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Tribological and electrical properties of ceramic matrix composites with carbon nanotubes

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Abstract

Tribological behaviur of carbon fibrous phases (nanofibers and nanotubes) containing composites with Si_3N_4, ZrO_2 and Al_2O_3 matrices was studied by pin-on-disk technique in conditions of dry sliding. Coefficients of friction and wear rates were measured, wear damage mechanisms were observed and identified. The resulting tribological behaviur was related to microstructure and mechanical properties of respective materials. Electrical conductivity was measured in wide range of frequencies by two-point method and effect of volume fraction and distribution of CNTs and CNFs on percolation threshold was evaluated. Both coefficient of friction and electrical resistivity decreased with increasing amount of carbon phases, in both cases the nanofibers were more efficient than the nanotubes. The wear resistance in most cases decreased but for Si_3N_4–CNT composite a certain optimum (~5 wt.% CNT) was found.

Keywords: B. Nanocomposites; C. Friction; C. Wear resistance; C. Electrical conductivity; Nanotubes

1. Introduction

In recent decades there has been a strong effort to develop better ceramics for structural applications. Among the most promising materials belong zirconia and silicon nitride based systems. Zirconia (ZrO_2) has potential to attain very high toughness due to transformation toughening [1–4]. It is considered as prime candidate for many cutting tool applications and as biomedical material. Silicon nitride (Si_3N_4) was developed in a search for fully dense, high strength and high toughness materials [5]. A prime driver for its development was to replace metals with ceramics in advanced turbine and reciprocating engines to give higher operating temperatures and efficiencies. Alumina (Al_2O_3), on the other hand, is one of the most cost effective and widely used materials in the family of engineering ceramics which can be found in a very wide range of applications.

However, the inherent brittleness is still the main limiting factor of these materials. This problem has been tried to overcome by creating appropriate composite materials [6]. In recent years, following discovery and suggestions of Iijima [7], novel composites with carbon nanotubes and fiber-like structures are being developed. The fibrous nature of the toughening elements could lead to higher fracture toughness. Moreover, unlike other non-metallic additives, also other functional (electrical) properties can be useful, thanks to carbon electrical conductivity.

In all mentioned applications the materials will have to face the challenge of friction and wear damage. Since wear is a complex problem, which cannot be easily predicted from basic mechanical properties [8] (hardness, fracture toughness) tribological studies of the developed materials are indispensable.

The aim of this work was to study tribological properties of the CNF/CNT containing CMCs based on ZrO_2, Al_2O_3 and Si_3N_4 and to determine the influence of content of the carbon phases on coefficient of friction and wear. Further purpose was to measure the electrical properties, namely the electrical conductivity of these materials with respect to the composition and microstructure, and to find the percolation thresholds for particular cases.

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2. Experimental details

2.1. Experimental materials

The procedure of preparation of the experimental materials and their microstructures were investigated and described in detail elsewhere [9–12].

To prepare the zirconia based materials [9,10] 3 mol.% Y-TZP powder (from Tosoh, Japan) was used. Carbon nanofibers were grade HTF150FF (Electrovac, Austria). According to producer these CNFs have a diameter of 80–150 nm, specific surface area in a range of 20–100 $m^2\ g^{-1}$, Young's modulus of ~500 GPa, tensile strength of ~7 GPa and electrical resistivity of 10^{-3}–$10^{-4}\ \Omega$ cm.

The CNFs were dispersed by milling in Millipore water with dodecylbenzenesulfonic acid solution (DBSA) as a dispersing agent. The mixture was ultrasonically dispersed for 10 min using a probe. Separately Y-TZP powder was also ball milled in Millipore water. The CNFs solution was added to the ceramic slip, and the mixture was then given a further ultrasonic treatment. This slip was continuously stirred and spray dried in a small laboratory spray dryer (model 190, Büchi, Germany). Composite powders were prepared with 2.0 and 3.3 vol.% CNF content.

The resultant powder granulates were die pressed into 20 mm diameter discs for hot pressing. These samples were hot pressed in an argon atmosphere at a dwell temperature 1300 °C for 30 min at a pressure of 41 MPa. For comparison monolithic ZrO_2 was prepared under similar conditions, e.g. hot pressed at 1300 °C for 30 min at a pressure of 41 MPa. For SPS, discs of 20 and 30 mm diameter were prepared. The SPS samples were sintered at different dwell temperatures. The samples with 2.0 vol.% CNFs were sintered at 1400 °C and the samples with 3.3 vol.% CNFs were sintered at 1500 °C in both cases for with a 5 min dwell time and at 60 MPa.

The Si_3N_4 based composites were prepared using starting powder mixtures of the investigated materials as follows [11]: 90 wt.% Si_3N_4, 4 wt.% Al_2O_3, and 6 wt.% Y_2O_3. In addition the batches of multi-walled carbon nanotubes were added. The powder mixtures were milled in distilled water in an HDDM attritor (Union Process) at 4000 rpm for 5 h. Zirconia agitator delta discs and zirconia grinding media with 1 mm diameter were used. Each batch contained zirconia as contamination from media and discs. Samples were compacted by dry pressing at 220 MPa. The materials were sintered at 1700 °C, 20 MPa, 3 h in high purity nitrogen by a two-step sinter-HIP method using BN embedding powder. The final compositions contained 0, 1, 3, 5 and 10 wt.% of multiwall CNTs.

The experimental materials based on alumina were prepared by spark plasma sintering at the Queen Mary, University of London, UK [12]. Multi-wall CNTs (NC-7000 by Nanocyl Inc., Belgium: average outer diameter 9.5 nm; lengths of up to 1.5 μm; and density 1.7 g cm^{-3}) were dispersed in dimethyl-formadie, DMF using high power sonication for 2 h and then hand-mixed with alumina nanopowder (Sigma–Aldrich, UK: gamma phase; particle size <50 nm; surface area 35–43 $m^2\ g^{-1}$; melting point 2040 °C; and density 3.97 g cm^{-3}) for 2 min.

Table 1
Experimental materials, their compositions and processing routes.

Material	Matrix	Toughening phase	Fraction (wt.%)	Process route
ZrO_2	ZrO_2	CNF	0	HP
HP ZrO_2–1%CNF		(Electrovac)	1	HP
HP ZrO_2–2%CNF			2	HP
SPS ZrO_2–1%CNF			1	SPS
SPS ZrO_2–1%CNF			2	SPS
Si_3N_4	90% Si_3N_4	CNT	0	HIP
Si_3N_4–1%CNT	+	(KFKI)	1	HIP
Si_3N_4–3%CNT	4% Al_2O_3		3	HIP
Si_3N_4–5%CNT	+		5	HIP
Si_3N_4–10%CNT	6% Y_2O_3		10	HIP
Al_2O_3	Al_2O_3	CNT	0	SPS
Al_2O_3–2%CNT		(Nanocyl)	2	SPS
Al_2O_3–3.5%CNT			3.5	SPS
Al_2O_3–5%CNT			5	SPS
Al_2O_3–10%CNT			10	SPS
Al_2O_3–2%CB		Carbon black	2	SPS
Al_2O_3–5%CB			5	SPS

The liquid mixture was rotation ball milled for 8 h. It was then dried at 75 °C for 12 h on a heating plate in air, and then for 3 days in a vacuum oven at 100 °C for 3 days. Alumina and nanocomposite pellets (diameter 20 mm and thickness 2 mm) were prepared by SPS in a HPD 25/1 (FCT Systeme, Germany) furnace at 1800 °C. A pressure of 100 MPa was applied, heating rate was 300 °C min^{-1}, the sintering period was 3 min. As a reference, also carbon black containing Al_2O_3 based nanocomposites were prepared in the same way.

The overview of all experimental materials is summarized in Table 1.

2.2. Experimental methods

Wear behaviur of the experimental materials was studied in dry sliding for self-mated pairs. The surfaces were carefully prepared by polishing down to surface roughness below 1 μm. The wear testing was carried out on the tribometer HTT by CSM Instruments in air at the room temperature using the pin-on-disk technique, where the tribological partner for each material was a highly polished ball with 6 mm diameter made out of ceramics corresponding to the ceramic matrix of the tested material. The applied loads varied between 1 and 5 N, the sliding speeds from 2.5 to 15 cm/s and the sliding distance was 100 m.

The tangential forces during the test were measured and friction coefficients calculated. The worn surfaces were subsequently observed and the wear regimes, damage type and micromechanisms were identified. The material losses due to wear were measured by a contact profilometer and then specific wear rates (W) were calculated in terms of the volume loss (V) per distance (L) and applied load (F_p) according to the standard ISO 20808 [13]:

$$W = \frac{V}{L \cdot F_p} \quad [\mathrm{mm}^3/(\mathrm{m\ N})]$$