



# Processing and mechanical properties of mullite and mullite–alumina composites reinforced with carbon nanofibers

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## Abstract

In the present work homogeneous mullite and mullite–alumina composites reinforced with carbon nanofibers (CNFs) were prepared. The stability of aqueous mullite and mullite–CNFs suspensions was studied through zeta potential and rheological behaviour of concentrated slurries. Slurries were optimised in terms of dispersant concentration and solids content and further used for the preparation of homogeneous mixtures of powders by freeze drying. The obtained powders were characterised through the determination of particle size distribution and morphology by Field Emission Scanning Electron Microscopy (FSEM) and Laser Scattering. The freeze dried powders were used to obtain dense composites by spark plasma sintering. Their microstructure and mechanical properties were evaluated. It was found that mullite and mullite–alumina composites reinforced with 2 vol.% CNFs maintain the strength and hardness of the ceramic matrix and presents higher values of toughness. The results indicated the homogeneous distribution and the survival of CNFs after consolidation.

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## 1. Introduction

Mullite ( $3\text{Al}_2\text{O}_3\text{--}2\text{SiO}_2$ ) has been largely studied because of their excellent properties, especially at high temperature, which make them suitable for a wide number of applications, including functional applications, such as electronic packaging and structural ones for high temperature engineering, for example high-temperature filters, lances for liquid metals and glasses, furnace elements and components, coatings for turbine aircraft engine hot-section components such as blades, combustors, etc. These properties make mullite very attractive as a matrix for advanced composites [1]. Although mullite ceramics are good candidates for high-temperature applications, their wider application is limited by their brittleness, low flaw tolerance and low reliability. Therefore the production of large, load-bearing, or

rapidly rotating parts, which are placed under high mechanical loads at high-temperatures, is very difficult. Consequently, the research work in recent years has been focused on increasing the strength and toughness of mullite through composite engineering. Different mullite matrix composites with enhanced toughness were recently developed. Much attention has been focused on improving the fracture resistance of mullite via microstructural design of ceramic–ceramic [2] or ceramic–metal composites [3,4] and layer structures [5,6].

The implementation of nanocarbon, in the form of carbon nanofibers (CNFs) or nanotubes (CNTs), into ceramic nanocomposites are very promising examples and have supposed a revolution in the design of reinforcing mechanisms for ceramics [7–13]. For successful nanocarbons/composite development, a number of key challenges must be met. First, nanocarbons must be obtained in reasonable quantity at acceptable cost. The nanocarbons must then be processed in such a way as to ensure a homogeneous dispersion with high degree of matrix densification, an appropriate degree of interfacial bonding, and without

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damage. Different approaches have been proposed to improve the dispersion of nanocarbons into ceramic matrices such as the use of ultrasounds in diluted suspensions, functionalisation by chemical routes, milling techniques, in situ catalytic chemical vapour deposition (CCVD), sol–gel technology or colloidal processing.

Colloidal processing is generally used to coat CNTs and CNFs with ceramic particles by adjusting their surface chemistry in order to prevent agglomeration and to facilitate their homogeneous dispersion throughout ceramic matrix grains [14,15]. This processing method have been used for a range of ceramic matrices including  $\text{Al}_2\text{O}_3$  [16–21],  $\text{SiO}_2$  [22–25],  $\text{ZrO}_2$  [26–29],  $\text{Si}_3\text{N}_4$  [30,31], and  $\text{SiC}$  [32]. However, as far as we know this processing method for the production of mullite/CNFs and mullite–alumina/CNFs composites have not been previously investigated in the literature. Wang et al. [33] prepared such materials by mixing powders of multi-walled CNTs (MWCNTs),  $\text{Al}_2\text{O}_3$  and  $\text{SiO}_2$  under wet conditions, followed by ultrasonication; the dried powder was then crushed and sieved, and finally densified by hot-pressing in argon. These authors reported a moderate increase in bending strength and a strong increase in toughness, compared with pure mullite. Fracture of MWNTs and pullout of MWNTs at interfaces were proposed as reinforcement mechanisms. Weibel et al. [34] processed CNTs–iron–mullite nanocomposite by spark plasma sintering of powders prepared by a direct method involving a reduction in  $\text{H}_2$ – $\text{CH}_4$  and without any mechanical mixing step. They did not observe any increase in fracture strength, but the SENB toughness was twice the one obtained for unreinforced mullite (3.3 vs. 1.6  $\text{MPa m}^{1/2}$ ). The mechanisms of carbon nanotube bundle pullout and large-scale crack-bridging were evidenced. However, CNTs and bundles agglomerated at the grain boundaries were observed in some areas and weakened the materials.

CNFs are carbon materials that present a cylindrical shape similar as CNTs but with differential structural and textural characteristics, diameters in the range of 100–200 nm, length up to 500  $\mu\text{m}$  and bundles with a diameter less than 1800  $\mu\text{m}$ . One positive characteristic of CNFs with respect to CNTs is the easier production technology existing for the former [35].

The aim of the present work was to prepare homogeneous mullite and mullite–alumina suspensions with CNFs as a carbonaceous nanodispersoid to obtain homogeneous mixtures of powders by freeze drying and to evaluate the microstructural features and mechanical performance of samples sintered by SPS.

## 2. Experimental

As starting materials the following commercially available powders were used:

(1) A mullite powder (SMSA, Baikowski, France) with nominal stoichiometry  $3\text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2$  and alumina enriched composition (73.7%  $\text{Al}_2\text{O}_3$  and 27.3%  $\text{SiO}_2$ ). According to the supplier this powder was obtained by thermal decomposition of ammonium and silica, further calcination and dry ball milling. The as-received mullite powder is relatively coarse (average particle size of 1.8  $\mu\text{m}$ , and specific surface area of 13.5  $\text{m}^2 \text{g}^{-1}$ )

Table 1  
Physicochemical characteristics of the starting materials.

	Mullite	Alumina	CNFs
$S_s$ ( $\text{m}^2 \text{g}^{-1}$ )	14.3	9.5	120–200
Average size, $d_{v,50}$ (mm)	0.5	0.35	<40 (length) <0.05 (diameter)
Density ( $\text{g cm}^{-3}$ )	3.1	3.97	>1.97
BET (mm)	0.13	0.16	–
Agglomeration factor	3.9	2.2	–

and hence it was attrition milled to enhance sinterability. Milling was performed in a Teflon coated attritor in isopropyl alcohol using alumina balls of 5 mm in diameter for 7 h. After milling, powder was dried for 6 h at 80 °C till constant weight and sieved down to 60  $\mu\text{m}$ . In the following, all results regarding mullite will refer to the milled one.

(2) A commercial high purity  $\alpha$ - $\text{Al}_2\text{O}_3$  (Condea HPA0.5, USA) with mean particle size of 0.35  $\mu\text{m}$  and 9.5  $\text{m}^2 \text{g}^{-1}$  of specific surface area. Mixtures of mullite and alumina were prepared to a relative volume ratio of 90:10.

(3) CNFs were supplied by Grupo Antolin Ingeniería (Burgos, Spain). CNFs were manufactured by vapour phase growth [36] through decomposition of hydrocarbons in the gas phase in the presence of catalytic particles of nickel. Nickel impurities (8–10 wt%) are observed in final CNFs derived from the manufacturing process.

The following materials were prepared: mullite (M), mullite–alumina (MA), mullite–CNFs (M–CNFs), mullite–alumina–CNFs (MA–CNFs). The volume ratio of M to A was fixed to 90:10; the CNFs were always added in a concentration of 2 vol.% referred to dry ceramic powders.

Table 1 summarises the main physicochemical characteristics of the raw materials. Fig. 1 shows the morphology of the milled mullite and alumina powders as observed by SEM. Particle size distribution was obtained by laser diffraction (Mastersizer S, Malvern, UK) for mullite and alumina powders. The particle size distribution of both powders is shown in Fig. 1, also. Monomodal, Gaussian distribution is obtained for mullite whereas for alumina a small fraction of larger particles is also observed. The average diameters are lower than 0.5  $\mu\text{m}$ . The morphology and dimensions of the CNFs were described in previous work [35]. The CNFs present an average size of about 0.6  $\mu\text{m}$  with shoulders at 0.35 and 0.7  $\mu\text{m}$ . These measurements were used as a rough estimation due to the elongated shape of CNFs very far from the sphericity assumed by diffraction techniques.

The colloidal stability of aqueous suspensions of mullite was studied by measuring the zeta potential as a function of pH and deflocculant content using a Zetasizer NanoZS instrument (Malvern, UK), based on the laser Doppler velocimetry technique. HCl and KOH were used to change the pH, and KCl  $10^{-2} \text{M}$  was used as an inert electrolyte. An ammonium salt of polyacrylic acid (PAA, Duramax D3005, Rohm & Haas, USA) was used as a deflocculant with additions of 0.5, 0.8, 1.0, and 1.5 wt.% (on a dry solids basis). Suspensions for zeta

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