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Original Research Paper

Effect of silane modification on CNTs/silica composites fabricated by a non-firing process to enhance interfacial property and dispersibility

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ABSTRACT

Carbon nanotube/ceramic composites have been in the spotlight thanks to their excellent properties. Sintering is the vital part of ceramics fabrication in terms of reliability, however sintering the carbon nanotube (CNT) based ceramic composites is a challenging task. In this study, interfacial bonding of silane functionalized CNT with silica ceramic is investigated by a non-firing sintering process. CNTs are first treated by a mixed acid with the aid of a silane 3-aminopropyl triethoxysilane (APTES), which improves the chemical bonding and dispersibility of CNT in ceramic bodies. The extent of APTES chemical functionalization and mechanical property of CNT/silica ceramic composites are characterized using Raman spectrometer, FT-IR analysis, X-ray photoelectron spectroscopy (XPS), scanning electron microscopy (SEM) and three-point bending strength measurement. Results show that composites are successfully prepared without sintering with stable CNT-silica interface, superior dispersibility, and good mechanical properties.

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1. Introduction

Carbon nanotubes (CNT) has emerged as a new kind of carbon based materials thanks to its superior structural, electrical, mechanical, and chemical properties [1] since its discovery in 1991 by Iijima [2]. Both theoretical and experimental results [3] show extremely high elastic modulus, 1 TPa, and high tensile strength, 200 GPa for CNTs thanks to which CNTs are considered as a potential material for being used for nanocomposites. However, most studies regarding CNT composites have been based on polymer-matrix materials for improvement of electrical conductivity, optical devices, and higher strength, and CNT reinforced ceramic matrix composites have been less studied. In-situ synthesis of carbon nanotubes to form carbon nanotubes/metal-oxide composites are reported [4] with a hot-press technique, however they lack good mechanical properties. Chang et al. [5] fabricated alumina matrix composites with 20 vol% of carbon nanotubes, which enhanced the fracture toughness up to 24% as compared to that of the alumina. In addition, hot-pressed CNT/SiC nanocomposite are prepared by mixing SiC particles and carbon nanotubes, leading to a 10% improvement in both strength and the fracture toughness as compared to monolithic SiC [6].

Dispersing CNTs throughout the matrix homogeneously is probably the most important part of preparing CNT based composites. In particular, when mechanical properties are required a good interfacial bonding across the CNT/matrix interface. In one approach, surfactants are used as an aid to enhance the dispersion of CNTs in epoxy resin, resulting in better mechanical properties [7]. Poly (vinylidene fluoride) is another approach to facilitate the dispersion of CNTs acting as a glue to increase the interfacial adhesion between CNT and poly (methyl methacrylate) [8].

CNTs usually tend to form bundles due to strong van der Waals forces, and separating them individually is still a challenging task. To obtain high-performance engineering ceramics with high reliability, it is of importance to prepare green compacts with high microstructural homogeneity. Some well-known approaches for preventing CNTs from making bundles are as follows. (I) surface modification on CNTs such as oxidation and deposition characterization [9,10]; (II) adding surfactant [11]; (III) in-situ synthesis [12]; (IV) applying silane coupling agents [13,14]. Among them, oxidation on CNT surface plays an important role in the modification of CNTs [15,16]. During modification, the hydroxyl and carboxyl groups that are introduced in the oxidation process can be used for further reacting with other chemical agents.

Sintering is the vital part of ceramics fabrication in terms of reliability of the final product because pore-free and high-strength materials can be obtained only by a proper sintering process.

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Low temperature sintering of ceramic materials has been long desired because high-temperature sintering is an energy-consuming and high-cost process, which can take several hours to several days in order to achieve a dense final product. Previously, our research group has proposed a facile “non-firing sintering” [17] process where no heating takes place for consolidation of the green compact. The main idea of non-firing sintering is to activate the surface of particles by ball milling and consolidate the particles via the joining of activated surfaces. By ball milling, the surface of particles is rubbed against balls, and the friction between particle/ball breaks the bonds of functional groups on the surface of particles. This method can be utilized for designing ceramic matrix composites, where heating the sample to higher temperatures causes problems like delamination or cracks due to the mismatch between fiber and ceramic.

In this study, a composite of SiO₂ (silica) nanoparticles and CNT is designed by a non-firing method. A silane coupling agent of 3-aminopropyl triethoxysilane (APTES) is also added because amino groups and the alkoxy groups on both sides of APTES molecule can bond to the silicon atoms and CNTs [18–20]. APTES functionalizing of CNTs not only improved the physical affinity of two materials but also enhance the dispersibility of CNTs and prevents agglomeration due to the high surface energy of CNTs.

2. Experimental details

2.1. Materials and preparation process

SiO₂ (Admatechs Co., Ltd) produced by Vaporized Metal Combustion Method with dozens of nanometer particle size was mechanically treated by ZrO₂ balls and vials (under the condition of 200 rpm, for 15 min). Polyhedral quartz powder (KS-100 s, F-plan) with the average surface area was also added for prevent the shrinkage during solidification. Multi-walled CNT (VGCF-H, Showa Denko) with an average fiber length of 10–20 μm and diameter of 150 nm was used as fillers. Silane coupling agent 3-aminopropyl triethoxysilane (APTES, obtained from Shin-Etsu Chemical Co., Ltd. KBE-930) and mixed acid H₂SO₄:HNO₃ = 3:1 (Kanto Chemical Co., Inc. and Wako Pure Chemical Industries, Ltd. Respectively) were used to modify the surface of MW-CNTs.

2 g of as-received MW-CNTs was acid-treated by 90 mL H₂SO₄ and 30 mL HNO₃. CNTs was added into the mixed acid and slightly stirred by a glass rod. Then, solution was slowly transferred into a three-necked flask in water bath for six hours at 80 °C. After water bath, the solution was filtered through distilled water several times until pH value reached neutral. The aqueous solution was dried at 60 °C overnight to obtain the acid-CNTs powder for further APTES functionalization. APTES was dropped into ethanol and stirred for 2 h at room temperature before adding pre-treated acid-CNTs into solution. The APTES and Acid-CNTs were sufficiently reacted with each other during string and finally dried to powder, APTES-CNTs. Amorphous SiO₂ was mechanically treated to obtain active sites on surface by planetary ball mill (Pulverisette.5, Frisch Japan Co. Ltd.), and then a moderate amount of quartz was mixed with activated SiO₂ to form the silica ceramic matrix material after 4 h tumbler ball milling with smaller zirconia balls (Φ50mm). After sufficient tumbler ball milling, silica powder was transferred into a plastic bottle for further mixing with APTES-CNTs. For ceramic composite fabrication, all the components were mixed together and the amount of CNT in controlled volume percentages gradually increased. Then 2.88 g of KOH solution with the concentration of 1 mol/L was added to the mixed powder for preparing the ceramic slurry by an electric mixer (ARV-930TWIN, THNKY), which could both mix and defoam slurry. Finally, obtained slurry was immediately poured into a mold keeping for 5 h at 80 °C. To investigate the

effect of the CNTs amount on the mechanical property of ceramic composite, different CNT concentrations from 0.25%, 0.5%, 0.75%, 1.0%, 4.0% and 6.0% in the blended silica powder were prepared.

2.2. Characterization

Raman spectra (NRS-3100, JASCO Corporation) was acquired with excitation wavelength of 523 nm to determine the phase compositions at room temperature. Fourier Transform Infrared Spectrometer (FT-IR) spectrum was recorded by JASCO Corporation FT/IR-6200 instrument with KBr discs. The X-ray photoelectron spectroscopy (XPS) of samples were obtained using the ESCALAB 250, Thermo Fisher Scientific, USA with the 150 W scanning X-ray source of monochromated Al Kα. Field emission scanning electron microscope (FE-SEM) micrographs were obtained at accelerating voltage of 10 kV by a JEOL JSM-7600F. Three-point bending strength was measured with an average of 5 samples by SHIMADZU AGS-G Series Universal/Tensile Tester with a constant pressure of 5 KN.

3. Results and discussion

Raman spectra of as-received raw CNT and H₂SO₄/HNO₃ treated CNT are shown in Fig. 1. Both samples consist of three typical bands, D-band (~1342 cm⁻¹), G-band (~1572 cm⁻¹) and 2D-band (~2684 cm⁻¹) [21]. D-band originates from the existence of amorphous carbon and disordered structure rather than seamless concentric tube structure formed by carbon atoms in the hexagonal arrangement. Defects of CNTs concentrate represent as vacancies of carbon atoms, heptagon instead of hexagon, and also kinds of functional groups modification on surface. G-band corresponding to the graphite structure of CNTs. The peak of 2D-band is the second order of the D-band peak, which is caused by the double resonant Raman scattering, involved by two-phonon emissions with opposite momentum.

In Fig. 1, it is obvious that intensity of the D-band is increased after acid treatment as compared with raw CNT. Corresponding ratio of D-band and G-band characteristic peak intensity ($R = I_D/I_G$) is also inset in Fig. 1, confirming a significant reduction of the R-value from 0.48 to 0.15. It seems that functional groups such as –COOH and –OH are introduced to CNT by acid treatment since sp³ hybrid carbon structure is affected, as well as introduction of defects on both walls and tips of CNT [22]. 2D-band revealed a

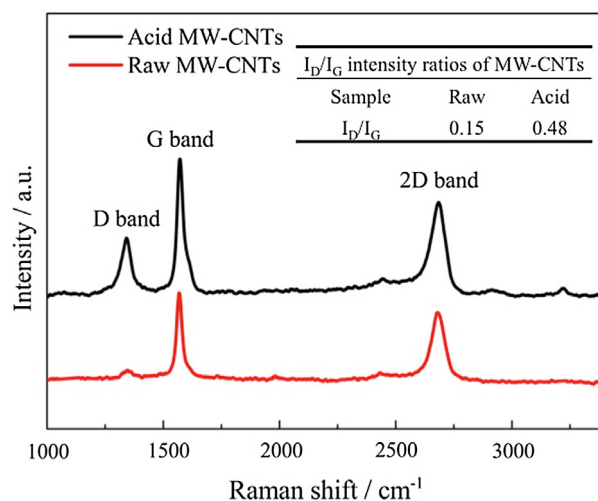


Fig. 1. Raman spectra and I_D/I_G intensity ratios of raw and acid treated MW-CNTs.

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