Application Research of Modified Activated Carbon Fiber in Nuclear Air Purification

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Abstract. Based on the disadvantages of the odine adsorber currently used, compare the performance differences between traditional granular activated carbon and new activated carbon fibers in adsorbing radioactive gaseous iodine. And explore the modification methods of new impregnated activated carbon fiber materials in terms of hydrophobicity, flame retardancy, and microporous structure adsorption. Provide theoretical and experimental data support for the development of new activated carbon fiber iodine adsorbers in the future.

Keywords: Iodide adsorber; Impregnation and modification; Triethylenediamine; Potassium iodide.

1 Introduction

Air filtration and purification in nuclear facilities are key measures to protect personnel and the environment from the hazards of radioactive gases. At present, the air filtration and purification function mainly relies on high-efficiency filters and iodine adsorbers installed in the ventilation system [1-4]. Among them, the ipdine adsorber is used to adsorb radioactive gaseous nuclides I-131 that are difficult to intercept [5-6].

At present, the nuclear grade iodine adsorber has some problems, such as low lower limit of purification capacity, purification efficiency being easily affected by site temperature and humidity, short service life [7], low purification capacity per unit volume and mass, and large ventilation resistance. Activated carbon fiber [8] is a type of carbon fiber containing an activated treatment. It changes its adsorption characteristics by treating a certain carbon fiber felt with high-temperature carbonization and activation, which results in a nanoscale pore size on the surface and increases the specific surface area. Compared to traditional granular activated carbon, activated carbon fiber has a rich internal microporous structure, which has a larger adsorption capacity and faster adsorption kinetics performance, and has higher adsorption efficiency for organic compounds and cations and anions in liquid and gas phases. It is resistant to acid and alkali, has good resistance to high temperature and high humidity, good conductivity and chemical stability, and is a more ideal environmental purification material. To further

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enhance the purification performance of activated carbon fiber, it also needs to be modified to enhance its purification stability under extreme conditions.

2 Principles of the Modification of Activated Carbon

2.1 Physical Adsorption

Activated carbon fiber has a high specific surface area, and when it adsorbs volatile organic compounds in the air, it mainly adsorbs physically. Physical adsorption is also the basis for subsequent chemical adsorption. When organic waste gases come into contact with activated carbon fiber, there is a type of interaction force between them - Van der Waals force, including dispersion force, induction force, and orientation force. Because iodine-containing molecules are polar molecules, the induction force and orientation force play an important role in physical adsorption.

The molecules on the adsorbent surface are left with free force fields to attract the adsorbate due to an imbalance in the force of attraction. Since this is adsorption caused by intermolecular forces, the binding force is weak, the adsorption heat is small, and the adsorption and desorption rates are both fast. The adsorbed substance is also easily desorbed, so physical adsorption to some extent is reversible. The characteristics of physical adsorption include: small adsorption heat, fast adsorption rate, no selectivity, and reversibility.

When studying the purification of nuclear air by activated carbon fiber, methyl iodide was chosen as the main adsorbent because it is the most difficult to capture in a nuclear accident scenario and poses the greatest threat to human health. Other gaseous radioactive substances can be effectively trapped and purified using high-efficiency particulate air (HEPA) filters.

Currently, the internal pores of commercially available activated carbon fibers are mainly 1-2 nm in size, accounting for more than 95% of the total pore volume. The molecular diameter of methyl iodide is approximately 5 Å(1 Å=0.1 nm). According to the literature, when the pore structure is 1.7-2 times the size of the adsorbate molecule [9], the adsorbate molecule can effectively accept the adsorption force field on the pore surface, which is beneficial for the adsorption of the adsorbate. Therefore, further chemical impregnation modification is needed to adjust the pore size of activated carbon fibers to maximize the physical adsorption capacity.

2.2 Chemical Complexation Adsorption

To avoid the disadvantage of easy desorption in physical adsorption, and to improve the purification efficiency of active carbon fibers for radioactive iodine gas, the active carbon fibers need to be modified with triethylenediamine (TEDA) impregnation [10]. Studies have shown that the adsorption mechanism of TEDA impregnant for methyl iodide is mainly that TEDA reacts with methyl iodide by SN2 nucleophilic substitution mechanism to form quaternary ammonium salt, thereby binding methyl iodide onto the active carbon. Because the terminal nitrogen atom of the tertiary amine is very active due to the σ-p supraconjugative effect of the three methylene groups connected to it,

the feature determines that it has a very high reaction rate constant when reacting with methyl iodide. Therefore, when methyl iodide molecules encounter the TEDA molecules impregnated on the surface of the active carbon fiber, they are easily subjected to chemical coordination reaction and trapped on the surface of the active carbon fiber. The reaction is shown in Figure 1.

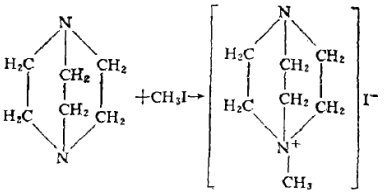

Fig. 1. Schematic representation of the reaction between methyl iodide molecules and TEDA to form quaternary ammonium salts.

The process requires the following steps: ① volume diffusion (diffusion of gas to the surface of the carbon particles); ② pore diffusion (diffusion of gas into the internal structure of the pores); ③ adsorption; ④ chemical reaction. This process requires fewer steps than the isotope exchange process and achieves a higher degree of purification in the same retention time compared to the isotope exchange process.

Due to the chemical reaction between methyl iodide and TEDA, a stable quaternary ammonium salt is formed and trapped within the pores of activated carbon fibers. Therefore, this adsorption is irreversible, and desorption is unlikely to occur again, belonging to a firm adsorption.

2.3 Isotope Replacement Adsorption

In the nuclear grade iodine adsorber, potassium iodide (K127I) is also impregnated to further improve the adsorption and purification effect on airborne radioactive iodine. The reaction process is as follows:

$$\begin{aligned} &CH_3^{131}I\ (gas) + K^{127}I\ (carbon\ surface) \\ &\rightarrow CH_3^{127}I\ (gas) + K^{131}I\ (carbon\ surface) \end{aligned} \tag{1}$$

Since the amount of 127I on the carbon surface is much greater than that of 131I, the exchange equilibrium proceeds greatly to the right. This phenomenon is called isotope replacement adsorption. As long as there is sufficient residence time, the decontamination coefficient of the replacement adsorption can also reach a very high level. It is generally believed that completing such a process requires seven steps: ① Volume diffusion (diffusion of gas to the surface of the carbon particles); ② Pore diffusion (diffusion of gas into the interior of the pore structure); ③ Adsorption; ④ Isotope exchange; ⑤ Desorption; ⑥ Diffusion from the pore structure to the surface; ⑦ Diffusion into the gas phase.

The process required for replacement adsorption is more complicated than that for chemical complexation adsorption. Thus, it is generally considered that the proportion of this adsorption effect is relatively small. However, it can serve as a supplement to chemical complexation adsorption to prevent the loss of purification capacity caused by the sublimation and loss of TEDA due to high temperature and long-term operation.

3 Modification Direction Selection

3.1 Moisture Resistance Treatment

Whether it is traditional granular activated carbon or activated carbon fiber, both are prone to absorbing water molecules in the air, leading to a decline in purification capacity. Granular activated carbon has three types of adsorption pore diameters: macropores, mesopores, and micropores. The role of mesopores is to guide the air containing the adsorbate to the micropores, and it is the micropores that truly play the adsorption role. However, water vapor is prone to form droplets at the mesopores, hindering the adsorbate from entering the micropores, thereby causing a significant decline in the purification capacity of granular activated carbon under high humidity working conditions. The pore structure of activated carbon fiber is mainly composed of micropores, and these micropores directly open to the air, enabling the adsorbate to directly enter the micropores without the need for the guidance of mesopores. Due to the effect of airflow, droplets are difficult to form and block the micropores on the outer surface of activated carbon fiber. Nevertheless, water molecules will still enter the micropores and occupy a certain physical adsorption surface area, thereby causing a decline in the purification capacity of activated carbon fiber under high humidity, but the extent of the decline will not be exacerbated. To minimize this influence as much as possible, it is highly necessary to conduct hydrophobic treatment on activated carbon fiber.

3.2 Flame Retardant Treatment

Conventional granular activated carbon lacks flame retardancy. The adsorbents employed in iodine adsorbers are typically coconut shell activated carbon that has undergone modification treatment with chemical reagents such as TEDA and potassium iodide (KI), and the addition of these chemical reagents reduces the ignition point of the activated carbon.

An important indicator that needs to be taken into account in the development of this activated carbon fiber is to furnish its flame-retardant characteristics, essentially averting the risk of fire.

4 Modification Method

To achieve the research goals, the impregnation modification process of traditional granular activated carbon and the knitting, carbonization, and activation processes of activated carbon fiber were investigated and screened. The preferred impregnation

modification process flow of activated carbon fiber was selected: optimization of activated carbon fiber → flame retardant treatment → hydrophobic treatment → TEDA impregnation modification → KI impregnation modification → vacuum drying.

4.1 Selection of Activated Carbon Fiber

After investigation, the commonly used active carbon fibers for the current materials are polyacrylonitrile-based, viscose-based, phenolic-based, and asphalt-based. They are compared and graded in terms of specific surface area, pore size, filament diameter, knitting density, and thickness. Among them, phenolic-based is the most expensive, which leads to a higher production cost of the activated carbon fiber felt. Asphalt-based is not suitable for the use environment of iodine adsorber due to its hygroscopicity, and its service life is short. In the end, polyacrylonitrile and viscose-based materials are selected for modification work.

4.2 Improvement of Flame Retardant Performance

The activated carbon fiber was treated with a bottom layer of flame retardant to improve its fire resistance. A mixture of molecular weight smaller sodium pyrophosphate and aluminum dihydrogen phosphate was selected and prepared in a certain proportion as a mixed flame retardant. The mixed flame retardant was then prepared into a water solution of a certain concentration and evenly sprayed on the surface of the activated carbon fiber. The activated carbon fiber was left to stand for 24 hours and then rinsed and dried.

4.3 Hydrophobicity Modification

The newly produced activated carbon fiber itself does not have hydrophobic properties. Properly selected ethyl orthosilicate and ethanol are used as hydrophobic agents, and a water solution is prepared by mixing them in a certain proportion. The activated carbon fiber is immersed in the water solution for more than 30 minutes, then removed and dried.

4.4 TEDA Vacuum Flash Explosion Impregnation Modification

The chemical complexation adsorption of TEDA is the main adsorption mechanism of activated carbon fibers for methyl iodide. To achieve the goal of impregnating TEDA into the interior of activated carbon fibers and coating it uniformly, the vacuum flash method was employed for the impregnation modification of TEDA. The principle is as follows: The activated carbon fibers are evacuated, and TEDA is heated under nitrogen protection to form gaseous molecular substances of a certain concentration. The TEDA vapor rapidly enters the sample chamber of the activated carbon fibers under high vacuum and is left to stand and cool down at atmospheric pressure.

4.5 KI Impregnation Modification

A water solution of KI was prepared at a certain concentration and uniformly sprayed onto the surface of the activated carbon fiber. After standing for 24 hours, it was air-dried.

4.6 Low Temperature Drying

The activated carbon fiber samples processed by the aforementioned solution were placed and dried in a low temperature and ventilated area.

5 Performance Test of Modified Sample

According to the above process, the flame retardant activated carbon fiber samples were preliminarily made. In order to study the adsorption capacity of activated carbon fiber samples for gaseous methyl iodine, the following performance tests were carried out.

5.1 Dynamic Purification Capacity Test of Methyl Iodine Under Different Humidity

Referring to the laboratory detection method for the performance of removing methyl iodide of granular activated carbon [6], a test bench was constructed, and non-radioactive methyl iodide [7] was employed to test the performance of activated carbon fiber in removing methyl iodide. The experimental device for the dynamic purification capacity of activated carbon fiber is depicted in Figure 2.

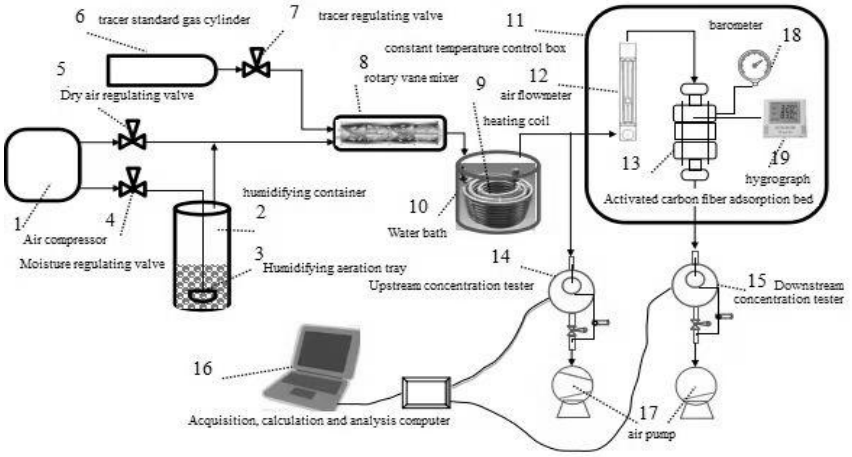

Fig. 2. Schematic diagram of the experimental device for dynamic purification capacity of activated carbon fiber

The activated carbon fiber samples after impregnation modification were cut into circular shapes that were compatible with the size of the experimental bed. Three layers were stacked, each approximately 3.5 mm thick, so that the total thickness of the activated carbon fibers was 11 mm. The samples were loaded into a dedicated adsorption bed and clamped, and then connected to the testing system. The overall air volume, temperature and humidity were controlled through the testing system to ensure that the experimental environment met the measurement requirements and achieved the functions of constant temperature and humidity adjustment. Adsorption performance tests under multiple humidity conditions were conducted. A methyl iodide standard gas cylinder was used as the tracer source, and a certain flow rate of the tracer was injected through the regulating valve. The tracer was uniformly mixed with moist air in the mixer to obtain a mixed gas of a certain concentration of the tracer for the experiment.

Using a dedicated test tool for non-radioactive methyl iodide for real-time measurement, the concentration of methyl iodide in the measured gas is converted into a voltage signal output, and the voltage signal can be read by computer-specific software to calculate the real-time concentration of methyl iodide in the system. The concentration of methyl iodide gas upstream and downstream of the activated carbon fiber can be measured, and the total amount of tracer released and the total leakage downstream after passing through the activated carbon fiber can be calculated by integrating and accumulating the tracer concentration upstream and downstream within the sampling time according to the system air volume. Thus, the purification coefficient or efficiency of the activated carbon fiber can be calculated. The calculation formula is:

$$E = \frac{\int CQdH}{\int cQdh} \tag{2}$$

$$\eta = \left(1 - \frac{1}{\mathrm{E}}\right) * 100\% \tag{3}$$

In the equation:

E represents the purification coefficient of activated carbon fiber, which is derived by integrating the sampling concentrations in the upstream and downstream over the sampling time to determine the total amount injected upstream and the total leakage downstream;

C and c represent the concentrations of methyl iodine in the upstream and downstream (in volume percentage) respectively;

Q and q represent the measured air volume of the system, in m^3/h;

H and h represent the sampling times in the upstream and downstream respectively;

η represents the purification efficiency.

Using the above methods and devices, the dynamic purification capacity of traditional nuclear grade Granular activated carbon, unimpregnated modified activated carbon fiber, and impregnated modified activated carbon fiber was tested under different humidity conditions, with an upstream injection concentration of 4-6 μmol/mol, an injection time of about 2 minutes, a sampling measurement time of 20 minutes, and each humidity condition stabilizing to a balance for about 20 hours. The results are shown in Table 1.

Table 1. Test Results of Dynamic Adsorption Efficiency of Different Types of Adsorbents

Humidity	Nuclear grade granular activated carbon: 25 mm thick, retention time 0.2 s	Unpregnated modified activated carbon fiber: 11 mm thick, retention time 0.18 s	Impregnated modified activated carbon fiber: 11 mm thick, retention time 0.18 s
15%	99.997%	99.988%	99.995%
40%	99.991%	62.264%	99.991%
60%	99.959%	49.512%	99.992%
80%	98.404%	/	99.942%
90%	83.976%	/	99.738%

It can be seen from table 1 that the purification efficiency of unimpregnated activated carbon fiber is far lower than 90% even at a humidity of 40%, and it cannot be directly applied to the purification of nuclear air.

Comparing the test data of nuclear grade granular activated carbon, unimpregnated modified activated carbon fiber, and impregnated modified activated carbon fiber on the same chart, as shown in Figure 3, the purification efficiency of granular activated carbon drops rapidly around 80% humidity; the impregnated modified activated carbon fiber maintains a high purification efficiency at high humidity.

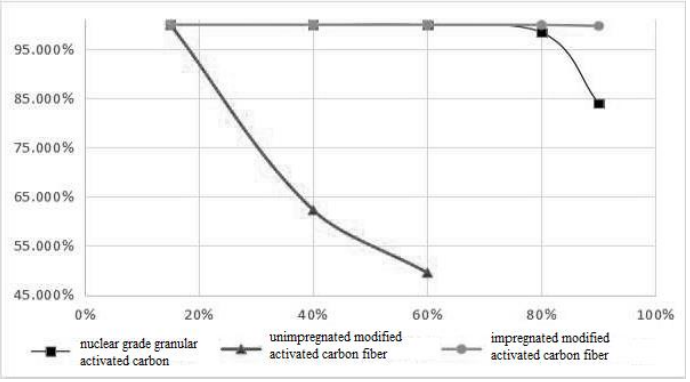

Fig. 3. Comparison of the purification efficiencies of the three types of activated carbon

For the humidity requirements during the operation of a nuclear power plant, the iodine adsorber must meet a purification efficiency of at least 99.9% for methyl iodine at a humidity of 60%. As shown in Table 1, the purification efficiency of 11mm thick impregnated modified activated carbon fiber for methyl iodine at a humidity of 60% is 99.992%, far exceeding the basic requirements of this study.

To further validate the dynamic purification capability of the impregnated modified activated carbon fiber, the sample underwent a purification capacity test for radioactive methyl iodine in accordance with the standard ASTM D3803–1991 "Standard Test Methods for Nuclear Grade Activated Carbon" [11] (2009 version). The test outcome was as follows: under the working condition of 86% humidity, the purification efficiency was 99.909%. The inspection and acceptance standard for Type III iodine

adsorber activated carbon in China stipulates that at 70% humidity, with a 100 mm thick carbon bed and a retention time of 0.5 s, the purification efficiency should not be lower than 99.5%. In contrast to the above-mentioned test results of the modified activated carbon fiber, the 11 mm thick activated carbon fiber, with a retention time of 0.18 s, has far met the requirements of this test.

It can be seen from Figure 3 that in a low-humidity environment, the impregnated modified activated carbon fiber maintains a high purification efficiency similar to that of nuclear grade granular activated carbon. However, when the humidity exceeds 80%, the purification efficiency of nuclear grade granular activated carbon drops rapidly compared to that of the impregnated modified activated carbon fiber. This indicates that the modified activated carbon fiber has a much superior purification capacity to nuclear grade granular activated carbon under higher humidity, thinner carbon bed thickness, and shorter retention time.

5.2 Static Adsorption Total Amount (Accelerated Aging Life) Test of Activated Carbon Fiber

Using the above dynamic test pipeline apparatus, a continuous flow of approximately 5 μmol/mol or 10 μmol/mol methyl iodine tracer gas is introduced into the tested adsorbent, and the upstream and downstream tracer concentrations are continuously tested and recorded. The upstream concentration N and downstream concentration M are recorded every 10 minutes, and the process purification efficiency $\eta=(1-M/N)\times100\%$ is calculated. The process purification efficiency is continued to be tested until it reaches approximately 99%, at which time the dynamic adsorption efficiency of the adsorbent can be roughly estimated to be about 99%. According to the acceptance standard for iodine filters in nuclear power plants, it has reached the service life and needs to be replaced. The accumulated deployment time is the accelerated aging time at the corresponding concentration. Three dynamic purification efficiency tests are conducted at the beginning, middle, and end of the service life to verify whether they conform to the performance aging law. The adsorbed methyl iodine mass can be calculated by integrating the upstream and downstream concentrations and the test volumetric flow rate over the test period.

Using the Granular activated carbon used in power plants as a comparison, the lifetime performance of the researched activated carbon fiber was verified. The test results are compared in Table 2.

Table 2. Life Test Data of Activated Carbon Fiber and Granular activated carbon

	Unimpregnated modified active garbon fiber 1	Impregnated modified active carbon fiber 2	Granular activated carbon
Initial dry mass/g	2.9617	2.8798	24.8101
Initial dynamic purification efficiency/%	99.954	99.966	99.969
Test humidity/%	85	60	59

Test flow/(m^3/h)	0.38	0.38	0.64
Average injection concentration/(μ mol/mol)	5.72	11.95	10.09
Mid life purification efficiency/%	99.769	99.512	99.693
Purification efficiency at the end of service life/%	98.898	99.101	99.388
End of life dry weight/g	3.1021	3.2011	25.0053
Theoretical methyl iodine absorption mass/g	0.1321	0.2698	0.1735
Actual methyl iodine absorption mass/g	0.1404	0.3213	0.1952
Accelerated aging time/min	620	640	300
Absorption ratio/(mg/g)	44.6	93.7	7.87

The process purification efficiency of the three adsorbents is shown in Figure 4.

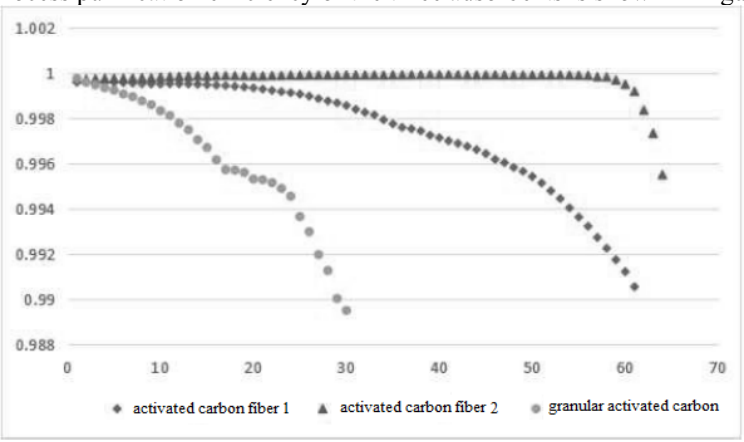

Fig. 4. purification efficiency of different adsorbents during test

From the above test results, it can be seen that:

(1) The mass of absorbed methyl iodine obtained through weighing and calculation is highly consistent, suggesting that the data from the accelerated aging calculation and analysis is trustworthy.

(2) The dynamic purification efficiency at the end of life is around 99%, which meets the replacement index of iodine adsorber in nuclear power plant;

(3) The absorption ratio of methyl iodide by activated carbon fibers is 93.7 mg/g (at 60% humidity), which is far higher than 7.87 mg/g of granular activated carbon under the same humidity condition. Even at 85% humidity, the adsorption ratio of methyl iodide by activated carbon fibers is still higher than that of granular activated carbon.

(4) Judging from the purification efficiency in the test process, activated carbon fibers have a long period of stable performance in the early stage. During this stable period, the decline in purification capacity is not significant. However, at the end of

their service life, the purification capacity drops rapidly due to approaching saturated adsorption. This phenomenon is not prominent in granular activated carbon.

(5) Subsequently, the total amount of methyl iodine adsorbed is about 656 g based on the calculation of 7 kg loading amount of activated carbon fiber filter. The dry activated carbon filling amount of granular activated carbon iodine adsorber is 40 kg, and the total amount of methyl iodine adsorbed is about 315 g. Under the same air volume of the system, the service life of the impregnated activated carbon fiber filter is about twice that of the granular activated carbon iodine adsorber.

Through the above accelerated aging life test, the impregnated modified activated carbon fiber has a higher absorption ratio for methyl iodide compared to nuclear grade granular activated carbon, a longer service life cycle, and higher purification efficiency during the same period than nuclear grade granular activated carbon.

5.3 Flame Retardant Performance Test

A third-party institution was commissioned to conduct a fire point test on the researched activated carbon fiber and the used granular activated carbon in nuclear power plants, according to the method of GB/T20450—2006 "Test Method for Ignition Point of Active Carbon" [12]. From the test results, it can be seen that the TEDA+KI impregnated granular activated carbon smoked at 225 °C, which was caused by the large amount of TEDA vaporizing and escaping from the activated carbon. The fire started at 271 °C.

The impregnated modified activated carbon fibers were divided into 2 samples, with flame retardant and hydrophobic modifications being consistent. Sample 1 was impregnated with TEDA+KI, while Sample 2 was impregnated with TEDA alone, without KI. The test results showed that the ignition point of the TEDA+KI modified sample was 432°C, while the ignition point of the TEDA modified sample was 426°C, with little difference between the two. However, the ignition point of the activated carbon fiber modified with granular activated carbon was significantly higher.

For further comparative tests, a flame gun was used to burn the granular activated carbon and modified activated carbon fibers, with the flame temperature set at over 1,100°C. The materials were heated for about 2 minutes. After the flame was removed, the activated carbon fiber immediately returned to its original color without any signs of burning or deformation. However, the granular activated carbon remained partially in a red or gray burning state after the flame was removed.

From the above tests, it can be concluded that the modified activated carbon fiber has a significantly higher ignition point than granular activated carbon, greatly reducing the potential risk of spontaneous combustion compared to granular activated carbon.

5.4 Pore Structure Test

The pore structure of the newly produced activated carbon fiber is mainly micropores, with 98% of the total pores smaller than 2 nm. The average pore diameter is 1.65 nm, which is slightly larger than the diameter of methyl iodide molecule (0.5 nm).

This study impregnated activated carbon fibers to modify them, allowing the modified impregnated material to enter the microscopic pores, thus reducing their size and

indirectly achieving the regulation of pore size. To evaluate the regulatory effect of this modification, the pore size of the impregnated activated carbon fibers was tested again, as shown in Figure 5.

The average pore size of activated carbon fiber after impregnation modification is 1.01 nm, which is twice the diameter of methyl iodide molecule, making it easier for activated carbon fiber to physically adsorb methyl iodide molecules.

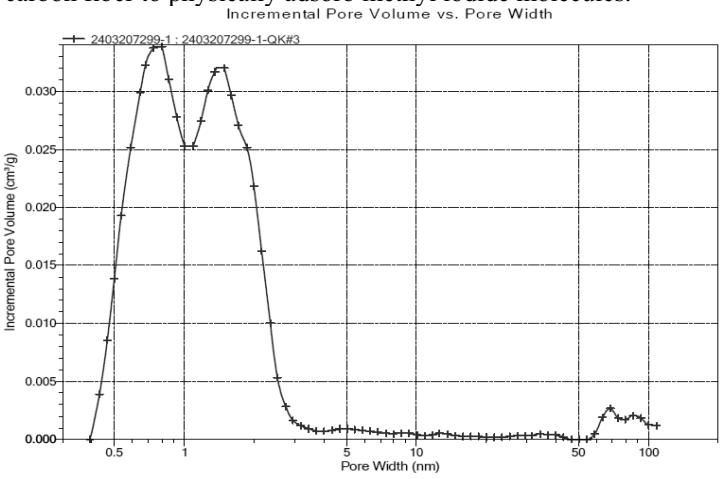

Fig. 5. Porosity Distribution of Modified Active Carbon Fibers after Impregnation

5.5 Microscopic Morphological Analysis Before and After Life Test

The microscopic morphology analysis of the modified activated carbon fiber before and after the life test was analyzed, and the SEM images are shown in Fig. 6 and Fig. 7.

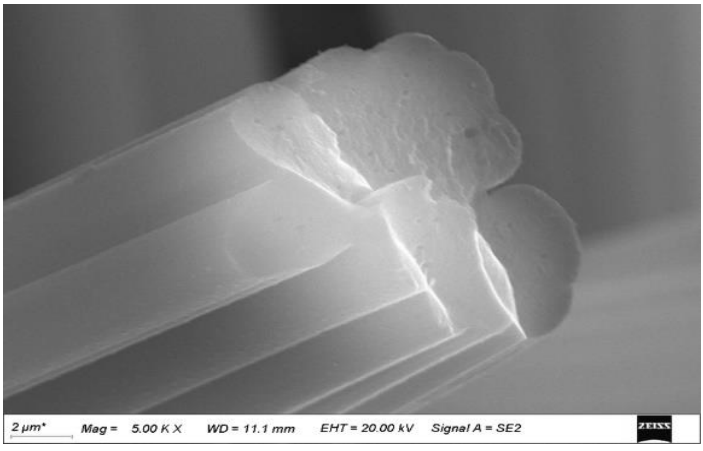

Fig. 6. SEM image before life test

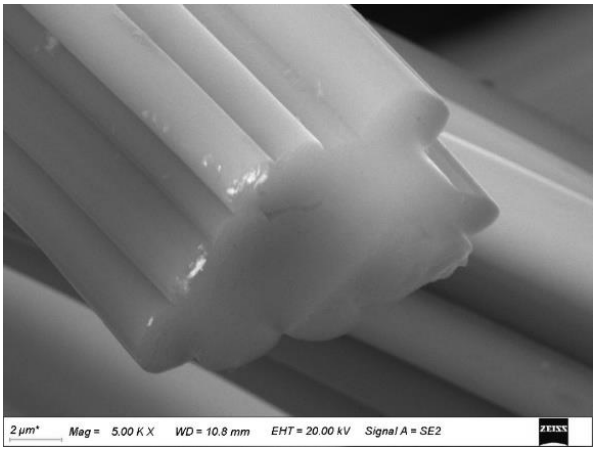

Fig. 7. SEM image after life test

From the SEM images before the life test, it can be seen that the active carbon fiber filament has a smooth appearance, no residual crystallization of impregnating material, and the impregnating modified substance has entered the interior of the active carbon fiber filament.

After the life test, the appearance of the activated carbon fiber filament remained smooth, with no obvious changes before and after the test. No external loose adherent crystalline phenomenon was observed, indicating that the methyl iodine gas adsorbed had entered the interior of the activated carbon fiber and was a firm adsorption.

5.6 Analysis of Microporous Volume Before and After Life Test

The pore volume of the activated carbon fiber before and after the life test was measured by the N_2 adsorption density functional theory (DFT) analysis method. The results are presented in Figures 8 to 11.

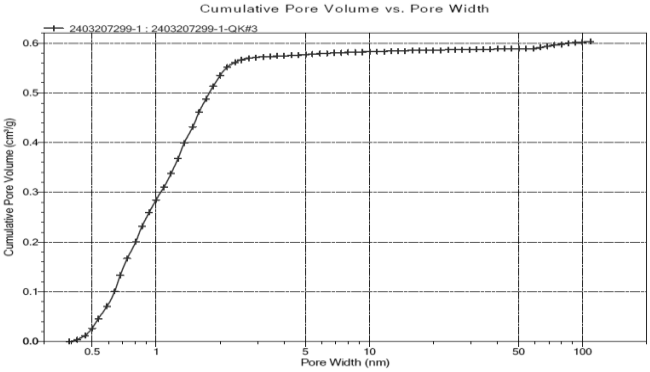

Fig. 8. Porosity Distribution Chart Before Life Test

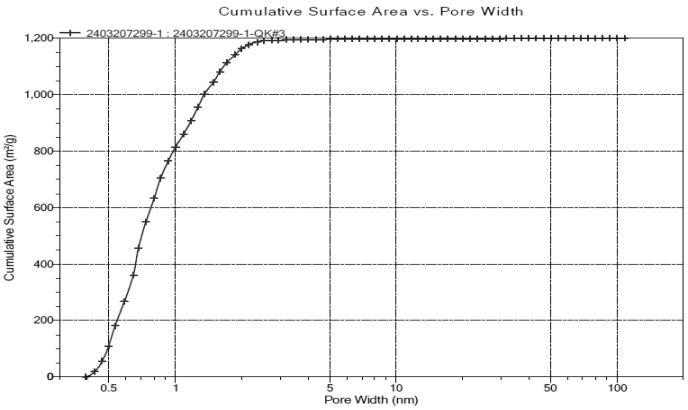

Fig. 9. specific surface area Chart before life test

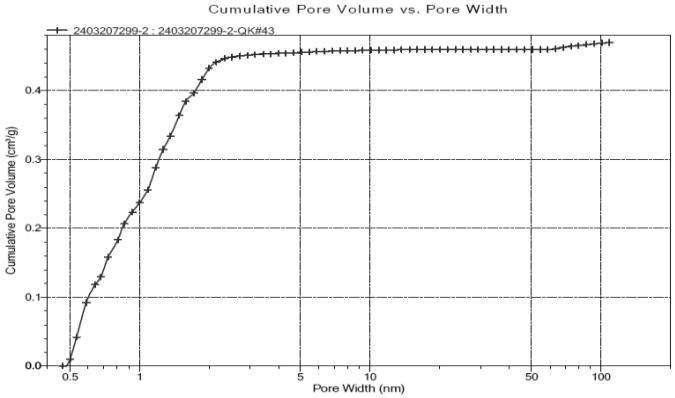

Fig. 10. Porosity Distribution Chart after life test

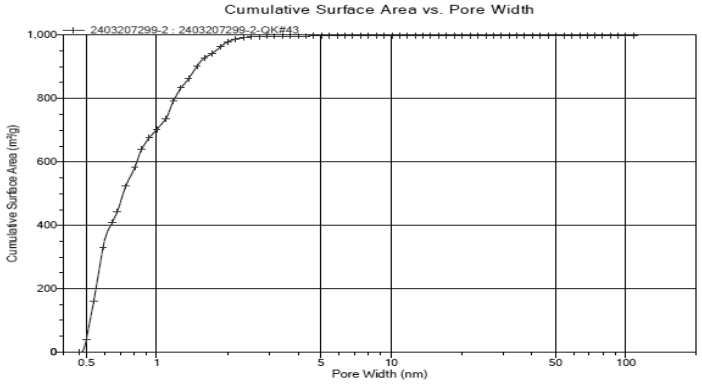

Fig. 11. specific surface area Chart after life test

Before the life test, the specific volume of pores smaller than 2 nm was 0.535 cm^3/g, accounting for 88.7% of the total specific volume of 0.603 cm^3/g. The specific surface area of pores smaller than 2 nm was 1162 cm^2/g, accounting for 96.9% of the total specific surface area of 1199 cm^2/g. After the life test, the specific volume of pores smaller than 2 nm was 0.432 cm^3/g, accounting for 91.9% of the total specific volume of 0.470 cm^3/g. The specific surface area of pores smaller than 2 nm was 979 cm^2/g, accounting for 98.0% of the total specific surface area of 999 cm^2/g.

It can be concluded that the specific volume and specific surface area of activated carbon fiber after adsorbing methyl iodide have decreased to some extent, with a reduction rate of about 19%, indicating that the methyl iodide gas has been adsorbed and occupied the pore volume of the activated carbon fiber. After the end of the service life test, although there was a considerable amount of unoccupied space for the activated carbon fiber to further adsorb methyl iodide, the dynamic purification adsorption efficiency was lower than 99%, and no longer met the requirements for nuclear power plants.

6 Conclusions

In this study, a series of impregnation modification techniques were carried out on the preferred commercially available activated carbon fibers, including flame retardant modification, hydrophobic treatment, TEDA impregnation modification, and KI impregnation modification. A comprehensive and detailed performance analysis and evaluation of the modified activated carbon fibers were conducted, and the conclusions are as follows:

(1) The dynamic purification capacity tests were conducted on activated carbon fibers. Under a humidity of 86%, the purification efficiency of radioactive methyl iodine was 99.909%. Under a humidity of 60%, the purification efficiency of non-radioactive methyl iodine was 99.992%, far exceeding the research goal of the project (the purification efficiency of methyl iodine at 60% humidity should be no less than 99.9%).

(2) The adsorption capacity of activated carbon fiber for methyl iodide is 93.7 mg/g (60% humidity), which is much higher than that of granular activated carbon at the same humidity condition (7.87 mg/g); the adsorption capacity of activated carbon fiber is also higher than that of granular activated carbon even at 85% humidity.

(3) The modified activated carbon fibers exhibit a significantly elevated ignition point compared to granular activated carbon. Moreover, there is an extremely small number of combustible substances in the activated carbon fibers. After being exposed to a flame, the non-combustible substances remain promptly, and the combustion ceases shortly. The activated carbon fiber filters have considerably mitigated the fire risk and fire consequences in contrast to the traditional granular activated carbon iodine adsorbers.

(4) From the analysis of the morphology and micropore volume and specific surface area of activated carbon fiber, the adsorption of methyl iodide by activated carbon fiber occurs in the micropores inside, and it is not easy to re-desorb. Compared with granular activated carbon, the micropore content of granular activated carbon is much smaller

than that of activated carbon fiber, and its physical adsorption of methyl iodide is not firm enough, making it more likely to desorb and release methyl iodide.

Based on the above research, impregnated modified activated carbon fiber has the advantages of high purification efficiency, long service life, high flame resistance, and high adsorption ratio compared to granular activated carbon. If used as the content of iodine adsorber in the ventilation system of nuclear power plants, it can greatly improve the operating efficiency of the ventilation system. And the iodine adsorber, as a radioactive solid waste, has a high adsorption ratio, which greatly reduces the radioactive solid waste of nuclear power plants and reduces the cost of solid waste treatment. Therefore, it has great promotion value. It is suggested to add a brief concluding remark at the end with an extension outlook: application practice, application prospect, promotion value, etc.

References

1. National Energy Administration(2011) Code on nuclear air and gas treatment Ventilation, air conditioning and cleaning—Part 11: Iodine adsorber (type I): NB/T 20039.11-2011, 1-15(in chinese).
2. National Energy Administration(2011) Code on nuclear air and gas treatment ventilation, air conditioning and air cleaning—Part 12: Iodine adsorber (type II): NB/T 20039.12-2011, 1-16(in chinese).
3. National Energy Administration(2012) Code on nuclear air and gas treatment ventilation, air conditioning and air cleaning—Part 12: Iodine adsorbers (type III): NB/T 20039.13-2012, 1-26(in chinese).
4. Jun Xiao, Lixin Zhu(2011) Application of iodine adsorber in ventilation system of nuclear power plant , Nuclear safety ,(2): 53-55, 63(in chinese).
5. NADIA B,ALA A,GORAN P,et al. (2001) ^{129}I from the nuclear reprocessing facilities traced in pre cipitation and runoff in northern Europe, Environmental Science & Technology, 35(8): 1579-1586.
6. Ying Ma, Qun Liu, Lei Dong, et al. (201) A review of methods for capturing gaseous radioactive iodine ,Journal of Shanxi University, 40 (3): 515-520(in chinese).
7. Liantao Yu, Yuqing Lin (2011) Operation experience feedback and analysis of nuclear air cleaning system in nuclear power plant,China Nuclear Society Progress report on China's nuclear science and Technology (Volume II), 5(in chinese).
8. Jingtang Zheng (2000) Activated carbon fiber,New carbon materials, 15 (2): 80(in chinese).
9. Rudzinski w, Steele W A, Zgrablich G.(1997) Equilibrium and Dynamics of Gas Adsorption on Heterogeneous Solid Surface. Vol.104, Elsevier Science B.C.
10. Jianlu Pei, Jia Wang, Bo Wu, et al (2021) Research status of influencing factors and mechanism of adsorption performance of TEDA impregnated activated carbon,Applied chemical industry, 50 (S2): 261-265(in chinese).
11. American Society for Testing Materials (2014)Standard Test Method for Nuclear-Grade Activated Carbon: ASTM D3803-91,1-18.
12. State Forestry Administration(2006)Test method for ignition point of activated carbon: GB/T 20450-2006,1-12(in chinese).