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Tribological behavior of silicon nitride and carbon based filler composites – a review

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Abstract

Silicon nitride is a structural ceramic material which has been applied in many engineering structures where hardness, high flexural strength, resistance to oxidation and brittleness are required. High brittleness, low fracture toughness and low electrical conductivity limit its use in many sectors. Silicon nitride is one of the best candidates for tribological application and can be used in many sectors such as automotive and aerospace sectors. Carbon nanofillers, e.g. graphene and carbon nanotubes (CNT), possess remarkable electrical and thermal properties as well as excellent mechanical properties compared to conventional ceramic composites. Tailoring the tribological properties of Si_3N_4 by introducing the carbon nanometric reinforcement is becoming the popular. This review paper summarizes the advances of carbon nanofiller (CNTs and graphene) reinforced Si_3N_4 composites which can assist the ceramists to develop the Si_3N_4 composites with improved tribological properties.

Keywords: Silicon nitride, CNTs, Graphene, Tribology

1. Introduction

The unique properties of ceramics as high hardness and lower wear rate had been exploited to employ them in tribological systems [1]. With the time, the research discovered more properties of ceramics which enabled them to use in structural applications. The metallic materials suffer in high temperature environment therefore advanced ceramic have attractive features which enable them to use in high temperature applications. The high strength, corrosion resistance, high hardness and lower wear rate allow them to operate under conditions unsuitable for metals [2]. The importance of tribology can be realized with the impact on the global economy. According to the calculation the 23% of the world's energy consumption is due to the tribological issues. The 20% of that is consumed to overcome the friction and 3% is used to reprocessing the worn parts [3].

Silicon nitride-based materials have been used in cutting tools, bearings, sealings and gas turbine engines due to its exceptional flexural strength, high hardness, resistance to oxidation and thermal properties [4-6]. There are two ways of employing carbon nanofillers in tribological systems in order to reduce the friction and wear. One is to use the carbon nanostructures as a solid lubrication and the other is to use carbon nanofillers as additives in the bulk material for self-lubricating. Development of self-lubricating materials got more attentions and the extensive studies have demonstrated the carbon nanofillers as additives in bulk materials to achieve good lubricity [7-9]. Carbon nanofillers added composites got great attention to avoid the continuous addition of lubrication in machine parts. Carbon nanotubes (CNT) and graphene are the extensively used in the silicon nitride composites for tribological applications.

2. Carbon nanostructures silicon nitride

2.1. Carbon nanotubes (CNTs)

Carbon nanotubes were prepared first time by Ijima in 1991 by using an arc discharge evaporation method [10]. Numerous researches have been conducted on CNT-based ceramic composites in order to improve the properties [11-18]. The carbon nanotubes have been divided into single and multi-walled carbon nanotubes [19]. Multi-walled carbon nanotubes (MWCNTs) consist of multiple concentric graphene cylinders while single-walled carbon nanotubes (SWCNTs) comprises of single layer of graphene cylinder. CNTs are one-dimensional (1D) with *sp^2-hybridization* and owing excellent as charge carrier mobility [20-21].

2.2. Graphene platelets (GPLs)

Graphene is one of the allotropes of carbon having two-dimensional (2D) hexagonal lattice single or multi-layered with *sp^2 hybridization.* Graphene nanoplatelets consist of small stacks (10 -100 sheets) of graphene that can replace carbon fiber, carbon nanotubes, or other compounds in many composite applications because there are cheap and easily available. The stack layers are easily slide with respect to each other and this property can be exploited as a lubrication in tribology. Graphene became a promising candidate for tribological application due to its mechanical, low surface energy, high thermal, electrical and lubricating properties [22-23]. Similarly as CNTs, an extensive study has been conducted on the graphene based composites for tribological properties [24-30].

2.3. Silicon nitride

Silicon nitride (Si_3N_4) is an advanced structural ceramic with high melting point, high hardness, good shock resistance and relatively chemical inert. Silicon nitride has been considered one of the best candidates in tribological application due to its higher hardness than metal, 80% less friction, three to ten times longer lifetime, 80% higher speed, 60% less weight, non-reactive to lubricant, high corrosion resistance and ability to work on higher temperature [31].

3. Influence of CNTs on Wear rate and coefficient of friction

Carbon nanotubes have been considered as one of the ideal reinforcements and intrinsic self-lubricant material in ceramics (especially in Si_3N_4) to reduce the friction and wear rates and plenty of research works have been done [16], [32-36].

Gonzalez-Julian et al. studied the tribological behavior of CNTs added silicon nitride composites under the ball-on-plate configuration test [33]. They reported the significant decrease of wear rate and coefficient of friction with the addition of carbon nanotubes. With the addition of 8.6 vol. % of CNTs, the friction coefficient and the wear volume decreased about 40% and 80%, respectively, as compared with the monolithic material. The enhanced tribological properties were attributed to the good dispersion of CNTs in Si_3N_4 matrix which served as a solid lubricant and helped in uniform distribution of stress field at the load contact. Gonzalez-Julian et al. prepared the *ex-situ* and *in-situ* carbon nanotube/silicon nitride composites and found the better tribological properties of both ex-situ and in-situ composites than that of monolithic silicon nitride [32].

Contrary to this, the higher wear rate was observed with the addition of CNTs to the silicon nitride matrix. Koszor at al. [36] reported the higher wear rate (3.34 x 10^{-5} mm^3/N/m) of CNTs reinforced silicon nitride than the wear rate (3.02 x 10^{-6} mm^3/N/m) of silicon nitride without CNTs. The reason of having higher wear rate was the agglomeration of carbon nanotubes in matrix. CNTs have tendency to be agglomerated due to the Van der Waals forces which cause porosity and degrade the mechanical properties of silicon nitride.

Hvizdos et al. [16] studied the tribological properties of MWCNTs added ceramics including Si_3N_4 under the pin-on-plate configuration test. Very fine nanocrystalline structure were received by suppressing the grain growth by the addition of MWCNTs. More than 60% reduction in friction coefficient was observed. Hvizdos et al. [37] reported the friction coefficient and wear rate of monolithic and CNTs reinforced Si_3N_4 composites. In monolithic materials the dominant wear mechanism was abrasion, in composites with CNT and with higher amount of CNTs (5 and 10%) fiber pull-out and lubricating by the carbon phases occurred which helped in reducing the friction and wear rate.

The influence of CNTs on wear rate and coefficient of friction reported by several researchers [16, 32, 33, 36, 46, 47, 50] and compiled in Figure 1 and 2; but it is difficult to compare the properties of composites produced and investigated by different authors. Because the authors used different processing techniques, parameters and testing methods for Si_3N_4/CNTs composite. In Figure 1 and 2, the CNTs content is given in weight percent (wt. %), a few researchers used CNTs by volume percent (vol. %), but we converted them in to wt. % to compare the results easily.

As a summary, four factors are important in enhancing the tribological properties of CNTs reinforced silicon nitride composites:

1) uniform distribution of CNTs in the matrix,
2) load transfer efficiency,
3) structure stability of CNTs during processing in the matrix,
4) interfacial bonding between CNTs and matrix.

In most of the cases, it was reported that wear rate decreased with the addition of CNTs. The benefits of addition of CNTs can be realized in tribological properties. The uniform dispersion of CNTs act as the lubrication and decrease the coefficient of friction and reduce the wear rate. The main wear mechanism in the nanocomposite is abrasion accompanied with a pull-out of the carbon nanofibers which act in the interface as a lubricating agent. As pulling-out effect is strengthening mechanism and enhance the fracture toughness of the composite, similarly, pulling-out effect is beneficial for tribology.

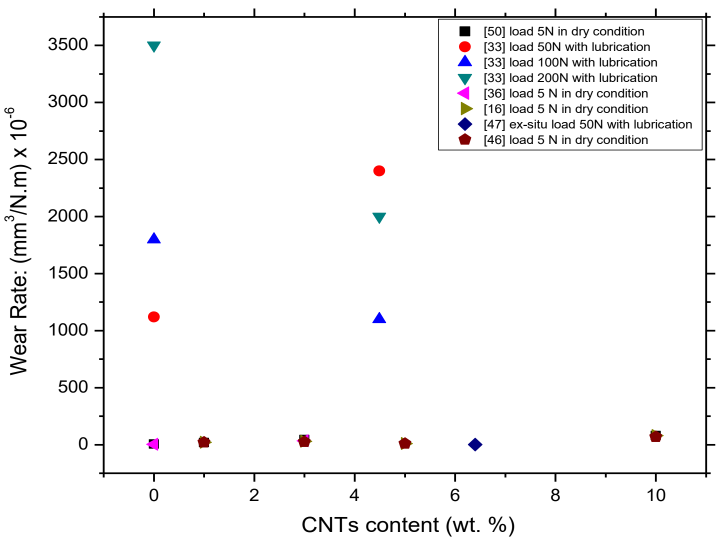

Figure 1 *- Effect of CNTs on the wear rate of silicon nitride ceramics reported by different researchers*

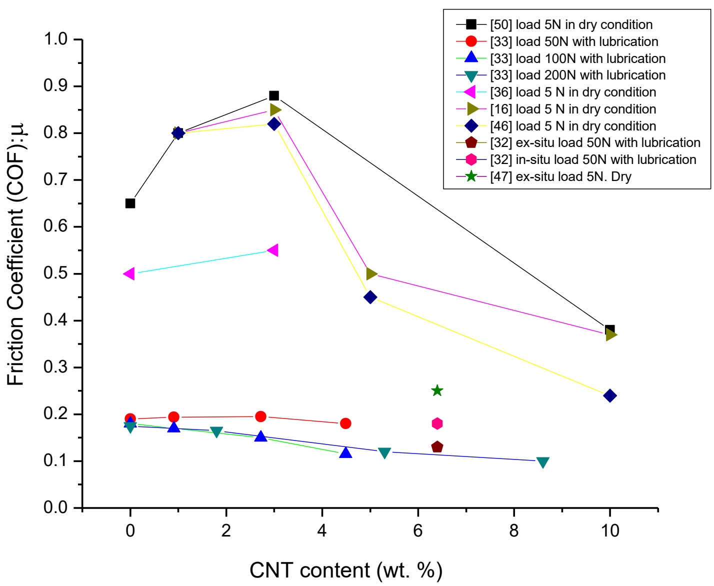

Figure 2 *- Coefficient of friction values with respect to CNTs content were collected from the literature and compiled in one plot for the comparison*

4. Influence of Graphene on Wear Rate and coefficient of friction

Similar to the CNTs-reinforced silicon nitride composites, a homogenous distribution of graphene in composites is critical to exploit of graphene as their anti-wear enhancement additives.

Many researches have been carried out to study the tribological properties of graphene based silicon nitride composites [24-26], [29], [38]. Likely CNTs, uniform dispersion of graphene in silicon nitride matrix is a challenge. Developing homogenized silicon nitride composite with even distribution of graphene is extremely difficult whilst avoiding the defects into graphene [39]. Some solvents are required (such as isopropyl alcohol, *N- methylpyrrolidone* and *dimetyloformamid*) during the colloidal processing to avoid the agglomeration [40]. Another method can be used to get a good dispersion is a hetero coagulation [41].

Belmonte et al. [42] studied the tribological properties of GNPs reinforced Si_3N_4 composites for the first time tested by a reciprocating ball-on-plate configuration under isooctane lubrication. Up to 56% enhancement in wear resistance as observed at high contact pressures. This enhancement was attributed through the exfoliation of nanoplatelets of graphene which would create an adhered protective tribo-film.

Balazsi et al. [24] prepared graphene reinforced silicon nitride by hot pressing for the tribological applications in aqueous environment and reported the decrease of wear with the increase of graphene content in the silicon nitride matrix. The presence of graphene in the wear debris acted as lubrication, reduced the friction and enhanced the wear resistance.

Figure 3 and 4 illustrate the graphene effect on the wear rate and coefficient of friction, the values were collected from the literature [24, 25, 26, 29, 48, 49, 50] and most of the researchers reported the positive effect of graphene on wear rate coefficient of friction. Graphene based silicon nitride composites had lower wear rate and friction than that of the monolithic silicon nitride. The positive effect attributed to the uniform dispersion of graphene nanofillers, exfoliation and good agreement between graphene nanoparticles and silicon nitride grains. Some researchers used graphene by vol. %, and we converted vol. % to wt. % for easier comparison.

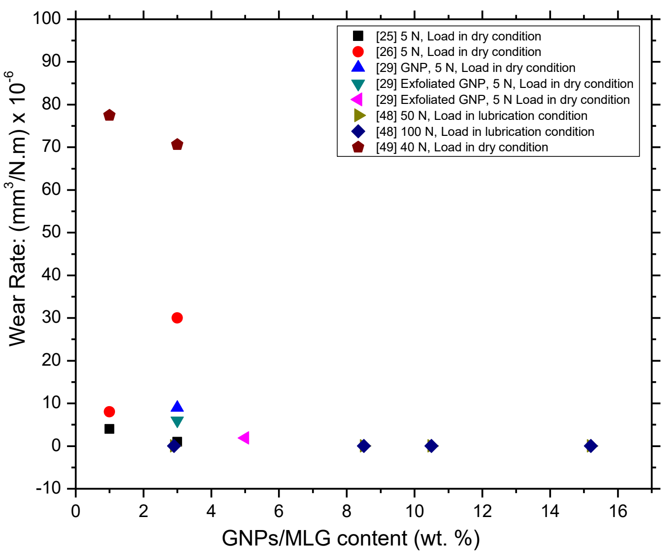

Figure 3 *Wear rate vs graphene content in the silicon nitride matrix composites reported by different researchers*

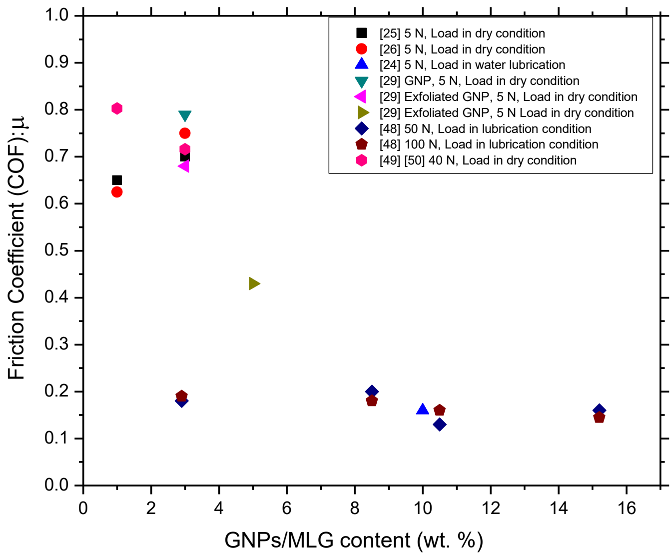

Figure 4 *- Coefficient of friction (COF) vs graphene content in the silicon nitride matrix reported by different researchers*

5. Wear mechanism of carbon nanostructured phases added Si_3N_4 composites

Wear is a deterioration process of a material and progressive loss of material happens during the relative motion of two solids in contact mode. In certain cases, hardness and fracture toughness of silicon nitride are considered the most important properties in meeting wear requirement [43].

The wear rate depends on the degree of abrasive penetration into the surface of the material under abrasion. Particles that cause wear usually have sharp edges to cut or shear the solid under wear [44]. Several wear mechanisms, such as abrasion, adhesion, micro-fracture and delamination, separate or combined, contribute to the wear damage in ceramic-ceramic sliding and rolling contacts [45]. Typical wear mechanism in ceramics can be seen in the Figure 5 [45].

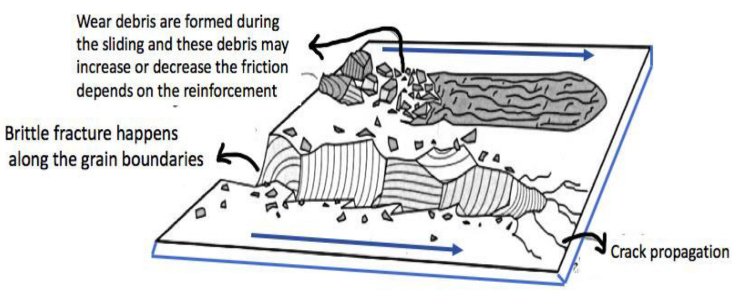

Figure 5 *- Typical wear mechanism in ceramics [45]*

The purpose of carbon nanofillers in the silicon nitride matrix is to reduce the friction and wear rate during the sliding of two surfaces and the carbon nanofillers should be enabled to act as lubrication. Lubrication has three main regimes e.g. fluid film lubrication, boundary lubrication and mixed lubrication. The graphene and CNTs may protect the surface from mechanical and chemical wear and promotes local hydrodynamic lift. This enables a gradual transition from mixed lubrication conditions to hydrodynamic lubrication in tribological system.

The general wear mechanism in monolithic silicon nitride composites, the grains are detached from the surface during the sliding and these grains cause the abrasion and pronounce the effect of wearing. In general, wear debris were formed by the action of the micro-abrasion mechanism, being compacted during the motion of the sliding pairs. If CNTs and graphene are present in the debris wear, then the debris wear serves as lubrication and overcomes friction. One of the examples was observed by the Gonzalez-Julian et al. in in-situ CNTs/Si_3N_4 composites, the debris areas appeared well adhered to the surface which protected it against wear [32].

6. Conclusion

To some extent, efforts have been done to exploit the carbon nanofillers reinforcement in silicon nitride to improve the tribological properties, there is still need of much efforts to optimize the amount of carbon nanofillers in to matrix, uniform dispersion and processing technologies. The use of nanofillers in silicon nitride ceramics to reduce the friction coefficient and wear rate is promising. In future, the materials scientists will be able to develop a robust and super wear resistant material with the addition carbon nanofillers reinforcement.

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