):

Table 4. Experimental results

Different methods	Method for catalytic removal of soot particles based on sol-gel method	Low-temperature plasma-based catalytic removal method of soot particles	Method for removing soot particles based on perovskite catalyst
Burning time/min	1200	1200	1200
Burning weight/mg	70	78	82
Catalyst content/%	100	100	100
The light-off temperature/°C	480	120	500
Maximum combustion temperature/°C	580	320	600

First: when the burning time reaches 1200 min, based on the sol-gel preparation methods of carbon smoke particle catalytic removal method, based on the low temperature plasma carbon smoke particle catalytic removal method, based on perovskite catalyst of carbon smoke particle removal method after removing carbon burning smoke gradually rising trend curve. Among them, the combustion weight of the soot

particle catalytic removal method based on the sol-gel method is 70 mg, the combustion weight of the soot particle catalytic removal method based on low-temperature plasma is 78 mg, and the soot particle catalytic removal based on the perovskite catalyst the burning weight of the method is 82 mg. After comparing, the perovskite catalyst based carbon smoke particle removal method of removal effect is best.

Second: when the catalyst content reaches 100%, the starting temperature of the method for catalytic removal of soot particles based on the sol-gel method is 480, and the maximum combustion temperature is 580, respectively. The start of the method for catalytic removal of soot particles based on low-temperature plasma the temperature is 120, the maximum combustion temperature is 320, the starting temperature of the perovskite catalyst-based soot particle removal method is 500, and the maximum combustion temperature is 600. It can be seen that, although the removal method of carbon smoke particles based on perovskite catalyst has the best removal effect, it requires higher starting temperature and maximum combustion temperature. The method of catalytic removal of carbon smoke particles based on low temperature plasma has the lowest requirements for the starting temperature and the maximum combustion temperature.

Thirdly, it is found that the potassium titanate catalyst is not easy to absorb water and has good stability and anti-aging performance. Moreover, the catalytic performance of carbon smoke is still good under loose contact. It is also found that a certain amount of NO_x catalysis is beneficial to the oxidation of carbon smoke.

Fourth, potassium titanate catalyst can still effectively catalyze the combustion of carbon smoke particles at low temperature even when it is in loose contact with carbon smoke, and has good catalytic activity, which fully proves that potassium titanate catalyst is an excellent catalyst for the oxidation of carbon smoke.

Fifthly, the mass ratio between catalyst and soot particles did not significantly change the catalytic oxidation of soot by potassium titanate through the experiment, which fully verified the good catalytic performance of the removal method of soot particles based on perovskite catalyst.

5 Conclusion

This paper presents a statistical analysis method based on big data for the catalytic removal of soot particles. Using big data technology, extract the catalytic removal method of soot particles, and compare and analyze the flue gas combustion performance of catalytic removal of soot particles according to different methods. The results show that the perovskite-type catalyst is more effective in removing soot particles than the sol-gel preparation method, and the low-temperature plasma catalyzed soot particle catalytic removal method has better combustion performance and better catalytic removal performance.

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