A Composite Purification Material Research of Indoor Purification for Particulate Matter and Gas Pollutants

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SUMMARY

Large air handling unit and air purifiers are increasingly appearing in people's lives in order to reduce indoor pollutants harm to human health. With a variety of pollutants emergence, people's requirements for air purification is not limited to remove particulate matter, but also the removal of gaseous pollutants function. It is necessary to study a composite purification material for indoor pollutants removal. This paper studied the "Physical Loading" method of nano-activated carbon combined with filter (F9/G4), and evaluated the loading state using Scanning Electron Microscopy (SEM). The G4 composite filter was selected to evaluate the loading amount of activated carbon at different loading time. Then the effects of the prepared G4 composite filter on the removal of ozone, PM2.5 and toluene were studied.

INTRODUCTION

In recent years, the national haze weather occurred frequently. Beijing and Tianjin as the representative of the northern region in the autumn and winter season is particularly serious. Haze pollution in Jing-Jin-Ji area comparable to the "fog" London in sixties of the last century. Haze caused widespread concern, and indoor VOC pollution events occurred frequently which leading to human cancer, leukemia and other diseases, also attracted people's attention at the same time. Another epidemic air pollution-ozone pollution has not attracted enough attention, but also slowly affect people's lives and health.

With the emergence of a variety of pollutants, people's requirements for air purification is not limited to remove particulate matter, more tend to have both the removal of particulate matter and gaseous pollutants function. Therefore it is necessary to study a composite purification material for simultaneous removal of particulate matter and gaseous contaminants in the interior of the building for the removal of indoor pollutants. The EPA's recommended technology for the removal of indoor gaseous pollutants is porous media adsorption technology. The "sandwich" form in which the packed bed and the fiber filter material combination is the main form of the porous media adsorption technology purification module. When dealing with large amounts of air, high concentrations of gaseous pollutants or in order to make the purification module has a longer service life, people often use such filters. Such filters generally use a larger amount of activated carbon up to several tens of kilograms, thus ensuring the efficiency and longevity of such filters. Its cost is expensive, the resistance is bigger, and service life is uncertain in the ordinary indoor environment (Fisk 2007). Therefore, such filter used in some occasion which has specific pollutants, and rarely used in the building environment.

Due to the large resistance of the above-mentioned filters, the practical application, such as household air purifiers, often combines the packed bed with the honeycomb structure to disperse a certain amount of adsorbent material in the honeycomb channel (Nilsson 1989; Lester and Homeyer 1996), or the equivalent diameter of about 2-4 mm of particulate adsorbent adhered to the honeycomb wall to increase the porosity of the packed bed, thereby reducing the resistance. But the diffusion time constant is much larger than the residence time of the airflow through the packed bed filter (Pei and Zhang 2011). Therefore, the large particle size of the adsorption particles often make the initial stage of the filling bed is very efficient. When the outermost adsorbent is saturated, the efficiency is obviously decreased due to the slow mass transfer within the particles, but the internal material of the particles is not fully used.

In fact, for indoor air purification, according to the quality balance and typical indoor environment parameters, 15-35% of the purification efficiency can make the indoor pollutant concentration to the target level (Wang et al. 2013), so there is no need to pay expensive high resistance of the high energy costs to achieve unnecessary high efficiency. In summary, the above-mentioned form of gaseous pollutant filtration device is not the best choice for indoor pollutant control.

At present, the more popular activated carbon filter material is sandwich activated carbon cloth. The main structure of the two layers of fiber filter paper sandwiched or adhere to the fine activated carbon particles, and then through the adhesive into one, and then made of a plate or a V-shaped structure. This filter reduces the amount of activated carbon, and has various forms of structure. It can be used flexibly and conveniently. It has the function of removal for particulate matter and gaseous pollutants. It is currently widely used in people's lives. However, the use of adhesives covered the porous adsorption medium on the surface of the adsorption hole reducing its adsorption capacity, and adhesion will introduce other VOCs into the indoor air, causing indoor pollution. The limited field test data for the actual performance of this material also makes such filter not widely recognized (Fisket al. 2002).

Based on the above considerations, it is necessary to develop a new type of gaseous pollutant purification filter, which should have a small particle size of the adsorption material to reduce the internal diffusion resistance, but also to avoid the traditional activated carbon filter production process to prevent the production process additive to affect the filter performance.

Research on fiber loading and purification materials abroad has been reported that Auburn University has studied a catalytic material for metal-fiber mat-supported noble metal
catalysts (Kalluri et al. 2009) which mainly using wet papermaking technology. The active ingredient particle size of preparation material is less than 500μm. The research process is complex and has not been widely used in indoor air purification. In addition, similar to the honeycomb structure, there are foam structures (Yang et al. 2013) shown in Figure 1. This kind of three-dimensional foam structure is similar to the honeycomb structure, the resistance is small, and because of its irregularity of the internal flow channel, the disturbance of the air flow and the turbulence are strengthened, which is beneficial to the mass transfer of the pollutant and the catalytic reaction.

Figure 1. The metal fiber of loading active ingredient (left) with the three-dimensional foam structure carrier (right)

Sidheswaran et al. (2011) load the laboratory-prepared MnOx catalyst on a general ventilated filter by physical method and tested its filtration efficiency for formaldehyde. They found that it was able to maintain a high efficiency of about 80% in 6-12 months. But there are no detailed reports for the specific load method. In fact, small particles can be caught by fiber materials only by physical means. Air conditioning system of construction commonly used this principle in the air filter to purify the particles in the air pollutants.

In this study, the micro-scale activated carbon adsorbent material was combined with the fiber-based filter medium by the method of using the traditional fiber filter to filter the particulate matter. The effective particle size was reduced to the micron level to reduce the internal diffusion resistance. Improve the efficiency at the same time not to sacrifice the cost of resistance, and to achieve the purpose of filtering PM2.5 and gaseous pollutants. It can improve people's living environment and protect their health. It is of particular significance in today's China. In this study, the evaluation and primary selection of particulate matter and gas purification materials were carried out. The removal performance of the prepared materials for ozone, VOCs (toluene) and particulate matter were studied in the fiber material test system.

METHODS
Materials and Appearance characterization

The loading tests were conducted in the developed loading test system using the selected particulate filter material (F9 glass fiber filter and G4 chemical fiber material) and gas purification material (micron-grade activated carbon powder). The filter materials were clamped in the production of the fixture. Then open the original Ebmpapst centrifugal experimental fan (R3G220-AC08-45), set a certain loading speed, after a certain loading time, you can achieve a certain loading amount. The loading conditions of the two filter media are shown in Figure 2 (F9 filter) and Figure 3 (G4 filter). The loading parameters is shown in Table 1.

<table>
<thead>
<tr>
<th>Filter material types</th>
<th>Loading speed (m/s)</th>
<th>Loading time (h)</th>
<th>Loading capacity (mg)</th>
</tr>
</thead>
<tbody>
<tr>
<td>F9</td>
<td>0.2</td>
<td>12</td>
<td>9.45</td>
</tr>
<tr>
<td>G4</td>
<td>0.2-0.3</td>
<td>3</td>
<td>35.6</td>
</tr>
</tbody>
</table>

It is found that the loading performance of G4 filter is better than that of F9 filter by comparing the loading parameters of the micron-grade activated carbon powder. In order to further evaluate the loading status of activated carbon loaded with different fiber filter materials, the loading filters was photographed using SEM (Scanning Electron Microscope) model of Hitachi S-4800 in the Tianjin University Materials Institute. Because different filter material has different characteristics, the loading status of the particles are also quite different after filtration, see Figure 4. SEM micrograph analysis was carried out by Nano Measurer 1.2 software (USA). It was found that F9 filter loading little activated carbon powder and was relatively smooth in most areas. The particulate matter size loading in F9 filter is predominantly 0.1-0.5 μm, and only a few such size particles are present in the purchased activated carbon powder. G4 filter loading activated carbon powder on a single fiber was increased, but there are still some areas are relatively smooth. The smooth part has a large loading potential, the loading particle size is mainly 1-3μm or so, accounting for 80% (more consistent with the particle size distribution range).

Figure 4. SEM micrograph of F9 filter loading activated carbon (left) and G4 filter loading activated carbon (right)

Ozone, particle and toluene generation and measurement
Ozone was generated by UV ozone generators (SOG-2, UVP, USA) with a UV lamp. The generation amount was controlled by adjusting the length of the UV lamp. Ozone concentration was monitored with a photometric ozone analyser (Model 202, 2B Technology, USA), with measuring accuracy of 0.1 ppb and measuring range of 1.5 ppb to 250 ppm. The sampling interval is 1 min at a flow rate of 0.6 L/min. Ozone concentration was set three levels (~90 ppb, ~160 ppb, ~280 ppb). And the accuracy of the basic is within ±10% which meet the experimental requirements.

Atmospheric dust concentration was regarded as the upstream particle concentration. Particle was directly measured using the Lighthouse (3016, US) which is a hand-held air dust particle counter. Lighthouse 3016 counts 6-channel synchronous particles and displays cumulative and monomeric particle count data as well as temperature and humidity data for its hand-held particle counter through its 3.8-inch (9.65 cm) touch screen.

The toluene was generated by the toluene generator developed using evaporation principle, through the mass flow controller to control the intake air flow, the evaporation of toluene liquid through the 3mm thick glass slowly penetrate into the gas flow area, thus controlling the occurrence of pollutants concentration stability. And the concentration was continuously monitored by a real-time PID sensor equipped with a 10.6eV lamp (ppbRAE, RAE Systems, USA). Continuous monitor will better grasp of the TVOCs change during the reaction process. It has a detection range of 1 ppb-10000ppm and a resolution of 1 ppb. The upstream concentrations were set as different levels (1.38ppm, 2.49ppm, 4.60ppm, 16.55ppm). The accuracy of the pollutant generation system is within ±10% or less to meet the experimental requirements.

**Methods and related parameters**

In the performance test and evaluation, analysis of different performance evaluation parameters is particularly important in the performance evaluation of ozone, particle and toluene. Filter efficiency and containing capacity are mainly the two parameters to evaluate the filtration performance.

The performance evaluation parameters are calculated as follows:

(1) Filter efficiency:

The formula for calculating the filter efficiency of the prepared material is:

\[
E = \frac{C_1 - C_2}{C_1} \times 100\% \quad (1)
\]

E: Filter efficiency;
C_1: Upstream concentrations of the fiber material test reactor;
C_2: Downstream concentrations of the fiber material test reactor;

(2) Capacity:

In the test of the above pollutants, different concentrations are chosen, so the capacity under different concentrations is calculated. The filter efficiency curve of the filter material is plotted with time according to the test data. It is calculated according to the complete efficiency breakthrough curve, terminated at 10% filter efficiency.

Defined t_0 as the starting time of the test; t_f as the termination time of the test; L as the horizontal line when the final removal efficiency is 10%; A as the left area of t_f below the curve, B as the left area of t_f over the curve.

The formula for calculating the capacity of the prepared filter material is:

\[
m_c = \frac{A}{A+B} \times m_t \quad (2)
\]

Where

m_c: The capacity of the testing filter;

m_t: The total mass of the upstream pollutant acting on the testing filter;

**RESULTS**

1. Experimental Study on Ozone Removal Performance of Prepared Filter

Different activated carbon loading amount, ozone concentration and wind speed will have an impact on the prepared filter removal performance. Different activated carbon loading amount filter were chosen for the ozone removal performance test. Then different ozone concentration (~90 ppb, ~160 ppb, ~280 ppb) and wind speeds (0.1 m/s, 0.2 m/s) were used to study the effect of the filter ozone removal performance.

1.1 Removal performance comparison of different loading amount filter

With the continuous occurrence of ozone, F9 filter penetration increased rapidly, and G4 filter is relatively slow. The difference of prepared F9 and G4 filter is actually the difference of activated carbon loading amount. Figure 5 shows penetration concentration increased slowly with the activated carbon loading amount increased, indicating that the ozone removal performance has improved. The results show that with the increase of the activated carbon loading amount, the filter final removal efficiency with large amount has improved 6% after 7.5h. Compared the single-layer G4 filter (S-G4 + AC-596g/m²) with the double-layer G4 filter (D-G4 + AC-249g/m²), the double-layer G4 filter loading amount is smaller than that of single-layer G4 filter. But the final removal efficiency is higher than that of single-layer G4 after 7.5 h of ozone removal. The residence time increase with the increase of layers number, and the pollutants obtain a better removal. Thereby it further improves the efficiency of the pollutants removal. Detailed removal performance comparison of different loading amount filter shown in Table 2.
1.2 Removal performance comparison of different ozone concentration

This study aimed at the serious ozone excess phenomena and high value events in the Yangtze River Delta and Jing-Jin-Ji area. The removal performance of different ozone concentration was studied. Based on the different ozone concentration levels in these areas, three ozone concentrations (~ 90ppb, ~160ppb, ~280ppb) were set as the upstream concentration. Two layers prepared filter (596g/m²) were used to do the experiment, and results were shown in Figure 6. It was found that ozone downstream concentration almost stabilized with the reaction time accumulation. Improve the ozone concentration at this time. Then prepared G4 filter removal efficiency decreased with the ozone concentration increased. Prepared G4 filter removal efficiency is of stability in a specific value in each ozone concentration range. The ozone removal efficiency of such filters is stabilized at about 90% when the ozone concentration is close to 300ppb as shown in Table 3. This shows that the prepared filter can really achieve a good removal of ozone.

![Figure 6. Prepared filters for ozone removal of different ozone concentrations](image)

**Table 3. Table of different ozone removal efficiency**

<table>
<thead>
<tr>
<th>Concentration (ppb)</th>
<th>Filter efficiency</th>
</tr>
</thead>
<tbody>
<tr>
<td>98</td>
<td>98.57%</td>
</tr>
<tr>
<td>160</td>
<td>91.16%</td>
</tr>
<tr>
<td>288</td>
<td>87.53%</td>
</tr>
</tbody>
</table>

1.3 Removal performance comparison under different wind speeds

In the recommended wind speed for air purifiers or large air conditioning units, the penetration of the filter (596g/m²) increased about twice with the wind speed increase twice. And the ozone removal efficiency is reduced by 10% as shown in Figure 7. In order to improve the efficiency of the prepared G4 filter, low wind speed was the best choice in normal use. Low wind speed can increase the retention time, thereby better to remove the pollutants from the flow.

![Figure 7. Effect of different wind speeds on ozone removal performance of filter media](image)

2. Experimental Study on Particulate Removal Performance of Prepared Filter

Atmospheric dust was chosen as the upstream dust source in particle removal performance test. Filter testing wind speed is about 0.1m/s-0.2m/s according to ISO standards and literature search. Therefore the wind speeds were set at 0.1m/s, 0.15m/s, 0.2m/s for the experiment, respectively. The upstream concentration was the average value of the different particle size range at three speeds. The prepared G4 filter was put into a fiber material test reactor and tested the upstream and downstream pollutant concentration, the sampling time were set as 30s, and interval 30s for a sampling. The particle concentration situation of upstream and downstream for G4 prepared filter at 0.1m/s was shown in Figure 8. At the beginning of the testing, the activated carbon powder loading on the prepared G4 filter was brought into the duct at 0.1m/s wind speed which count as a particulate source. The downstream concentration was comparable to the upstream concentration of particle size more than 0.3μm. The downstream concentration of particulates in 0.3-0.5μm, 0.5-1.0μm and 1.0-2.5μm is higher than the upstream concentration, but the particles concentration soon decreased with the passage of air. Downstream concentration is equivalent to upstream concentration by a few minutes.

![Figure 8. Particle concentration situation of upstream and downstream for G4 prepared filter at 0.1m/s](image)

Figure 9 showed that at the three different wind speed (0.1m/s, 0.15m/s, and 0.2m/s) , the downstream particle concentration is comparable to the upstream particle concentration for several minutes after testing. The total mass concentration of the particulate matter contained in the downstream duct is determined to be 0.001μg/m² calculated by the mass concentration calculation formula of the particulate matter which converted number concentration into the mass concentration. The indoor PM2.5 concentration reference the standard of "ambient air quality standards" (2012) which the average daily concentration is about 35μg/m³. This found that the amount of soot blowing at this time is much smaller than the indoor PM2.5 standard.

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**Table 2. Detailed removal performance comparison of different loading amount filter**

<table>
<thead>
<tr>
<th>Filter</th>
<th>Initial filter eff.</th>
<th>7.5h filter eff.</th>
</tr>
</thead>
<tbody>
<tr>
<td>S-F9</td>
<td>13g/m²</td>
<td>0.50</td>
</tr>
<tr>
<td>S-G4</td>
<td>51g/m²</td>
<td>1.00</td>
</tr>
<tr>
<td>S-G4</td>
<td>245g/m²</td>
<td>1.00</td>
</tr>
<tr>
<td>S-G4</td>
<td>596g/m²</td>
<td>1.00</td>
</tr>
<tr>
<td>D-G4</td>
<td>249g/m²</td>
<td>1.00</td>
</tr>
</tbody>
</table>
requirements. Therefore the secondary soot problem of the prepared filter was with a short time and the little amount of soot blowing which has little effect on the human

![Comparison of the upper and lower concentration of G4 prepared filter at different wind speeds](image1)

Figure 9. Comparison of the upper and lower concentration of G4 prepared filter at different wind speeds

3. Experimental Study on Toluene Removal Performance of Prepared Filter

The filter efficiency at different toluene concentration gradient was plotted in Figure 10. All the capacity at different concentrations gradient was calculated based on the above method (formula 2), and results listed in Table 4. It was found that with the increase of the upstream concentration, the capacity \( m_c \) of the activated carbon increased obviously, and increased from 17.83mg/g of 1.38ppm to 63.98mg/g in 16.55ppm. There is a better linear relationship between the capacity \( m_c \) and the upstream concentration.

![Filter efficiency at different toluene concentration gradient](image2)

Figure 10. Filter efficiency at different toluene concentration gradient

<table>
<thead>
<tr>
<th>Upstream conc. (ppm)</th>
<th>Flow rate (l/min)</th>
<th>90% Penetrates time ( t_{90%} ) (min)</th>
<th>( m_t ) (mg)</th>
<th>( m_c ) (mg)</th>
<th>AC Amt. M(g)</th>
<th>( m_c ) M (mg/g)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.38</td>
<td>4.25</td>
<td>832</td>
<td>18.76</td>
<td>3.51</td>
<td>0.197</td>
<td>17.83</td>
</tr>
<tr>
<td>2.49</td>
<td>4.25</td>
<td>380</td>
<td>15.48</td>
<td>3.58</td>
<td>0.191</td>
<td>18.73</td>
</tr>
<tr>
<td>4.60</td>
<td>4.27</td>
<td>202</td>
<td>15.31</td>
<td>3.24</td>
<td>0.197</td>
<td>16.39</td>
</tr>
<tr>
<td>16.55</td>
<td>4.2</td>
<td>168</td>
<td>28.29</td>
<td>10.30</td>
<td>0.161</td>
<td>63.98</td>
</tr>
</tbody>
</table>

Table 4. Adsorption performance parameters at different toluene concentration gradient

CONCLUSIONS

1) The prepared filter removal performance improved with the increases of active carbon loading amount, finally ozone remove efficiency for the maximum loading amount filter increased by 6% in the same time;

2) Under different concentration of ozone (~900ppb, ~1600ppb, ~2800ppb), the removal efficiency of stability of the preparation of filter material in the same value (99%, 91%, 88%), under different velocity (0.1m/s, 0.2m/s), when velocity increased about doubled, the penetrability increased twice as much value of low velocity, and removal efficiency declined by 10%;

3) Prepared G4 filter do have the secondary blowing problem, but after a few minutes under the wind speed of 0.1 m/s the downstream concentration is at the stable stage, the study found that the time of secondary blowing problem is short and only small amount of ash fall off from the prepared G4 filter.

4) The removal of toluene for the preparation of G4 filter is more objective, a few hours after the removal efficiency decreased to 10%. With the increase of toluene concentration in the upstream (1.38-16.55ppm), the capacity \( m_c \) is also greatly improved (17.83-63.98mg/g). There is a better linear relationship between the capacity \( m_c \) and the upstream concentration.

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