Ultrathin graphene tribofilm formation during wear of Al₂O₃–graphene composites

Cheng Zhang

PhD student, Plasma Forming Laboratory, Department of Mechanical and Materials Engineering, Florida International University, Miami, FL, USA

Andy Nieto

PhD student, Plasma Forming Laboratory, Department of Mechanical and Materials Engineering, Florida International University, Miami, FL, USA

Arvind Agarwal*

Professor, Plasma Forming Laboratory, Department of Mechanical and Materials Engineering, Florida International University, Miami, FL, USA

Graphene nanoplatelet (GNP)-reinforced aluminum oxide (Al₂O₃) composites were sintered by spark plasma sintering in three different compositions (0, 0.5, 5 vol.% GNPs). To investigate the effects of graphene addition on the composites' wear resistance, ball-on-disk wear tests were conducted under very high normal load (40 N) by using a 3-mm-dia. ceramic counterface. Aluminum oxide–0.5 vol.% GNP exhibited 65% improvement in the wear resistance, while aluminum oxide–5 vol.% GNP displayed 53% poorer wear resistance as compared with aluminum oxide. The coefficient of friction was 0.45 for aluminum oxide–0.5 vol.% GNP, 0.40 for aluminum oxide–5 vol.% GNP and 0.60 for aluminum oxide. The highest wear resistance of aluminum oxide–0.5 vol.% GNP is attributed to formation of a continuous, protective and ultrathin graphene tribofilm on the wear surface. Tribofilm formation occurs due to the high shear forces induced by countersurface movement and localized heating, which causes GNP's delamination, overlap and welding together. In the case of aluminum oxide–5 vol.% GNP, poor dispersion and agglomeration of GNP results in a thick and discontinuous graphene tribofilm, which does not protect from the brittle fracture of aluminum oxide grains during wear.

1. Introduction

Graphene exhibits extraordinary mechanical properties, electrical conductivity and thermal conductivity, which allow it to become a potential filler material for ceramic matrix composites.^{1,2} Structural ceramics such as silicon nitride (Si₃N₄), tantalum carbide (TaC), yttria-stabilized zirconia, aluminum hydroxide (Al₂O₃), hydroxyapatite, silicon dioxide (SiO₂), silicon carbide (SiC), titanium dioxide (TiO₂), zirconium diboride (ZrB₂) and aluminum nitride (AlN) have been reinforced with graphene.^{3–27,28} Most of these studies focused on improving the electrical conductivity and the fracture toughness of the ceramic matrix by adding graphene as a nanofiller. The new electrical paths created by graphene's superior electrical conductivity have been attributed as the cause of the increase in electrical conductivity,^{15–17} whereas grain wrapping, crack deflection, bridging, sheet sliding and kink formation are the causes of the toughening of ceramics.^{3,11,14,17–24}

Friction is one of the major forms of energy loss. In the automobile industry for example, one-third of the fuel energy is used to overcome friction.²⁵ Hence, improving wear resistance

and reducing the coefficient of friction (COF) are critical to energy-saving requirements. Graphene has also proven to be an excellent lubricant additive due to its two-dimensional layered structure.^{9,11–13,26–29} It has been shown that a small amount of graphene solution added to a steel surface displays a four-order-of-magnitude reduction in wear during sliding. The significance of graphene addition was also observed in the reduction in the COF by a factor of 6, which was attributed to a conformal protective graphene coating formation on the sliding steel surface.²⁸ A few researchers have also investigated the tribological behavior of graphene addition to ceramic matrices.^{9,11–13,26–29} Belmonte *et al.*²⁶ studied the tribological property of a spark plasma-sintered silicon nitride–3 wt.% graphene nanoplatelet (GNP) composite for gasoline direct injection system by using a reciprocating ball-on-plate configuration under isooctane lubrication. Their results showed a 56% increase in the wear resistance as compared with that for bare silicon nitride, which was attributed to lubricating tribofilm formation. However, this study reported a marginal improvement in COF.²⁶ In a relatively contradictory finding, Hvizdos *et al.*²⁷ reported the tribological behavior of different types of graphene nanoplate additives to the silicon nitride matrix.

*Corresponding author e-mail address: agarwala@fiu.edu

It was concluded that the carbon phase did not participate directly in the lubrication process and hence no significant change in COF was observed. The increase in wear resistance was attributed to the increase in the fracture toughness of silicon nitride due to graphene addition. In another recent study, Kim *et al.*²⁹ reported one-order-of-magnitude wear improvement in a aluminum oxide/graphene system due to the increased fracture toughness and flexural strength. It was pointed out that only a small amount of GNP (0.25–0.5 vol.%) was needed to reinforce the composite and the improvement would be greater if the graphene phase was used instead of partially reduced graphene oxide.²⁹

The improvement in wear resistance of graphene-reinforced ceramic has been attributed two main reasons, namely improved fracture toughness/flexural strength and a protective graphene tribofilm formation during wear process. The improved fracture toughness and flexural strength are easier to measure and visualize. However, none of the studies on ceramic-graphene composites have provided a mechanism of graphene tribofilm formation during wear. This has resulted in contradictory conclusions without a clear understanding of what is really happening to graphene during the wear process. Based on this scenario, the prime objective of this study is to investigate graphene tribofilm formation and understand the mechanism of formation during wear in ceramic-graphene composites. In this study, aluminum oxide-GNP composites were prepared by spark plasma sintering (SPS) in 0, 0.5 and 5 vol.% GNP compositions. The wear behavior of aluminum oxide-GNP composites was characterized by a ball-on-disk tribometer in dry sliding condition. An extremely high normal load of 40 N using a 3-mm-dia. aluminum oxide counterface was used, which resulted in an extremely high contact pressure.

2. Experimental procedure

2.1 Powder processing

Spray-dried alpha phase aluminum oxide powder (Inframat Advanced Materials, CT, USA) with ultrahigh purity (99.99%) was used as the starting powder. The average agglomerate size was 20–50 μm with individual particle size of 100 nm. Grade M-5 GNPs were obtained from XG Sciences, MI, USA. The average plate thickness was 6–8 nm, the diameter was 15 μm and the typical surface area was 120–150 m^2/g . The GNPs and the aluminum oxide powder were mixed by wet chemistry method. The GNPs were ultrasonicated in acetone for 60 min followed by addition of aluminum oxide powders into the solution and were further sonication for 30 min. The mixed slurry was dried in the oven at 85°C for 2 h. Two different compositions were made: aluminum oxide–0.5 vol.% GNP (aluminum oxide–0.5G) and aluminum oxide–5 vol.% GNP (aluminum oxide–5G) and compared with pure aluminum oxide.

2.2 Spark plasma sintering

The dried powders were placed into a graphite die (20 mm dia.) and sintered in a vacuum chamber by using an SPS furnace by

SPS Nanoceramics Inc., IL, USA. The residual pressure inside the chamber was less than 10 Pa. The powders were preheated to 300°C and held for 2 min. The powders were again heated to 1400°C at a heating rate of 100°C/min with 70 MPa pressure applied to the powders when the temperature reached 1300°C. A temperature of 1400°C and a pressure of 70 MPa were held for 5 min before turning the SPS power off. The samples were cooled down naturally in the furnace chamber.

2.3 Wear tests

The wear tests were conducted by using a ball-on-disk tribometer (Nanovea, Irvine, CA, USA), using a static 3-mm-dia. alumina ball as the counter material. The sintered samples were rotated at 50 rpm, making a 4-mm-dia. wear track for a total period of 60 min; the total linear distance was 37.7 m. A very high normal load of 40 N was applied. A real-time COF was also recorded. After wear, the tracks were profiled by a non-contact three-dimensional (3D) PS50 optical profilometer (Nanovea). Wear depth and width were obtained by using line profiles from 3D profiles of the wear track and analyzed by using the SPIP software (Image Metrology A/S, Hørsholm, Denmark). Wear volume was computed by multiplying the cross-sectional area of the wear track with the wear depth.

2.4 Characterization

The sintered samples' density was measured by using a helium pycnometer (Accupyc II 1340, Micromeritics Instrument Corporation, Norcross, GA, USA). The microstructure of the starting powder, mixed powders, sintered samples and wear tracks were examined by using a JEOL JSM 6330F field emission scanning electron microscope (SEM) (JEOL Ltd, Tokyo, Japan). A Hysitron TI 900 triboindenter was used for nanoscale hardness measurement. Average grain size measurements were obtained from the fracture surface images by using the ImageJ software.

3. Results and discussion

3.1 Microstructure

The fracture surfaces of the spark plasma-sintered aluminum oxide, aluminum oxide–0.5G and aluminum oxide–5G samples are shown in Figures 1 and 2. The densities of the sintered pellets are shown in Table 1. The densification percentage was calculated by taking the ratio of the measured value to the theoretical value. The theoretical densities of graphene and aluminum oxide were assumed to be 1.82 and 4.02 g/cm^3 respectively.³ It is clear from the fracture surfaces and Table 1 that all samples achieved a very high densification (~98–99%). Pure aluminum oxide shows a dense structure with a fine grain size of $0.96 \pm 0.15 \mu\text{m}$ (Figure 1(a)). Aluminum oxide–0.5G also shows a very dense structure with dispersed and aligned GNP (Figure 1(b)). The average grain size of aluminum oxide–0.5G reduces to $0.80 \pm 0.11 \mu\text{m}$. The grain refinement is attributed to the addition of GNPs that wrap around the grains and inhibit the grain growth.^{23,30} Figure 2(b) is a high-magnification SEM image of aluminum oxide–0.5G, which shows several uniformly dispersed GNPs embedded between

Figure 1. Low-magnification fracture surfaces of spark plasma-sintered (a) aluminum oxide, (b) aluminum oxide-0.5G and (c) aluminum oxide-5G (the arrows mark the agglomeration of GNPs)

Figure 2. High-magnification fracture surfaces of spark plasma-sintered (a) aluminum oxide, (b) aluminum oxide-0.5G and (c) aluminum oxide-5G

aluminum oxide grains. The GNPs shows severe kinking, bending and folds without any cracks. These energy dissipation mechanism of GNP lead to improved fracture toughness of ceramics as observed in our previous study.^{23,31} Figure 1(c) shows fracture surface of aluminum oxide-5G. The overall structure is

dense; however severe GNP agglomeration is observed which is attributed to its poor dispersion at high concentration of 5 vol.%. Figure 2(c) shows a cluster of highly wrinkled GNP. A few of GNP are sandwiched between aluminum oxide grains whereas others are loosely held. The average grain size of aluminum

Sample	Densification: %	Grain size: μm	Nanohardness: GPa
Aluminum oxide	98.9	0.96 ± 0.15	18 ± 3.5
Aluminum oxide-0.5G	98.5	0.80 ± 0.11	23 ± 3.4
Aluminum oxide-5G	98.3	0.77 ± 0.18	12.6 ± 4.1

Table 1. Densities, grain sizes and nanohardnesses of aluminum oxide, aluminum oxide-0.5G and aluminum oxide-5G

oxide-5GNP is $0.77 \pm 0.18 \mu\text{m}$, which is almost similar to aluminum oxide-0.5G.

Table 1 also lists hardness values obtained from nanoindentation of sintered samples. aluminum oxide-0.5G has the highest hardness ($23 \pm 3.4 \text{ GPa}$), which is attributed to GNP inhibiting the grain growth compared with pure aluminum oxide.²³ Aluminum oxide-5G shows the lowest hardness of $12.6 \pm 4.1 \text{ GPa}$. Although the aluminum oxide-5G sample had an average grain size similar to that of aluminum oxide-0.5G, the large presence of agglomerated graphene between the grain boundaries (Figure 2(c)) weakens the bonding forces, resulting in the lowest hardness. These findings suggest that there could be an optimum GNP composition between 0 and 5 vol.% for a more homogeneous and effective dispersion in the aluminum oxide matrix. A similar behavior regarding an optimum GNP content has also been observed in polymer-GNP composites.³² No phase transformation was observed during SPS. All samples displayed identical peaks belonging to the α -alumina phase, which is same as the starting powder (X-ray diffraction data not included here).

3.2 Wear behavior

Figure 3 shows the wear depth and 3D wear track profiles of the aluminum oxide, aluminum oxide-0.5G and aluminum oxide-5G samples. The highest wear depth ($30 \mu\text{m}$) is observed for aluminum oxide-5G, whereas aluminum oxide-0.5G shows the

Figure 3. Wear depths and track profiles of aluminum oxide, aluminum oxide-0.5G and aluminum oxide-5G

lowest wear depth ($15 \mu\text{m}$). The wear volume loss was computed as described in Section 2.3. Figure 4 shows the wear volume loss from aluminum oxide-GNP composites under a dry sliding ball-on-disk test at 40 N normal load. The addition of 0.5 vol.% GNP resulted in a 65% improvement in the wear resistance of pure aluminum oxide. However, the addition of 5 vol.% GNP resulted in 53% degradation in wear resistance as compared with pure aluminum oxide. The wear volume loss difference can be attributed to the difference in hardness among the three samples (Table 1) as per Archard's equation which illustrates that, the harder the material, the better will be the wear resistance³³

$$1. \quad dV = \frac{kP}{H} dx$$

where dV is the wear volume loss, P and H are the load and the hardness respectively, k is called the wear resistance factor and dx is the wear distance. It must be recalled that aluminum oxide-5G shows the lowest hardness (Table 1) due to GNP agglomeration (Figure 1(c)). By using Archard's equation, calculations were carried out to see if the improved wear volume loss in aluminum oxide-0.5G was solely due to the difference in hardness. Wear resistance in aluminum oxide-0.5G would improve by 21.7% if hardness was the only contributing factor. However in the reality, the wear resistance improved by 65%.

Figure 4. Wear volume losses of aluminum oxide, aluminum oxide-0.5G and aluminum oxide-5G

Also, Archard's equation predicted a 42.3% reduction in the wear resistance of aluminum oxide-5G sample. However, the experimental results showed 53% reduction in wear resistance of aluminum oxide-5G. These observations suggest there may be additional factor(s) which may be contributing toward the improved wear resistance of aluminum oxide-0.5G.

The wear mechanism associated with volume loss can be elucidated by observing the wear tracks. Figure 5(a) shows a low-magnification SEM image of the wear track of pure aluminum oxide. The wear track shows a small amount of debris, but a large number of cracks are found on the surface, which is a sign of abrasive wear. The wavy marks shown in Figure 6(b) could be attributed to a run-in period where partial slippage may occur due to the uneven surface. During this period, the protuberances from the initial unworn aluminum oxide surface were either pushed down or broken into small debris. As the wear test continues, the wear of aluminum oxide would go to the next stage, forming a smoother surface. Localized heat generated due to high normal

Figure 5. Wear surface SEM pictures of pure aluminum oxide at (a) low and (b) high magnification

Figure 6. Wear surface SEM pictures of aluminum oxide-0.5G at (a) low and (b) high magnification

stress plays an important role as it creates a temperature gradient within the sample surface due to the ceramic's low thermal conductivity. This temperature difference results in large mismatch from the thermal expansion, and when it was combined with high stress, the sample surface arched up and cracked. Figure 5(b) shows that the unworn fracture surface become exposed, from which it can be inferred that that the cracked layer would peel off if the wear test time was extended. A similar behavior would be expected to occur in aluminum oxide-0.5G and aluminum oxide-5G samples; however, they behave significantly different from pure aluminum oxide as shown in Figures 6 and 7.

Figure 6(a) shows a clean and smooth wear track with insignificant wear debris in the aluminum oxide-0.5G sample. At a higher-magnification view (Figure 6(b)) of the wear track, a very interesting feature of an electron-transparent graphene tribofilm is observed on the surface. Due to the high strength of graphene in the planar direction, this tribofilm acts as a protective film during sliding wear and does not fracture easily. It is emphasized that even when the tribofilm did break at a few spots,

Figure 7. Wear surface SEM pictures of aluminum oxide-5G at (a) low magnification (cracks marked by arrows) and (b) high magnification

instead of falling into pieces, the film curled up and rolled due to its ultrathin and strong nature. The grains of aluminum oxide lying below the fractured graphene tribofilm do not have a conductive surface and become electrically charged as observed in Figure 6(b). The wear surface of aluminum oxide-5G in Figure 7 shows bimodal features including smooth and rough regions. The smooth surface shows formation of a graphene tribofilm similar to that for the aluminum oxide-0.5G sample; however, the film in the aluminum oxide-5G sample is scattered with relatively high thickness. Figure 7(b) shows a high-magnification view of the rough region showing mixed debris of aluminum oxide and agglomerated graphene. Such wear behavior is attributed to severe agglomeration of GNPs at the high concentration of 5 vol.%. During wear of aluminum oxide-5G, it is envisaged that a graphene tribofilm would form first around the graphene-rich region. However, the agglomerated GNPs serve as defects that weaken the structure and induce fracture. As a result, the regions adjacent to the thick graphene films would collapse and cave in. This explains the cracks found on the surface. Most cracks are located around the interface between the smooth and rough regions (Figure 7(a)). This also explains why there is no continuous graphene tribofilm forming in aluminum oxide-5G such as the one in aluminum oxide-0.5G.

To understand further the mechanism of graphene tribofilm formation, the normal stress exerted during the wear tests for the

Figure 8. Normal pressure against revolution during the wear tests for aluminum oxide, aluminum oxide-0.5G and aluminum oxide-5G

three samples is plotted as a function of the number of revolutions (Figure 8). The total number of revolutions in each wear test was 3000 (60 min $\times$ 50 rpm). The normal stress was computed by dividing the normal load (40 N) by the effective contact area between the 3-mm alumina ball and the underlying sample at any given point of time.³⁴ As the wear depth increases, the effective contact area increases, resulting in lower normal stress. The stress exerted during the initial 50 revolutions (run-in period) is not plotted because the area of contact was very small, resulting in an extremely high instantaneous pressure. It can be seen that the average normal stress is highest for aluminum oxide-0.5G, which is at least twice of aluminum oxide-5G.

It is proposed that under severe high normal stress and localized heat during wear test, graphene sheets from nanoplatelets are pulled under shear and become welded with overlapping areas to form an ultrathin graphene tribofilm. The proposed mechanism is schematically shown in Figure 9(a). Since GNPs are homogeneously distributed at a lower concentration of 0.5 vol.%, a thin and uniform graphene tribofilm forms in aluminum oxide-0.5G (Figure 9(b)) as compared with aluminum oxide-5G, which has a thicker and discontinuous tribofilm (Figure 7(b)). The difference in thickness between the tribofilms can be gauged by comparing their degrees of transparent behavior. Aluminum oxide grains under the graphene tribofilm can be clearly seen in aluminum oxide-0.5G (Figure 9(b)) due to its higher degree of transparent behavior, whereas no aluminum oxide grains are observed in aluminum oxide-5G (Figure 7(b)). In a recent study by Zhu *et al.*,³⁵ it was shown that the transmittance of multilayer graphene decreases with increasing number of layers. This finding further corroborates the authors' conclusion that the transparent graphene tribofilm formed in aluminum oxide-0.5G is much thinner than that in aluminum oxide-5G. Also, the protective graphene tribofilm is a major contributing factor in improving the wear resistance, in addition to the contribution from hardness.

Figure 9. (a) Schematic illustration of graphene tribofilm formation during wear; (b) transparent and thin graphene tribofilm in aluminum oxide-0.5G sample showing underlying aluminum oxide grains below grey region

Figure 10 shows the COF for the entire duration of the wear test for the three samples. Both aluminum oxide and aluminum oxide-5G shows greater a variation in friction behavior, which is attributed to the chipping and cracking of aluminum oxide as described earlier. Aluminum oxide-0.5G shows a continuous and more uniform COF value of 0.45, which is attributed to homogeneous graphene tribofilm formation over the wear track (Figure 6). The lowest COF value (0.35–4) is observed for aluminum oxide-5G, which is attributed to the highest concentration of GNPs, which also serve as a solid lubricant.³⁶ The highest COF (0.5–0.6) is observed for pure aluminum oxide, which does not have a lubricant additive such as graphene. Based on the above discussion, it can be safely concluded that the addition of GNP to aluminum oxide below a certain composition will increase the wear resistance by forming a protective tribofilm of graphene. This ultrathin graphene film can serve as both a solid lubricant and a protecting film that can provide toughness and contain wear damage. However, beyond the threshold of GNP content, wear resistance may decrease due to their agglomeration.

4. Conclusions

Aluminum oxide composite reinforced with 0, 0.5 and 5 vol.% GNPs were synthesized by SPS. The aluminum oxide-0.5 vol.% GNP sample exhibited a 65% improvement in wear resistance, while aluminum oxide-5 vol.% GNP showed 53% poorer wear

Figure 10. COFs of aluminum oxide, aluminum oxide-0.5G and aluminum oxide-5G samples

resistance compared with pure aluminum oxide. The highest wear resistance of aluminum oxide-0.5 vol.% GNP is attributed to higher hardness and the formation of a continuous, protective and ultrathin graphene tribofilm. It is shown that under severe high normal contact stress and localized heat during wear test, graphene sheets from nanoplatelets are pulled under shear and become welded with overlapping areas to form an ultrathin graphene tribofilm. Due to the high strength of graphene in the planar direction, the tribofilm acts as a protective film during wear test and does not fracture easily. Graphene film serves both as a solid lubricant and as a protecting film and contains wear damage. However, the graphene film is discontinuous due to agglomeration at the higher volume fraction of 5 vol.% and does not protect from brittle fracture of aluminum oxide grains during wear. The significant reduction in friction and the improvement in wear resistance suggest the great potential of adding GNPs as an energy-saving approach.

Acknowledgements

The authors would like to acknowledge Dr Ali Sayir, program manager of High Temperature Aerospace Materials at the Air Force Office of Scientific Research under grant FA9550-12-1-0263. The authors also acknowledge the support of the Advanced Materials Engineering Research Institute (AMERI) at the Florida International University.

REFERENCES

1. Novoselov KS, Blake P and Katsnelson MI (2008) Graphene: electronic properties. In *Encyclopedia of Materials: Science and Technology*, 2nd edn (Buschow KHJ, Cahn RW, Flemings MC *et al.* (eds)). Elsevier, Oxford, UK, pp. 1–6.
2. Lee C, Wei X, Kysar JW and Hone J (2008) Measurement of the elastic properties and intrinsic strength of monolayer graphene. *Science* **321**(5887): 385–388.

3. Nieto A, Lahiri D and Agarwal A (2012) Synthesis and properties of bulk graphene nanoplatelets consolidated by spark plasma sintering. *Carbon* **50**(11): 4068–4077.
4. Shin JH and Hong SH (2014) Fabrication and properties of reduced graphene oxide reinforced yttria-stabilized zirconia composite ceramics. *Journal of the European Ceramic Society* **34**(5): 1297–1302.
5. Azhari A, Toyserkani E and Villain C (2015) Additive manufacturing of graphene-hydroxyapatite nanocomposite structures. *International Journal of Applied Ceramic Technology* **12**(1): 8–17.
6. Zhang C, Chaudhary U, Lahiri D, Godavarty A and Agarwal A (2013) Photocatalytic activity of spark plasma sintered TiO₂-graphene nanoplatelet composite. *Scripta Materialia* **68**(9): 719–722.
7. Gao Z, Wan Y and Luo H (2014) An efficient route for the synthesis of aluminum nitride/graphene nanohybrids. *Journal of the American Ceramic Society* **97**(6): 1966–1970.
8. Yadhukulakrishnan GB, Karumuri S, Rahman A et al. (2013) Spark plasma sintering of graphene reinforced zirconium diboride ultra-high temperature ceramic composites. *Ceramics International* **39**(6): 6637–6646.
9. Porwal H, Tatarko P, Saggarr R et al. (2014) Tribological properties of silica-graphene nano-platelet composites. *Ceramics International* **40**(8): 12067–12074.
10. Roman-Manso B, Sanchez-Gonzales E, Ortiz AL et al. (2014) Contact-mechanical properties at pre-creep temperatures of fine-grained graphene/SiC composites prepared in situ by spark plasma sintering. *Journal of the European Ceramic Society* **34**(5): 1433–1438.
11. Rutkowski P, Stobierski L, Zientara D et al. (2015) The influence of the graphene additive on mechanical properties and wear of hot-pressed Si₃N₄ matrix composites. *Journal of the European Ceramic Society* **35**(1): 87–94.
12. Li H, Xie Y, Li K et al. (2014) Microstructure and wear behavior of graphene nanosheets-reinforced zirconia coating. *Ceramics International* **40**(8): 12821–12829.
13. Balko J, Hvizdos P, Dusza J, Balazsi C and Gamcova J (2014) Wear damage of Si₃N₄-graphene nanocomposites at room and elevated temperatures. *Journal of the European Ceramic Society* **34**(14): 3309–3317.
14. Ramirez C, Miranzo P, Belmonte M et al. (2014) Extraordinary toughening enhancement and flexural strength in Si₃N₄ composites using graphene sheets. *Journal of the European Ceramic Society* **34**(2): 161–169.
15. Ramirez C, Figueiredo FM, Miranzo P, Poza P and Osendi MI (2012) Graphene nanoplatelet/silicon nitride composites with high electrical conductivity. *Carbon* **50**(10): 3607–3615.
16. Fan Y, Wang L, Li J et al. (2010) Preparation and electrical properties of graphene nanosheet/Al₂O₃ composites. *Carbon* **48**(6): 1743–1749.
17. Centeno A, Rocha VG, Alonso B et al. (2013) Graphene for tough and electroconductive alumina ceramics. *Journal of the European Ceramic Society* **33**(15–16): 3201–3210.
18. Inam F, Vo T and Bhat BR (2014) Structural stability studies of graphene on sintered ceramic nanocomposites. *Ceramics International* **40**(10): 16227–16233.
19. Porwal H, Tatarko P, Grasso S et al. (2013) Graphene reinforced alumina nano-composites. *Carbon* **64**: 359–369.
20. Liu J, Yan H and Jiang K (2013) Mechanical properties of graphene platelet-reinforced alumina ceramic composites. *Ceramics International* **39**(6): 6215–6221.
21. Fan Y, Estili M, Igarashi G, Jiang W and Kawasaki A (2014) The effect of homogeneously dispersed few-layer graphene on microstructure and mechanical properties of Al₂O₃ nanocomposites. *Journal of the European Ceramic Society* **34**(2): 443–451.
22. Walker LS, Marotto VR, Raflee MA, Koratkar N and Corral EL (2011) Toughening in graphene ceramic composites. *ACS Nano* **5**(4): 3182–3190.
23. Nieto A, Lahiri D and Agarwal A (2013) Graphene nanoplatelets reinforced tantalum carbide consolidated by spark plasma sintering. *Materials Science and Engineering: A* **582**: 338–346.
24. Wang K, Wang Y, Fan Z, Yan J and Wei T (2011) Preparation of graphene nanosheet/alumina composites by spark plasma sintering. *Materials Research Bulletin* **46**(2): 315–318.
25. Holmberg K, Andersson P and Erdemir A (2012) Global energy consumption due to friction in passenger car. *Tribology International* **47**: 221–234.
26. Belmonte M, Ramirez C, Julian JG et al. (2013) The beneficial effect of graphene nanofillers on the tribological performance of ceramics. *Carbon* **61**: 431–435.
27. Hvizdos P, Dusza J and Balazsi C (2013) Tribological properties of Si₃N₄-graphene nanocomposites. *Journal of the European Ceramic Society* **33**(12): 2359–2364.
28. Berman D, Erdemir A and Sumant AV (2013) Few layer graphene to reduce wear and friction on sliding steel surfaces. *Carbon* **54**: 454–459.
29. Kim HJ, Lee SM, Oh YS et al. (2014) Unoxidized graphene/alumina nanocomposite: fracture- and wear-resistance effects of graphene on alumina matrix. *Scientific Reports* **4**: 5176.
30. Nieto A, Huang L, Han YH and Schoenung JM (2015) Sintering behavior of spark plasma sintered alumina with graphene nanoplatelet reinforcement. *Ceramics International* **41**(4): 5926–5936.
31. Rudolf C, Boesl B and Agarwal A (2015) In situ indentation behavior of bulk multi-layer graphene flakes with respect to orientation. *Carbon* **94**: 872–878.
32. Lahiri D, Dua R, Zhang C et al. (2012) Graphene nanoplatelet-induced strengthening of ultrahigh molecular weight polyethylene and biocompatibility in vitro. *ACS Applied Materials & Interfaces* **4**(4): 2234–2241.
33. Archard JF (1953) Contact and rubbing of flat surfaces. *Journal of Applied Physics* **24**: 981–988.
34. Lahiri D, Singh V, Keshri AK, Seal S and Agarwal A (2010) Carbon nanotube toughened hydroxyapatite by spark plasma

-
- sintering: microstructural evolution and multiscale tribological properties. *Carbon* **48(11)**: 3103–3120.
35. Zhu S, Yuan S and Janssen GCAM (2014) Optical transmittance of multilayer graphene. *EPL* **108(1)**: 17007.
36. Lahiri D, Hec F, Thiesse M *et al.* (2014) Nanotribological behavior of graphene nanoplatelet reinforced ultra high molecular weight polyethylene composites. *Tribology International* **70**: 165–169.

WHAT DO YOU THINK?

To discuss this paper, please submit up to 500 words to the managing editor at journals@ice.org.uk.

Your contribution will be forwarded to the author(s) for a reply and, if considered appropriate by the editor-in-chief, will be published as a discussion in a future issue of the journal.

ICE Science journals rely entirely on contributions sent in by professionals, academics and students coming from the field of materials science and engineering. Articles should be within 5000–7000 words long (short communications and opinion articles should be within 2000 words long), with adequate illustrations and references. To access our author guidelines and how to submit your paper, please refer to the journal website at www.icevirtuallibrary.com/nme