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Assessing the electrical property of carbon nanotube reinforced oxide ceramic matrix composites produced by ceramic injection moulding

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Keywords

Carbon Nanotubes, Ceramic Injection Moulding, Electrical Property, Oxide Ceramic Matrix Composite

Abstract

Owing to its remarkable properties, multiwalled carbon nanotubes (MWCNTs) are attracting the interest for realization of sensors.

The potential of MWCNTs for high temperature sensing applications was investigated by integration within reinforced aluminium oxide ceramic composite. MWCNTs up to 2 wt% were mixed with reinforced aluminum oxide ceramic composite using solution mixing method to ensure good homogeneilty of the oxide ceramic composite powders. Specimens were realized using ceramic injection moulding process (CIM) followed by debinding and sintering procedures. The topography of specimens were examined using atomic force microscopy (AFM). Electrical measurements were also carried out.

The results show good demouldability at high MWCNTs concentration and homogenous distribution of the MWCNTs within the oxide ceramic matrix. AFM images illustrate the reduction of surface roughness by increasing the MWCNTs content which demonstrates the role of MWCNTs to improve the fracture resistance of the oxide ceramic matrix composite. As well, the electrical resistance of the feedstocks were reduced sharply. After sintering process, the resistance range drops enormously from M Ω to reach 12.1 Ω at low MWCNTs content; which make them suitable to be as electrode with high temperature capability. The electrical resistance temperature dependency shows a negative temperature coefficient behaviour with negligible resistance change.

1 Introduction

Nowadays, the interest in the development of advanced materials that can withstand high temperature up to 1350 °C and have excellent mechanical and thermochemical properties as well mechanical fatigue resistance and corrosion resistance is getting increased especially in aeronautic application such as in turbine blade [1]. To this aim, extensive research efforts have been devoted towards the realization of these novel materials. Among these materials, ceramics are attracting a lot of attention due to their lighter weight in comparison to metal conventional materials [2]. In particular, oxide ceramics such as aluminium oxide (Al_2O_3), aluminium titanate (Al_2TiO_5), and zirconium oxide (ZrO_2) show great potential to be applied for long term applications where thermal and oxidising atmosphere is existing [1]. However,

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the brittleness of oxide ceramic materials is the issue. Therefore, fibre-reinforced oxide ceramics (OCMC) are gaining great of attention compared to monolithic oxide ceramics owing to their higher fracture toughness and damage tolerance under thermal and mechanical load [3].

Several progresses are driven toward the enhancement of these properties using several type of reinforcements. In fact, two types of fiber reinforcements were frequently used in the literature [3]. Oxide fibers such as alumina fiber were incorporated within oxide ceramics as they offer excellent oxidation resistance and good mechanical properties but creep faster at high temperature comparing to non-oxide fibers. Furthermore, the addition of the non-oxide fiber reinforcements is not only beneficial to enhance material ductility but also to develop functional structure by including conductive reinforcement such as carbon black, carbon nanotube (CNT) [4]. The integration of conductive reinforcement will enable continuous structural health monitoring capabilities at high temperature by exploiting the change of internal electrical resistance of composite caused by strain and /or damage [4, 5]. Recently, carbon nanotube is used to convert insulating ceramics into conductive composite due to their remarkable electrical conductivity [6]. In this work, hybrid oxide ceramic composites were developed as potentially material for strain sensing application in high temperature. The realized composites have fixed concentration of alumina fiber and different amount of MWCNTs. The specimens were realized using ceramic injection moulding process followed by debinding and sintering. Indeed, feedstock materials were prepared via solution mixing method to enable good dispersibility of the MWCNTs within the reinforced oxide ceramic matrix (Al_2O_3) containing alumina fiber. Topography measurements were conducted to investigate the quality of the realized specimens. The electrical response of the composites was investigated before and after sintering in order to define the percolation threshold. Furthermore, the temperature dependency of a specimen was also investigated.

2 Materials and methods

2.1 Materials

In this work, MWCNTs with a purity of +90 %, average length of about 1.5 µm and average diameter of 9.5 nm were used and purchased from Nanocyl SA. MWCNTs are produced via the Catalytic Chemical Vapor Deposition (Cat-CVD) process.

The feedstock **FAMR7040** used for the production of oxide ceramic composite via CIM consists of alumina powder “Martoxid® MR70” from Martinswerk GmbH, Bergheim, Germany, reinforced with 40 Vol-% of “Nextel 610 chopped alumina fibres” with polyvinyl alcohol (PVA) sizing (~2.5 mm in length and 10 to 11 µm in diameter) from 3M Deutschland GmbH, Neuss, Germany. The binding system consists of paraffin wax (PW) “Sasolwax® 6403” from Sasol Performance Chemicals, Hamburg, Germany, low-density polyethylene (PE) “Lupolen® 1800H” from LyondellBasell Industries N.V.; Frankfurt, Germany, and stearic acid (>98 %) from Carl Roth GmbH & Co. KG, Karlsruhe, Germany.

2.2 Preparation of MWCNTs oxide ceramic composite feedstock

The feedstocks with different weight percentages of MWCNTs (from 0.5 wt.% to 2 wt.%) were prepared using a simple fabrication process. First, oxide ceramic composite feedstock **FAMR7040** was grinded and dissolved in tetrahydrofuran (THF) solvent using magnetic stirrer at 120 °C and 1200 rpm for 1 h in order to guarantee good dissolving.

Then, MWCNTs were added to the dispersion as shown in Fig. 1 and mixed using magnetic stirrer. To enhance the distribution of the MWCNTs in the ceramic matrix composite, the mixture was sonicated for 15 min under 20 % amplitude with Sonoplus HD7300 horn sonicator. At the end, the as-obtained mixtures were dried at room temperature.

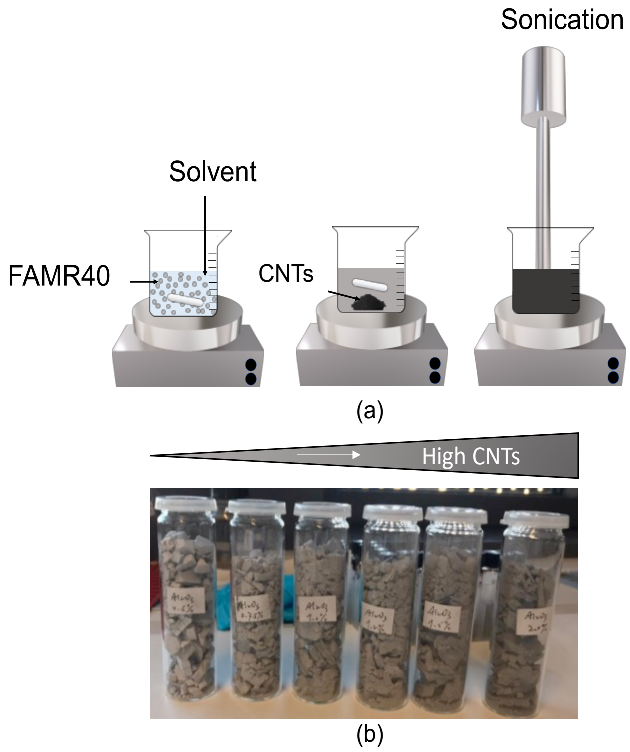


Figure 1: Realization of CMC feedstocks (a) Solution mixing for realization of CMC feedstocks and (b) CMC feedstocks with different MWCNTs concentration

2.3 Ceramic injection moulding for realization of MWCNTs oxide ceramic composite specimens

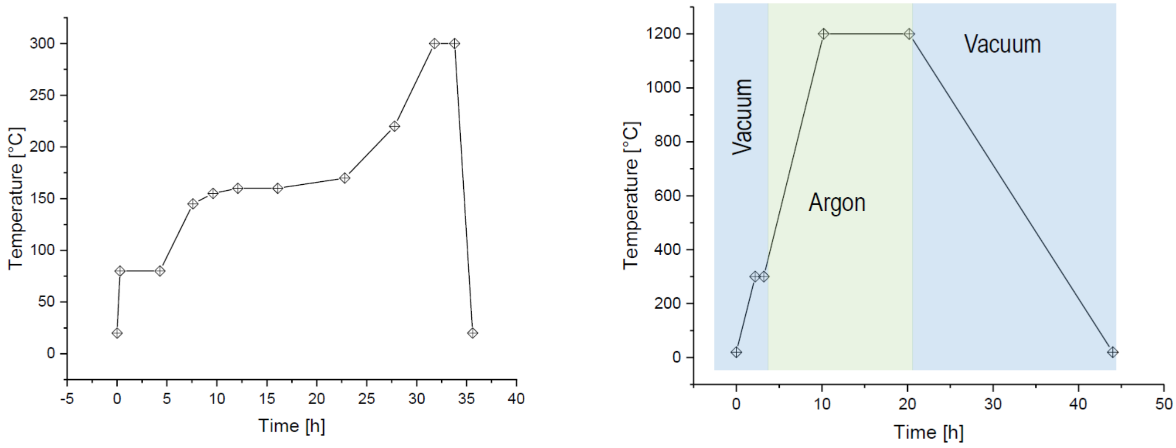
Generally, the ceramic injection moulding process is well established in the production of monolithic oxide ceramics and was also transferred to the manufacturing of OCMCs [3]. Furthermore, this process offers the opportunity to produce components with complex geometries to be manufactured cost-effectively in large quantities.In this study, this process is adapted as alternative to manual production process known for fabrication of fiber reinforced oxide ceramics. Samples with rectangular prism shape (47×7×3 mm) is obtained using HAAKE MiniJet II (Thermo Scientific) injection moulding machine. Details about injection moulding parameters are listed in Table 1.

Table 1:Applied CIM parameters

Parameter [unit]	
$T_{Cylinder}$ [°C]	130
T_{Mould} [°C]	30
$p_{Injection}$ [bar]	300
$t_{Injection}$ [s]	2
$p_{Holding}$ [bar]	300
$t_{Holding}$ [bar]	7

2.4 Debinding and sintering

The respective green bodies were first thermally debinded following the temperature profile shown in Fig. 2(a) to remove the major part of the organic binder. Subsequently, a sintering process was carried out at 1200 °C for 10 hours under inert conditions (see Fig. 2(b)) to avoid the decomposition of the MWCNTs.



(a) (b)

Figure 2: (a) Temperature profile for the thermal debinding and (b) temperature profile during sintering

2.5 Topography measurement

In order to investigate the impact of sintering on the quality of specimens and microcracks propagation, topography measurements were carried out using AFM Agilent LS5600. The images were extracted in tapping mode using a silicon nitride probe mounted on a cantilever of 40 N/m with a resonance frequency of 300 kHz.

2.6 Electrical characterization

The electrical resistance was firstly measured for the realized feedstocks using Keithley 2602A sourcemeter connected to Labview software interfaced to a computer in order to estimate the percolation threshold.

Later, the internal resistance of the produced OCMC probes were measured to verify the effect of the sintering on the percolation threshold. To this aim, a two-point probe method was employed. The respective OCMC samples covered in their two end with copper tape were connected to PlamSens 4.

The I–V characteristics were measured by applying a voltage from -1 to 1 V. The appropriate resistance of the films was calculated from the current and the voltage values. The electrical temperature dependency, the stability and the homogeneity of MWCNTs in the respective samples were investigated using DC measurements, whereby the effect of the temperature was studied in a temperature range from 30 °C to 80 °C using an electronic oven.

3 Results

The fibre reinforced oxide ceramic feedstocks shown in Fig. 1(b) have a gradiant gray color indicating different amounts of MWCNTs. A darker gray is observed with an increase of MWCNTs.

Furthermore, MWCNTs were well distributed within the fibre reinforced oxide ceramic feedstock containing the 40 Vol-% of alumina fibers. So far, such high concentrations of the reinforcements with fibres and MWCNTs were not achieved to the best of our knowledge indicating that the binder is very well suited for this material system regarding the viscosity of the binder as well the compatibility of the used solvent for MWCNTs integration.

Fig. 3 further confirms the choice of the binder system because the brown bodies have a sufficient strength and form stability retaining their shape without damage also after sintering.

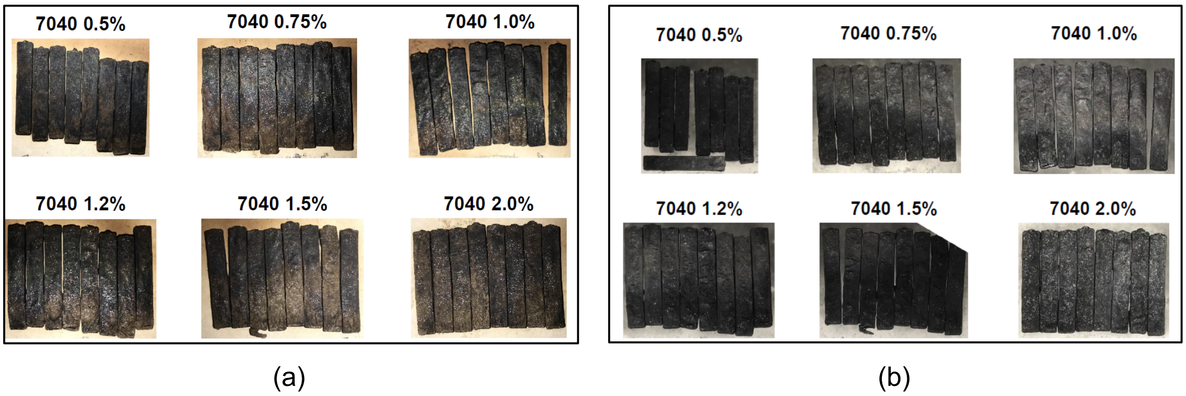


Figure 3: MWCNTs fiber-reinforced oxide ceramic composite specimens containing different MWCNTs concentration (a) after debinding and (b) after sintering

To characterize the surface microstructure of specimens after thermal treatment, AFM measurements were performed. The AFM images of the specimens with different amounts of MWCNTs are shown in Fig. 4 revealing the presence of alumina grain with size less than 1 µm. The surface roughness is reduced from 0.539 µm to 0.439 µm for the sample containing high amounts of MWCNTs. This observation indicates a positive effect of the MWCNTs regarding the mechanical properties of the composite. Furthemore, the shrinkage of the structure is decreased boosting their resistance to fracture during thermal treatment.

The electrical resistance of the feedstocks depend on the MWCNTs concentration and is reduced by increasing the amount of MWCNTs owing to the formation of more conductive pathways within the composite (Fig. 5(a)). However, the measured resistance was in the range of M Ω.

In addition, the homogeneity of the respective specimens is considered. The measured IV curves after CIM and sintering process are plotted in Fig. 5(b). It is obvious that the slope is higher by increasing the amount of MWCNTs illustrating the improved conductivity.

The resistance range drops enormously to reach 12.1 Ω corresponding to a specific resistance $\rho = 54.06\ 10^{-4}\Omega.\mathrm{m}$ at low MWCNTs concentration around 0.5 wt.% as demonstrated in Fig. 5(c). The stability of the prepared specimens were also verified as shown in Table 2. The standard deviation for each concentration was calculated. It was found that increasing the amount of MWCNTs lead to a better resistance stability which can be explained by the existence of microcracks within the samples with low MWCNTs concentration. Therefore, the developed materials are suitable to be used as electrode in high temperature applications.

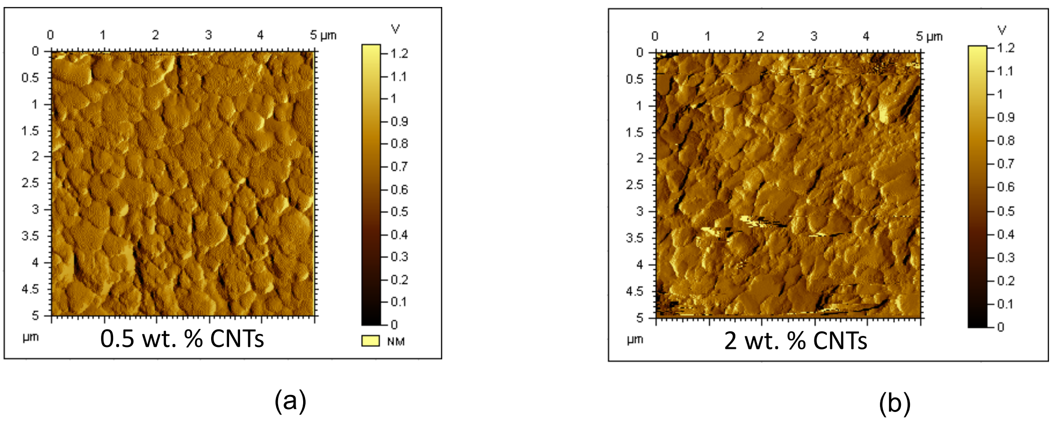


Figure 4: AFM images of OCMCs specimens containing different MWCNTs concentration (a) 0.5 wt.% MWCNTs and (b) 2 wt.% MWCNTs

For that reason, the investigation of the electrical behaviour at high temperature was required. A negative temperature coefficient behaviour was observed. The resistance is decreased with increasing temperature with a negligible resistance change of 0.0036 Ω/°C.

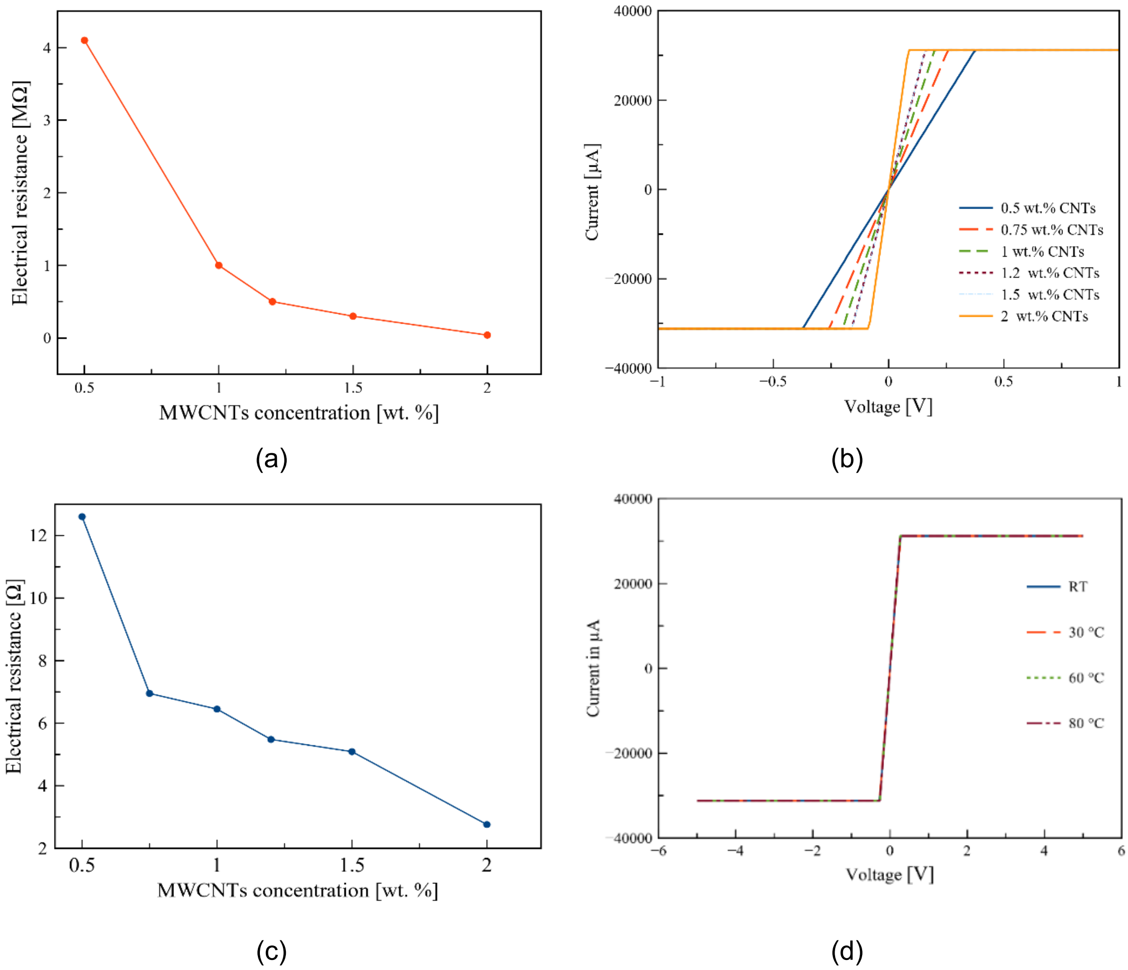


Figure 5: Electrical measurement of OCMCs at different stages (green bodies, sintered samples) and conditions (a) electrical resistance of feedstocks depending on MWCNTs concentration (b) IV characteristic after sintering depending on MWCNTs concentration, (c) calculated electrical resistance and (d) IV characteristic depending on the temperature

Table 2: Stability of the prepared specimens depending on MWCNTs concentration

MWCNTs concentration [wt.%]	R [Ω]	Standard deviation
0.50	12.10	1.259
0.75	6.95	1.141
1.00	6.45	0.676
1.20	5.48	0.239
1.50	5.09	0.497
2.00	2.76	0.100

4 Discussion and conclusion

In this study, we have examined the potential of MWCNTs reinforced oxide ceramic matrix composite for sensing applications. Therefore, specimens with different amounts of MWCNTs ranging from 0.5 to 2 wt.% were prepared and investigated based on the topography and the electrical measurements.

The results show the increase of the electrical conductivity with increasing amount of MWCNTs in addition to the low electrical resistance in the range of Ohm even at 0.5 wt.%. Therefore, the prepared specimens have great potential to be used as electrode for strain sensor in high temperature applications. In fact, a negligeable change in resistance of 0.0036 Ω/°C is observed. To realize sensing materials, lower MWCNTs concentration must be used.

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