

N-situ growing of platinum catalyst on activated carbon for formaldehyde purification at room temperature

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Abstract. The activated carbon (AC) particles with different microstructures were used to support platinum active component by in-situ growing method, so as to prepare a series of Pt/AC structured catalysts. Compared to pure AC samples, the as-prepared Pt/AC catalysts do not have the limit of saturated adsorption capacity in the field of formaldehyde (HCHO) purification, and can be used for many times to eliminate HCHO pollutants at room temperature. Moreover, all the Pt/AC catalysts can maintain a high HCHO conversion of 75-85% in the multi-use test, indicating that the HCHO purification mechanism on the catalysts is a synergistic function of physical adsorption and catalytic oxidation. Furthermore, the HCHO oxidation activities of Pt/AC catalysts are also influenced by the microstructures of AC substrates, such as BET surface area, mesopore content and BJH pore volume.

1. Introduction

Formaldehyde (HCHO) is one of the major indoor pollutants, which usually emits from decorative and building materials [1]. Due to its harmful effects on human health, it is necessary to find suitable method to reduce the HCHO concentration at room temperature in doors. Activated carbon (AC) are widely used in the field of air purification because of its large specific surface area and good adsorption capacity [2]. However, since the AC sample purifies air pollutants (such as HCHO) just by physical adsorption, it will lose purification ability when the pollutant amount is over its saturated adsorption capacity.

Currently, catalytic oxidation is considered as one of the most effective and economical feasible methods to eliminate air pollutants (such as volatile organic compounds, VOCs) [3]. Especially the noble metal catalysts exhibit higher catalytic oxidation activity for air pollutants under lower reaction temperature [4, 5]. And the catalysts can be used for infinitely many times in theory. In this study, the AC was used as the substrate to support the active component of Pt to prepare high-performance oxidation catalysts by in-situ growing method. And the as-prepared catalysts exhibit good HCHO purification performance at room temperature.



2. Experimental

The AC sample was pretreated by using a nitric acid solution. Then a chloroplatinic acid solution with certain concentration and pH value of 8-9 was used as platinum precursor to support on the AC sample by wetness impregnation. After that, the sodium borohydride solution was added dropwise to in-situ reduce the platinum precursor to Pt metal. Finally, the obtained catalyst sample was dried at 100°C for 3 h, and was denoted as Pt/ AC.

The AC sample's structure was analyzed by Nitrogen adsorption-desorption isotherms, Brunner-Emmet-Teller (BET) measurements and Barrett-Joyner-Halenda (BJH) measurements (Micromeritics ASAP 2020 HD88), and scanning electron microscopy (SEM, Hitachi SU8010) with energy dispersive spectrometer (EDS).

The Pt/AC catalyst particles were filled in a $7 \times 7 \times 0.5$ cm PP grid, and then was placed into a closed formaldehyde simulation box ($0.7 \times 0.7 \times 0.5$ m, about 245 L) to test. The gas in the box was sampled using a TYQ-1000 sampler, and the HCHO concentration was determined by the phenol reagent method according to GB/T 18204.2-2014. The calculated HCHO conversions were used to evaluate HCHO purification activity on the catalysts.

3. Results and Discussion

Three AC samples were chosen from different manufactures. Among them, C3-002 sample is obtained after surface modification treatment, but C1-001 and C4-003 samples are not. From the results of EDX analysis, the three AC samples are proved to have only C element. And the surface morphologies of the three AC samples are shown in Fig. 1. From Fig. 1 a and c, it can be observed that the surfaces of C1-001 and C4-003 are all relatively smooth, but C4-003 are composed of smaller particles. The result indicates that C4-003 might have more mesopores and higher surface area in contrast to C1-001. It can also be seen from Fig. 1b that the C3-002 has a fuzzy thin layer on the outmost surface because of its surface modification treatment, which can effectively increase its surface area and mesopores content.

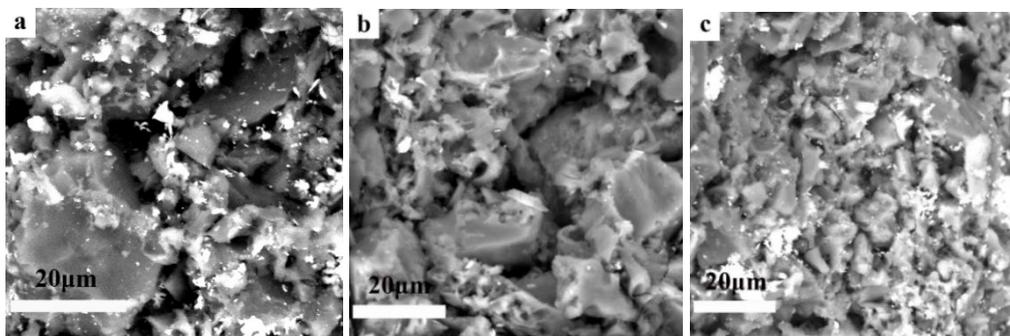


Figure 1. SEM images of (a) C1-001, (b) C3-002 and (c) C4-003.

The surface area and pore structure of the AC samples were analysed by N₂ adsorption-desorption measurements, as shown in Fig. 2. It is identified from the figure that the isotherms of three AC samples were all type I, indicating that the adsorption process for the samples were dominated by the single-layer adsorption mode. The specific texture parameters of AC samples are summarized in Table 1. According to Fig. 2 and Table 1, it can be found that the three AC samples are all composed with micropores and mesopores, but the BET surface area, mesopore content and BJH mesopore volume all follow the order of C1-001 < C4-003 < C3-002. This behaviour can be addressed by the surface morphology difference for the three AC samples, which is consistent with the foregoing SEM results in Fig. 1.

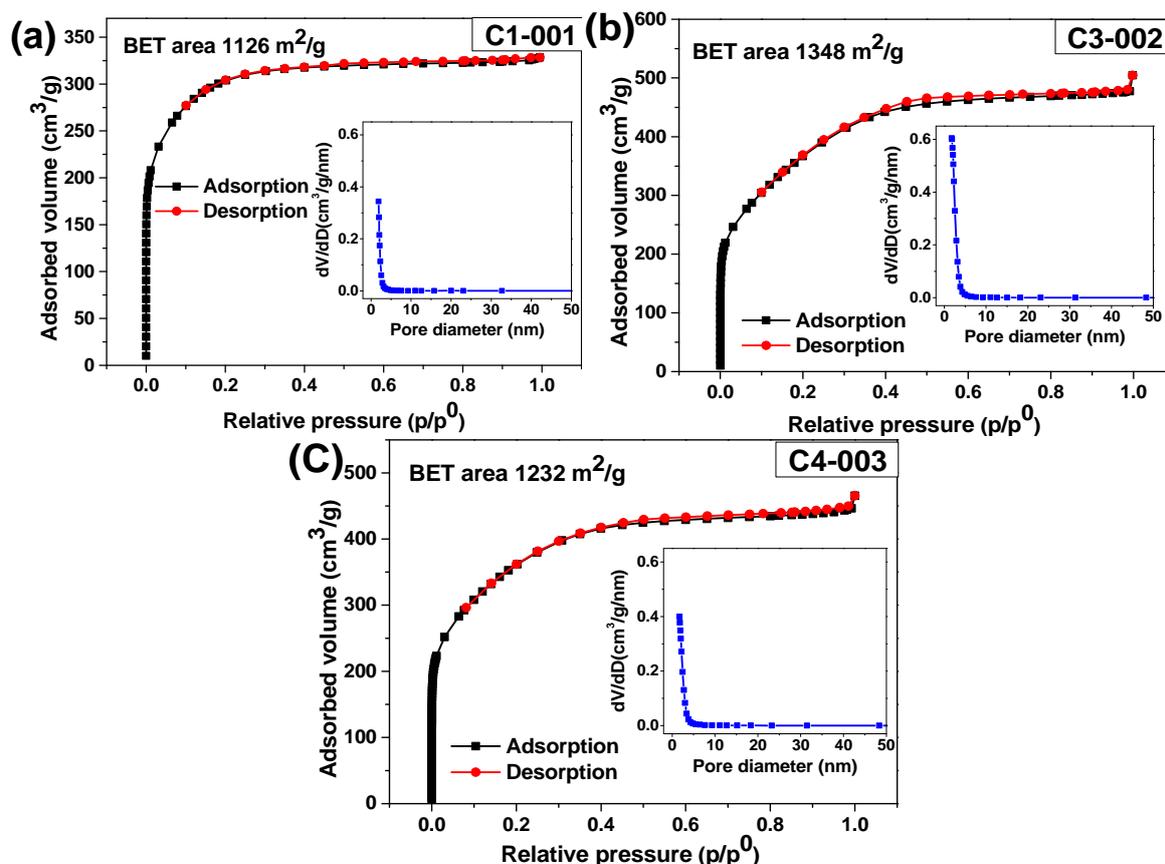


Figure 2. Nitrogen adsorption–desorption isotherms and corresponding mesopore size distribution curves (inset) of (a) C1-001, (b) C3-002 and (c) C4-003.

Table 1. Textural parameters of the three AC samples.

Sample	BET (m ² /g)	BJH surface area (m ² /g)	Mesopore content (%)	BJH pore volume (cm ³ /g)
C1-001	1126	300	26.6	0.180
C4-003	1232	675	54.8	0.437
C3-002	1348	1096	81.3	0.711

The HCHO purification test for the AC sample of C1-001 is shown in Fig. 3a. It can be found that the HCHO conversion of pure C1-001 sample was as high as 60% in the 1st-use test, but greatly decreased to 18% in the 2nd-use test. And the other AC samples of C3-002 and C4-003 exhibited similar HCHO purification performance. Such behaviors suggest that the HCHO purification activity for pure AC samples just depends on their physical adsorption characteristics. That is to say, the AC sample will lose the HCHO purification ability when the HCHO amount is over its saturated adsorption capacity. So the pure AC samples cannot be used multiple times.

The three AC samples were used to support Pt active component to prepare Pt/AC catalysts by in-situ growing method. The HCHO purification activity on the as-prepared Pt/AC catalysts are presented in Fig. 3(b-d). Compared to pure AC sample, the three Pt/AC catalysts can all maintain the high HCHO conversion of 75%-85% in the multi-use test, indicating that the HCHO purification mechanism for the as-synthesized Pt/AC catalysts is a synergistic function of physical adsorption and catalytic oxidation. That is to say, the as-prepared Pt/AC catalysts can be used forever theoretically. Moreover, compared to Pt/C1-001 catalyst, the Pt/C3-002 and Pt/C4-003 catalysts exhibit superior HCHO purification

performance ---- higher and more stable HCHO conversion for multi-use test. This can be addressed by the different microstructures of AC supporters in the three Pt/AC catalysts. Because C3-002 and C4-003 have higher BET surface area, mesopore content and BJH pore volume in contrast to C1-001, they will be not only more favourable for Pt active component uniformly dispersed on the surface of AC supporters, but also more favour for reactants entering into mesopore channel to extend contact time with Pt active sites, leading to better HCHO purification activity.

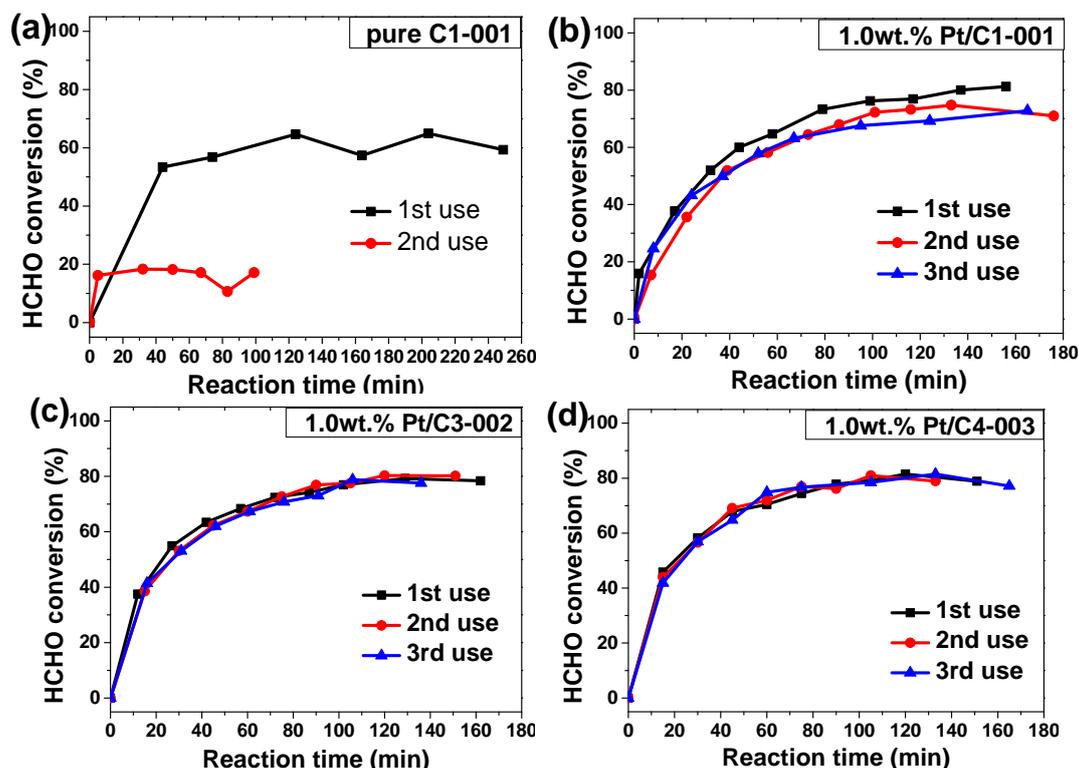


Figure 3. HCHO purification activity on (a) pure C1-001 sample, (b) Pt/C1-001, (c) Pt/C3-002 and (d) Pt/C4-003 catalysts for multi-use test at room temperature.

4. Conclusion

The as-prepared Pt/AC catalysts exhibit good HCHO purification performance at room temperature. The HCHO conversion on the three Pt/AC catalysts can always maintain as high as 75-85% in the multi-use test. That is to say, the as-prepared Pt/AC catalysts can be used to effectively eliminate HCHO pollutant for infinitely many times theoretically. Moreover, the microstructures of AC substrates have effect on the HCHO purification activity of Pt/AC catalysts. Higher BET surface area, mesopore content and BJH pore volume for AC substrate are favorable for as-prepared Pt/AC catalysts to obtain better HCHO purification activity at room temperature.

Acknowledgements

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