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TiO_2-SiO_2 nanocomposite aerogel loaded in melamine-impregnated paper for multi-functionalization: Formaldehyde degradation and smoke suppression

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HIGHLIGHTS

- Multi-functions of formaldehyde degradation and smoke suppression were achieved.
- Both high gaseous and liquid formaldehyde degradation rates (56.6% and 65.8%) were achieved.
- The coated melamine-impregnated paper is easy to realize industrial production.

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ABSTRACT

A raw decorative paper was coated with a mixture of TiO_2-SiO_2 nanocomposite aerogel and melamine-formaldehyde resin to achieve dual functions, namely formaldehyde degradation and smoke suppression. The physical and chemical structure of TiO_2-SiO_2 nanocomposite aerogel and its photocatalytic activity on liquid formaldehyde degradation were revealed. Moreover, the coated melamine-impregnated paper was further decorated into the surface of a poplar wood veneer to prepare a real indoor panel. A self-designed sealed box and cone calorimeter were used to test its photocatalytic activity on gaseous formaldehyde degradation and smoke suppression. The results showed that the presence of massive mesopores in TiO_2-SiO_2 nanocomposite aerogel and the appearance of anatase nanocrystal of TiO_2 synergistically lead to both the high liquid and gaseous formaldehyde degradation rates of 65.8% and 56.6%. In addition, the porous structure and incombustibility of TiO_2-SiO_2 nanocomposite aerogel also result in high smoke suppression, demonstrating the maximum decrease in CO and CO_2 production by 56.2% and 33.2%. The preparation process of the coated melamine-impregnated paper is easy to realize industrial production. After decorated into the surface of wood-based panels, it can be used as the indoor materials of wall, floor, and furniture.

1. Introduction

Melamine-impregnated paper is widely applied to decorate the surface of wood-based panels such as particleboard, fiberboard, and plywood. It can improve the dimensional stability of wood-based panels via resisting naturally absorption of humidity [1]. Physical and mechanical properties can be also enhanced especially on the modulus of elasticity [2]. Moreover, the panel surface coated with multi-layer of melamine-impregnated paper results in the significant reduction in formaldehyde emissions, since the paper provides a physical isolation layer to prevent the emission of gaseous formaldehyde from panel itself [3]. However, melamine-impregnated paper is easy to be damaged during the process of application, and thus the gaseous formaldehyde generated from wood-based panels would be released into external environment. In addition, melamine-impregnated paper has no

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effect on the degradation of gaseous formaldehyde from external environment.

During the last decade, TiO_2 was doped into the nano-SiO_2 structure in order to synthesize TiO_2-SiO_2 nanocomposite aerogel. Its photocatalytic activity is significantly enhanced owing to the improvement on specific surface area for TiO_2. Early studies have demonstrated the high efficiency of TiO_2-SiO_2 nanocomposite aerogel on the decomposition of hazardous chemicals, especially for liquid phase [9–11]. Yoda has found that TiO_2-SiO_2 composite aerogel can lead to the effective decomposition of benzene and the decomposition degree mainly depends on the crystallinity of TiO_2 [4]. Deng has prepared the TiO_2-SiO_2 composite aerogel via the sol-gel method and reported that phenol decomposition rate of 34% was achieved [5]. Anderson has studied the synergistical effect between TiO_2 and SiO_2 during the photocatalytic decomposition of organic compounds, which revealed that the particle size of SiO_2 and its distribution on composite structure have a significant effect on the photocatalytic activity of TiO_2 [6]. In addition, Su has proved that the formation of network structure based on cross-linked Si-O-Ti bond and oxygen vacancies in TiO_2 has a synergetic effect on both phases [7]. The photocatalytic decomposition effect of TiO_2-SiO_2 composite aerogel on organic chemicals of indoor air pollutants becomes the recent research topic, since the most part of furniture and indoor decorating materials will release hazardous organic chemicals, especially gaseous formaldehyde [12,13]. Suligoj has applied the TiO_2-SiO_2 films to decompose gaseous formaldehyde and the decomposition rate reached 91% [8]. Therefore, melamine-impregnated paper coated with TiO_2-SiO_2 nanocomposite aerogel can be applied in gaseous formaldehyde degradation, since high specific surface area and porous structure lead to high adsorption capacity which can block the emission of gaseous formaldehyde from the wood-based panel itself. In addition, its high photocatalytic activity can also result in the effective degradation on gaseous formaldehyde in indoor air. The effect of TiO_2-SiO_2 nanocomposite aerogel on liquid formaldehyde degradation is also worth studying since it can reveal its degradation mechanism comprehensively.

The wood-based panel is easy to burn under a fire accident. Its combustion would also release the toxic fumes including carbon monoxide, nitrogen oxide, and char power which can result in the death of human life. Many methods were applied in the wood-based panel to achieve high smoke suppression. Especially, previous studies have reported the good smoke suppression of the wood composite coated with SiO_2 or TiO_2, respectively [14,15]. Mahr has found that single and double layered sol-gel derived TiO_2 and SiO_2-wood composites have excellent smoke suppression [16]. The incombustibility of SiO_2 and TiO_2 can form the stable and closed barrier layer during burning. Especially, the porous structure of SiO_2 can adsorb the hazardous volatiles generated from wood combustion and thus reduce the emission of smoke. Therefore, it can be expected that a raw decorative paper coated with the mixture of TiO_2-SiO_2 nanocomposite aerogel and melamine-formaldehyde resin can achieve high smoke suppression when it is used to decorate the surface of inflammable wood-based panel.

In the present study, TiO_2-SiO_2 nanocomposite aerogel was prepared via a sol-gel method and characterized by a series of characterization methods to reveal its physical and chemical structure. It was further mixed with melamine-formaldehyde resin and coated in a decorative paper which was subsequently decorated into the surface of a poplar wood veneer in order to prepare a real indoor panel for achieving multi-functions of formaldehyde degradation and smoke suppression. Such understanding is essential for the development of its utilization as the indoor materials of wall, floor, and furniture.

2. Experimental

2.1. Preparation of TiO_2-SiO_2 nanocomposite aerogel

All reagents and substances used in this study are of analytical grade and purchased from a local chemical factory in Nanjing city. In order to obtain TiO_2-SiO_2 nanocomposite aerogel with uniform structure, TiO_2 sol solution and SiO_2 sol solution were prepared, respectively. Acetic acid, distilled water, and 1/3 of absolute ethyl alcohol were mixed in order to prepare solution A. Meanwhile, tetrabutyl titanate and absolute ethyl alcohol were mixed to obtain solution B. Subsequently, solution A was added into solution B with a speed of 60 drops/min and stirred in order to obtain uniform sol solution. The total molar ratio of $n(Ti(OC_4H_9)_4)$: $n(C_2H_5OH)$: $n(H_2O)$: $n(CH_3COOH)$ was 1: 18: 3: 0.5. Similarly, SiO_2 sol solution was prepared under the total molar ratio of $n(Si(C_2H_5)_4)$: $n(C_2H_5OH)$: $n(H_2O)$: $n(CH_3COOH)$ = 1: 5: 2: 0.5. The as-prepared TiO_2 sol solution and SiO_2 sol solution were mixed and stirred under the molar ratio of n(Ti)/n(Si) = 4. Then, methanamide was added into the mixed solution with a molar ratio of $n(CH_3NO)/[(n(Ti) + n(Si))] = 0.25$ and subsequently dispersed using ultrasonic for 10 min. The mixture was subjected to magnetic stirring until the gel solution appears. Absolute ethyl alcohol was used to replace the water in as-prepared gel solution which was subsequently placed into the mixed solution (tetraethyl orthosilicate/absolute ethyl alcohol with a volume ratio of 2: 1). Absolute ethyl alcohol was used once again to remove the tetraethyl orthosilicate. Finally, the TiO_2-SiO_2 nanocomposite aerogel was successfully prepared after applying supercritical drying method and calcination for the appearance of anatase type TiO_2.

2.2. Characterization of TiO_2-SiO_2 nanocomposite aerogel

Fourier transform infrared spectroscopy (FT-IR) was carried out on the NEXUS 870 to study the chemical structure of as-prepared TiO_2-SiO_2 nanocomposite aerogel. The spectrum was recorded in the wavenumber region of 400–4000 cm^{-1} under a resolution of 4 cm^{-1}. Scanning electron microscope (SEM) was conducted on JSM-6360LV to observe the surface morphology of TiO_2-SiO_2 nanocomposite aerogel which was first coated with gold using a coating machine in order to decrease the charging effect. The surface morphology was further revealed by transmission electron microscope (TEM, Tecnai G220). X-ray diffraction (XRD) was performed on an Ultima-IV to investigate the crystalline structure of as-prepared TiO_2-SiO_2 nanocomposite aerogel. A Cu Kα radiation (40 kV, 200 mA) was applied to record the diffraction angle of 2θ in the range of 10–80° under a step rate of $0.02° \cdot s^{-1}$. The automated surface analyzer was applied on ASAP 2020 in order to investigate the Brunauer-Emmett-Teller (BET) surface area and pore distribution of TiO_2-SiO_2 nanocomposite aerogel. The N_2 adsorption/desorption isotherms were both recorded. The BET surface area and pore size distribution were calculated and determined by applying the BET standard method and Barrett-Joyner-Halenda (BJH) method, respectively.

2.3. Liquid and gaseous formaldehyde degradation

Aerogel was added into the melamine-formaldehyde resin under a desired mass ratio and dispersed by ultrasonic for 30 min. A raw decorative paper was subsequently dipped into the mixed solution for 2 min and removed out. Then, a glass rod was used to remove the excessive mixed solution on the paper surface. Finally, the loaded amounts of the resin and TiO_2-SiO_2 nanocomposite aerogels were determined by the difference between raw paper and the coated one. The coated melamine-impregnated