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An in-situ thermally regenerated air purifier for indoor formaldehyde removal

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Abstract

Formaldehyde is a common indoor pollutant that is an irritant and has been classified as carcinogen to humans. Adsorption technology is safe and stable and removes formaldehyde efficiently, but its short life span and low adsorption capacity limit its indoor application. To overcome these limitations, we propose an in-situ thermally regenerated air purifier (TRAP) which self-regenerates as needed. This purifier has four working modes: cleaning mode, regeneration mode, exhaust mode, and outdoor air in-take mode, all of which are operated by valve switching. We developed a real-scale TRAP prototype with activated carbon as adsorbent. The experimental testing showed that the regeneration ratios for formaldehyde of TRAP were greater than 90% during 5 cycles of adsorption-regeneration and that through the 5 cycles, there was no damage to the adsorption material as confirmed by scanning electron microscope (SEM) and Brunauer-Emmett-Teller (BET) tests. The total energy consumption by the prototype for purifying 1000 m^3 indoor air was 0.26 kWh. This in-situ thermal-regeneration method can recover the purifier's adsorption ability through at least five cycles.

KEYWORDS
adsorption, air cleaning, formaldehyde, indoor air quality, performance evaluation, regeneration

1 | INTRODUCTION

The main source of indoor formaldehyde pollution is urea-formaldehyde resins in wood-based construction materials.[1] More than 65% of the formaldehyde produced in China is used to synthesize these resins.[2,3] The urban residential building area in China grew from 4 to 21 billion m^2 from 1990 to 2010, and more than 1 50 000 km^2 of synthetic wood-based decorating materials were produced in 2010.[4,5] This construction boom has resulted in formaldehyde pollution in Chinese homes, especially in newly decorated homes. Tang et al reported that the average formaldehyde concentration was 0.238 mg/m^3 with a range of 0-5.59 mg/m^3 in 5905 newly remodeled Chinese homes and 0.256 mg/m^3 with a range of 0-3.39 mg/m^3 in 351 new Chinese office buildings, far higher than China's indoor formaldehyde standard, 80 μg/m^3.[1,6] Formaldehyde is an irritant which causes sensory irritation including eye and nasal irritation even at low concentrations, at 0.25-0.35 mg/m^3.[7] What's more, formaldehyde has been classified as a carcinogen by the International Agency for Research on Cancer. Du et al[8] showed that formaldehyde has become the largest mean contributing pollutant to inhalation cancer risk for adults in urban China.

There are three approaches to reducing indoor formaldehyde concentration: source control, ventilation and air cleaning.[9] Source control is clearly the most preferred method.[9] However, the main source of indoor formaldehyde, urea-formaldehyde adhesives, are the most commonly used products in manufacturing wood-based materials and furniture because they cure rapidly, are compatible with additives, and relatively inexpensive.[3] Ventilation is effective only at very high air change rates, which requires great energy consumption.[9] So besides effective regulations to control formaldehyde emissions from such products and ventilation, more effective air cleaning technologies for formaldehyde removal are also needed to reduce indoor formaldehyde concentration.

The most commonly used technologies for indoor air cleaning are catalytic oxidation, photocatalytic oxidation (PCO), and adsorption.[10,11] Catalytic oxidation materials generally need much higher temperatures than indoor temperature to achieve high removal efficiency.[12] Catalytic oxidation and PCO have the additional problem that the removal material can be poisoned and produce potentially harmful by-products.[13] Moreover, little is known about the practical performance and long-term performance of catalytic and PCO air cleaning in real situations.[11] In contrast, adsorption technology is cheap, stable, and does not produce secondary pollution, but the life span of adsorbents is short and the adsorption capacity is limited.[10] Presently, activated carbon (AC) is the most widely used adsorbent for gas-phase contaminants in buildings.[14] Kumagai et al[15] reported the formaldehyde adsorption capacity of activated carbon from heat-treated rice-husks to be 0.1 mg/g for an inlet formaldehyde concentration of 1.2 mg/m^3. Rengga et al[16] found formaldehyde adsorption capacity of commercial coal-based carbon at inlet concentration of 1.2 mg/m^3 to be 21 mg/g. In comparison, catalysts have been found to have much greater removal capacity. For example, the Pt/MnO_x-CeO_2 catalyst tested by Tang et al[17] removed more than 144.6 g/g formaldehyde for a 36.8 mg/m^3 inlet formaldehyde concentration.

There are two ways to improve adsorbents' formaldehyde adsorption performance. The most widely used method is using additives that generate formaldehyde-adsorbing function groups on the adsorbent surface as well as increase the attractive force of adsorbent surface.[16] Adsorption on these materials is non-reversible. P-amino benzoic acid increased formaldehyde adsorption ability of a rayon-based activated carbon fiber by 2.27 times and by 2.64-3.68 times with heat treatment.[18] Rezaee et al[19] impregnated activated carbon from bone char with acetic acid solution, increasing the adsorption capacity slightly, up to 1.06 times of the initial value. Shin and Song added silver nanoparticles to activated carbon, which increased its adsorptive capacity from 0.458 mg/g to 1.56 mg/g.[20] However, modified activated carbon requires large energy consumption and can be chemically polluting. Besides, the safety of modified materials remains unexamined.

In contrast, few researchers have explored ways to make adsorption reversible. The most commonly studied adsorbent for reversible adsorption is activated carbon fibers (ACF).[21-24] However, ACF is much more expensive than commonly used adsorbents and its preparation process is more complex and difficult.[25] Moreover, loaded ACF is desorbed by direct current (DC) heating.[21-24] Desorption by DC heating depends on the electrical resistance of the adsorbent, which depends on the material geometry and material pre-treatment method, and is generally not practical for indoor purification.[26] Additionally, regenerating ACF using DC heating requires high energy consumption. Sidheswaran et al[21] set up an indoor air cleaning system that regenerates activated carbon fiber alternatively in two sets of air ducts for a mixture of indoor VOCs at ppb level. Their single-pass DC heating method requires 3.08-125.9 kWh to process 1000 m^3 air, depending on the regeneration condition. Subrenat and Cloirec found 0.01 kWh per regeneration period was required to desorb volatile organochloride compounds on 66 g ACF when adsorption period lasts 20 minutes and desorption period lasts 10 minutes, alternatively.[23] High energy consumption has limited the indoor application of DC heating regeneration.

Practical Implications

- Most previous studies have focused on developing new materials with high adsorption ability in non-reversible process. But the improvement of adsorption ability is limited, and those adsorbents are expensive. We have developed an in-situ thermally regenerated air purifier for indoor formaldehyde removal to repeatedly recover the purifier's adsorption ability and thus prolong the life span of purifier. As thermal regeneration is performed on an "as needed" basis, the energy cost of our method is considerably less than the present regeneration method. We use a mechanical method to regenerate the adsorbents so that cheap materials with low adsorption capacity can be satisfactory for indoor purification.

If regeneration can be accomplished with sufficiently low energy consumption, then reversible adsorption on common adsorption materials will be practical for indoor application. In this study, we have developed and evaluated an in-situ thermally regenerated air purifier for indoor formaldehyde removal.

2 | DESIGN AND METHODS

2.1 | Principle of the purifier (TRAP)

Most regeneration methods use continuous single-pass heating method to increase the inlet air temperature and then to regenerate the adsorbents. This method is energy intensive. To find a more energy-efficient process, we noted that regeneration of adsorbent is a coupled heat and mass transfer process. The Lewis number (*Le*) in Equation (1) describes the velocity of thermal diffusion compared to mass (formaldehyde) diffusion inside the adsorbent.

$$Le = \frac{\text{Thermal Diffusion}}{\text{Mass Diffusion}} = \frac{\alpha}{D_{AB}} = \frac{\lambda/(\rho C_p)}{D_{AB}}, \quad (1)$$

where α is the thermal diffusivity of adsorbent, m/s^2; λ is the thermal conductivity of adsorbent, W/(m·K); ρ is the density of adsorbent, kg/m^3; C_p is the specific heat of adsorbent, J/(kg·K); and D_{AB} is the diffusion coefficient of formaldehyde in adsorbent, m/s^2. Table 1 lists Lewis numbers for three common adsorbents at 80°C. All are three to four orders of magnitude greater than 1. This indicates that heat transfers much faster than mass (formaldehyde) at 80°C for these adsorbents. When heated air passes over the adsorbent, the temperature of the adsorption particles will rise fast by thermal conduction, while the release of formaldehyde in the adsorbent will be relatively slow. Therefore, the commonly used single-pass heating method is not

TABLE 1 Lewis numbers of common adsorbents at 80 °C

Adsorbents	Thermal conductivity,W/(m·K)	Bulk density, kg/m^3	Specific heat, J/(kg·K)	Diffusion coefficient of formaldehyde, m/s^2	Lewis number
Activated carbon	0.15[a]	706.7*	890[c]	$8.8 \times 10^{-11,**}$	2710
Silica	0.13-0.17[b]	718*	821.6[d]	$3.15 \times 10^{-12,**}$	69961-91488
Molecular sieve	0.13-0.36[b]	940.1*	1046[e]	$2.19 \times 10^{-11,**}$	6036-16716

Data source: a[27]; b[28]; c[29]; d[30]; e[31];
*Obtained by mercury test.
**Obtained by our lab using the Xu et al method.[32]

necessary, as its main purpose is to provide low-concentration inlet air for desorption. A better method is to keep the adsorbent at a specific high temperature without continuously heating the air through the adsorbent.

We developed an in-situ Thermally Regenerated Air Purifier (TRAP) for indoor formaldehyde removal. The TRAP uses a cycling heating mode to decrease the energy expense of regeneration. TRAP can be embedded in the building envelope, and Figure 1 shows its configuration. The purifier has two ducts: the main duct is open to indoor air by ports P1 and P3, while the side duct is connected to the outdoor air by port P5 through an additional outdoor air in-take duct inside the wall. The main and side ducts are connected by two ports P2 and P4. A primary particulate matter (PM) filter, an adsorbent filter, a fan and a heater are placed in sequence in the main duct from the bottom up. Four operation modes are achieved by switching 3 valves (V1-V3), and each valve can switch to two positions to control the air flow paths as represented by the arrows in Figure 1. The darker arrow represents the higher formaldehyde concentration in the air flow.

Here is a description of each operation mode. (A) Cleaning mode: ports P1 and P3 are opened, and ports P2, P4, P5 are closed by switching valves. Indoor polluted air is drawn into the purifier and passes through the PM filter, adsorbent and fan, respectively. The indoor formaldehyde is removed by the adsorbent. (B) Regeneration mode: ports P2 and P4 are opened, and ports P1, P3, P5 are closed. The air inside the thermal-insulated ducts continuously cycles and is heated by an electrical heater, which desorbs formaldehyde from the adsorbent filter. (C) Exhaust mode: ports P2, P3 and P5 are opened, and ports P1, P4 are closed. The purifier is connected to outdoors so the polluted air inside the purifier is exhausted. (D) Outdoor air in-take mode: ports P1, P4 and P5 are opened, and ports P2, P3 are closed. Outdoor air is drawn indoors to dilute indoor pollutants such as carbon dioxide. During the regeneration process, the regeneration mode (B) and exhaust mode (C) will operate alternatively to prevent formaldehyde concentration inside the purifier growing too high. The operation mode of TRAP, including the interval time for the four modes and the heating temperature, is controlled by a programmable logic controller (PLC). The outdoor air–in-take mode is an additional function that can be operated to provide outdoor air when residents perceive indoor air to be "stuffy" or the CO_2 concentration in room exceeds a benchmark value.

We built a real-scale prototype of TRAP as shown in Figure 2A. The unit is covered with insulating materials and connected to outside air by a plastic tube. Its basic parameters are described in Table 2. This prototype uses activated carbon obtained from Shanxi Xinhua Chemical Co. Ltd, which is treated with $CuSO_4$ solution, washed and with pH adjusted to neutral before it is used as an adsorbent. The results of X-ray diffraction (XRD) tests (Rigaku Corp., Japan) and energy-dispersive X-ray spectroscopy (EDX) tests (EDAX Inc., America) in Figure S3 and Table S1. show that Cu, S, and O are the most abundant elements. Treatment with Cu^{2+} increases the percentage of mesopores and consequently the diffusion velocity into the activated carbon particle for formaldehyde.[33] This treatment also increased the surface polarity and attractiveness of the activated carbon surface.[16] Detailed information about the material is shown in Table 3.

2.2 | Experiment procedure

The formaldehyde removal performance test was carried out in a 3.5 m × 3.4 m × 2.5 m full-scale 30 m^3 airtight chamber. The chamber is made of glass with a stainless steel framework and the internal arrangement inside the chamber is shown as Figure 2B. A ventilating system is built inside the chamber to mix the air in the chamber and exhaust air out. A detailed description is in the China national standard "Indoor Air Cleaner."[6]

Single-pass efficiency and clean air delivery rate (CADR) were used to evaluate the purifying performance of the purifier in this study.[34] To test the performance of the purifier, we used a "pull-down" procedure similar to that described by Chen et al.[35] Prior to each experiment, the test chamber was ventilated for about 1 hour until formaldehyde concentration dropped to less than 0.01 mg/m^3. A quantity of formalin solution with 37% formaldehyde by mass was then evaporated into the chamber using a heating beaker to form a stable initial concentration of 1 mg/m^3, in accordance with the performance test method in China national standard for purifiers.[6] The initial temperature and humidity inside the chamber were adjusted to 25 ± 2°C and 50% ± 10%. Then the purifier at the center of the chamber was turned on for 90 minutes. With this setup and chamber operation, we obtained the formaldehyde concentration decay curve for the purifier and calculated the CADR and single-pass efficiency of the purifier. We also obtained a natural formaldehyde decay curve for natural formaldehyde removal mechanisms such as surface deposition and chamber leakage before the formal experiment.

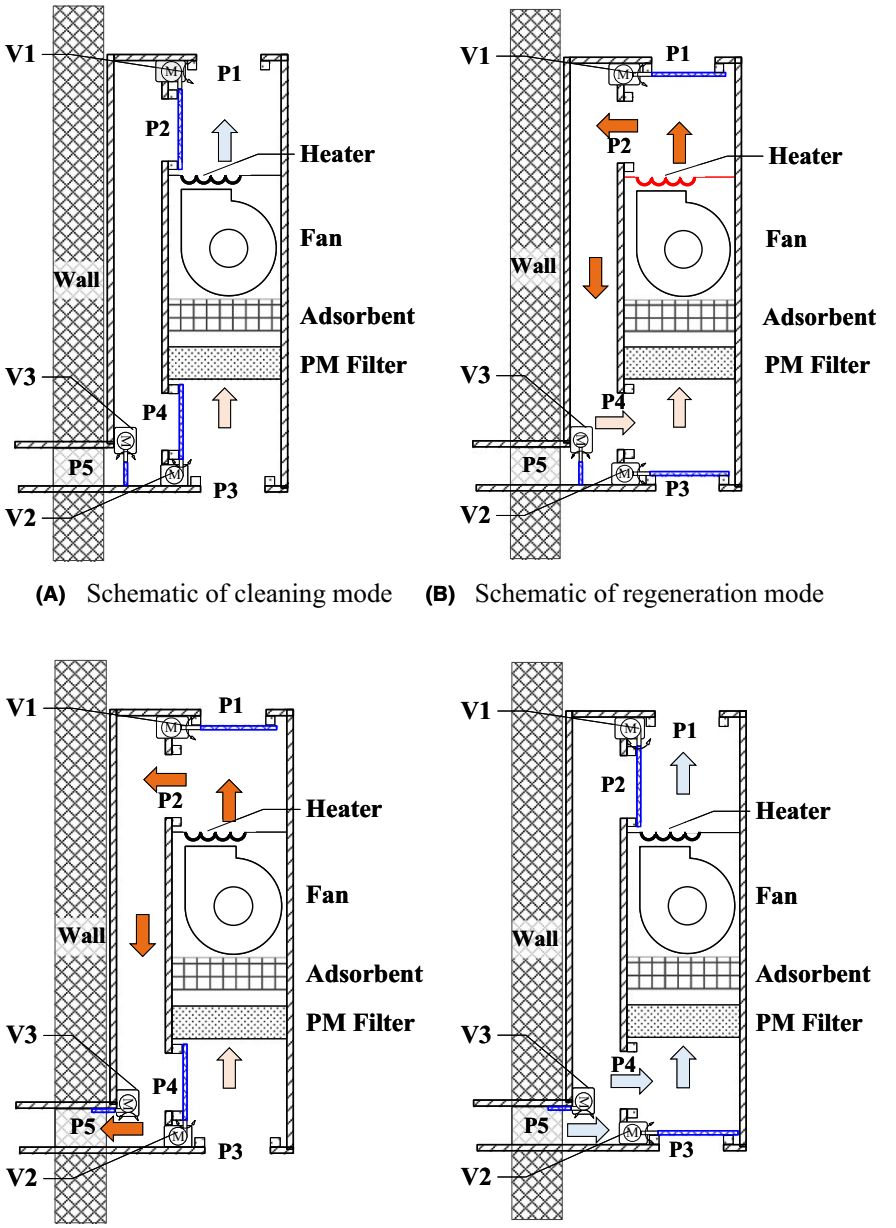
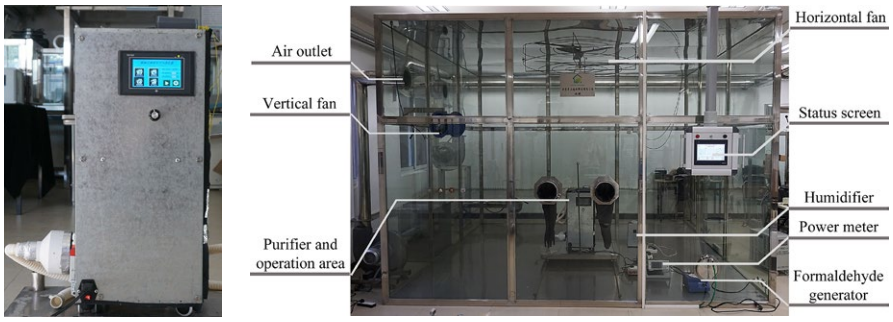


FIGURE 1 Schematic of the in-situ thermally regenerated air purifier (TRAP)

To test the performance stability of the purifier after regeneration, we repeated the experimental procedure for 5 repeated cycles with 3 stages for each cycle. Figure 3 shows the timeline with purifier working modes for each cycle. The working process is shown above the labels, and working modes of the purifier are shown below the label.

Stage 1 is the performance test, which is the same as the "pull-down" procedure described above. Stage 2 is the aging process. The formaldehyde removal device was aged by 10-hour continuously constant formaldehyde dosing and 2-hour standby period, during which the formaldehyde dosing was stopped and the purifier continued working. The formaldehyde was generated by a micro-syringe-pump-based generator using a 37% formaldehyde solution in the chamber.[36] During this stage, the purifier was in cleaning mode and a total of 1350 mg formaldehyde was injected into the chamber. The formaldehyde loading on activated carbon was 2.61 mg/g after an aging period. Another "pull-down" test was performed to obtain the CADR and single-pass efficiency of the purifier after the aging stage in the first cycle. Stage 3 is the regeneration process, in which the purifier was alternatively operated between regeneration and exhaust mode for 6 regeneration cycles. In each regeneration cycle, the regeneration mode was 25 minutes with the air temperature inside the purifier set at 80°C and the exhaust mode was 5 minutes. After 6 regeneration cycles, the purifier was kept at exhaust mode for 30 minutes to cool down the heated adsorbent. The chamber was ventilated during the entire regeneration process to ensure that formaldehyde exhausted from the purifier was completely removed from the chamber. During the entire experiment, the formaldehyde concentration in the chamber was monitored in real time by a gas analyzer (INNOVA



(A) Photo of prototype **(B)** Photo of experiment chamber

FIGURE 2 Photo of real prototype and experiment chamber

TABLE 2 Basic parameters of the TRAP prototype

Parameters	Details
Inner size	0.3 m × 0.2 m × 0.7 m
Inner volume	0.042 m^3
Airflow rate	101.7 m^3/h
Face velocity through filter	0.71 m/s
Duct material	Stainless steel
Adsorbent	0.5 kg activated carbon
Insulation material	Rubber insulation cotton
Heater power (maximum)	400 W

TABLE 3 Basic parameters of activated carbon used in the prototype

Parameters	Details
Ash content [a]	15%
Particle diameter	Cylinder of Φ2 mm × 4-8 mm
Density	608.8 kg/m^3
BET surface area[b]	804.6 m^2/g
Pore volume [b]	0.362 m^3/g

[a]Tested by combustion method.
[b]Tested by Brunauer-Emmett-Teller (BET) test.

1312, AirTech, Denmark), which was calibrated using the MBTH (3-m ethyl-2-benzothiazolinonehydrazone hydrochloride) spectrophotometry method with UV spectrophotometer (Unico, WFJ7200) in advance.

All 3 stages were included in each test cycle. After the i^{th} regeneration process, the performance test in the $i + 1^{th}$ cycle began. Thus, the performance in the first stage of the i^{th} cycle represents the adsorption performance of the adsorbent after *i-1* times of adsorption-regeneration cycles.

2.3 | Mass conservation of formaldehyde in the test chamber

Assuming that the chamber air is well mixed and that other formaldehyde removal mechanisms such as surface deposition and chamber leakage can be characterized by a first-order ratio constant k_n, the mass

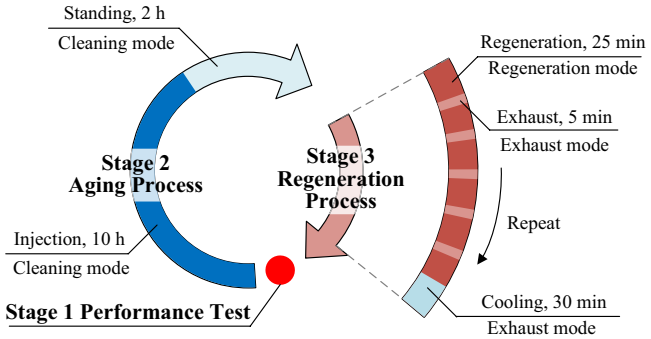


FIGURE 3 Timeline of performance stability test

conservation of formaldehyde in each "pull-down" test can be written as Equation (2). Assuming that CADR does not change during the test period, the CADR of the purifier can be determined by linear regression of ln (*C*) vs time from the measured concentration decay curve, Equation (3). The single-pass efficiency η can be calculated as Equation (4),

$$V\frac{dC}{dt} = -(k_n V + \mathrm{CADR})C, \tag{2}$$

$$\ln(C) = -(k_n + \frac{\mathrm{CADR}}{V})t + \ln(C_0), \tag{3}$$

$$\eta = \frac{\mathrm{CADR}}{Q}, \tag{4}$$

where *V* is the volume of the chamber, 30 m^3; *C* is formaldehyde concentration in the test chamber, mg/m^3; k_n is the formaldehyde concentration decay rate without the air purifier operating, h^{-1}; *Q* is the volumetric flow rate through the purifier, m^3/h; η is the single-pass efficiency of the purifier, %.

Because of the uncertainty of linear regression, we estimated the CADR error using Equation (5), which is obtained using Equation (3) and the error propagation principle:

$$\mathrm{CADR} = V \times [(k - k_n) \pm (\Delta k + \Delta k_n)] \tag{5}$$

where *k* represents the slope of linear regression, h^{-1}; Δk is the uncertainty of slope, h^{-1}; and Δk_n is the uncertainty of formaldehyde concentration decay rate without the air purifier operating, h^{-1}.

3 | RESULTS AND DISCUSSION

3.1 | Performance of single adsorption-regeneration cycle

Figure 4 shows the fitting curve of ln (*C*) vs time for three "pull-down" tests of the purifier in a single adsorption-regeneration cycle, including initial performance and the performances after aging and regeneration. The formaldehyde concentration decay curves are shown in Figure S1. The initial CADR for formaldehyde of the purifier was 66.3 m^3/h, and its initial removal efficiency was 65.2%, as calculated by Equations (3) and (4). The formaldehyde concentration was reduced to less than the China national standard, 0.08 mg/m^3, after 60 minutes.

After aging, formaldehyde concentration inside the test chamber decreased more slowly in the "pull-down procedure," and the CADR value of the purifier dropped to 39.3 m^3/h. This indicates that the adsorption ability of the purifier decreased due to the large amount of formaldehyde adsorbed on activated carbon in the aging process. However, after regeneration for 3 hours at 80°C, the decay curve of formaldehyde was almost the same as in the initial decay curve, and its CADR value was 69.6 m^3/h, signifying full recovery of adsorption capacity. Thus, the thermal-regeneration process can effectively remove the adsorbed formaldehyde from the activated carbon in the purifier.

The temperature of activated carbon material and external surface of purifier was monitored by thermocouple (AZ88598, Hengxin Company, China) in regeneration process and is shown in Figure S4. The energy consumption during the adsorption and regeneration process was monitored by an electrical power meter (PA310, ZhiYuan Company, China). Total energy consumption is the sum of energy consumed for heating, fan and valve operations. During the 3-hour regeneration process, the total energy consumption was 0.325 kWh and the energy used to keep air temperature inside the purifier at 80°C was 0.073 kWh. Assuming the purifier works 12 hours and is regenerated for 3 hours every day, the total energy consumption based on the prototype is 0.26 kWh to purify 1000 m^3 indoor air per regeneration process. Energy consumption in the regeneration process is mainly for conducting heat to the environment, heating adsorbent material and heating air in the duct. Because the air inside the duct is sealed and cycles within the duct, energy consumption for heating air is less than that required for single-pass heating.

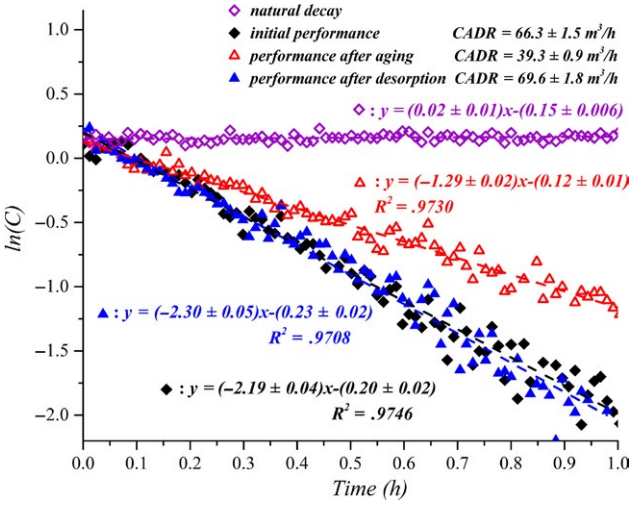


FIGURE 4 Fitting curves of decay curve for a single adsorption-regeneration cycle

3.2 | CADR and regeneration ratio of multi-cycles

The CADR value in each adsorption-regeneration cycle can be calculated by Equation (2) and (3). The regeneration ratio of the i^{th} cycle is defined as:

$$r_i = \frac{CADR_i}{CADR_0}, \tag{6}$$

where r_i represents the regeneration ratio of the i^{th} cycle; $CADR_0$ represents the initial CADR of the purifier; and $CADR_i$ represents the CADR in the first stage of the i^{th} cycle. The fitting curve of "pull-down" tests, ln (*C*) vs time in five repeated cycles, is shown in Figure S2. The CADR values in different cycles and the regeneration ratios are summarized in Figure 5.

The linear fitting between the CADR value and the repeating cycle is described by Equation (7).

$$CADR_i = \beta_0 + \beta_1 \cdot \text{cycle}_i, i = 0,1,\cdots,n \tag{7}$$

where $CADR_0$ is the initial CADR value; $CADR_i$ is the CADR value in the performance test stage of the i^{th} cycle; β_1 is the slope of the fitting curve; β_0 is the intercept of the fitting curve; n is the number of the cycle.

The null hypothesis H_0: β_1=0 is validated with a t distribution test at confidence probability of .05 using Equation (8), where the standard deviation of the slope $S(\beta_1)$ is given by Equation (9); $\overline{CADR}$ and $\overline{cycle}$ are the average values of *CADR* and *cycle*, separately. In our study, the test statistic t_1 is 0.837, which is less than the t-test, $t_{0.05/2}$ (n-2) = 2.776. Thus, the hypothesis H_0 is accepted, which means the slope β_1 is not significant. It concluded that the CADR value remains stable over 5 adsorption-regeneration cycles.

$$t_1 = \frac{\hat{\beta}_1}{S(\hat{\beta}_1)} \tag{8}$$

$$S(\hat{\beta}_1) = \sqrt{\frac{\sum_{i=1}^{n}(CADR_i - \overline{CADR})^2}{(n-2)\sum_{i=1}^{n}(cycle_i - \overline{cycle})^2}} \tag{9}$$

The dots in Figure 5 are the regeneration ratios calculated using Equation (6). They were all above 90%, which indicates that most formaldehyde adsorbed in the aging process was removed by 3-hour

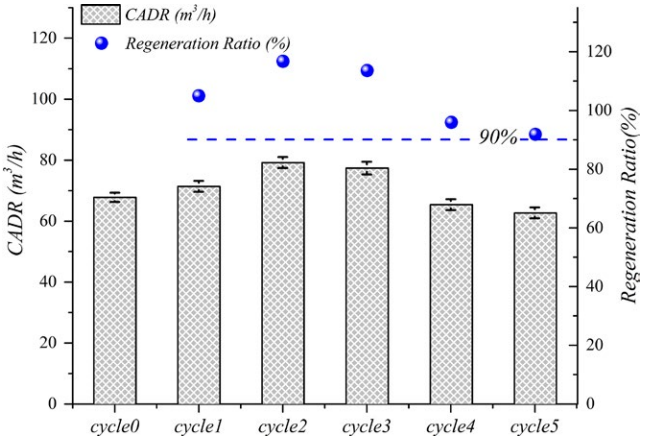


FIGURE 5 Comparison of CADR values and regeneration ratios of the purifier in different cycles

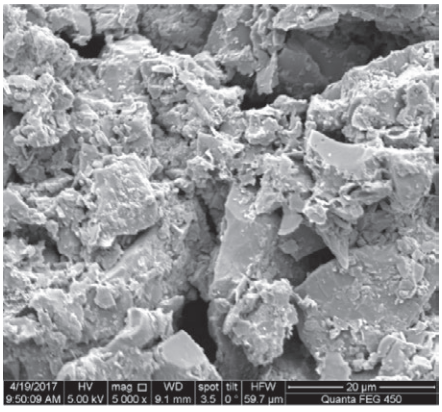
regeneration at 80°C. From Figure 5, we can conclude that the purifier can effectively self-regenerate by the adsorption-regeneration cycle, thus extending the purifier's life span.

3.3 | Comparison of material characteristics before and after regeneration

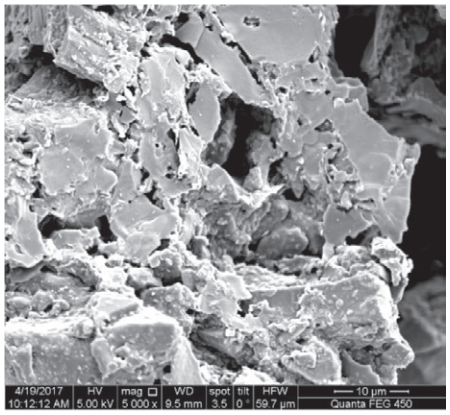
We also explored the structure of the activated carbon before and after regeneration using a scanning electron microscope (SEM) to observe surface morphology and the Brunauer-Emmett-Teller (BET) test to test the material structure.

The activated carbon used in this study was ground and sieved through 80-mesh sieves. SEM images before and after 5 cycles of adsorption-regeneration were obtained by scanning electron microscopy (Quanta, FEI), as shown in Figure 6A,B. The magnification of the SEM images is 5000 × at 5 kV. We also examined an SEM picture of the same activated carbon heated at 380°C for 4 hours at magnification of 10000x, 5 kV, as shown in Figure 6C. Figure 6A,B shows that abundant 1 μm ~ 10 μm pores are widely distributed on the surface of activated carbon. The pore size and pore distribution before and after adsorption-regeneration cycle are similar. From Figure 6C, the structural destruction caused by heating, namely a densified surface, is apparent in many particles. But for particles after 5 cycles of regeneration at 80°C in Figure 6A,B, there is no evidence of this kind of destruction. The structure and surface morphology appear to be stable after regeneration at 80 °C.

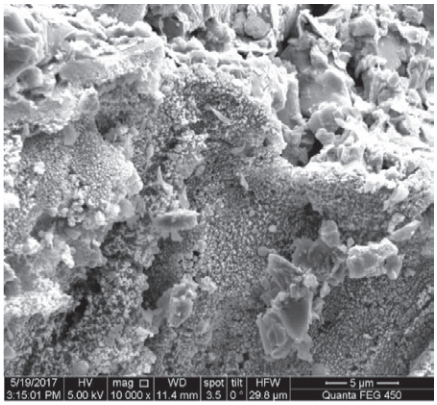
Nitrogen adsorption isotherms of 14 samples, including 9 samples before and 5 samples after 5 adsorption-regeneration cycles were measured by Micromeritics ASAP 2020 accelerated surface area and porosimetry apparatus (Micromeritics Ins., Corp.) at 77.404 K and at a relative pressure range from 7×10^{-6} to 1 atmosphere. The pore size distribution of samples was tested using density functional theory (DFT) as shown in Figure 7.[37] The *x* axis represents pore diameter, the left and right *y* axis represent differential pore volume and cumulative pore volume. Figure 7 shows that the differential pore volume and



(A) SEM image before regeneration (5000×)



(B) SEM image after regeneration (5000×)



(C) SEM image after being treated for 4 h at 380 °C

FIGURE 6 The surface SEM images before and after regeneration

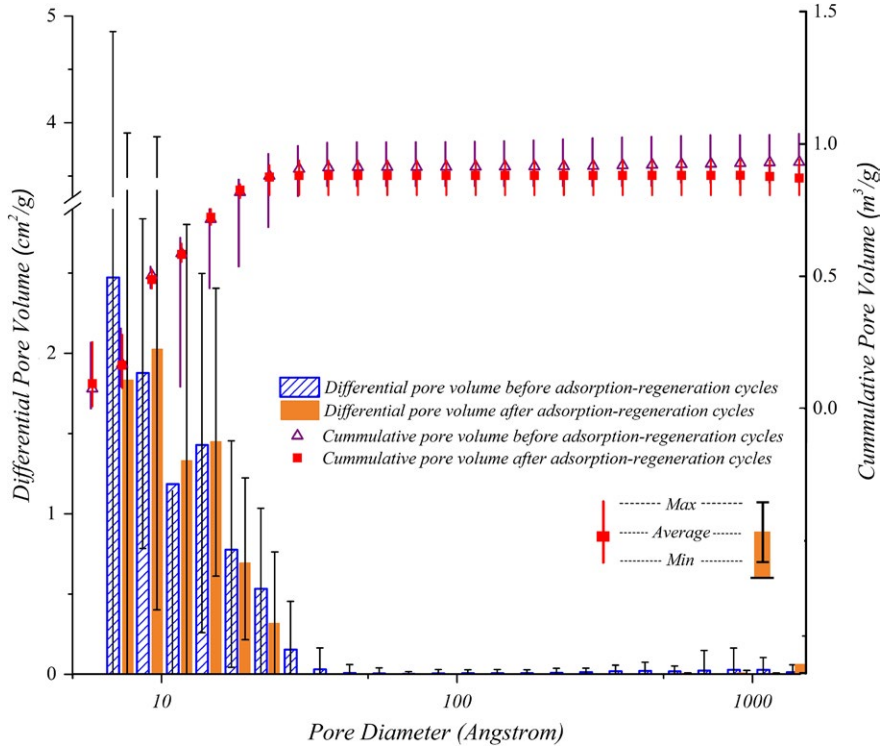


FIGURE 7 Comparison of pore distribution before and after adsorption-regeneration cycles

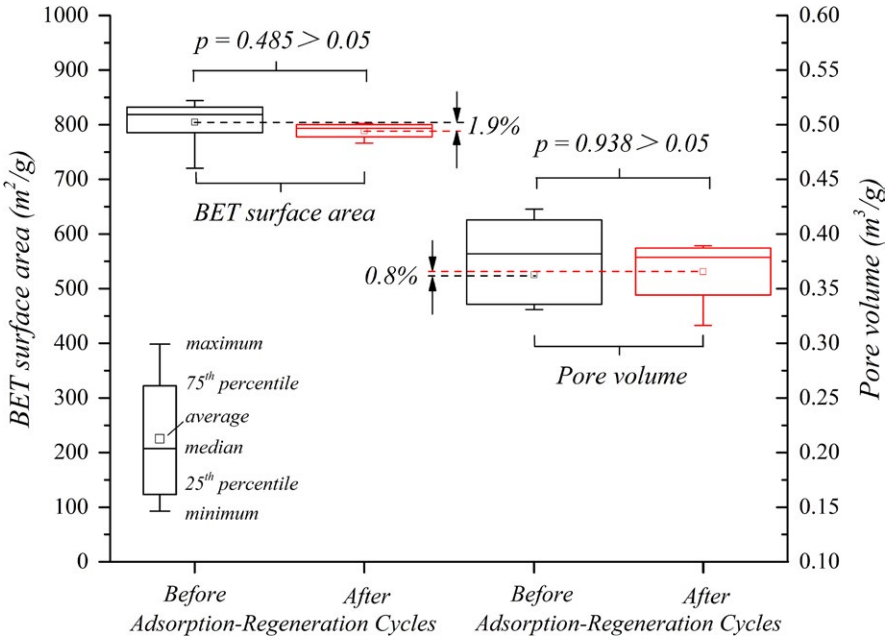


FIGURE 8 BET surface area and pore volume before and after adsorption-regeneration cycles

cumulative pore volume in each range of pore diameter remained the same after the adsorption-regeneration process.

Figure 8 shows the specific surface area and pore volume calculated by the standard BET method.[38] The average BET surface area of material before and after adsorption-regeneration cycles decreased from 804.6 m^2/g to 789.0 m^2/g, that is, by 1.9%. The average pore volume of material before and after the adsorption-regeneration cycles increased from 0.362 m^3/g to 0.366 m^3/g, that is, by 0.8%. The significance of these differences was tested with a *t*-test with significance accepted for *P*-values ≤ .05. We obtained *P*-values ≥.05 and concluded that the BET surface area and pore volumes did not change significantly during the experiment. Thus, both SEM examination and BET test give evidence that the adsorbent was not significantly changed by TRAP regeneration.

4 | CONCLUSIONS AND FUTURE WORK

We have presented TRAP, an in-situ thermally regenerated air purifier in which the adsorbent can be regenerated following air purification

and can be used repeatedly. In 5 cycles of adsorption-regeneration, regeneration ratios exceeded 90% with no damage to the material as detected by SEM and BET tests. Compared to formaldehyde removal devices without the regeneration module, the proposed in-situ thermal-regeneration method prolongs the life span of adsorbents by regeneration, thus decreasing the frequency of filter replacement. Importantly, the regeneration process based on a cycling heating mode is energy-efficient. We use a mechanical method to reduce the requirement for adsorbents and to make cheap materials with low adsorption capacity usable for indoor air purification.

This kind of purifier has wide applications. In addition to being used as a wall-hanging air cleaner or a portable air cleaner, it can also be used as an in-duct component in central HVAC systems. In real situations, the performance of the purifier may be influenced by environmental conditions such as temperature, humidity, and competitive adsorption of other volatile organic compounds.[39] For example, the adsorbed water vapor on the adsorbent may enhance the adsorption of formaldehyde due to the water solubility of formaldehyde. Therefore, to obtain better adsorption and thermal-regeneration performance, more studies of adsorbent materials, regeneration temperatures, regeneration intervals, and the influence of competitive adsorption should be performed. Another issue that deserves further investigation is the combination of in-situ thermal regeneration with adsorption-catalytic materials. Many catalytic materials need high temperature to achieve high decomposition efficiency for indoor gaseous contaminants, and the cost of catalysts is high.[12] Catalysts and adsorbents can be combined to overcome this problem. The adsorbent will adsorb the indoor gaseous contaminants first and when the adsorbent reaches saturation, the catalyst can be in-situ heated to achieve efficient decomposition of the adsorbed contaminants. However, for catalytic materials, adsorption/desorption must be tested through a sufficient number of cycles to evaluate performance stability. More investigations on this issue are needed.

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REFERENCES

1. Tang XJ, Bai Y, Duong A, et al. Formaldehyde in China: production, consumption, exposure levels, and health effects. *Environ Int*. 2009;35:1210-1224.
2. Bohm M, Salem MZM, Srba J. Formaldehyde emission monitoring from a variety of solid wood, plywood, blockboard and flooring products manufactured for building and furnishing materials. *J Hazard Mater*. 2012;221:68-79.
3. Salthammer T, Mentese S, Marutzky R. Formaldehyde in the indoor environment. *Chem Rev*. 2010;110:2536-2572.
4. China Statistical Bureau. *China Statistical Yearbook*. Beijing, China: Chinese Statistical Bureau; 2010.
5. Zhang YP, Mo JH, Weschler CJ. Reducing health risks from indoor exposures in rapidly developing urban china. *Environ Health Persp*. 2013;121:751.
6. GB/T 18801-2015. *Air Cleaners*. Beijing, China: Standardization Administration of the People's Republic of China; 2015.
7. Lang I, Bruckner T, Triebig G. Formaldehyde and chemosensory irritation in humans: a controlled human exposure study. *Regul Toxicol Pharm*. 2008;50:23-36.
8. Du ZJ, Mo JH, Zhang YP. Risk assessment of population inhalation exposure to volatile organic compounds and carbonyls in urban China. *Environ Int*. 2014;73:33-45.
9. Fanger O. What is IAQ? *Indoor Air*. 2006;16:328-334.
10. Zhang YP, Mo JH, Li YG, et al. Can commonly-used fan-driven air cleaning technologies improve indoor air quality? A Literature Review *Atmos Environ*. 2011;45:4329-4343.
11. Gallego E, Roca FJ, Perales JF, et al. Experimental evaluation of VOC removal efficiency of a coconut shell activated carbon filter for indoor air quality enhancement. *Build Environ*. 2013;67:14-25.
12. Pei JJ, Zhang JS. Critical review of catalytic oxidization and chemisorption methods for indoor formaldehyde removal. *HVAC&R Res*. 2011;17:476-503.
13. Mo JH, Zhang YP, Xu QJ, et al. Determination and risk assessment of by-products resulting from photocatalytic oxidation of toluene. *Appl Catal B-Environ*. 2009;89:570-576.
14. Vanosdell DW, Owen MK, Jaffe LB, et al. VOC removal at low contaminant concentrations using granular activated carbon. *J Air Waste Manage*. 1996;46:883-890.
15. Kumagai S, Sasaki K, Shimizu Y, et al. Formaldehyde and acetaldehyde adsorption properties of heat-treated rice husks. *Sep Purif Technol*. 2008;61:398-403.
16. Rengga P, Sudibandriyo M, Nasikin M. Development of formaldehyde adsorption using modified activated carbon-a review. *Int J Renew Energ Dev*. 2012;1:75.
17. Tang XF, Chen JL, Huang XM, et al. Pt/MnO_x-CeO_2 catalysts for the complete oxidation of formaldehyde at ambient temperature. *Appl Catal B-Environ*. 2008;81:115-121.
18. Rong HQ, Liu ZY, Wu QL, et al. Formaldehyde removal by rayon-based activated carbon fibers modified by P-aminobenzoic acid. *Cellulose*. 2010;17:205-214.
19. Rezaee A, Rangkooy H, Jonidi-Jafari A, et al. Surface modification of bone char for removal of formaldehyde from air. *Appl Surf Sci*. 2013;286:235-239.
20. Shin S, Song J. Modeling and simulations of the removal of formaldehyde using silver nano-particles attached to granular activated carbon. *J Hazard Mater*. 2011;194:385-392.
21. Sidheswaran MA, Destaillats H, Sullivan DP, et al. Energy efficient indoor VOC air cleaning with activated carbon fiber (ACF) filters. *Build Environ*. 2012;47:357-367.
22. Yao M, Zhang Q, Hand DW, et al. Adsorption and regeneration on activated carbon fiber cloth for volatile organic compounds at indoor concentration levels. *J Air Waste Manage*. 2009;59:31-36.
23. Subrenat A, Le Cloirec P. Adsorption onto activated carbon cloths and electrothermal regeneration: its potential industrial applications. *J Environ Eng*. 2004;130:249-257.
24. Das D, Gaur V, Verma N. Removal of volatile organic compound by activated carbon fiber. *Carbon*. 2004;42:2949-2962.
25. Lee T, Ooi CH, Othman R, et al. Activated carbon fiber-the hybrid of carbon fiber and activated carbon. *Rev Adv Mater Sci*. 2014;36:118-136.

26. Subrenat A, Le Cloirec P. Thermal behavior of activated carbon cloths heated by joule effect. *J Environ Eng-Asce*. 2003;129:1077-1084.
27. Tamainot-Telto Z, Mayet D. Thermal conductivity measurement of activated carbon with granular graphite. *Chem Sci Eng Ser*. 2016;4:105-105.
28. Vyas RK, Kumar S. Estimation of temperature-dependent thermal conductivity of a packed bed of 13X molecular sieves. *Ind Eng Chem Res*. 1995;34:4058-4062.
29. Dobrosavljevic A, Perovic N, Maglic K. Thermophysical properties of POCO A×M-5Q1 graphite in the 300 to 1800 K range. *High Temp-High Press*. 1987;19:303-310.
30. Chase MW, Curnutt J, Prophet H, et al. Janaf thermochemical tables, 1975 supplement. *J Phys Chem Ref Data*. 1975;4:1-176.
31. Burkholder HR, Fanslow GE, Bluhm DD. Recovery of ethanol from a molecular sieve by using dielectric heating. *Ind Eng Chem Fund*. 1986;25:414-416.
32. Xu QJ, Zhang YP, Mo JH, et al. How to select adsorption material for removing gas phase indoor air pollutants: a new parameter and approach. *Indoor Built Environ*. 2013;22:30-38.
33. Zhang SS, Li W, Zhao X, et al. Influence of pore structure and surface chemical properties on removal performance of formaldehyde by the copper loaded activated carbon. *Chem Ind Forest Prod*. 2015;35:1-7.
34. Siegel J. Primary and secondary consequences of indoor air cleaners. *Indoor Air*. 2016;26:88-96.
35. Chen WH, Zhang JS, Zhang ZB. Performance of air cleaners for removing multiple volatile organic compounds in indoor air. *ASHRAE Trans*. 2005;111:1101-1114.
36. Wan HL, Mo JH, Xiang JB, et al. A dynamic generator of gaseous formaldehyde for the life-span evaluation of household air cleaners. Proceedings of the 9th International Symposium on Heating Ventilation and Air Conditioning and the 3rd International Conference on Building Energy and Environment, Tianjin, China; 2015: Paper ID: 903.
37. Ryu ZY, Zheng JT, Wang MZ, et al. Characterization of pore size distributions on carbonaceous adsorbents by DFT. *Carbon*. 1999;37:1257-1264.
38. Gregg SJ, Sing KSW, Salzberg H. Adsorption surface area and porosity. *J Electrochem Soc*. 1967;114:279C-279C.
39. Fairen-Jimenez D, Carrasco-Marin F, Moreno-Castilla C. Adsorption of benzene, toluene, and xylenes on monolithic carbon aerogels from dry air flows. *Langmuir*. 2007;23:10095-10101.

SUPPORTING INFORMATION

Additional Supporting Information may be found online in the supporting information tab for this article.

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