Modelling and implementation of an in-situ thermally regenerated adsorption module for removing gaseous xylene

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Keywords:
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Mathematical modelling
Air cleaning
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A B S T R A C T
The technology of in-situ thermally regenerated air purifiers (TRAP) can effectively recover adsorbents' adsorption capacity, which is promising for low-concentration uncontinuous waste gas treatments in light pollution industries, such as furniture industries and tire factories. This study aims to provide a design model for the adsorption behaviours of the TRAP module under various practical situations in the furniture industry. We propose a dimensionless parameter (the Number of Mass Transfer Units, NTU_m) to predict the one-pass efficiency. A TRAP module consisting of multiple flexible and thin adsorption boards was developed to validate the model experimentally. The polluted air with xylene flows through the module and is adsorbed by the boards. When the adsorption boards are saturated, the heating plate will be electrically heated to about 80 °C, releasing xylene from the boards and regenerating their adsorption performance. The modelling results agree with the experimental results, which indicate that the NTU_m is a crucial and helpful parameter in enhancing the efficiency of the TRAP module.

1. Introduction
Volatile organic compounds (VOCs) are the essential precursors of near-surface ozone and secondary organic aerosols, significantly contributing to urban chemical smog and haze [1–3]. VOCs will carry out photochemical action in the atmosphere, which is the main reason for forming particulate matter [4]. In addition, VOCs such as toluene and formaldehyde emitted from industry manufacturing are harmful to humans [5], which will cause sensory irritation [6], allergies [7], asthma [8], and even leukaemia [9]. In China, industrial emissions are the largest source of atmospheric VOCs [10–12]. The industrial release of VOCs mainly comes from stationary industry sources, typically including furniture manufacturing, vehicle manufacturing, printing, equipment coating, electronic manufacturing, and biological pharmacies. Wang et al. reported that furniture manufacturing contributed immensely to VOCs emissions and ozone formation potential [13]. Thus, the emissions from furniture manufacturing were estimated to have substantial health risks and should be priority-controlled.

Commonly used methods for VOCs emission control include regenerative combustion, photocatalysis, adsorption and other technologies, which are limited by the VOCs concentration, species and air volume. Regenerative combustion is mainly suitable for treating medium- and high-concentration continuous waste gas. The advantages of this technology are that VOCs can be quickly and efficiently decomposed, with an efficiency of more than 99% [14,15]. However, oxidation chamber temperature and outlet temperature are the main factors affecting the VOCs decomposition elimination of the regenerative thermal oxidizer [16]. In practical applications, it is necessary to further design the operating conditions and device structure according to VOCs composition [15,17,18]. Thus, the equipment investment and operating costs of the regenerative combustion method are high. Among the approaches to removing VOCs from the air, photocatalytic oxidation (PCO) is a promising method [19,20]. However, some reactants will generate partial oxidation products that are relatively more harmful to people’s health, limiting this technology’s application and development [21].

For low-concentration uncontinuous waste gas treatments in light pollution industries, such as furniture industries and tire factories, adsorption technology is widely used for VOCs purification due to its low cost and high efficiency [22,23]. Although the purifying performance of adsorbents decreases fast during operation time, which would
lead to the industry’s emissions not being reduced in the long-term application, thermal regeneration can effectively recover the adsorption capacity of adsorbents [24–26]. Therefore, the adsorbent can maintain a higher purification capacity during long-term application. Xiao et al. [24] presented an in-situ thermally regenerated air purifier (TRAP) in which the adsorbent can be regenersted following air purification and used repeatedly. However, the purifier still adopted the granular packed bed, resulting in a slow mass transfer rate and long regeneration time in the regeneration process. At the same time, much energy is consumed to heat the inflow air [24,25,27]. Chen et al. [28] demonstrated an in-situ thermal regeneration lamellar air purification board with adjustable surface temperature fabricated with activated carbon, polyimide, and copper foil. The technology developed a micron-scale purification regeneration coating technology, which can achieve rapid in-situ recovery of adsorption material properties through uniform electric heating of each lamellar layer supplemented by clean airflow. Furthermore, Chen et al. [29] proposed a modified method to change the surface topography by preparing macroscopic channels perpendicular to the board. The purpose of the macroscopic channels is to enhance vertical mass transfer into the board. However, all the studies mentioned above focus on purifying residential VOCs. The VOC types and concentrations in industry buildings differ significantly from those in residential environments. In addition, due to the large air volume of the air duct in the industrial plant, the size of the module also needs to be increased, so the board fabrication technology also needs to be upgraded. It is essential to develop a novel TRAP module for the furniture industry.

In addition, most current research focuses on fabricating high-performance adsorption materials. However, there is an intrinsic gap between high-performance materials to high-performance modules in real applications. The purification module is often faced with different working conditions, such as velocities and concentrations, which lead to the deviation of material results when applied to different situations. In a word, it is necessary to build a model to predict the module performance and put forward dimensionless parameters for quick judgment. A design method is also needed to guide the construction of the actual TRAP module based on the physical parameters of the purification module.

In this study, we expected to get a tool to evaluate the performance of TRAP modules quickly. We set up a model to simulate the module performance. According to the simulation results, we designed and implemented an in-situ TRAP module to reduce the typical pollutant, xylene, released from the acrylic painting process on wooden furniture. The one-pass removal efficiency and long-term performance of the module for xylene were tested under typical concentrations and air velocities. In addition, the regeneration efficiency and energy consumption of the module were also evaluated.

2. Methods

2.1. Design of the TRAP module

The TRAP module consists of multiple in-situ surface temperature adjustable laminated adsorption boards developed by Chen et al. [28] to
shorten the regeneration time and reduce the airflow resistance, as shown in Fig. 1. To facilitate the installation, we used thread screws through the adsorption boards to string all the boards together (Fig. 1a).

The overall width of the module composed of all boards is \( W \); the overall height of the module composed of the board and the shell is \( H \); \( L \) represents the length of the board, m. Upstream air with pollutants passes evenly through each adsorption board, where the air velocity, \( u \), and the concentration of pollutants in the air, \( C_p \), passing over each adsorption board can be considered the same.

The boards are kept parallel to each other to form an air channel (Fig. 1b). Narrow aluminium gasket strips were added to the upper and lower edges between the boards. Thus, the spacing distance between the boards, \( d \), remained unchanged at the specific design value. When the polluted air passes through the parallel air channels composed of these lamellae, it is adsorbed and purified by the thin coating layer of adsorbent on the lamellae (Fig. 1c). The polyimide film with etched copper was adopted as a heating plate. Because the adsorption board contains conductive copper, the adsorption board will be heated directly from the inside of the adsorbent, with no need to heat air, reducing the heat transfer process. The parallel air channel structure and the direct heating of the adsorbent ensure low airflow pressure drop and short heating regeneration time, respectively, reducing the energy consumption of the ventilation fan and the regeneration heat.

As detailed in previous research [28,29], the thin coating layer of adsorbent on the lamellae was fabricated using glue adhesion. The adsorbents (activated carbon) are bonded to each other and the surface of the lamellae through adhesives (Fig. 1d). We ground and sieved the activated carbon (KZ15-6 Coal-based impregnated activated carbon from Shaxi Xinhua Chemical Co. Ltd., China) into powder with diameters of 0.15–0.25 mm. The sieved activated carbon powder, adsorbents (ethylene-vinyl acetate copolymer), and deionized water with a thickener (methylcellulose and sodium dodecyl sulfonate) were mixed at a mass ratio of 1:0.1:2 and dispersed by a magnetic stirrer for 30 min at 800 r/min. Unlike the previous preparation [28,29], because this preparation required a more considerable amount of activated carbon, the surfactant SDS (sodium dodecyl sulfonate [30,31]) was used to disperse the entire mixture more evenly. The mixture was then uniformly coated onto the surface of the heating plates and dried at 120 °C for 2 h. The fabricated adsorption board is shown in Supplementary Fig. S1.

After designing such a structure, it is crucial to determine the parameters related to the module, such as the spacing, the length of the lamellae, and the wind velocity skimming the boards’ surface.

### 2.2. Modelling of the TRAP module

#### 2.2.1. Model establishment

Chen et al. [32] have developed a concrete mathematical model to predict the adsorption behaviours of the board in various practical situations. However, there are two shortcomings with the model. On the one hand, the model is built based on two separate lamellae and needs to be expanded into a whole module. On the other hand, comparing the performance of two modules can only be done through model calculation but cannot be quickly judged by the structure and boundary conditions of modules. So there is still a lack of a dimensionless parameter to guide the design of the module structure. We need to propose a new integration parameter to guide the structural design.

The Number of Heat Transfer Units (NTU) is an index reflecting the ratio of the heat transfer capacity of the heat exchanger and the specific heat capacity of the fluid. It is defined as Eq. (1).

\[
NTU = \frac{\dot{Q}}{(G \cdot c_p)_{\text{heat}}} \tag{1}
\]

where, \( \dot{Q} \) is the overall heat transfer coefficient, W/(m\(^2\)·K); \( A \) is the total heat transfer area, m\(^2\); \( G \) is the mass flow rate, kg/s; \( c_p \) is the specific heat of the fluid, kJ/(kg·°C). If the heat transfer coefficient \( \dot{Q} \) and the heat capacity \((G \cdot c_p)\) are particular, the value of NTU is determined by the size of the heat exchanger. Therefore, it is a dimensionless parameter for designing and calculating heat exchanger structure characteristics.

The mass transfer process can be analogous to the heat transfer process, so the module essentially acted as a mass exchanger to remove pollutants and can also be structured with a similar parameter. We established a model for the mass transfer process of pollutant adsorption and purification through the module by analogy with the heat transfer process, as shown in Fig. 1.

The mass transfer process between each board is the same, so two boards can be selected as the object of the equation. The airflow direction is along the z-axis, and the perpendicular direction is the x-axis. At the same time, some of the same assumptions are detailed in Chen’s study [32] and will not be repeated here.

Based on the above simplification, the mass transfer equation for the polluted air can be expressed by Eq. (2).

\[
\frac{dC_p}{dt} + \frac{u dC_p}{dz} = -\frac{2h_{nt}}{d} \left( C_p - C \right), \text{ at } 0 \leq z \leq L \tag{2}
\]

where, \( C_p \) is the concentration of the target pollutant in the air channel, mg/m\(^3\); \( t \) is the time, s; \( h_{nt} \) is the convective mass transfer coefficient, m/s; \( C \) is the concentration of the target pollutant in the adsorbent coating, mg/m\(^3\); \( K \) is the partition coefficient.

Eq. (2) can be dimensionless, as shown in Eqs. (3)–(5).

\[
\frac{dC_p}{dt} + \frac{C_p}{Lh_{nt}} = -\frac{2h_{nt}}{d} \left( C - C_s \right), \text{ at } 0 \leq z' \leq 1 \tag{3}
\]

\[
r' = \frac{tu}{L} \tag{4}
\]

\[
z' = \frac{z}{L} \tag{5}
\]

A dimensionless parameter Number of Mass Transfer Units (NTU\(_{\text{nt}}\)), as shown in Eq. (6), similar to Mo’s study [33], is applied to better design and calculate the module structure characteristics.

\[
NTU_{\text{nt}} = \frac{2h_{nt}}{d} \tag{6}
\]

#### 2.2.2. Validation of the NTU\(_{\text{nt}}\)

It is necessary to verify the obtained dimensionless parameter to further guide us in implementing the module and conducting subsequent adsorption and purification experiments. Chen’s model [32] can accurately predict module efficiency, so the model can be used to verify whether the efficiency results of modules with different size parameters but the same NTU\(_{\text{nt}}\) are equal. Table 1 shows the one-pass adsorption efficiency of modules of different structural sizes at the same inlet pollutant concentration. The larger NTU\(_{\text{nt}}\) means greater mass transfer capacity and higher one-pass adsorption efficiency. On the other hand, the same NTU\(_{\text{nt}}\) presents the same mass transfer capacity. Although the coating length and the height between two parallel boards of two modules are different, the one-pass efficiency is basically identical. Thus, NTU\(_{\text{nt}}\) can be used as an improved parameter to guide the

<table>
<thead>
<tr>
<th>Parameters</th>
<th>NTU(_{\text{nt}}) = 1</th>
<th>NTU(_{\text{nt}}) = 2</th>
</tr>
</thead>
<tbody>
<tr>
<td>Coating length (m)</td>
<td>0.16</td>
<td>0.32</td>
</tr>
<tr>
<td>Height between two parallel boards (m)</td>
<td>0.005</td>
<td>0.005</td>
</tr>
<tr>
<td>Convective mass transfer coefficient (m/s)</td>
<td>1.54 × 7.69 × 1.47 × 7.33</td>
<td>10(^{-2}) × 10(^{-3}) × 10(^{-2}) × 10(^{-3})</td>
</tr>
<tr>
<td>Face velocity of the module (m/s)</td>
<td>1</td>
<td>0.5</td>
</tr>
<tr>
<td>One-pass efficiency</td>
<td>64.24%</td>
<td>64.26%</td>
</tr>
<tr>
<td></td>
<td>86.53%</td>
<td>86.56%</td>
</tr>
</tbody>
</table>
structural design of TRAP modules. However, whether the model can be used for dynamic simulation for the whole module rather than the two boards needs further experimental verification.

2.3. Characterisation

The microstructure of adsorption boards was characterized to evaluate their removal performance. The surface morphology images were obtained using Scanning electron microscopy (SEM, MERLIN VP Compact, Zeiss) with an acceleration voltage of 15 keV. Nitrogen adsorption/desorption isotherms were obtained using a standard BET method. Both the BET surface area and porosity of adsorbents decrease slightly after the fabrication and regeneration. After heating or clogging caused by the adhesive added during fabrication. Thus, the wrapped adhesive may block some gaps, especially the slit channels between particles, leading to a slight decrease in the adsorbents’ porosity. From Fig. 3c, after 8 h of regeneration at 80 °C, there is no evidence of structural destruction caused by heating. The structure and surface morphology appear stable after regeneration at 80 °C.

By increasing the magnification, we can see the material’s internal structure. Images of the internal structure also show that the material’s porous nature has not changed. Compared with Fig. 3d, e and f only have a layer of gelatinous material on the surface, and the pore size and pore distribution before and after the adsorption-regeneration cycle are similar; the structure is not destructed by heating.

Micropore and macropore structural characteristics were obtained from the nitrogen adsorption/desorption and mercury intrusion tests. The specific surface area and pore size distribution of activated carbon before fabrication, after fabrication and after adsorption-regeneration was tested and shown in Fig. 4. The point plot in Fig. 4 shows that the surface area of the activated carbon material is not much reduced by the heating or clogging caused by the adhesive added during fabrication. Further, the fill colour areas in Fig. 4 represent the pore volume density distributions over the pore diameter. The diameter distribution of the pores did not change significantly after the fabrication.

Table 2 shows the specific surface area and porosity calculated by the standard BET method. Both the BET surface area and porosity of adsorbents decrease slightly after the fabrication and regeneration. After adding adhesive and heating, the average BET surface area of materials decreased from 692 m²/g to 670 m²/g and 654 m²/g, respectively. After adding adhesive and heating, the porosity of micropores decreased from 77.7% to 69.3% and 68.3%, respectively. Thus, the SEM and BET tests show that the fabrication process did not significantly change the adsorbent.

3. Results and discussion

3.1. Characterization of activated carbon

Fig. 3 shows that the surface morphologies of activated carbon before and after coating on the fabricated adsorption boards did not change significantly. Fig. 3b shows that activated carbon particles are wrapped by adhesive because the adhesive and particles were stirred evenly during the fabrication process. Thus, the wrapped adhesive may block some gaps, especially the slit channels between particles, leading to a slight decrease in the adsorbents’ porosity. From Fig. 3c, after 8 h of regeneration at 80 °C, there is no evidence of structural destruction caused by heating. The structure and surface morphology appear stable after regeneration at 80 °C.

By increasing the magnification, we can see the material’s internal structure. Images of the internal structure also show that the material’s porous nature has not changed. Compared with Fig. 3d, e and f only have a layer of gelatinous material on the surface, and the pore size and pore distribution before and after the adsorption-regeneration cycle are similar; the structure is not destructed by heating.

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3.2. Prediction and experiment of module VOCs removal capability

3.2.1. One-pass adsorption efficiency of the module

As described in Section 2.2 above, the NTUₘ determines the ideal efficiency of a module. NTUₘ is related to several parameters, but the only physical parameter related to the actual implementation of the module is the spacing between the adsorption boards. Therefore, we need to focus on the parameter of the adsorption boards. At the same time, considering the practical factors, we also need to consider the change in air velocities.

In order to compare the influence of different parameters on the module performance of xylene, we simulated the variation curves of efficiency and resistance under different air velocities and different lamellar spacing. As shown in Fig. 5, with constant spacing, the one-pass efficiency will gradually decrease with the air velocity increase, while the pressure drop will increase by degrees. From the perspective of comprehensive efficiency and resistance, we need the module to have a high adsorption efficiency to ensure the removal of pollutants. On the other hand, we also need to consider the impact of resistance on the cost, so we choose the spacing d of 5 mm as the parameter of the final module.
After completing the module according to the above parameters, the one-pass removal efficiency was tested to investigate the TRAP module’s target gaseous pollutants removal ability. Fig. 6 shows the module’s one-pass adsorption efficiency at different face air velocities. The data for each box in Fig. 6 consists of the average efficiencies of 20 groups per 3 min in 1 h. The inlet concentration of xylene was maintained at 1.0 ppm. The one-pass efficiency of the module for xylene is over 50% at 1 m/s face air velocity. With the increase of face air velocity, the adsorption performance of the module for the target gaseous pollutants gradually decreased because the residence time of pollutants in the board surface became shorter than before, and the contact time between pollutants and adsorbent became shorter, too. However, the one-pass efficiency of xylene decreased slightly with the increase in face air velocity. Through the average efficiency within 8 h, we can obtain the one-pass adsorption efficiencies of the modules under three different face air velocities for xylene.

The relevant parameters of the module were substituted into the model for simulation, and the simulation results obtained are shown in Table 3. Among the related parameters, the diffusion coefficient of xylene in air related to the convective mass transfer coefficient can be obtained from the relevant literature [34]. By comparison, it can be found that the simulated initial efficiency results are in good agreement with the experimental values, and the relative errors are 5.3%, 1.6% and 10.3%, respectively. As can be seen from the error values, the simulation results are credible. To be clear, according to previous research, the one-pass efficiency experimental results for xylene demonstrated that the fabricated process and parallel air channel structure did not affect the adsorption performance of the board for gaseous contaminants.

### 3.2.2. Long-term adsorption efficiency and regeneration efficiency of the module

In order to further explore the purification capacity of the module, long-term experiments were carried out on xylene, which is the most abundant in factory test results. The inlet concentrations of target gaseous pollutants were also maintained at 1.0 ppm, and the face air velocity through the module was set at 1 m/s. The adsorption experiments were conducted in a controlled environment described in Section 2.4.

The long-term efficiency test results (Fig. 7) showed some fluctuations, but the efficiency still showed a downward trend on the whole. Each box in Fig. 7 represents a set of 1-h average efficiencies in one day. Significantly after 20 days of the adsorption experiment, the activated carbon on the TRAP module gradually saturated, and the adsorption efficiency declined to a greater extent. The above results implied that the module has an excellent long-term removal capacity for the target gaseous pollutants and can undertake the purification task of the exhaust gas of the furniture factory so that the exhaust gas can meet the emission standard.

In order to verify the accuracy of the model’s prediction of module adsorption capacity, we also need to put relevant parameters of the
module produced and experimented with into the model to obtain long-term efficiency curves and compare them with the experimental results. Among the relevant parameters, the diffusion coefficient and the distribution coefficient between xylene and the adsorbent can be obtained from Chen’s study [32]. Fig. 7 shows that the predicted efficiencies of xylene were consistent with the experimental data. Modelled values present the same trend as experimental values and can assess that our assumption on mass transfer is justified. The similarity between results during almost the entire experiment suggests that the model can be used for steady-state removal rates and dynamic conditions. The average relative error between the measured and predicted value was 7.4%, and the maximum relative error was 13.3%. Hence, we can conclude that the model fits well with the experimental data for the adsorption process.

At the same time, to evaluate the influence of thermal regeneration on the performance of the TRAP module, the removal efficiency for xylene was monitored following adsorption and desorption cycles. In the desorption process, the adsorbed xylene was rapidly desorbed from the module as the surface temperature of the board increased from room temperature to 80 °C with 30 V applied to the copper foil. Thus, the xylene concentration in the outlet became higher than that at the inlet. Fig. 7 shows that after just 8 h of thermal regeneration, the removal efficiency of the module adsorbed for a month recovered by 90%.

An electrical power meter (PA310, ZhiYuan Company, China) monitored the energy consumption during the adsorption and regeneration. Total energy consumption is the sum of energy consumed for heating and fan. During the 8-h regeneration process, the total energy consumption was 2.7 kWh. Assuming the module works for a month and is regenerated for 8 h, the total energy consumption based on the module is only 128 kWh to purify 40000 m³ of polluted air per regeneration process and month.

4. Conclusion

The modelling of the adsorption of xylene, the most abundant waste gas produced by the furniture industry, was conducted on an in-situ TRAP module. The model is based on the mass transfer of pollutants on an ideal infinite plate. It considers the saturation of the activated carbon layer and the diffusion coefficient of mass transfer at different air

Table 3
The one-pass removal efficiency comparison between the simulation results and experimental results.

<table>
<thead>
<tr>
<th>Air velocity (m/s)</th>
<th>The simulation results (%)</th>
<th>The experimental results (%)</th>
<th>The relative error (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>59.8</td>
<td>56.8</td>
<td>5.3</td>
</tr>
<tr>
<td>2</td>
<td>47.0</td>
<td>47.7</td>
<td>1.6</td>
</tr>
<tr>
<td>3</td>
<td>37.3</td>
<td>41.6</td>
<td>10.3</td>
</tr>
</tbody>
</table>

Fig. 5. The variation curves of (a) efficiency and (b) pressure drop under different air velocities and lamellar spacing.

Fig. 6. The one-pass adsorption efficiency of the TRAP module; The two ends of the box whiskers are the maximum and minimum values in a data set. In addition, the top and bottom of the box represent 75% and 25% of the data, respectively. The horizontal line in the middle of the box represents the average value of the data.

Fig. 7. Long-term adsorption efficiency of the TRAP module for initial 1 ppm xylene at 1 m/s and regeneration efficiency of the module; The gray curve represents the continuous predicted efficiency results; The red box plots represent the adsorption results; The blue box plots represent the regeneration results. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)
velocities of the board. The model is validated by comparison with experimental results generated on a TRAP module. The developed model tends to show that the modelling results are close to the experimental results. It is mainly the case for one-pass efficiency where results from modelling are excellent and accurate with experiments. The modelled results for the long-term efficiency of the xylene also present close trends with the experimental results. The model illustrates that particular attention should be addressed to the NTUmodelling are excellent and accurate with experiments. The modelled tends to show that the modelling results are close to the experimental velocities of the board. The model is validated by comparison with X. Lei et al. org/10.1016/j.buildenv.2023.110275.

Data availability
The work reported in this paper.

Declaration of competing interest
The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability
No data was used for the research described in the article.

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Appendix A. Supplementary data
Supplementary data to this article can be found online at https://doi.org/10.1016/j.buildenv.2023.110275.

References


