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Supplementary Information

CNT-enhanced Amino-functional Graphene Aerogel Adsorbent for Highly Efficient Removal of Formaldehyde

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1. Adsorption calculation

The breakthrough time is defined as the period between the decrease in formaldehyde concentration after passage through the adsorption column and the increase in formaldehyde concentration to the safety threshold according to the WHO guidelines (0.08 ppm). The adsorption experiment is concluded when the output concentration (C) of formaldehyde reaches 80% of the input value (C_0). Another critical parameter for judging the adsorption performance of adsorbents is adsorptive capacity (W). The amount of pollutants adsorbed was calculated according to the following equation:

$$W = \frac{P \times M}{R \times T} \times L \times t \times 10^6 \quad (1)$$

P is the atmospheric pressure (Pa).

M is the molecular weight of formaldehyde (30.01 g·mol^{-1}).

R is the ideal gas constant (8.314 J.K^{-1}·mol^{-1}).

T is the temperature (K).

L is the volume flow rate of nitrogen (L·min^{-1}).

t is the breakthrough time of the adsorbent sample (min·g^{-1}).

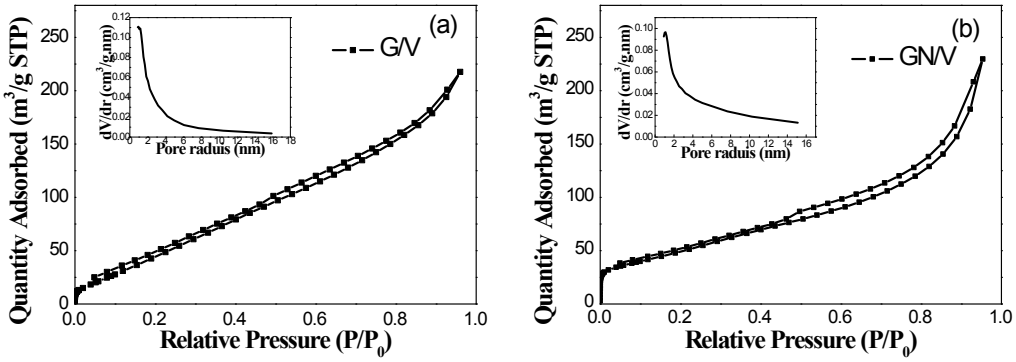

Fig. S1 The N_2 adsorption-desorption isotherm and pore volume distribution of G/V (a) and GN/V (b)

The N_2 adsorption-desorption curve and the pore size distribution (PSD) analysis of CNT-enhanced graphene aerogel is shown in Fig. S1. The N_2 adsorption-desorption curves are consistent with the type IV adsorption curve, thus indicating that the pore structure of the adsorbents is mainly mesoporous. The distribution of the pore size illustrates that the materials contain micropores because the population is concentrated below ~2 nm, in addition to larger sized pores.

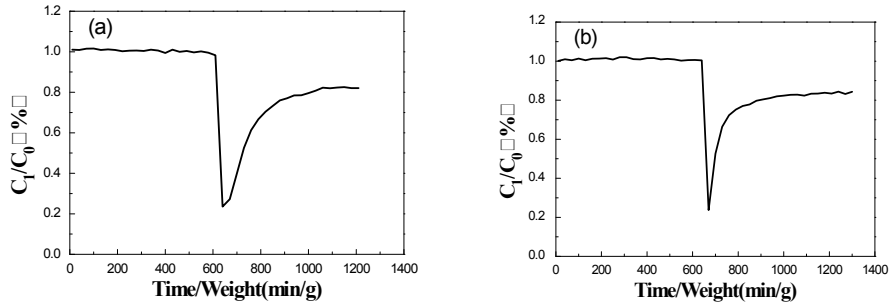

Fig. S2 Breakthrough curves of formaldehyde adsorption for G/V (a), and GN/V (b).

Fig. S2 shows that G/V and GN/V have a weak effect on the adsorption of formaldehyde. The initial concentration of formaldehyde C_0 was stable at 3.7 ppm and underwent rapid decrease after the adsorbents were embedded. Thus, the three adsorbents adsorb formaldehyde, although without a specific breakthrough time. The composites of CNTs and graphene have a more amendatory structure than that of pure CNTs or graphene aerogels. In contrast, the adsorption of formaldehyde by GN/V still depends on the VDW forces between the aerogel structure and formaldehyde molecules, which is a process of physical adsorption and has a limited adsorption capacity.

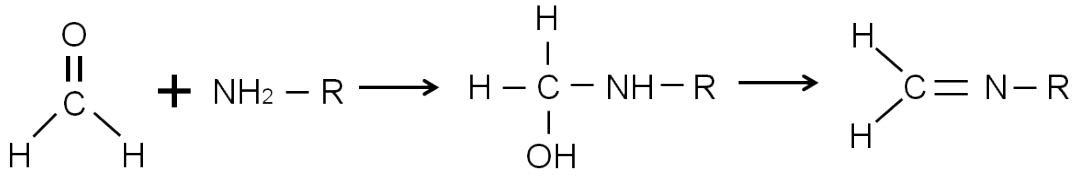

Fig. S3 The reaction equation of formaldehyde and amid

Amino-functional graphene aerogel and CNT-enhanced amino-functional graphene aerogel were prepared by using EDA as a reducing agent. During the reaction process, the amino groups (-NH2) were modified on the adsorbent and contributed to the removal of formaldehyde by reacting with the carbonyl group in formaldehyde. The reaction between –NH2 and formaldehyde is shown in Fig. S3.

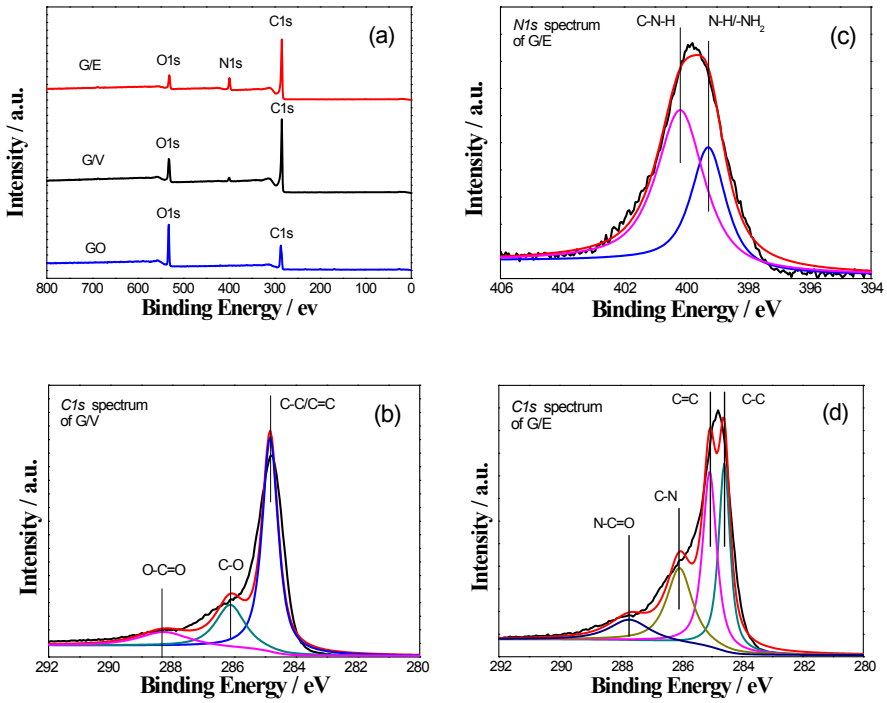

Fig. S4 The XPS spectrums of graphene aerogels

The comparison among the general XPS spectra of G/V, G/E and GO is shown in Fig S4. A detailed analysis of N1s in G/E and C1s in G/V and G/E is shown to illustrate the amino modification of the graphene aerogels.

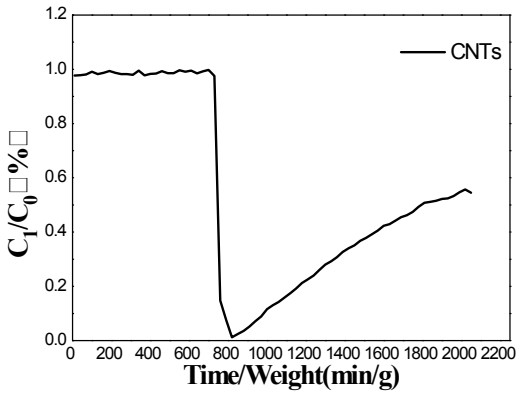

Fig. S5 The breakthrough curve of formaldehyde adsorption for CNTs

In this study of formaldehyde removal, graphene aerogels modified with CNT addition were prepared. Fig. S5 shows the adsorption breakthrough curve of CNTs under a formaldehyde concentration of 3.7 ppm and that the adsorption capacity of CNTs is relatively weak. It is concluded that the improvement of the adsorption capacity of the CNT-enhanced amino-functional graphene aerogel (GN/E), compared with amino-functional graphene aerogel (G/E), is due to the development of the microstructure by CNT modification.