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Synthesis of Si/O/C/N quaternary composite aerogels with micro/mesoporous structures and their selective adsorption property for volatile carbonyl compounds in cigarette smoke

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ABSTRACT

A novel micro/mesoporous Si/O/C/N quaternary composite aerogels were prepared for the first time and possessed efficient synergistic effect of physisorption and chemisorption. In the preparation process, a simple sol-gel method was applied to integrating nitrogen rich chitosan sol and tetraethoxysilane (TEOS) sol skillfully. The 2 mm-thick Si/O/C/N composite adsorption layer was sandwiched into the acetate filter rod of control sample to form a test sample with high removal efficiency for volatile carbonyl compounds, especially for crotonaldehyde (75.9%) in mainstream cigarette smoke (MCS). The micro/mesoporous Si/O/C/N quaternary composite aerogels have brilliant prospect in the application of cigarette smoke filtration, water purification, etc.

1. Introduction

Aerogels are of immense importance in various applications such as adsorption, sensing and catalysis owing to their high surface area, low porosity, small poresize and adjustable framework [1]. Further, silica aerogels are open foam type materials and become quite popular because of a number of excellent physichemical properties such as high specific surface area (>500 m^2/g), high porosity (>90%), low bulk density (<0.3 g/cm^3), low thermal conductivity (~0.005 W/m K), ultra low dielectric constant (k = 1.0–2.0) and low index of refraction (~1.05) [2–5]. Due to all of those unsual characteristics, silica aerogels are used in several technological application including thermal insulation materials [6,7], acoustic barrier materials [8], battery electrodes [9], catalyst supports [10], oxygen and humidity sensor [11] and adsorbent [12]. Usages of aerogels as adsorbent have become more widespread because of its specifications such as high porosity and specific surface area. Silica aerogels show excellent selective adsorption for BTEX (Benzene, Toluene, Ethyl benzene, Xylene) [13], volatile organic compounds (VOCs) from polluted gas stream [14], toxic organic compounds, oil and heavy metal from water [15–18]. However, many technical applications of silica aerogels have been restricted because of their extreme fragility, poor mechanical properties, weak high temperature stability, single functionality and poor selectivity [19]. Generally, how to overcome the shortcoming of silica aerogels in specific application and how to combine all the advantages of silica aerogels in complex application are the focus of current research.

Traditional optimization methods for silica aerogels are mostly the composition of binary and ternary composites, including the addition of nanoparticles [20], polymers, fibers [21,22], and carbon nanomaterials include carbon nanotubes [23,24], carbon fibers [25,26], graphene [27,28] and carbon aerogels [29–31]. These methods could get desired functional properties and improve the strength of the aerogel [32,33]. Furthermore, hierarchically structured carbon-silica aerogel composites have high affinity towards aromatic molecules and fast adsorption kinetics, excellent performance of dynamic adsorption capacity and high mass transfer efficiency arising from the well-developed microporosity as well as open foam mesostructure in the silica-carbon composites [34]. Silica aerogel-granulated activated carbon is superior in terms of removing uranium from a stock solution in comparison with activated carbon alone [29]. The combination of micropores and mesopores can also improve the adsorption efficiency of materials. Moreover, the introduction of functional groups in the surface of sample can greatly

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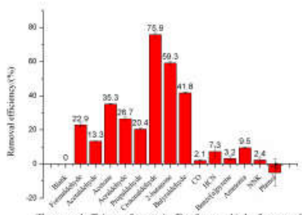
ABSTRACT BODY:

Abstract: Many kinds of compounds in cigarette smoke can effect human health，and some may be carcinogenic or mutagenic to humans. The adsorb materials in cigarette filters are important for the reduction of the harmful compounds in cigarette smoke. In this study, we present a functional composite aerogels, a series of N-C-SiO_2 composite aerogels with different nitrogen content are prepared via one step sol-gel technique. The porosity of N-C-SiO_2 composite aerogels depends on the content of chitosan and the calcining procedure. Nitrogen atoms are successfully introduced into silica aerogels through carbon atoms and the N-C-SiO_2 composite aerogels possess higher specific surface area (681 m^2/g). We sandwich the composite aerogels in the acetate filter rod of control samples to get a sandwich structure filter and the control samples named Hong Shuang Xi are purchased from market. The cigarettes with composite filters and control samples are smoked in a smoking machine, and the mainstream smoke is collected and analyzed. Seven kinds of compounds include carbonyls (formaldehyde, acetaldehyde, acetone, propaldehyde, crotonaldehyde, 2-butanone, butyraldehyde), CO, HCN，benzo[a]pyene, ammonia, NNK and phenol examined in our study. The removal efficiency of composite filter for seven kinds of compounds in cigarette smoke is calculated as a formula (Removal efficiency = $(V_{sample} – V_{control}) / V_{control}$). Totally, the composite filter shows high removal efficiency for seven kinds of compounds, especially for carbonyls and the removal efficiency for crotonaldehyde is 75.9% . Sensory evaluation test shows the aroma of the sample with composite filter is smoother and softer than the control cigarette. The N-C-SiO_2 composite aerogels have a good application prospect in cigarette smoke filtration.



The removal efficiency of composite filter for seven kinds of compounds in cigarette smoke. (Removal efficiency = $(V_{sample} - V_{control}) / V_{control}$)

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