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Three-dimensional mullite fiber network with controllable hierarchical structure for effective soot combustion

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ABSTRACT

It has been proved that the secondary structure assembled on each fiber in the porous fiber network can catch the ultrafine particles easily in filtration process. In this work, three different networks without (naked fiber) or with different secondary structure (mullite whisker/mullite fiber or corundum plate/mullite fiber) were prepared to investigate the influence of secondary structure on the high temperature mechanical property and the soot oxidation. The mullite whisker/mullite fiber (MW/MF) network exhibited the best high temperature compress resistence and lowest soot oxidation temperature. The soot oxidation start temperatures were 434 °C and 443 °C in the mullite whisker/mullite fiber (MW/MF) and corundum plate/mullite fiber (CP/MF) webs respectively, which was lower than that in the Naked mullite fiber (MF) web (473 °C). Thus, fiber webs assembled with secondary structures have been shown to be promising filter with effective soot oxidation activity.

1. Introduction

The diesel engine is one of the most environmentally-friendly vehicle devices, because it emits less carbon dioxide and is more fuel efficient than the universal gasoline stoichiometric engine [1]. However, exhaust gases which emitted by these diesel engines own very high concentration of particulate materials and nitrogen oxides, especially ultrafine particles with smaller than 0.1 μm in diameter. The gases have a greater surface area, getting into lung tissue, the brain, and possibly the blood with toxic results [2], which is tremendously harmful to human health [3–6]. To control the emission of fine particulate matter from diesel engines and to meet the stated effluent standards, different types of diesel particulate filter (DPF) systems have been introduced [2].

The most widely studied PM controllers such as ceramic foam [7], wall-flow monolith [8,9] and ceramic fiber filter [10] can physically capture soot by their non-catalytic trapping structure as the exhaust gas passes through its porous wall. In order to consistently maintain the filter performance, the accumulated soot inside the filters should be burned off via passive or active regenerations [1,11]. An ambitious way for diesel particulate removal is by using filters carrying a suitable catalyst, which enables simultaneous soot filtration and combustion [12,13].

A number of filter supported catalysts have been reported, however the non-uniform dispersion of catalysts in the filters is the most serious defect. For example, the cordierite monolith containing catalysts can induce catalyst agglomeration [14] that would reduce the soot-catalyst contact surface area, which impacts largely negative on the catalytic activity [15]. As is known, contact between soot and catalyst particles is of great importance in soot oxidation [16]. To increase the number of the contact points between the catalyst and soot, catalyst with different topology and texture structures has been produced [17]. For example, catalyst nanofibers have been fabricated into the three-dimensional networks to increase soot-catalyst interactions [1,18]. However, their chemical, thermal and mechanical properties are not stable enough to withstand corrosion operating conditions at elevated temperatures [3,7].

Ceramic fibrous materials are the good choice to consider for DPFs because of their excellent corrosion resistance, high mechanical strength, high heat and creep resistance at high temperatures [19]. They are also inherently tougher than their porous monolithic counterparts, which makes them possible to design DPFs with high porosity and high fracture toughness [20].

Inspired by the inexpensive, environmentally friendly, and renewable character offered by natural plants, the useful biological resource (pollen structure) has been applied to synthesize the hybrid inorganic material with multiple scales and hierarchical

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structure which promote catalytic function by increasing the interaction between carbon and inorganic compound [21]. Moreover, in the bovine stomach, the papillary bulge on the surface of the stomach can promote the mixing of about food and the gastric juice. Herein inspired by the bovine stomach structure, the secondary structure (mullite whiskers or hexagon alumina sheets) synthesized by vapor-phase reaction [22,23] like the papillary bulge on the mullite fiber surface, which can not only improve the filtration efficiency of nano-particles, but also increase the number of soot-catalyst contact points.

$La_{1-x}Sr_xCoO_3$ [24,25] was coated on fiber paper by vacuum impregnation. (MF(naked mullite fiber web), CP/MF (corundum plate/mullite fiber web) and MW/MF (mullite whisker/mullite fiber web)) have been investigated to make clear the effects of novel mullite fiber network with hierarchical structure on the filtration efficiency, the dispersing of soot and catalyst and the efficient of the oxidation of soot.

2. Experimental

2.1. Chemicals and preparation of catalytic ceramic paper

The short polycrystalline mullite fibers (MF, Hongda Crystal Fiber Co., Ltd., Zhejiang, China) were approximately 5–10 μm in diameter and 50:1 to 60:1in length. The aluminum fluoride powder (AlF_3, analytically pure) and silica powder (SiO_2, analytically pure) were ball-milled using ethanol as medium (500r/min, 5 h) in a planetary ball milling machine to produce the W-active powder (which was used to fabricate mullite whisker during the reaction) in which the Al/Si molar ratio was 3/1. The C-active powder (used to fabricate corundum plate during the reaction) was the AlF_3 powder which was ball milled with ethanol (500r/min, 5 h) to make the particle smaller and then dried in an oven.

The fabrication process of the three different network-samples is illustrated in Fig. 1. In Fig. 1(a), the polycrystal mullite fiber (45 g) was mixed with silica sol solution (5 wt%, 400 ml) by stirring for 10 min, then poured into a Buchner funnel and filtrated by a vacuum pump. Finally the network was formed after calcination at 1200 °C, 2 h. In Fig. 1(b,c), the C-mullite and W-mullite active powders were dispersed respectively in water (400 ml) for uniform mixing. 9 ml poly diallyldimethylammonium chloride (PDADMAC used as cationic polyelectrolyte, Aladdin, China, 15 mg/ml) and 6 ml polyacrylamide (A-PAM used as anionic polyelectrolyte, Jiangtian Chemical Co., Tianjin, China, 7.5 mg/ml) was added to form the dual polyelectrolyte retention system to increase the binding force between the active powders and the mullite fibers as we discussed in our earlier study [26,27]. After pumping filtration, the MF, CP/MF and MW/MF samples were calcinated at 1200 °C, holding 2 h in airtight condition.

According to the stoichiometric ratios in the $La_{0.9}Sr_{0.1}CoO_3$ catalyst, 4.671 g of Lanthanum (III) nitrate hexahydrate ($La(NO_3)\cdot 6H_2O$, 99%), 0.254 g of strontium nitrate (II) ($Sr(NO_3)_2$, 99%), 2.989 g of cobaltous acetate tetrahydrate ($Co(CH_3COO)_2\cdot 4H_2O$, 99%) and 4.751 g of glucose (99%) were dissolved in 128 ml of distilled water to get the catalyst solution which was vigorously stirred at room temperature for 1 h as the earlier study reported [28]. To fabricated catalytic ceramic papers, the three different green networks were immersed in the catalytic solution for 30 min. To remove excess solution that had not penetrated into the webs, the sample was pump filtrated and dried at 80 °C for 5 h. Then they were calcinated at 800 °C for2 h at the speed of 5 °C min^{-1}.

2.2. Characterization

Phases of the samples before and after support catalysts were characterized by X-ray diffraction (XRD, Rigaku D/Max 2500). The microstructure of the three-dimensional webs and the catalyst distribution were analyzed using scanning electron microscope (SEM, Nanosem430 from FEI). The macropore size distributions were measured by Automatic Mercury Porosimeter (Micromeritics Autopore9500).

The filtration efficiency was calculated as follows:

$$\eta = \frac{Q_1 - Q_2}{Q_1}$$

where Q_1 and Q_2 were the inlet and outlet particle numbers in the air respectively and the gas flow was 28.3L/min. The number of the particles with diameter in 0.1–10.0 μm was counted by Laser Particle Counter (MetOne 2400). The apparent porosity of the samples was calculated by Archimedes method namely the water-immersion technique:

$$P = \frac{m_3 - m_1}{m_3 - m_2}$$

where m_1 was the weight of the dry sample, m_3 was the weight of the sample after the pores of the sample was full filled with water and m_2 was the weight of the sample under water. The compressive strength of the mullite fiber brick at room temperature and 800 °C (holding 2 h) were tested respectively by electronic universal testing machine (CSS-44001) according to the standardized testing method GB/T8489-2006.

2.3. Soot combustion measurements

In order to consider the soot oxidation activity of various catalytic materials, catalysts and soot were mechanically mixed in loose-contact (LC) and tight-contact (TC) modes for laboratory-scale studies [29]. However, in a practical situation, soot is continuously fed into the filter under loose contact conditions [30]. So to emulate the actual soot-catalytic contact condition, in our study the catalytic fiber webs were immersed in carbon black solution (PRINTEX-U,2000 ppm, ethanol as solvent) and were subsequently treated by an ultrasonic at 40 kHz for 10 min to help the carbon black disperse uniformly into pores of the webs without aggregation. To study the effect of hierarchical structures on the soot oxidation performance, differential scanning calorimetry (DSC) and thermal gravity analysis (TG) (NETZSCH STA 449F3) were performed in an air atmosphere (21%O_2 and 79N_2, 100 ml min^{-1}). The temperature range was 200–800 °C and the heating rate was 10 °C min^{-1}.

3. Results and discussion

3.1. Mechanical property of the networks at high temperature

An excellent diesel particulate filters are ones which have high filtration efficiency and also be able to withstand high temperature peaks associated with thermal regeneration, and during the thermal regeneration the accumulated soot is burned away [31]. As for the diesel engine, outlet gases temperature were usually 200–600 °C, and the transient temperature was almost reached 800 °C during the regeneration process [32]. The mechanical stability at elevated temperatures was the important requirement in their applications. In order to evaluate the mechanical stability of the filter at the high temperature, the mechanical behaviors of the three different networks were tested at room temperature and 800 °C (after exposing for 2 h).