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Molecular level mixing: An approach for synthesis of homogenous hybrid ceramic nanocomposite powders

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ABSTRACT

An approach for the production of homogenous Al_2O_3–5SiC-1CNTs hybrid nanocomposite via molecular level mixing is reported. Aluminum nitrate nonahydrate, dimethylformamide, SiC_β nanoparticles, and functionalized CNTs were magnetically stirred and sonicated to prepare the desired slurry. Heating of the dried mixture in normal atmosphere at 400 °C for 4 h resulted in the formation of amorphous Al_2O_3 matrix, imbedded with CNTs and SiC nanoparticles, which crystallized during subsequent sintering. FT-IR was used to characterize bonds formed during powder synthesis. The uniform distribution of CNTs and SiC in the synthesized powder and consolidated sample was ascertained using x-ray mapping, FE-SEM, and TEM. Dense alumina and homogeneous Al_2O_3–5SiC-1CNT nanocomposite were consolidated by spark plasma sintering at 1500 °C for 10 min. The composite possessed improved fracture toughness (by ~33%) and slightly reduced hardness (by ~4%) with respect to monolithic Al_2O_3.

1. Introduction

Ceramic materials [1] have high elastic modulus and hardness compared to metals, which made them attractive for many applications. However, their use in structural applications is often limited because of their intrinsic low fracture toughness. Therefore, significant scientific effort has been directed towards the improvement of fracture toughness of ceramics through design of their microstructures [2]. This includes designing microstructures with elongated grains; incorporation of fibers or whiskers; and reinforcing with second phase particles at the microscopic and nanoscopic length scales to produce ceramic composites and nanocomposites, respectively. However, obtaining uniform distribution of the nanoreinforcement(s) in the matrix remains a key issue in developing inorganic composite materials with the desired properties [3]. Fortunately, the use of promising methods such as molecular-level mixing process [4] enabled researchers to uniformly disperse carbon nanotubes or graphene in metal [4,5,6] and ceramic [7,8] matrices.

Alumina [9] is one of the most used advanced ceramics for production of cutting tools [10], biomedical materials [11], and electrical insulators [12]. Addition of a single nanoscale reinforcement to alumina was reported to improve its physical [13–16] and mechanical [6,8,17] properties. The availability of nanoscale reinforcements with different morphologies such as SiC nanoparticles [18], carbon nanotubes [19], and graphene platelets [20,21] has facilitated the design of new hybrid alumina ceramic nanocomposites with tailored nanostructures [22–27]. The powders of these hybrid materials were mainly synthesized using sonication and ball milling followed by drying [22–27]; and consolidated using hot-pressing [22,25], uniaxial pressing followed by furnace sintering [23], and spark plasma sintering [24,26,27].

Sonication, ball milling, and drying were used to prepare Al_2O_3 containing 0, 0.1, 0.5 or 1.0 wt.% multi-walled carbon nanotubes (MWCNTs) with/without the addition of 25 wt.% SiC whisker [22]. The authors reported that the effects of MWCNTs addition on the mechanical properties of Al_2O_3 were trivial, whereas those of SiC were significant. They attributed the insignificant contribution of MWCNTs on the mechanical properties to the retarded sintering of Al_2O_3 by the MWCNTs and incomplete dispersion in the composites [22]. Homogeneous mixtures of Al_2O_3 with additions of 0.1 wt.% of MWCNTs, 0.1 wt.% SWCNTs, or with 0.05 wt.% MWCNTs + 0.05 wt.% SWCNTs were prepared by sonication, drying, and grinding [23]. A decrease in hardness and fracture toughness for the Al_2O_3 + 0.05 wt.% SWCNTs + 0.05 wt.% MWCNTs nanocomposite was reported. This was attributed to the poor and inhomogeneous dispersion of CNTs in the matrix, which impeded full densification. The authors observed inhibition of grain growth in Al_2O_3 − 0.1 wt.% MWCNTs composite due to pinning effect. The addition of 0.1 wt.% SWCNTs to Al_2O_3 led to grain growth because CNTs where residing within the grains, which contributed to higher mobility of grain boundaries [23].

Liu and co-workers [24] prepared alumina ceramic composites powders reinforced with graphene platelets (0.38 vol% GPL) and SiC nanoparticles (1, 3, and 5 vol% SiC) using wet dispersion, sonication,

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followed by ball milling, then drying and grinding. Yazdani and coworkers [25] reinforced Al_2O_3 by graphene nanoplatelets (GNPs) and carbon nanotubes (CNTs) using a combination of wet dispersion and probe sonication. Sonication and ball milling were used to disperse: 1 vol% of SiC nanoparticles and CNTs (0, 5, 7, and 10 vol%) [26]; 5 vol% of MWNT and SiC nanoparticles (1, 2, 3 vol%) [27] in alumina. In the above few studies [22–27], the authors reported noticeable and marginal improvements or even degradation of properties. The marginal improvement or degradation of properties was attributed to the non-homogeneous distribution of the reinforcements, growth of the matrix grain size, and inhibition of densification.

The properties of nanocomposites depend not only on the properties of the individual constituents i.e. the matrix and reinforcement as well as the interface between them, but also on the extent of nanoreinforcement dispersion [3]. The molecular-level mixing process was successfully used to develop homogenous alumina-reduced graphene oxide [7] and Al_2O_3-CNTs [8] composites with good interface between the matrix and reinforcement. However, so far the process was not explored in order to produce hybrid ceramic nanocomposites. Our approach is to extend the use of this novel process to synthesize homogenous Al_2O_3 hybrid nanocomposites with uniform distribution of CNTs and SiC nanoparticles. The procedure involves bonding between functionalized CNTs and metallic ions. CNTs are functionalized to generate $COOH^-$ functional groups on the surface of the tubes. The metallic ions are obtained through the dissociation of a metallic salt in an organic solvent. High-energy probe sonication is used to disperse SiC nanoparticles in the prepared slurry. Heating of the dried mixture in normal atmosphere leads to the formation of amorphous Al_2O_3 matrix, imbedded with CNTs and SiC nanoparticles, which crystallizes during subsequent sintering. Finally, the powders are consolidated using spark plasma sintering process [28,29].

2. Materials and experimental procedures

2.1. Starting materials

Al_2O_3–5SiC-1CNT hybrid nanocomposite powder was synthesized from the following precursors and powders: aluminum nitrate nonahydrate ($Al(NO_3)_3 \cdot 9H_2O$), CVD synthesized MWCNT, and SiC_β (45–55 nm), 97.5% purity, obtained from nanostructured and amorphous material. On the other hand, α-Al_2O_3 (average particle size of 150 nm), 99.85% purity, procured from ChemPUR, Germany, was consolidated to prepare a reference material.

2.2. Nanocomposite powder synthesis

Al_2O_3–5SiC-1CNTs nanocomposite powder was synthesized through molecular-level mixing (MLM) and high-energy probe sonication. Carbon nanotubes (CNTs) produced using chemical vapor deposition (CVD) were functionalized to decorate them with negatively charged organic groups such as OH^- and/or $COOH^-$. CNTs were dispersed in concentrated nitric acid and refluxed for 48 h at 120 °C, followed by

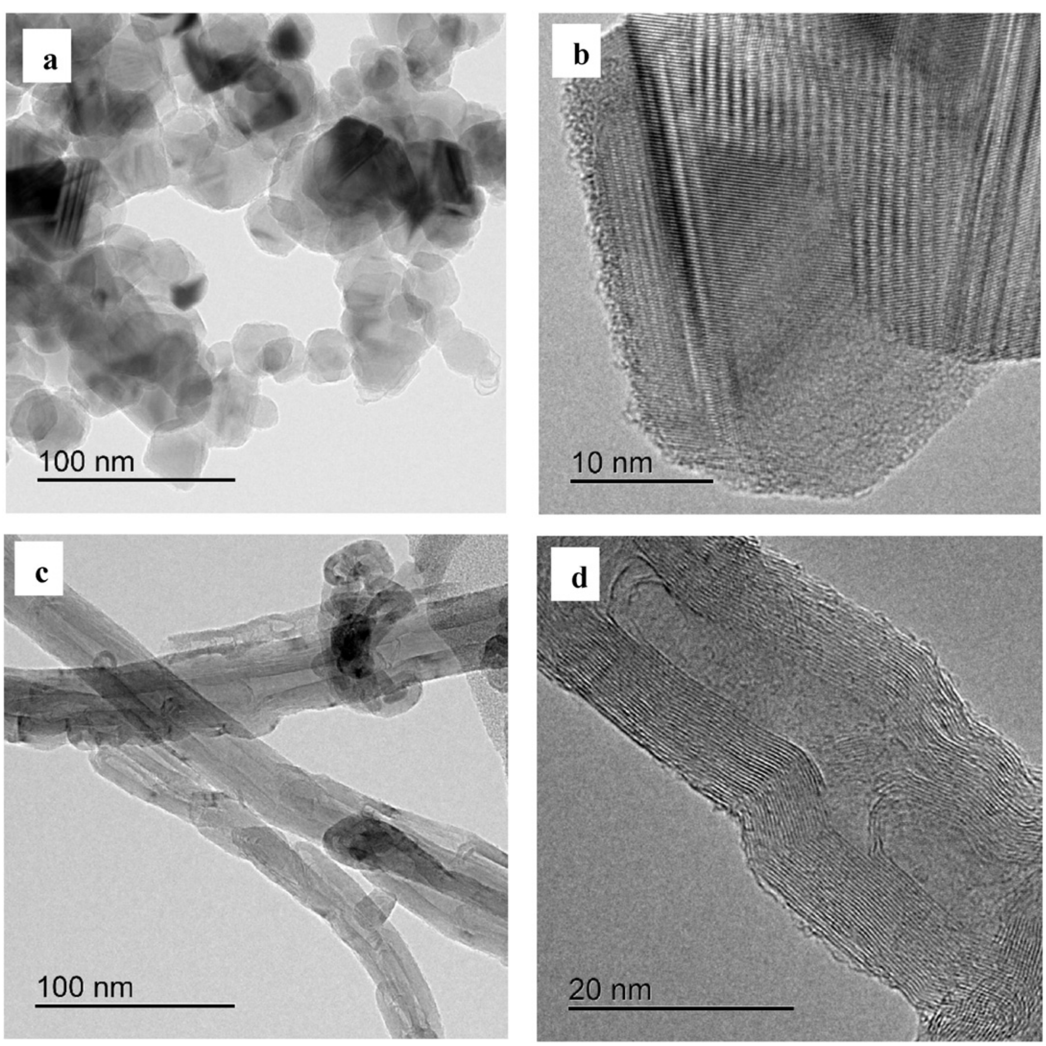

Fig. 1. TEM images of the starting powders (a, b) SiC, (c, d) CNTs.