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Development and Status of Spontaneous Coal Combustion Research in China

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Abstract Spontaneous coal combustion is the result of physical and chemical interactions between coal and oxygen over a long period of time. It is a complex physical and chemical process that only occurs under certain conditions (such as low-temperature oxidation and heat accumulation), during which the temperature of the coal rises, eventually reaching the ignition point. It is also a common cause of major disasters in coal mines, as it threatens mine safety, coal production, and coal applications. In order to deeply grasp the research and development status of coal spontaneous combustion in China and point out the direction for future research, firstly, this paper introduces the energy status of China's coal resources, then expounds the development process and theory of coal spontaneous combustion, summarizes the characters and related problems of different spontaneous combustion test methods, and thirdly introduces the main institutions and personnel of coal spontaneous combustion and the corresponding results, finally this paper discusses the existing problems and future development direction of coal spontaneous combustion in China.

Keywords: *spontaneous coal combustion, research institutions, representative figures, representative results*

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1. Introduction

China's total energy consumption reached 4.86 billion tons of standard coal in 2019, with an average annual growth rate of 2.9%, and the use of standard coal increased by 5.3 billion tons compared to 2015. Coal accounted for approximately 57.7% of the total primary energy consumption in 2019 [1]. The energy supply issue is not only related to China's security and economic development, but also to environmental protection and climate change responses.

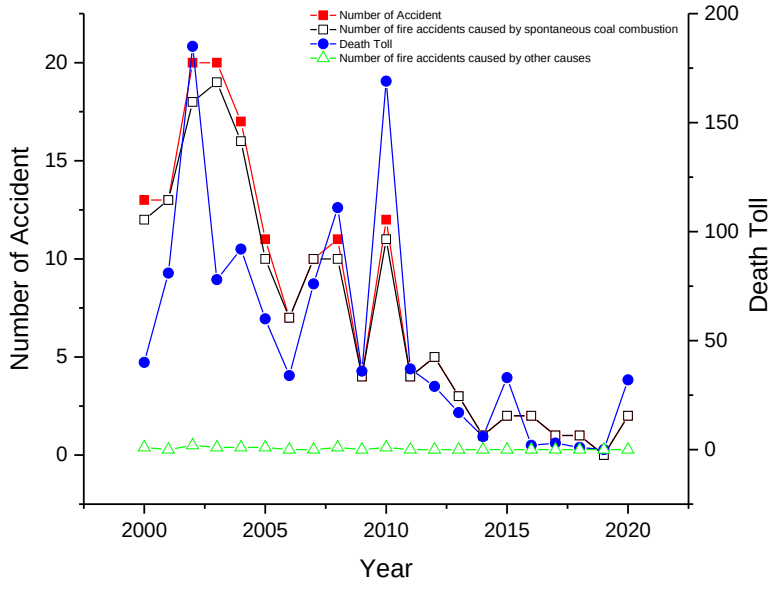

Figure 1. Coal mine fire accident statistics

However, with the continued growth of coal production, various types of coal mine accidents such as mine fires and gas and coal dust explosions have occurred, resulting in high economic losses and a high number of casualties. In recent years, the number of coal mine fire accidents and the resulting deaths have decreased annually through the prevention and control of coal mine fires (Figure 1), however, major fire accidents still occur [2]. Through the accident data query system of the National Mine Safety Administration [3], we counted the number of coal mine fire accidents from 2000 to 2020. We found that accidents caused by spontaneous coal combustion accounted for the vast majority of coal mine fire accidents. In the past 10 years, the direct cause of coal mine fires in China was spontaneous coal combustion.

2. Development Process of Spontaneous Coal Combustion

2.1. Development and Status

Spontaneous coal combustion generally refers to a small amount of heat generated by physical adsorption, chemical adsorption, and oxidation reactions between coal and oxygen in air at room temperature. Under certain conditions, the oxidation heat generation rate is greater than the rate of heat dissipation to the environment, and heat accumulation causes the coal temperature to rise slowly but continuously. When the critical self-heating temperature of coal reaches, the oxidation heating rate accelerates, and finally, the ignition temperature is reached, and the coal is burned. According to existing researches, the oxidation and spontaneous combustion of coal is considered basic chain reactions, and the process of spontaneous coal combustion is generally divided into three stages: low-temperature oxidation, self-heating, and combustion.

2.2. Analysis and Summary

On the basis of summarizing the research results of experts at home and abroad, our team summarized and simplified the sectional characters of coal spontaneous combustion by studying the heat production, oxygen consumption, products and so on in the process of coal spontaneous combustion. The details of that research are summarized in Table 1.

Table 1. Comprehensive characteristics of spontaneous coal combustion

Research Stage	Heat Characteristics	Oxygen Consumption and Gas Product Formation
Slow Temperature Rise	The heat which releases during the slow temperature rise stage is very small, up to 20% of total heat releases during the whole reaction process.	The oxygen consumption of each coal sample at this stage is very small, almost negligible. At lower temperatures CO, CO_2, and CH_4 are produced, of which the CO and CO_2 content is relatively large. In addition to CH_4, the generation temperature of other gases in alkanes is higher, and with the increase in the number of C-atom alkanes, the occurrence time is gradually increased.
High Temperature Rapid Heating	The heat which releases during the high-temperature rapid heating stage is very large, accounting for more than 80% of total heat release during the whole reaction process.	The oxygen consumption and the production of gas products increased significantly, and the increase degree of different coal types are also different. The more easily spontaneous combustion of coal, the greater oxygen consumption, and the corresponding temperature is also lower when consumption begin to increase significantly.

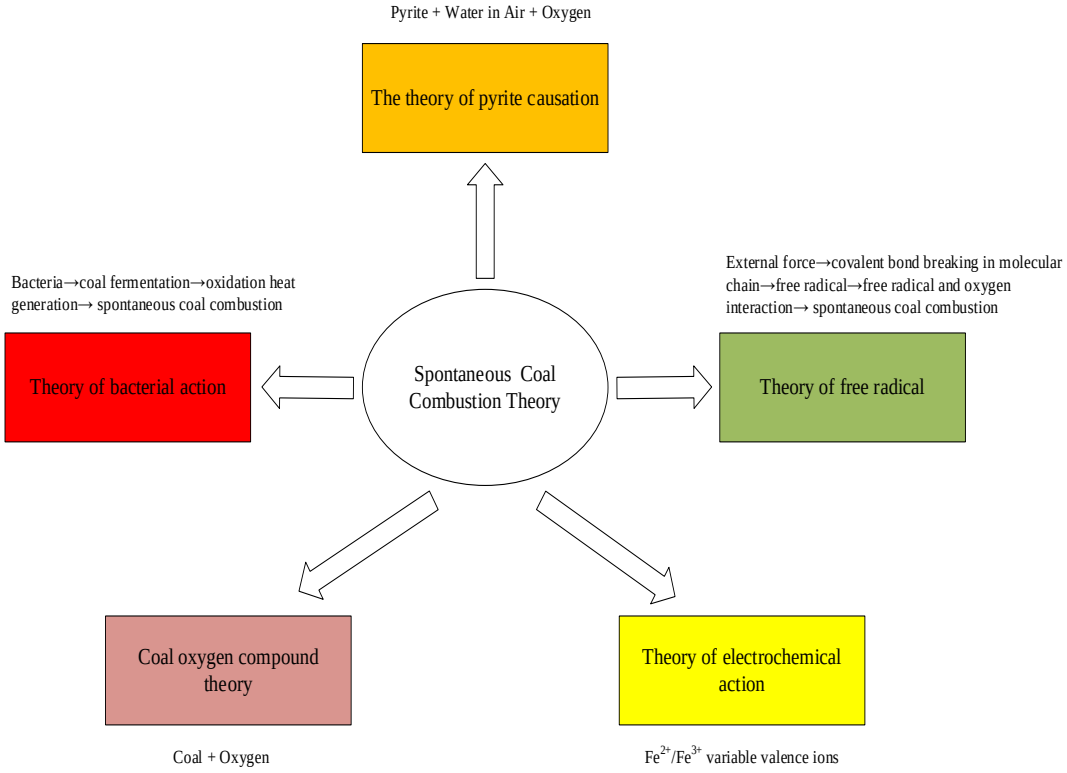

Figure 2. Development of the spontaneous coal combustion theory

3. Development of The Spontaneous Coal Combustion Theory

3.1. Development and Status

Plott and Berzehus first proposed the theory of pyrite genesis in the 17th century [4]. Potter et al. believed that a certain amount of heat released from coal during the fermentation of bacteria plays a decisive role in its self-ignition [4]. In 1940, Soviet scholar TponoB proposed that the self-heating of coal was due to the strong adsorption of oxygen in the air by unsaturated phenolic compounds in the coal, while releasing a certain amount of heat [4]. Drehm proposed the theory of electrochemical action in 1990 [5], and in 1996, Professor Li Zenghua of China University of Mining and Technology observed that coal broke under the action of an external force, resulting in a large number of cracks, which inevitably caused the fracture of the coal molecules [6]. In 1998, Lopez et al. proposed the hydrogen atom interaction theory [7]. In 1870, Rachtan discovered through experiments that the oxygen uptake per 1 g of coal in 24 hours was 0.1–0.5 ml, while that of lignite was 0.12 ml [7]. In 1945, Jones proposed that the oxygen uptake of bituminous coal can reach 0.4 ml/g in air at room temperature [7]. In the 1960s, the Fushun Coal Research Institute used the oxygen uptake of coal as an indicator to distinguish the spontaneous combustion tendency of coal. These theories are summarized in Figure 2.

3.2. Analysis and Summary

The pyrite action theory holds that the spontaneous combustion of coal is caused by the presence of pyrite in the coal seam, as pyrite interacts with water and oxygen in air and causes thermal reactions. However, further research found that spontaneous coal combustion still occurs without the presence of pyrite, thus setting limits on this theory and ultimately disproved it.

Bacteria may play a role in the self-heating process of coal, but they are likely not the main cause of spontaneous combustion, as the spontaneous combustion of coal is based on a chemical chain reaction process rather than on bacterial action.

Phenol gene theory clarifies that the coal-oxygen composite is the primary cause of spontaneous combustion, which can be considered as a supplement to the coal-oxygen composite theory.

The theory of electrochemical action that coal contains variable valence iron ions, and the redox system Fe2+ and Fe3+ play a catalytic role in the oxidation of coal, causing an electrochemical reaction in the coal and producing a chain reaction with chemical activity, thus greatly accelerating the automatic oxidation process of coal, causing spontaneous combustion.

Free radical theory analyzes the reaction process of various organic compounds in coal and reveals that organic compounds in coal can act in pure oxygen or other oxidants. This theory can be used as an extension of the coal-oxygen composite theory, but this theory has not been widely applied.

The coal-oxygen composite theory of coal-oxygen composite has several problems, primarily a lack of understanding regarding the initial cause of the coal-oxygen composite, the process of coal-oxygen composite, how to determine various critical parameters, how to determine the thermal effect in the low-temperature stage, how to determine the shortest spontaneous combustion period of coal, how to transport oxygen in coal.

4. Determination of the Spontaneous Coal Combustion Method

4.1. Development and Status

When conditions are appropriate, self-heating occurs, and heat continues to accumulate. When the temperature reaches the ignition point, spontaneous combustion occurs. Such fire-related phenomena not only leads to the loss of available calorific value, but also causes serious safety issues in coal mining, processing, and utilization. In recent years, according to the specific process of spontaneous coal combustion, several major coal-producing countries created methods to detect the tendency of their coal to spontaneously combust and this led to the formation of national standards (Table 2).

Table 2. Test methods of spontaneous coal combustion

Research Establishment	Main Methods Used	Application
Australian Mining Safety Research and Testing Center	Self-heating temperature method (SHT; adiabatic R_{70} test; gas composition analysis; traditional cross-point temperature method	Currently in the commercial testing phase [8]
Department of Chemistry and Materials Engineering, University of Auckland, New Zealand	New cross-point temperature method	CPT method was proposed and some materials were tested [9]
Krakhov Mine, Poland	Activation energy method	Industry standard [10]
College of Environmental and Materials Engineering, Leeds University, UK	New cross-point temperature method	Test on spontaneous combustion characteristics of Indonesian coal by CPT method [11]
College of Engineering, University of Aberdeen, UK	Improved heat release（HR）method; micro calorimeter method.	The standard ignition temperature method is questioned: using single activation energy will lead to greater uncertainty [12]
Labor Department of the United States American	SHT	No longer tested; SPONCOM software developed [13]
Turkish Middle East University of Technology	Cross-point temperature method	Most commonly used method [14]
Fushun Research Institute of China Coal Technology Group	Dynamic oxygen inhalation method	National standard [15]
China University of Mining & Technology	Oxidation kinetics test method	Industry standard [16]

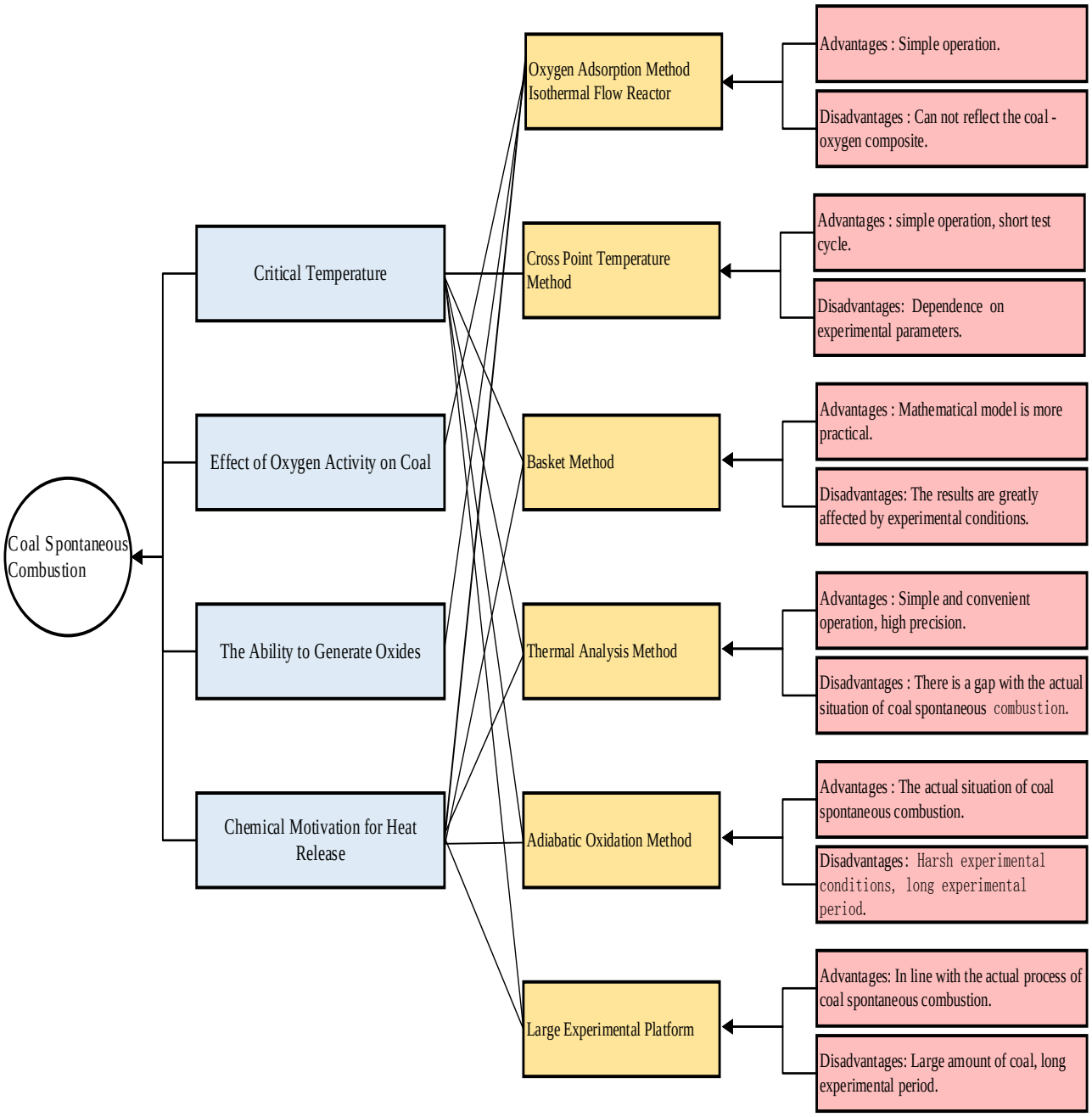

Figure 3. Summary of test methods

4.2. Analysis and Summary

The methods described in Table 2 evaluate spontaneous coal combustion and the strength of the combustion tendency by using different approaches. Different test methods have their own functional characteristics, which can only reflect the spontaneous combustion tendency of coal to a certain extent. Overall, the test indices for each method do not form a unified standard. In practical applications, choosing a method becomes difficult, therefore, this study aims to summarize the commonly used methods to detect spontaneous coal combustion both in China and abroad. These methods are divided into four categories according to the types and test methods of evaluation and development indicators; the characteristics and current problems of these methods are also analyzed and compared (Figure 3).

5. Current Status of Spontaneous Coal Combustion Research in China

5.1. Primary Institutions and Researchers

After consulting the relevant literature, we identified several primary research institutions that have been investigating spontaneous coal combustion measurements in China in recent years. These institutions include six national key laboratories, three provincial and ministerial key laboratories, two ministerial key laboratories, two provincial key laboratories, and one other research institution. The institutions location are mainly distributed in the central, eastern and western parts of China (Figure 4). The specific research contents are shown in Table 3.

Figure 4. Locations of research institutions

Table 3. Primary research institutions for determination of spontaneous coal combustion in China

Organization Name	Representative Figures	Representative Results
State Key Laboratory of Coal Mine Disaster Dynamics and Control Henan Polytechnic University	Minggao Yu	The coal caving model of the working face is established by using a self-made experimental crushing device [17].
State Key Laboratory of Fire Science	Haihui Wang	The methods for studying the interaction effect of coal and oxygen and the subsequent determination methods of spontaneous coal combustion tendency were summarized, their advantages and disadvantages are expounded [18].
State Key Laboratory of Coal Resources and Safe Mining Key Laboratory of Gas and Fire Control for Coal Mines	Deming Wang Fubao Zhou Botao Qin	The 13 elementary reactions in the spontaneous coal combustion process and their reaction order and secondary relationships were established [19].
State Key Laboratory of Coal Mine Safety Technology	Yuntao Liang	The determination method of hidden high-temperature zone layers under the influence of complex catastrophic geological bodies and an early warning index for spontaneous combustion fire was proposed [20].
	Haizhu Luo	The flow state chromatography oxygen adsorption identification method of spontaneous coal combustion tendency was established.
State Key Laboratory of Efficient Mining and Clean Utilization of Coal Resources	Shaowu Pan	Fourier transform infrared spectroscopy and peak separation technology were used to quantitatively study the functional group characteristics of long flame coal and anthracite with different metamorphic degrees [21].
State Key Laboratory of Clean Energy Utilization	Jianzhong Liu	The characteristics and existing problems of different test methods were summarized. The development trend of test methods of spontaneous coal combustion tendency was prospected [22].
Shanxi Mine Ventilation and Fire Prevention Engineering Technology Research Center Ministry of Education of Coal Science and Technology and Shanxi Key Laboratory	Jianming Wu	The feasibility of predicting spontaneous coal combustion by hydrogen combined with various index gases was discussed [23].
	Weiren Bao	Fourier transform infrared spectroscopy and the nitrogen adsorption method were used to analyze and test the functional groups and pore structure parameters of coal samples.
State Key Laboratory of Ministry of Education by Provincial Department of Coal Mine Safety and Efficient Mining	Guanlong Dai	The law of self-heating, low-temperature oxidation of lignite, gas coal, gas-fertilizer coal, and anthracite were revealed by combining macro and micro ways [24].
Key Laboratory of Western Mines and Hazard Prevention, Ministry of Education	Jingcai Xu	The theory of determining the risk area of spontaneous coal combustion is proposed. The colloid fire prevention and extinguishing theory of coal seam fire was established [25].
State Key Laboratory of Coal Fire Disaster Prevention in Shaanxi Province	Jun Deng	The unsteady mathematical model of the coalfield fire area physics and the coupling process of heat transfer, mass transfer, chemical reaction, stress strain and thermal failure in the coalfield fire area were established.
	Hu Wen	A coal seam spontaneous combustion monitoring and multivariate information fusion analysis system was developed to realize the quantitative identification of spontaneous combustion location, temperature, and time in goaf.
Key Laboratory of Mine of thermo-motive Disaster and Prevention Ministry of Education	Jiren Wang	The chemical structure models of coal organic macromolecules and low molecular compounds were constructed [26].
State Key Laboratory of Mine Disaster Prevention in Hebei Province	Jianfang Zhu	The distribution characteristics of the main functional groups in the low-temperature oxidation process and the change law with temperature were compared and analyzed by an infrared spectroscopy experiment [27].
State Key Laboratory of Mining Development and Safety Technology in Hebei Province	Fu sheng Wang	The low-temperature oxidation characteristics of five different metamorphic coal samples were studied, and the reasons for the difference in the comprehensive determination index of oxidation kinetics were analyzed.

Table 4. Spontaneous coal combustion test equipment

Equipment Name	Usage
Fourier Transform Infrared Spectrometer (FTIR)	According to the absorption peak position and position movement of various functional groups in coal, the composition and category of functional groups in coal were determined by infrared spectrum analysis. The type and content of functional groups determine the position and intensity of the spectral absorption peaks.
Differential Scanning Calorimetry (DSC)	DSC is used to study changes in the physical quantity of materials undergoing changes in temperature, allowing for the relationship between power difference, mass difference, and temperature of the sample and the reference material to be measured at the same time.
ThermoGravimetry (TG, DTG)	
Nuclear Magnetic Resonance (NMR)	The hydrogen and carbon spectra are primarily used to study coal structure information. However, due to the complexities of the structure of coal, the NMR test, and spectrum analysis process, there have been some technical defects in past coal structure constructions.
Electron Spin Resonance (ESR)	This is used to qualitatively and quantitatively detect unpaired electrons in atoms or molecules of matter and to explore the structural characteristics of surrounding environments.
Scanning Electron Microscopy (SEM)	SEM is used to observe the details of each area of coal sample surface.
Gas Chromatography	A gas chromatograph is mainly used to determine CO, CH4, C2H4, H2, and other gases generated during spontaneous coal combustion.
Laser Granularity Analyzer	This instrument is used to determine the particle size and distribution of coal.
Specific Surface and Pore Size Analyzer	This instrument is used to determine the specific surface area and pore size distribution of coal samples.

5.2. Modern Testing Equipment Used by Research Institutions

Through the analysis of journals, dissertations, and public data released by the laboratory's official website (Table 3), we concluded that the primary means of studying the characteristics of the coal low-temperature oxidation process in China is the spontaneous coal combustion characteristics test device (thermal reaction furnace) developed independently or jointly by various research institutions. This device was used to carry out experiments, and parameters such as oxygen consumption, products, and temperature change of coal was primarily measured macroscopically. With further development of coal science and technology, as well as coal testing technology, some measurements of microstructure changes, such as functional groups and free radicals, are now widely used by major research institutions in China. The characteristics of the devices are listed in Table 4.

Chinese scholars [28] have found the active sites of coal by modern testing and quantum chemical calculation methods and have mastered and obtained the active site structure and thermodynamic parameters of each element. This has revealed the elemental reaction process of coal oxidation and combustion and established the chain cycle process of the elemental reaction path in spontaneous coal combustion (Figure 5).

5.3. Key Findings of Research Institutions

Due to numerous cross-wise tasks, projects, invention patents, software copyrights, and related papers of the above research institutions, in order to facilitate statistical collation, this paper only makes a statistical collation of the representative national and provincial longitudinal projects and national, provincial, and association awards of the research institutions in the past ten years, as shown in Table 5.

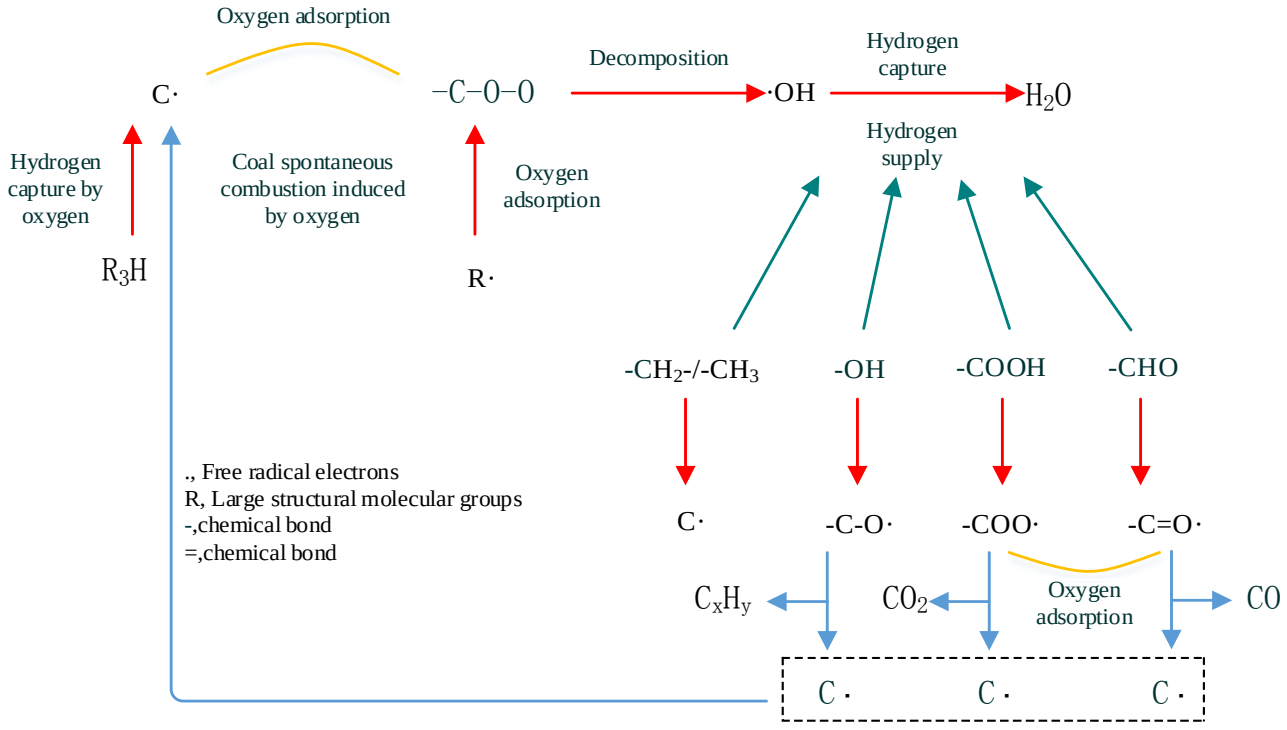

Figure 5. Spontaneous coal combustion in the chemical reaction diagram

Table 5. The achievements of spontaneous coal combustion research institutions

Organization name	Subordinate units	National vertical projects	Provincial or ministerial vertical projects	National awards	Provincial or ministerial awards	Industry association awards
State Key Laboratory of Coal Mine Disaster Dynamics and Control	Chongqing University	National Key Research and Development Program of China(1) Key Program of National Natural Science Foundation of China (1)	——	——	——	——
State Key Laboratory of Coal Resources and Safety Mining Key Laboratory of Gas and Fire Control for Coal Mines	China University of Mining & Technology	National Natural Science Foundations of China(2)	Ministry of Education Innovation Team Research Project(1)	National Award for Science and Technology Progress(1) National Award for technology invention(1)	——	——
State Key Laboratory of Coal Mine Safety Technology	Shenyang Research Institute of China Coal Technology & Engineering Group	Program of National Natural Science Foundations of China(10)	Provincial scientific research projects(6)	——	Sichuan Science and Technology Progress Award(1)	Science and Technology Awards of China Coal Industry Association(2) China Safety Production Association Award(2)
State Key Laboratory of Clean Energy Utilization	Zhejiang University	National Key Research and Development Program of China(1) National Natural Science Foundations of China(2) National Program on Key Basic Research Project (973 Program)(1)		National Awards for Science and Technology Progress(3)	Zhejiang Science and Technology Progress Award(3)	
State Key Laboratory of Ministry of Education by Provincial Department of Coal Mine Safety and Efficient Mining	Anhui University of Science and Technology	National Natural Science Foundations of China(3)	——	——	——	Science and Technology Awards of China Coal Industry Association(7)
Key Laboratory of Western Mine expltitation and hazard Prevention, Ministry of Education State Key Laboratory of Coal Fire Disaster Prevention in Shanxi province	Xi`an University of Science and Technology	National Natural Science Foundations of China(3)	Provincial scientific research projects(4)	National Award for Science and Technology Progress(1)	Shanxi Science and Technology Progress Award(1) Sichuan Science and Technology Progress Award(1)	Science and Technology Award of China Coal Industry Association(1)
Key Laboratory of Mine of thermo-motive Disaster and Prevention, ministry of education	Liaoning Technical University	National Science and Technology Support Program(1) Program of National Natural Science Foundations of China(9)	——	National Awards for Science and Technology Progress(2)	——	——
State Key Laboratory of Mine Disaster Prevention in Hebei Province	North China Institute of Science and Technology	——	Natural Science Foundations of Hebei province(1) Fundamental Research Funds for the Central Universities(1)	——	——	——
State Key Laboratory of Mining Development and Safety Technology in Hebei Province	North China University of Science and Technology	Programs of National Natural Science Foundations of China(2) Key Project of Chinese Ministry of Education(1)	Natural Science Foundations of Hebei province(1) Hebei Education Department Project(1)	——	Provincial or ministerial awards(3)	National Safety Production Supervision Administration Safety Science and Technology Achievement Award(1)

5.4. Analysis and Summary

China's coal reserves are widely distributed, which is the main form of energy supply in China, and this trend will continue for a long time. Correspondingly, China's research institutions for coal spontaneous combustion are widely distributed. In addition, China's economy continues to develop rapidly, and the country continues to increase investment in scientific research. The scientific and technological human resources in the field of coal spontaneous combustion are abundant, and the types and quantities of scientific research equipment are numerous. The research fields are comprehensive, diverse and in-depth. From the point of view of the scientific research projects obtained, the state, industries and enterprises continue to invest in this field, and the research funds are sufficient and the sources are diversified. From the scientific research awards obtained, scientific research projects and achievements have made great progress, great achievements, and continued growth.

6. Conclusion

The spontaneous coal combustion process is a combination of solid pyrolysis and gas combustion chemical reactions. The associated heat and mass transfer processes are complex and difficult to control, resulting in a diversity of spontaneous coal combustion test methods, among which results are often contradictory. Our study on the methods of testing the spontaneous combustion tendency of coal seams provides support and a basis for the targeted formulation of fire prevention and extinguishing measures. The theoretical and practical problems of the current spontaneous coal combustion tendency test methods need to be studied further.

1) The current chromatographic oxygen absorption identification method of spontaneous coal combustion tendency in China has good repeatability, accurate quantification, and easy standardization of tests. However, owing to the unreasonable principle and time-consuming operation, the test results are often inconsistent with the actual situation.

2) The determination method of oxidation kinetics of spontaneous coal combustion tendency is more exact than the principle of the chromatographic oxygen absorption method, but the instrument used in this method is a non-general instrument, which somewhat affects its use.

3) The apparent activation energy can reflect the oxidation dynamic characteristics of spontaneous coal combustion and has the advantage of simple, intuitive, and unified classification. However, this calculation method is insufficient.

Therefore, future researches on spontaneous coal combustion tendency in China should include the detailed kinetic process and characterization method of coal-oxygen chemical reactions, the application of new thermal analysis technology in spontaneous coal combustion tendency tests, and the use of composite indexes to reflect the intrinsic spontaneous combustion tendency of coal.

Acknowledgements

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