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Carbon Aerogel - Application as Toxic Vapor Adsorbent in Respirator

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In order to protect rescue personnel from the harmful effects of vapors during fire or chemical incidents, respirators are used as a mandatory requirement. The toxic gas adsorbent is a very important component of the respirator. In this study, carbon aerogel filter with a nonwoven fabric (polypropylene - PP, polyethylene terephthalate - PET) was fabricated by a sol-gel method incorporating thermal processes. Carbon nanomaterials are determined specific surface area by Brunauer-Emmett-Teller (BET), material surface shape and structure by Scanning Electron Microscopy (SEM), density. Experimentally evaluate the benzene vapor adsorption capacity of the filter plate at the initial benzene concentration of 5ppm, the temperature of 40 °C and the gas flow up to 30 $L.min^{-1}$. The resulting carbon aerogel filter has a surface area of 720.5–805.6 (m^2/g), porosity (97.5-98.9 vol%) and pore size (6.1-22.2 Å). CAF-PP and CAF-PET filters have capacity equivalent adsorption, 2.3 and 2.4 mg/g. Breakthrough time for both materials is more than 30 min. CAF-PET filter has shown to be more suitable for use in respirator than CA-PP filter when have lower pressure loss. The results show that the carbon aerogel is suitable for use as an adsorbent for respirator.

1. Introduction

In recent years, volatile organic compounds (VOCs), which are considered serious problems for the environment, such as ozone depletion, greenhouse effect, photochemical pollution and health humans (Zhang et al., 2017). Benzene, which is an important VOCs in the gas phase, is a potential precursor to photochemical pollution and a variety of toxic by-products, extremely dangerous to human health and the environment, even at very low concentrations (Kutluay et al., 2019a). Benzene causes both indoor and outdoor air pollution leading to many health problems including skin, eye or throat irritation, tired, headache, nausea, dizziness, respiratory disease, cancer and mortality from long-term exposure to these pollutants (Saha et al., 2018). Effective benzene reduction technologies such as adsorption, oxidation and biodegradation have been studied. Adsorption is considered as one of the best technologies widely used due to its applicability to remove many pollutants and simplicity in design and operation (Basaleh et al. 2019). Studies have been performed to investigate the adsorption of VOCs on a variety of carbon materials, including the newly designed carbon adsorbent (Zhang et al., 2017). Activated carbon (AC), produced from Elaeagnus angustifolia seeds by physical activation method, to investigate the gas-phase adsorption processes of benzene. The results show that adsorption time is 74.98 min, initial benzene concentration is 16.68 ppm, temperature is 26.97 °C and maximum adsorption capacity is 437.36 mg/g (Baytar et al., 2020). Carbon-silica aerogel composites synthesized from liquid glass and granular activated carbon through post-synthetic surface modification with trimethylchlorosilane give good benzene adsorption efficiency. The maximum adsorption capacity achieved was 395.3 mg/g at a controlled benzene concentration at 700 ppm, a flow rate of 100 $mL.min^{-1}$ and a temperature of 35 °C (Dou et al., 2011). Carbon xerogel activated with NH_4Cl 5% w/v combined with pyrolysis in nitrogen gas at 850 °C for 2 h at a flow rate of 10 $mL.min^{-1}$, achieving a specific surface area is 1008 m^2/g, shows high benzene adsorption capacity. The adsorption capacity was in the range of 109-206 mg/g when initial benzene concentration of 100-400 ppm with a flow rate of only 0.3 $L.min^{-1}$, at laboratory temperature (Rastegar et al., 2020). Tests were performed with very

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high initial concentrations of benzene at low flow rates. In practice, the concentration of benzene dispersed in the air is usually very low. The benzene adsorption efficiency of carbon materials shows the potential for their application in the manufacture of personal protective equipment, especially respiratory protective equipment.
In respirators, granular activated carbon (GAC) is the most common adsorbent used to combat gaseous contaminants, such as volatile organic compounds (Okrasa et al., 2019). The GAC has several limitations, including the need for containment, consumption of granular materials, and particle entrainment, which need to be contained in a box, which adds to the weight and bulkiness of the respirator, contributing to annoying for users. This leads to high rates of non-compliance with wearing personal protective equipment when necessary, with discomfort being the primary cause.
Carbon aerogel (CAF) cloth is considered an alternative adsorbent - to control VOCs - which overcomes some of the disadvantages of GAC (Okrasa et al., 2019). The CAF obtained from the carbonization and activation of aerogel fabric (AF) can be prepared from precursors resorcinol, formaldehyde, sodium carbonate, non-woven fabrics (Gan et al., 2019). CAF has small density, large specific surface area, rich pore structure, good chemical stability and environmental compatibility, high porosity and extremely low density due to its pore structure. medium. Carbon surface can be chemically modified to improve hydrophobicity and selectivity towards target specific compounds, such as VOCs (Gan et al., 2019). The low density of CAF should reduce the overall mass of the respirator. CAF is easier to handle than GAC because it can be produced in a variety of forms, such as woven fabrics and unwoven felts. These advantages make CAF a good adsorbent for the development of thin, light and effective respirators that can be used as short-term protection for first responders and the public in the event of a catastrophic event (Okrasa et al., 2019).
In this study, a carbon aerogel filter (CAF) was synthesized from resorcinol and formaldehyde combined with non-woven fabric (polypropylene - PP, polyethylene terephthalate - PET) by sol-gel combined with freeze-dried, and pyrolysis. Evaluation of the benzene vapor adsorption capacity of the CA filter by a laboratory gas adsorption system combined with the pressure loss assessment of the filter.

2. Material and Methods

2.1 Materials

Carbon aerogel synthesis from resorcinol, formaldehyde, deionized water and sodium carbonate (Xilong, China). Non-woven fabric from polypropylene and polyethylene terephthalate as the base for the toxic gas filter. Benzene vapor is generated in the laboratory for the adsorption experiments.

2.2 Characterization of CAF

CAF are determined density, specific surface area by BET method, material surface shape and structure by Scanning electron microscope, thermal stability evaluation by TGA analysis.

2.3 Preparation of CAF

The CAF is synthesized according to the process shown in Figure 1. Nonwoven fabric (PP, PET) is washed with deionized water 3-4 times to remove fabric fibers and dirt, then dries the surface by oven at 80 °C. Then cut into sheets of size diameter 120 mm and thick 0.5 mm.
Prepare a mixture of resorcinol (R), formaldehyde (F) and sodium carbonate (C) in the ratio R/F = 0.5, R/C = 1000, RF/water = 40 wt%. Cover sol-gel mixture on a prepared nonwoven sheet and leave for 24 h at 80 °C. The fabric is then freeze-dried for 24-48 h at -50 °C. The cloth was heated in a pyrolysis furnace and activated according to N_2 and CO_2 gas flow conditions at 800 °C for 30 min, with a flow rate of 200 mL.min^{-1}. The product is cleaned to remove coal dust, obtained a CAF and measured and analyzed physico-chemical properties.

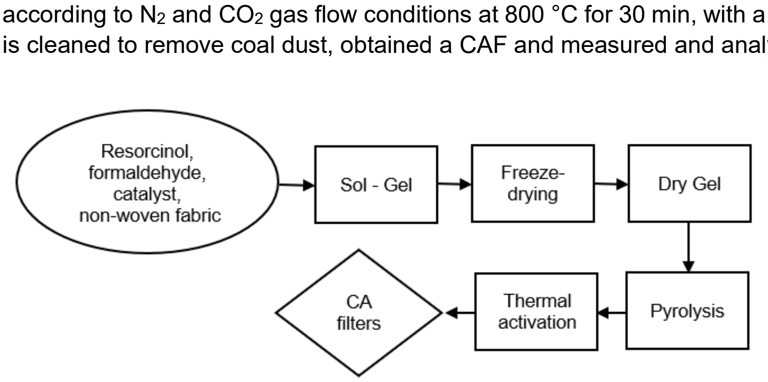

Figure 1: The process of synthesizing carbon aerogel filters

2.4 Adsorption experiments

The adsorption test was carried out on an isometric gas adsorption system to evaluate the filter pressure drop and the gas adsorption efficiency (Figure 2). Benzene vapor (5 ppm) was prepared for the test. The concentrations of benzene vapor before and after the filter were determined by chromatography-mass spectrometry (GC/MS) using a capillary column. Evaluate the number of filter layers from 1, 5, 10, 15 and 20. Air flow rate 30 L.min^{-1}, temperature 40 °C according to TCVN/QS 1223:2010. The adsorption efficiency was evaluated according to the activity retention time and the benzene vapor adsorption efficiency of the adsorbent.

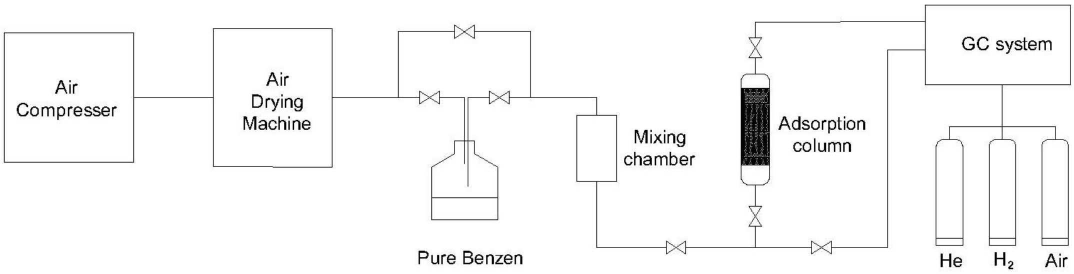

Figure 2: Experimental setup for gas adsorption system

3. Results and Discussion

3.1 Properties of the CAF

Table 1 shows the properties of CAF such as density, specific surface area, pore size and porosity. CAF has a low density (0.12–0.35 g.cm^{-3}) and high porosities (97.5-98.9 vol%). The specific surface area of CAF reaches 720.5–805.6 m^2/g with pore size of 6.1-22.2 Å, which is very suitable as an adsorbent. CAF is structured mainly of micropores with 71.98 vol% micropore, 12 vol% mesopore and 16 vol% macropore.

Table 1: Properties of the CAF

Properties	CAF
Density (g.cm^{-3})	0.12–0.35
Porosity (%)	97.5-98.9
BET (m^2/g)	720.5–805.6
Pore size (Å)	6.1-22.2
Particle size (Å)	77.02
V_{total} (cm^3/g)	0.4408
V_{mic} (cm^3/g)	0.3173
V_{mes} (cm^3/g)	0.0547
V_{mac} (cm^3/g)	0.0688
V_{mic} (%)	71.98
V_{mes} (%)	12.40
V_{mac} (%)	15.62

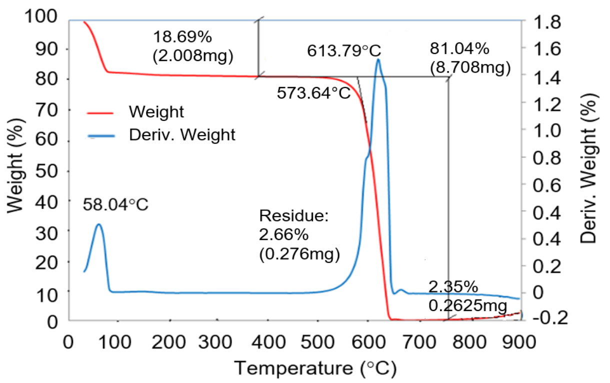

Figure 3: Thermogravimetric analysis (TGA) results for CAF

The thermal behavior of CAF was evaluated by thermogravimetric analysis (TGA) in air (Figure 3). The analysis results show that CAF has the ability to be applied in high temperature conditions up to 550 °C. The TGA graph shows two main peaks at about 58 °C and 614 °C. The mass change at low temperatures (below 100 °C) is mainly the evaporation of water vapor in the CAF. Since CAF is a porous material, it easily adsorbs water vapor in the atmosphere during storage, about 18 wt% water. The appearance of the second peak with great intensity in the temperature range of 550-650 °C shows that the CAF structure has a strong change, the mass is reduced by 81 wt%. At this temperature range occurs the combustion reaction of carbon with oxygen (600-800 °C) in the air, destroying the structure of CAF leading to a sharp decrease in mass.
Figure 4 shows the surface structure of the CAF samples. The morphology of the CAF particles formed is spherical or almost spherical. The CAF particles bond to the latter to form long chains of beads, and partially agglomerate together to create the porous structure of the CAF in three dimensions. The porous structure of CAF is not uniform in the whole material, the locations of the well-connected beads create a structural framework with higher porosity, larger pore size; while the positions of CAF particles agglomerate together to create a less porous structure, small pores. The results of scanning microscopy showed that CAF-PP pore size in the nanoscale is larger than that of CAF-PET.

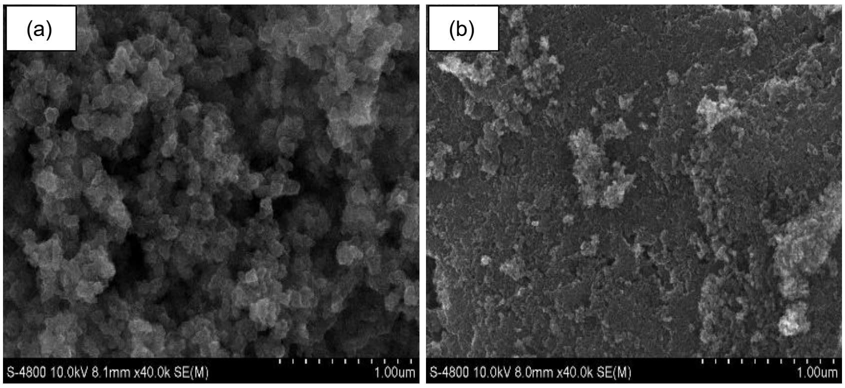

Figure 4: SEM images of CAF: CAF-PP (a) and CAF-PET (b)

The N_2 adsorption-desorption isotherms of the material samples are presented in the following Figure 5. Divide the adsorption isotherm into 3 regions corresponding to 3 relative pressure ranges P/P_o: region I ($P/P_o \leq 0.2$), region II ($0.2 < P/P_o < 0.7$), zone III ($P/P_o > 0.7$).

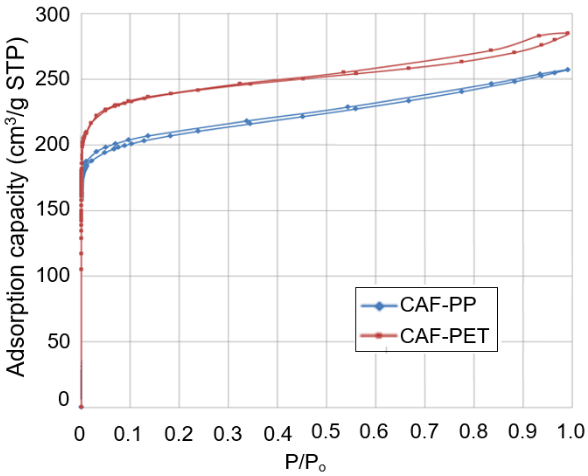

Figure 5: Adsorption desorption isotherms of N_2 on 2 samples: CAF-PP and CAF-PET

For activated carbon aerogel materials, the adsorption isotherm curve depends on type I, region I, the amount of N_2 increases suddenly in the direction of increasing relative pressure P/P_o, this shows that the studied material contains a large number of microcapillaries (ultra-micropore, capillary diameter less than 1 nm). In Zone II, the amount of N_2 increases but relatively slowly when the relative pressure P/P_o increases, so the research material contains medium capillary with small pore size. In Region III, the amount of N_2 also increases very little in the direction of increasing pressure. In this region a small hysteresis loop appears, indicating that there is a small amount of medium capillary with small pore width in the material. Both CAF-PP and CAF-PET have uniform adsorption and desorption curves indicating the reusability of the materials. CAF-PET has a higher adsorption and desorption curves of CAF-PP, showing that the adsorption capacity of CAF-PET is better than that of CAF-PP.

3.2 Evaluate the pressure drop and adsorption capacity of CAF

The Figure 6 shows the pressure drop of CAF by different number of filter layers. In this study, air flow rate of 30 L.min^{-1} at 40 °C, the filter from PP non-woven fabric has a higher-pressure loss than the filter from PET non-woven fabric. As the number of filter layers increases, the pressure drop increases linearly. According to TCVN/ QS 1223:2010, filter resistance is less than 215.75 Pa. When using a CA-PP filter can only use up to 20 layers (198.5 Pa). With the CA-PET filter has a very low-pressure loss (82.3 Pa), which allows up to 50 layers to be used while still meeting the filter resistance requirements. The results show that testing with 50 layers of CA-PET filters has a filter pressure drop of 205 Pa. The density of PP fibers is much higher than that of PET fibers, which increases the pressure loss. This result shows that the use of PET to make CAF filter is better than that of PP. The report by Zuo and Yao (2015) also shows that the pressure loss of the carbon nanotube filter, the doctor filter and the N95 filter is 84, 62 and 139 Pa, with a gas flow rate of 3 L.min^{-1}. This is a large pressure loss when testing at low air flow rates.

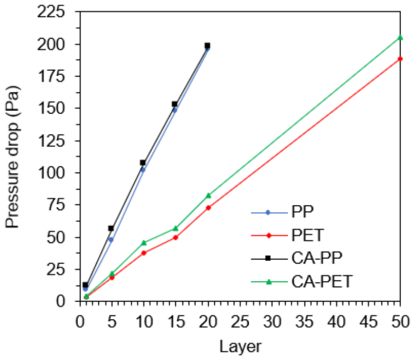

Figure 6: Pressure drop of non-woven fabric (PP, PET) and CAF (CA-PP, CA-PET) with different number of filter layers

Experimental evaluation of the benzene vapor adsorption capacity of CAFs shows in Figure 7. During the initial 30 min, the adsorption capacity increased rapidly. After 30 min, the adsorption equilibrium was reached at air flow rate of 30 L.min^{-1} at 40 °C, benzene concentration of 5 ppm.

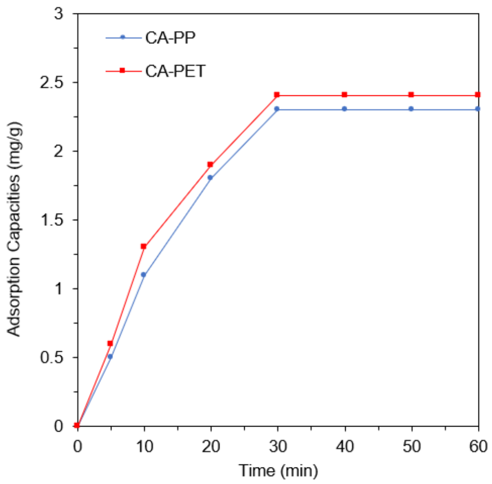

Figure 7: Adsorption capacity of CA-PP and CA-PET

The adsorption capacity from CA-PP and CA-PET filters did not have a clear difference. The results show that the benzene vapor adsorption efficiency is mainly due to the carbon aerogel composition in the filter plate. While non-woven fabric does not affect the adsorption capacity of the filter. Both filters with a CA mass of approximately 2 g have adsorption capacities of 2.3 mg/g and 2.4 mg/g. The results show that the adsorption capacity is higher than that of Huong et al. (2016) when testing benzene vapor adsorption with activated carbon from tea with an adsorption capacity of 0.9 mg/g at air flow rate of 0.3 $L.min^{-1}$, at 40 °C with an benzene initial concentration of 20-40 ppm. The adsorption test of Rastegar et al. (2020) was performed at a very high initial concentration of benzene up to 400 ppm with a low gas flow rate of only 0.3 $L.min^{-1}$ reaching an adsorption capacity of 206 mg/g. This result shows that when increasing the initial benzene concentration, the adsorption capacity also increases significantly (Okrasa et al., 2019).

4. Conclusion

Carbon aerogel filter from nonwoven fabric is fabricated with high specific surface area, small pore size and is mainly made of microparticles based on their type I nitrogen adsorption isotherms, making CAFs are suitable for benzene adsorption at low levels. Comparison of benzene vapor adsorption capacity and pressure loss of CAF-PP and CAF-PET filters was performed in the study. The adsorption capacity of both materials is similar, while CAF-PET has lower pressure loss than CAF-PP. With the advantages of CAF showing promise in the development of short-term disposable respirators that protect against benzene, and possibly other VOCs. Based on the pressure drop and adsorption capacity, CAF-PET is the best adsorbent candidate for the development of thinner, lighter and more efficient respirators. Further research is needed in the respirator practice to continue to evaluate the use of CAFs for respiratory protection.

Acknowledgments

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