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# Near carbon-zero cycle from VOCs capture to carbon fixation

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## Abstract

A new technical route of organic matter capture and carbon fixation is proposed in response of the increasingly strict emission standards of volatile organic compounds (VOCs) in petrochemical industry and the Chinese national strategic development goals of carbon peak and carbon neutralization. A closed loop from raw materials to adsorbents for gas treatment can be achieved by two key technical characteristics: (1) construct a new mesoporous adsorbent with complete desorption and regeneration function by carbon nanotubes (CNTs); (2) convert gaseous organic matter which cannot be recycled in liquid/gas state to CNTs. It realizes the resource integration of "turning waste into treasure" and maximizes the carbon emission reduction effect of waste gas treatment process without consuming extra precious fossil fuel, compared with the traditional technologies of VOCs treatments, including combustion or catalytic oxidation. What's more, the increase in supply of various green electricity is expected to change the current situation of large investment and heavy cost burden of environmental protection technology, and make a great contribution to the national carbon peak and carbon neutrality policy.

**Keywords:** Carbon nanotubes, Volatile organic compounds, Waste gas treatment, Carbon emissions, Closed loop

## 1 Introduction

Carbon neutral is an important issue recognized globally for the sustainable development of human society [1, 2]. The current focus is mainly on the controlled reduction of CO<sub>2</sub> in huge amount in many industries from fossil fuels by adopting various new technologies and the compensation from biomass (plants) or the trade with carbon tax [3]. Among many processes producing CO<sub>2</sub>, the treatment of volatile organic compounds (VOCs) is a special one, which involves low concentration of organics in the gas but huge gross amount of the released, as well as arises the undesirable secondary pollution of photochemical smog [4, 5]. For instance, the amount of the released VOCs per year across China is estimated to be around 20 million tons, similar to those amounts of SO<sub>x</sub> or NO<sub>x</sub>. In detail, the emission of CO<sub>2</sub> in the treatment

of VOCs includes the direct emission in several technical routes (i.e., direct combustion or catalytic degradation [6–8]) and the indirect emission (by using fossil fuel or grey electricity as the energy supply) in all technical routes (such as combustion, catalytic degradation, adsorption-desorption, cooling etc.). Big challenges lie in that when trying to suppress the concentration of directly released VOCs, in order to reduce direct emission of CO<sub>2</sub> and curb secondary pollution, the indirect CO<sub>2</sub> emission from energy consumption would increase drastically due to increasing technical difficulties. From this perspective, the fabrication of a near-zero carbon cycle for the treatment of VOCs remains a great challenge, but of high imperative importance.

Storage of CO<sub>2</sub> is a key step of carbon capture, utilization and storage (CCUS) for carbon neutral, but full of technique and economical challenge [9, 10] and limitation of available storing space [11, 12]. In this work, we proposed the carbon fixation of organics (from the adsorption-desorption unit) to production of carbon nanotubes (CNTs) by chemical vapor deposition (CVD)

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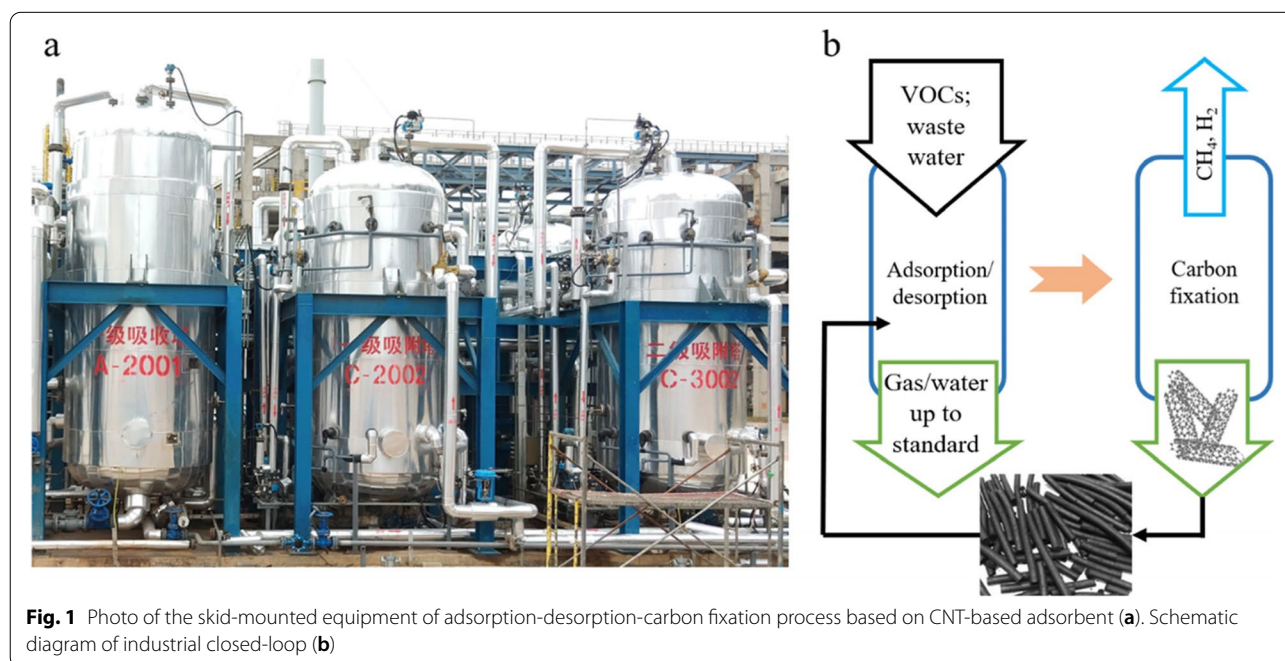
method, in which the organic matters decomposed mainly by transition metal-based catalysts under high temperature to CNTs and  $H_2$  [2–8]. In addition, we performed many studies on the CNT-based adsorbents for the treatment of VOCs and organic-containing water previously [13–18]. These allowed us to build a cycle of adsorption-desorption-carbon fixation technology route, in which CNT-based adsorbent was used for efficient adsorption and desorption of organic matter, and the desorbed organic matter was then transformed into CNTs by high-temperature catalyst. By this means, the direct carbon emissions were reduced largely and the carbon element was changed from pollution source to new material resource. In addition, the era of carbon neutral called for the use of green electricity (from hydro, solar, wind and photovoltaics power etc.) as an energy supply for the emission reduction, aiming to reduce the indirect carbon emissions mentioned above [19, 20]. In this case, it allowed us to develop an industrial closed loop based on the exhaust gas. For the quantitative comparison, we presented the comprehensive analysis of different technologies (including direct combustion, catalytic oxidization, adsorption-desorption-combustion, and adsorption-desorption-carbon fixation) for the treatment of VOCs from aromatics storage tank from 3000–65,000  $mg/m^3$  to 12  $mg/m^3$ . Their energy consumption and carbon emissions varied significantly. The use of green electricity as energy supply would make a great contribution to reduce carbon emissions because the green electricity generating process would not burn coal or other energy sources and

produced no  $CO_2$ . Furthermore, the combination of our technology and green electricity showed the potential to produce a near carbon-zero carbon emissions cycle, as well as large quantities of valuable carbon products. This evaluation would provide a new insight into the carbon emissions of VOCs treatment and offer a new choice for any chemical processes toward carbon zero.

## 2 A new closed-loop technology route constructed by CNTs and its outstanding carbon reduction effect

### 2.1 A new technical route of organic matter capture and carbon fixation

In previous work, we developed a CNT-based adsorbent for revisable adsorption-desorption of organics from VOCs or waste water [13–18]. There are also many reports on the conversion of organics into CNTs [21–27]. At 2020, we, for the first time, integrated these apparatuses together in a skid-mounted equipment for VOCs treatment with the capacity of 900  $Nm^3/h$  (Fig. 1a). The equipment mainly included the adsorption tower (packed with CNT-based adsorbent, operating at 5–10 °C), the carbon fixation column (packed with nanosized iron catalyst, operating at 700–800 °C) and the necessary heat exchanger, pump and compressor. The equipment is capable of continuous operation of adsorption-desorption and carbon fixation. According to estimates (S2), at least 6.3 kg of toluene, 1.1 kg of CNT products (about 659 dollars), as well as 0.1  $Nm^3$   $CH_4$  and 2.4  $Nm^3$   $H_2$ /900



**Fig. 1** Photo of the skid-mounted equipment of adsorption-desorption-carbon fixation process based on CNT-based adsorbent (a). Schematic diagram of industrial closed-loop (b)

$\text{Nm}^3$  VOCs gas were obtained in the carbon fixation process at low concentration.

Thus, we are able to fabricate a closed loop in which the organic matter was completely captured by a new type of CNT-based adsorbent and converted to CNTs by a new catalytic technology (Fig. 1b). The investment of VOCs treatment would be greatly reduced since the produced CNTs could be used as raw materials to fabricate adsorbent. Most of the carbon in the organic matter in VOCs gas was recycled in liquid form or immobilized in CNT products, reducing the  $\text{CO}_2$  emission greatly. However, the processes of adsorption-desorption and conversion of gaseous organic matter to CNTs required energy. The carbon fixation process produces methane and hydrogen at the same time as CNTs. Using methane and hydrogen as heating sources can reduce additional energy consumption and further reduce emission of  $\text{CO}_2$  (Figure S10).

## 2.2 The performance of adsorption–desorption of VOCs by CNT-based adsorbents

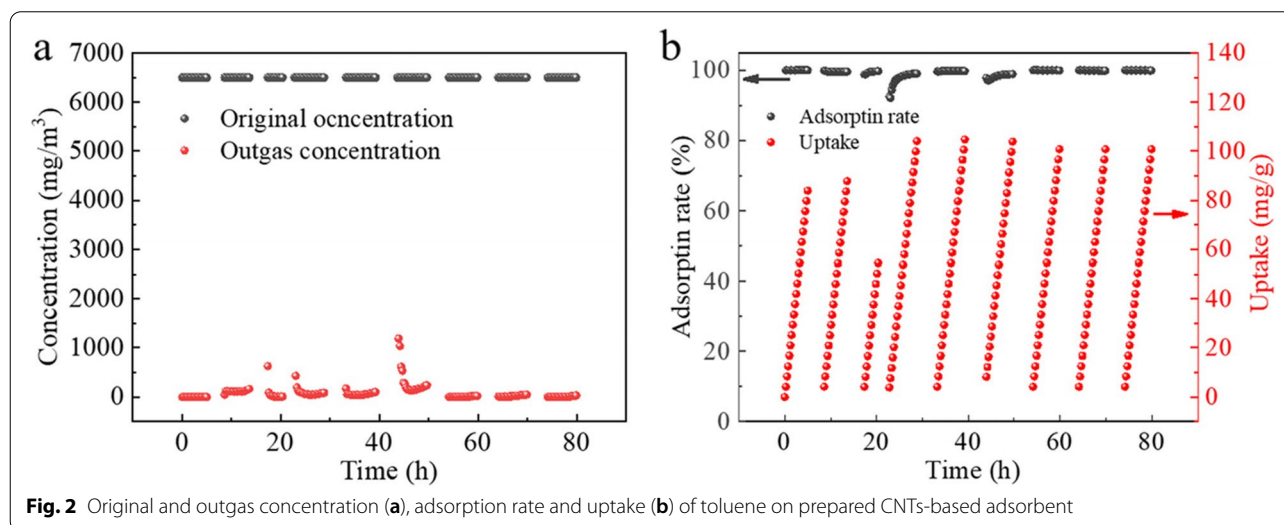
CNTs exhibited the convex pore structure, large mesoporous capacity and high chemical stability ( $\text{sp}^2$  hybrid carbon), enabling rapid adsorption and desorption for VOCs [13, 15–17]. Through the exploration and optimization of the formula and mold methods, we prepared CNT-based adsorbents with high strength, high mesoporous ratio, large specific surface area and high conductivity [14], which behaved rapid desorption rate, high desorption ratio and good recycling performance compared with activated carbon (AC) [13, 15]. In addition, the high thermal and electrical conductivities of CNTs provide an effective heat conduction advantage for cooling and heating operations in the process of

adsorption and desorption when used in industrial installations [18, 28, 29].

After the successful adsorption and desorption, macromolecular aromatic substances are the representative refractory substances of microporous AC. The most obvious advantages of the CNT-based adsorbent are excellent cyclic performance and large adsorption capacity. Taking toluene as an example, the adsorption-desorption cycle test of the prepared CNT-based adsorbents was conducted for the gas system of toluene ( $56,000 \text{ mg/m}^3$ ) and  $\text{N}_2$ . As shown in the cycles in the adsorption isotherm (Fig. 2a), the adsorption has good cyclic performance with an outgas concentration kept below  $12 \text{ mg/m}^3$ . The adsorption rates were all above 99.99%, the desorption rates were near 100%, and the uptake on the adsorbent increased linearly and was far from saturation (Fig. 2b). The outstanding cycle performance provides great possibility of infinite recycling. The far-from-saturation uptake indicate a potential of the adsorbents to work in a primary adsorption process for the treatment of some extremely high concentration gas, providing its usage in another way.

## 2.3 Conversion of gaseous organics into CNTs

In the process of VOCs treatment, there are two outlets for the concentrated organic matter after desorption. For the macromolecules that are easy to liquefy, they are often recycled in liquid form. Smaller fractions of light hydrocarbons cannot be recycled, which is why many scenarios have to use combustion to convert these light hydrocarbons into  $\text{CO}_2$  and meet VOCs emission standards. This is not only high in energy consumption, but also very bad for  $\text{CO}_2$  reduction.



In general, the preparation of CNTs from various carbon sources was very mature [30–33]. Based on our nano-metal catalysts, CNTs were successfully prepared from the transformation of butane, pentane and other hydrocarbons (such as gasoline, ethanol and acetone, showed in S2) to CNTs efficiently. The technology ensured the total amount of NMHC in tail gas well below the national emission standards. The flameless processing technology was realized.

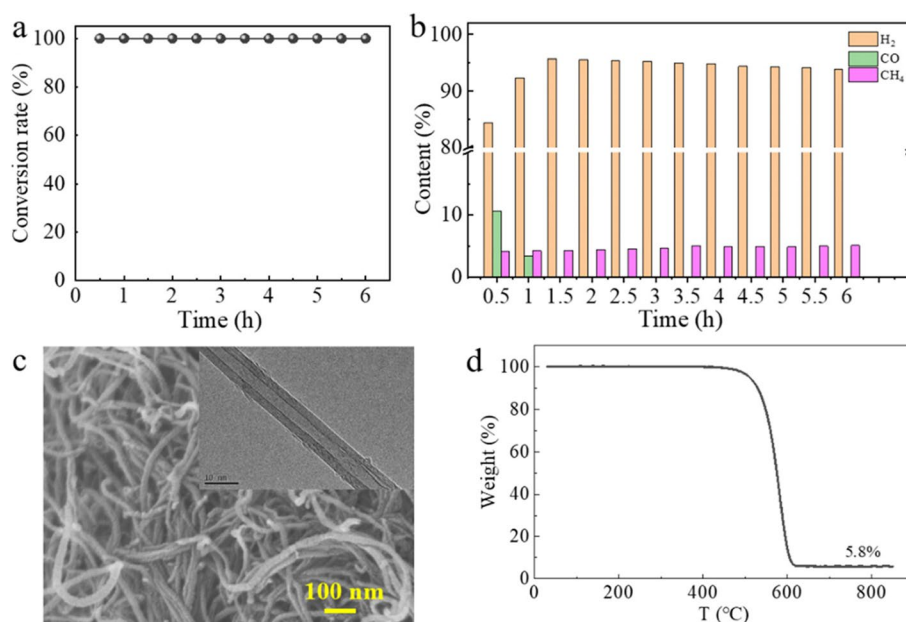
The conversion of pentane was presented here as an example (Fig. 3a). The conversion rate of pentane was 100% during the 6 h reaction. The outlet gas was mainly  $\text{CH}_4$  and  $\text{H}_2$  after 1 h reaction. High selectivity for hydrogen and methane was sustained during the entire reaction period (Fig. 3b). Apparently, the amount of NMHC were very few considering the highly efficient conversion with the nanosized metal catalyst in the present work. SEM image of carbon product had characteristic morphology of CNTs (Fig. 3c). TEM image (Fig. 3c inserted) suggested that the as-produced CNTs had an outer diameter of 8–15 nm. The purity (>90%) (Fig. 3d) was suitable for the fabrication of CNT-based adsorbents, following the method proposed in reference [34].

### 3 Energy consumption and $\text{CO}_2$ emissions of closed-loop technology route compared to traditional VOCs treatment methods

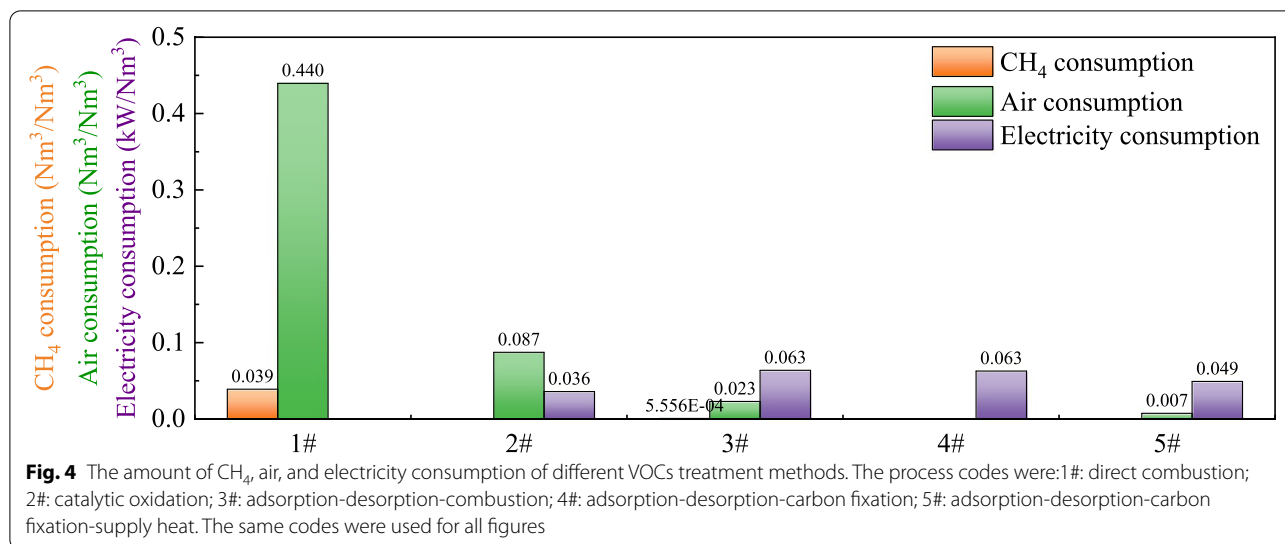
The VOCs in the typical aromatics storage area mentioned above was taken as the object, taking pentane and toluene in nitrogen as the main components, and

their concentrations were  $1500 \text{ mg/m}^3$  and  $6500 \text{ mg/m}^3$ , respectively. Based on the flow and the experimental data, we were able to compare the new adsorption-desorption-carbon fixation process based on CNTs with many conventional VOCs treatment methods (direct combustion process, catalytic oxidation process, and adsorption-desorption-combustion process). The carbon emission,  $\text{CH}_4$  consumption ( $\text{CH}_4$  as ancillary fuel for energy supply), air consumption (air as an assisted medium for burning) and electricity consumption (electricity as energy supply in cooling or heating) were calculated based on the mass balance and energy balance (the calculation details of was showed in SI).

As shown in Fig. 4, only the direct combustion used the fuel of  $\text{CH}_4$  and air in large amount. The preheating of the whole exhaust gas to the burning temperature ( $800^\circ\text{C}$  or above) consumed energy in large amount. In comparison, the catalytic oxidation worked at around  $300^\circ\text{C}$ . Only air was needed to oxidize carbon in organic matter. Electricity was considered here for the heating of the system to the oxidation temperature. For the integrated process of adsorption-desorption-combustion, where organics were condensed, the amount of air and electricity (aiming to energy consumption) were both significantly reduced, compared to the direct combustion. Air was needed for the oxidization of organic matter and  $\text{CH}_4$  which acted as igniter. Electric energy can be used for cooling the adsorption section at  $-5^\circ\text{C}$  and for heating at desorption section at  $200^\circ\text{C}$ . And the burning exhausted gases was



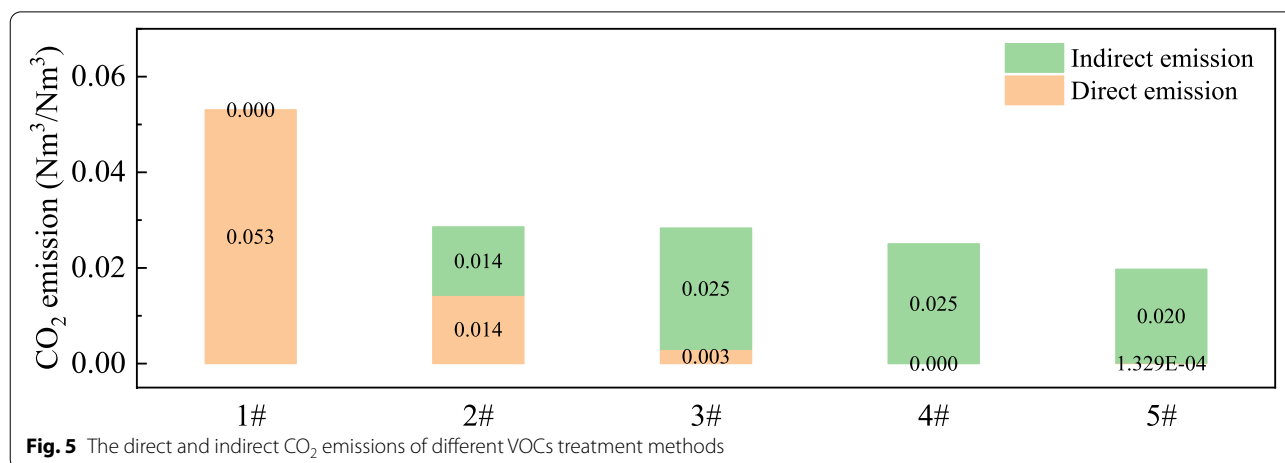
**Fig. 3** The conversion rate of pentane (a) and selectivity for hydrogen and methane during the 6 h reaction. Scanning electron microscopy (SEM) image and transmission electron microscope (TEM) image (the inserted) (c), and thermogravimetric analysis (TGA) (d) of CNTs after carbon fixation reaction



not used for the heat exchange in desorption section. For the adsorption-desorption-carbon fixation process, electricity was further used in heating for the CNTs growth at 700 °C and the high temperature gas (H<sub>2</sub>, CH<sub>4</sub>) was not used as part of energy source for desorption section or carbon fixation section. In this case, the electricity consumption amount was high among these processes. However, if the produced gases (H<sub>2</sub>, CH<sub>4</sub>) were burned as energy supply, the amount of electricity would drop drastically from 56.4 kW to 44.1 kW/900 Nm<sup>3</sup> VOCs gas, and the air consumption was only 6.5 Nm<sup>3</sup>/900 Nm<sup>3</sup> VOCs gas, far lower than 78.4 Nm<sup>3</sup>/900 Nm<sup>3</sup> VOCs gas in catalytic oxidation, since most of carbon from organics was fixed as CNTs. From the comparison, we would state that the direct combustion processes needed fossil fuel in the most amount, while the adsorption-desorption-carbon fixation process represented the process needing electricity in the most amount. There were plenty of

choices to balance the fossil fuel consumption and electricity consumption between these two processes.

As follows, we calculated the CO<sub>2</sub> emission of different processes (Fig. 5). The direct combustion and catalytic oxidation featured the direct emission of CO<sub>2</sub> (released by the oxidation of organics). While the adsorption-desorption-combustion process and processes with carbon fixation mainly featured the indirect emission from the energy supply. Conventional treatment methods of VOCs gave high direct CO<sub>2</sub> emission which cannot be reduced. Seemingly, the indirect emission in carbon fixation exceeds the direct emission in traditional ways. However, the amount of the indirect emission of CO<sub>2</sub> can be decline tremendously if green electricity is applied. In other words, the green potential in carbon fixation process is great, advantage over the other processes. In addition, only the processes with carbon fixation contributed to the valuable solid product (CNTs) with the



yield of 1.1 kg/900 Nm<sup>3</sup> VOCs gas and valuable gaseous products (H<sub>2</sub>, CH<sub>4</sub>), whose value goes far beyond the cost of electricity. Even valuable gaseous products (H<sub>2</sub>, CH<sub>4</sub>) are combusted for energy supply, it reduces the indirect emission of CO<sub>2</sub> by saving electricity. Direct emission of the adsorption-desorption-carbon fixation process is insignificant. In this case, the present carbon fixation as solid product is better than the CCUS where carbon elements are captured and stored in the form of CO<sub>2</sub>.

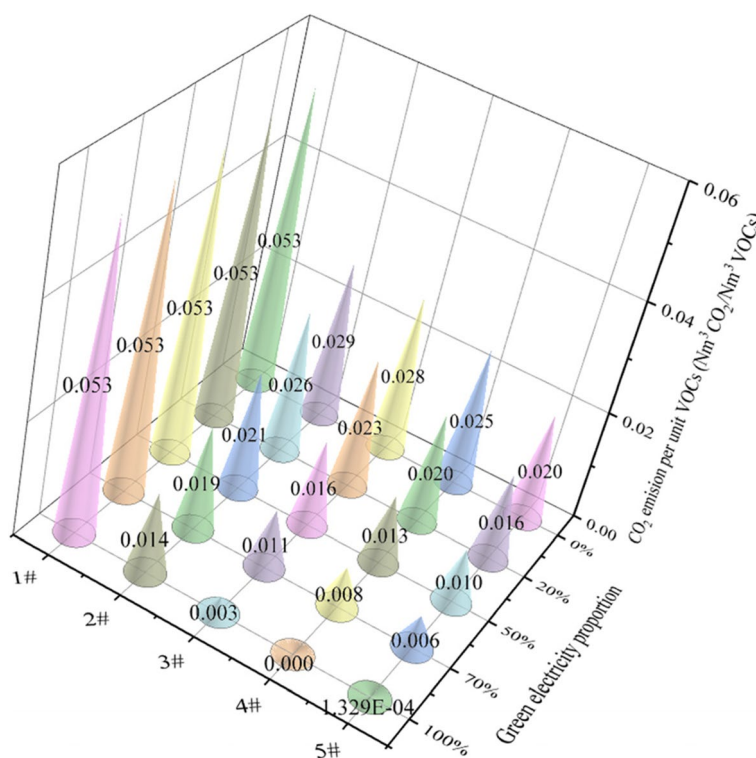
#### 4 The influence of green electricity ratio on CO<sub>2</sub> emissions

Considering current electricity supply contain a mixed source of grey electricity from the fossil fuel and green electricity from renewable energy, it is essential to evaluate the possibility of the reduction of indirect emission with the change of electricity ratio. When the green/grey electricity ratio changes, CH<sub>4</sub>, electric energy, and air consumption by the different processes are stable (Fig. 4), while CO<sub>2</sub> emission changes (Fig. 6). The CO<sub>2</sub> emission of direct combustion is fixed since it needs no electricity. However, if 100% green electricity is used, the carbon reduction effect of the catalytic oxidation process is obvious. Its CO<sub>2</sub> emission of grey electricity will be reduced by 49.9%, and its total CO<sub>2</sub> emission is only 27.0% of the

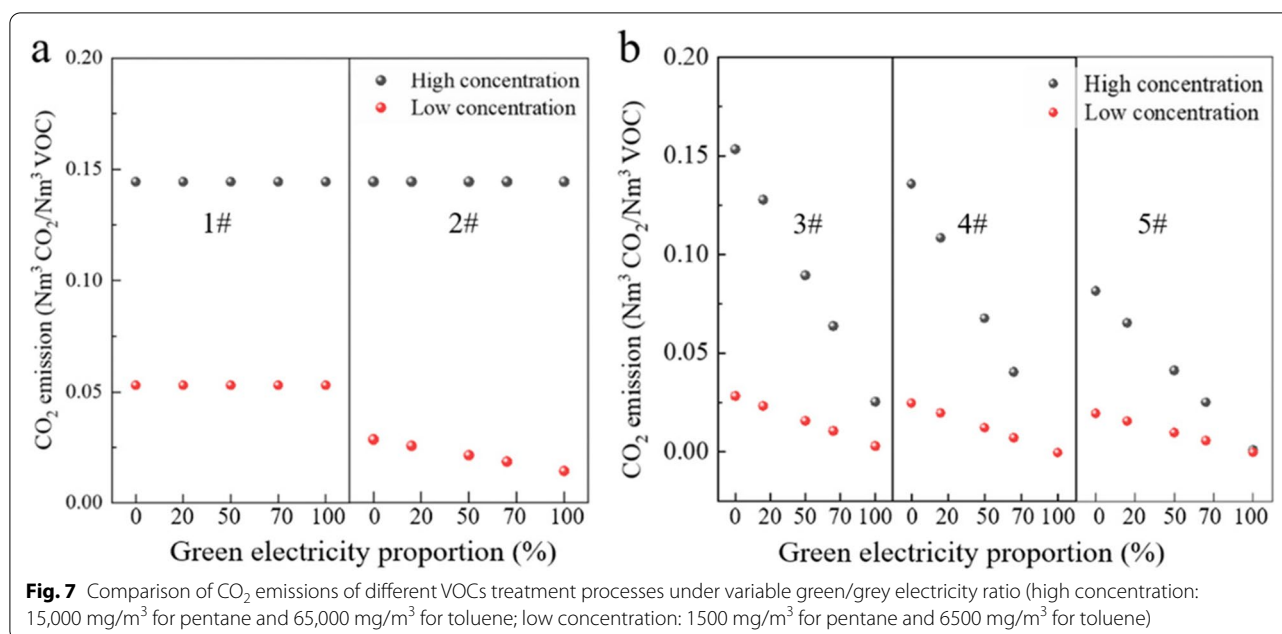
direct combustion process. Also, the increase of green electricity ratio will lead to a significant reduction of CO<sub>2</sub> emission in the adsorption and desorption processes (Figure S13) in the adsorption-desorption-combustion method, and CO<sub>2</sub> emission exists only in the combustion process when green electricity ratio is 100%.

At the same time, the advantages of green electricity are also fully reflected in the process of adsorption-desorption-carbon fixation in which CO<sub>2</sub> is mostly emitted indirectly. The CO<sub>2</sub> emissions of adsorption, desorption and carbon fixation all tend to be zero when green electricity is used, and the CH<sub>4</sub> and H<sub>2</sub> produced by this process can be used as raw materials for other chemical process (Figure S14).

In addition, if further increasing the initial concentration of VOCs (for example 15000 mg/m<sup>3</sup> for pentane and 65,000 mg/m<sup>3</sup> for toluene), which represents other tough-treated VOCs, it is able to obtain the associated CO<sub>2</sub> emission data (Fig. 7). The CO<sub>2</sub> emission trends in the processes of direct combustion and catalytic oxidation are similar. The CO<sub>2</sub> emission ratio of high and low concentration is almost steady when direct CO<sub>2</sub> emission is dominate (Fig. 7a). In comparison, when indirect CO<sub>2</sub> emission is dominate, their CO<sub>2</sub> output of high and low



**Fig. 6** Comparison of CO<sub>2</sub> emissions from different VOCs treatment processes under variable green/grey electricity ratio (initial concentration: 1500 mg/m<sup>3</sup> for pentane and 6500 mg/m<sup>3</sup> for toluene)



concentration decline sharply with the increase of green electricity (Fig. 7b).

It is indicated that whether a process has green potential is the key to CO<sub>2</sub> emission reduction in the future. The technology route of adsorption-desorption-carbon fixation which resulted in zero total CO<sub>2</sub> emissions is extremely attractive.

From the discussion above, it is clear that a near carbon-zero loop of adsorption-desorption-carbon fixation depends directly on the supply of green electricity. The adsorption-desorption-carbon fixation-supply heat loop would have flexibility when green electricity is in short or unstable supply.

Note that CNTs based adsorbent exhibited long life time. The amount of as-produced CNTs in carbon fixation section for long time operation would far exceed the amount of CNTs used in adsorption section. The situation would result in the significant reduction of the CNTs cost, which, in turn, stimulate the expanded application of CNT-based adsorbent in treatment of VOCs, waste water, and other fields.

## 5 Conclusions and prospects

The adsorption-desorption-carbon fixation process has great greening potential to effectively reduce the total CO<sub>2</sub> emission as well as produce valuable by-products. The CNT products generated in the carbon fixation process can be used as adsorbent raw materials in the adsorption process, and the carbon element closed loop chain is formed with the CNTs based adsorbent as the bridge. By further using CH<sub>4</sub> and H<sub>2</sub> as the heat supplier,

the CO<sub>2</sub> emission could be further reduced. As the proportion of green electricity increases, the process of adsorption-desorption-carbon fixation process shows its advantage on carbon emission reduction and becomes a green cycle. Obviously, the concept of adsorption-desorption-carbon fixation process is also applicable to the green and low carbonization treatment of organic wastewater. The trend is to develop some more efficient catalysts which can convert all VOCs to CNTs and hydrogen without methane. This technology embodies the development of the cleanest process and has a bright future. In the context of global carbon emission reduction and China's "dual carbon" era, this process will provide technical support for the reduction of carbon tax/carbon emission trading quota, and further delay the arrival of a negative carbon economy [35].

### Abbreviations

VOCs: Volatile organic compounds; CVD: Chemical vapor deposition; CNTs: Carbon nanotubes; CCUS: Carbon capture, utilization and storage; SEM: Scanning electron microscopy; TEM: Transmission electron microscope.

### Supplementary Information

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Additional file 1.

### Authors' contributions

ZFY: Writing—original draft, Writing—review & editing, Investigation, Formal analysis, Data curation. CJC: Project administration, Funding acquisition, Writing—review & editing, Conceptualization, Investigation. XY: Investigation, Formal analysis. WHZ, DXL and YZ: Project administration, Resources,

Investigation. KL: Resources, Investigation. WZQ: Supervision, Project administration, Writing—review & editing, Conceptualization, Investigation. The author(s) read and approved the final manuscript.

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#### Availability of data and materials

The data that support the findings of this study are available from the corresponding author upon reasonable request.

#### Declarations

#### Ethics approval and consent to participate

The research does not require ethical approval.

#### Consent for publication

We agree to publish the work.

#### Competing interests

The authors declare no conflict of interest.

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