BAJOPAS Volume 15 Number 1, June, 2022

http://dx.doi.org/10.4314/bajopas.v15i1.19

Bayero Journal of Pure and Applied Sciences, 15(1): 136 - 140
Received: February, 2022
Accepted: April, 2022
ISSN 2006 – 6996

HIGH TEMPERATURE APPLICATIONS OF CARBON NANOTUBES (CNTs) [V]: THERMAL CONDUCTIVITY OF CNTs REINFORCED SILICA NANOCOMPOSITE

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ABSTRACT

Consolidated functionalized carbon nanotubes/silica refractory ceramic nanocomposites (FCNTs/silica) were fabricated by pressureless sintering technique. Thermal conductivity of the nanocomposites with various amounts of carbon nanotubes (0, 1, and 4 wt.%) were investigated. The thermal conductivity increases with temperature, 1 wt. % FCNTs/silica nanocomposite gave the highest thermal conductivity. Therefore, it can be concluded that the carbon nanotubes (CNTs) are promising reinforcement for improving thermal conductivity of the silica refractory ceramics.

Keywords: Silica; carbon nanotubes; pressureless sintering; nanocomposites; thermal conductivity.

INTRODUCTION

Due to their remarkable functional properties; such as excellent electrical conductivity (Bandaru, 2007; Kaushik & Majumder, 2015; Lekawa-Raus *et al.,* 2014), and high thermal conductivity (Berber *et al.,* 2000; Kwon & Kim, 2006); for instance, electrical conductivity; 10^6 S/m at 300 K for SWNTs, $>10^5$ S/m for MWNTs, and high thermal conductivity; 6600 W/ m K for singlewalled carbon nanotubes (SWNTs), and >3000 W/ m K for multiwalled carbon nanotubes (MWNTs) (Baughman *et al.,* 2002; Biercuk *et al.,* 2002), carbon nanotubes (CNTs) are considered to be among the most promising reinforcement employed in many applications such as fabrication of composites, where the CNTs serve as fillers and binders to enhance the mechanical, electrical, and thermal properties (Ajayan *et al.,* 2000; Asl *et al.,* 2016; Biercuk *et al.,* 2002; Islam *et al.,* 2018; Kilbride *et al.,* 2002; Kumari *et al.,* 2008; Wan *et al.,* 2015).

Silica refractory material depending on its physical and functional properties can be considered as a good heat conductor (at high temperatures) and could be used extensively in the construction of coke ovens. Some reported thermal conductivities of silica refractory bricks are: 1.26 W/ m K, 1.67 W/ m K, and 2.09 W/ m K at 300°C, 700°C, and 1100°C respectively (Chesti, 1986). It has been reported that increased in density of the silica refractory bricks, would definitely improve the thermal conductivity property of the bricks with the addition of fumed silica micro-particles, titania micro/nano-particles, controlling the ratio of tridymite/cristobalite to approximately 1 – 1.2 on a coarse-crystalline raw material basis, and modifying the brick manufacturing process; by using a granular mineral having at least 95 wt. % of SiO_2 with maximum grain size of 3 mm and no more than 3 wt. % of CaO, by increasing the molding time or applying molding pressure gradually while molding, and by gradual increase in temperature when firing and exposing the brick to long curing time at the highest possible sintering temperature (Brunk, 2000; Mahler *et al.,* 1971; Manivasakan *et al.,* 2010; Mccreight *et al.,* 1964). Apart from enhancement in thermal conductivity, other additional benefits that could be derived due to the improved densification are, a greater strength, increased abrasion resistance, and low porosity (Mccreight *et al.,* 1964).

The vast majority of the most recent researches conducted on CNTs-reinforced ceramic composite dwelled on the engineering ceramics and investigated the effect of CNTs addition to mechanical and electrical properties (Bai & Xie, 2017; Barmin *et al.,* 2016; Chen *et al.,* 2018; Chen *et al.,* 2015; Chen *et al.,* 2015; Han *et al.,* 2017; Jin *et al.,* 2018; Lin *et al.,* 2016; Lin *et al.,* 2016; Lin *et al.,* 2018; Lin *et al.,* 2018; Mei *et al.,* 2017; Yang *et al.,* 2018; Yang *et al.,* 2016).

Only a few studies were conducted on thermal properties; heat capacity, thermal diffusivity, and thermal conductivity of CNTs/ceramic nanocomposites (Cao *et al.,* 2020; Chen *et al.,* 2017; Jiang & Gao, 2008; Kumari *et al.,* 2008; Ning *et al.,* 2003; Pöhls *et al.,* 2019; Shin *et al.,* 2018; Sivakumar *et al.,* 2007; Zhan & Mukherjee, 2004), out of which very few are of recent. For most of these SWNTs/MWNTs-reinforced ceramic composites, for evaluation of the thermal properties, the nanocomposites were fabricated by spark plasma sintering (SPS). As a novel study, this is the first research that attempts to determine thermal conductivity of the CNTs/crystalline silica ceramic employing also for the first time pressureless sintering technique for consolidation. CaO has been utilized as a potential binder and mineralizer in the manufacture of conventional silica refractory (Brunk, 2000; Brunk, 2001; Manivasakan *et al.,* 2010). Also, it has been employed as a versatile and suitable catalyst support in the synthesis of carbon nanotubes from carbon dioxide feedstock (Xu & Huang, 2007). Hence, CaO would be chemically compatible and can be a potential binder for the silica matrix and the CNTs.

MATERIALS AND METHODS

Materials

Pristine multiwalled carbon nanotubes (PMWNTs) (CVD-grown, > 98 % purity, 10 – 20 nm diameter, and 10 – 30 µm length) were received from the Chengdu Organic Chemicals Co., Ltd, Chinese Academy of Sciences (Sichuan Sheng, China). As-mined silica and clay both obtained from Gezawa quarry (Kano, Nigeria); the silica has been purified, crushed, ground, and pulverized (processed) using crushers, ball mill, and pulverizer at National metallurgical development centre NMDC (Farar-gada, Jos, Nigeria). CaO powder (ChemPur®, SYSTERM) was purchased from Classic Chemicals Sdn Bhd (Selangor, Malaysia).

Production of FCNTs/silica nanocomposite pellets

As-processed and graded ($d_{(0.5)}$ = 20.166 µm, using Malvern particle analyzer) silica, clay with size < 75 µm and CaO powder were added to portion of the stable suspension of FCNTs as prepared elsewhere (Tijjani *et al.,* 2018). These were thoroughly mixed, cold pressed under 90 – 100 MPa for 2 min, into specimen pellets dimensioned $(\emptyset 11mm \times 2mm\, height)$ using uni-axial hydraulic press and dried at 150°C for 24 hours. Then, the as-fabricated nanocomposite pellets; 0 wt. % FCNTs + S (SC*-0), 1 wt. % FCNTs + S (SC*-1), and 4 wt. % FCNTs + S (SC*-4) were subjected to thermal diffusivity and specific heat capacity tests. Where S stands for silica plus sintering aids (CaO + clay); to hasten the polymorphic conversion of quartz to cristobalite and tridimite. C for functionalized carbon nanotubes, * means all pellets are unsintered and without * means all sintered.

Determination of thermal diffusivity (α^2) of densified nanocomposite pellets:

The as-dried pellets dimensioned $\emptyset 11mm \times 2mm\, height$; for (SC*-0), (SC*-1) and (SC*-4) were consolidated by pressureless sintering technique under argon atmosphere at 1450°C for 2 h with a heating rate of 5°C/min. The sintered pellets were then subjected to thermal diffusivity test using Laser Flash Apparatus LFA 457 Micro Flash. The procedure and the values obtained are as explained by Tijjani (2018).

Determination of specific heat capacity (C_p) of densified nanocomposite pellets:

Thermo-analytical measurements were performed on the unsintered powdered FCNTs/silica $(\emptyset 11mm \times 2mm\, height)$ pellets; SC*-0, SC*-1, and SC*-4 by TGA/DSC analyzer, in a dynamic air atmosphere, using respective weights of 10.1428 mg, 10.1151 mg, and 10.0158 mg in aluminium crucible 70 μL. Heating has been done at the rate of 20 K min^{-1} from room temperature (RT) to 1450°C. The procedure and the values obtained are as reported by Tijjani (2018).

Determination of bulk density (ρ) of densified nanocomposite pellets:

The bulk density was determined by Archimedes' method. The values obtained for the densified samples; SC-0, SC-1 and SC-4 are $1.74 \pm 0.00\, g/cm^3$, $1.74 \pm 0.01\, g/cm^3$ and $1.72 \pm 0.00\, g/cm^3$ $respectively$ (Tijjani, 2018).

Determination of thermal conductivity (k) of densified nanocomposite pellets:

The thermal conductivities (*k*) of the nanocomposites were determined by calculation using Equation 1 and the values of α^2, C_p and ρ above for SC-0, SC-1 and SC-4.

$$k = \alpha^2 C_p \rho \quad (1)$$

RESULTS AND DISCUSSION

Thermal Conductivity of FCNTs/silica bricks

The thermal conductivities of FCNTs/silica nanocomposites are calculated based on the equation 1. The plots are as shown in Figure 1. Thermal conductivity increases with increased temperature for all FCNTs/silica bricks under the temperature range considered. SC-1 has the highest thermal conductivity of the three blends. It reported an enhancement of more than 130% as compared to that of unreinforced silica brick (SC-0) at 117°C. At 163°C, this amount is almost doubled > 240%. The same trend has been reported for MWNTs-TiN composite; in which, at 100°C, a lesser increment of 11 % thermal conductivity was exhibited as compared to plain

TiN and at 430°C, this amount was enhanced to 97 % for 5 wt. % MWNTs loading (Jiang & Gao, 2008). For SC-4, at 117°C, the thermal conductivity decreases by 78 % as compared to that of a conventional brick. This amount is steadily decreased to about 74 % at 163°C. The reason behind the remarkable improvement in thermal conductivity with 1 wt. % FCNTs addition can be attributed to the following outstanding properties of the nanocomposite: The high thermal conductivity of the of the MWNTs (3000 W m^{-1} K^{-1}) as compared to the negligible value of the silica matrix 2.35 W m^{-1} K^{-1} at 1000°C (Brunk, 2001). This may justify the extraordinary enhancement due to the incorporation of CNTs in the matrix. Better dispersion of the lowest loading CNTs may be another reason. It could be due to an excellent covalent combination of the CNTs and silica which reduces the tube-matrix thermal boundary resistance and hence improving the thermal conductivity. Also, the increment may be due to the polymorphic formation of more tridymite with crystal growth as compared to that in conventional brick (Brunk, 2000). The high reduction of the thermal conductivity with 4 wt. % FCNTs could be attributed to agglomeration that leads to subsequent deterioration of thermal property as the percentage of CNTs increase.

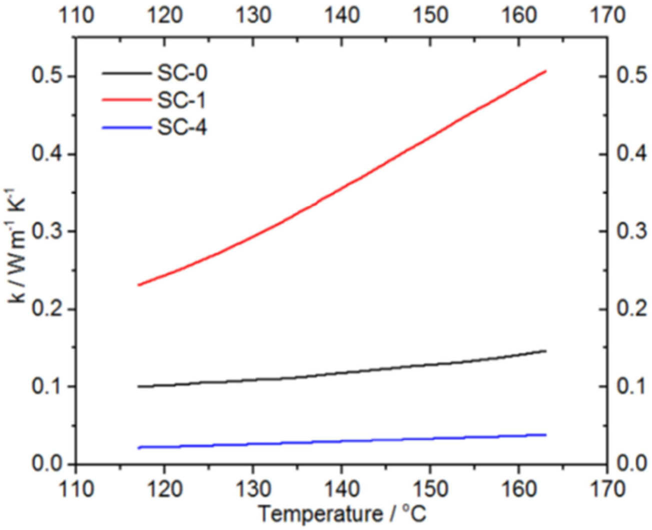

Figure 1: The thermal conductivity of the FCNTs/silica bricks

CONCLUSION

Thermal conductivities of fuctionalized CNTs-reinforced silica nanocomposite bricks for varied CNTs loading 0, 1, and 4 wt. % have been determined. 1 wt. % FCNTs/silica nanocomposite reports the highest thermal conductivity. At 117°C and 163°C the thermal conductivity has been enhanced remarkably to more than 130 % and in excess of 240 % respectively, as compared to that of the conventional un-reinforced silica brick.

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