EFFECT OF GRAPHITE ADDITION ON MECHANICAL PROPERTIES OF UHMWPE FOR USE AS TIBIA INSERT BIOCOMPOSITE MATERIALS

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Received: January 05, 2017; Revised: February 22, 2017; Accepted: April 01, 2017

Abstract

In this work, graphite was used as a filler material for ultra-high molecular weight polyethylene (UHMWPE) to prepare graphite/UHMWPE composites for a potential alternative tibia insert for total knee replacement, because graphite has been shown to have excellent solid lubrication and biocompatibility. Graphite powder was blended with 5, 10, 20, 30, and 40 wt% into the UHMWPE matrix. Then, it was compression molded at 250°C for 30 minutes under a pressure of 10 MPa. The mechanical and tribological properties, and the coefficient of the friction of the composite samples were investigated. As the results show, the highest hardness (Shore D) of 62.31, the highest impact strength of 78.51 kJ/m^2, and the highest coefficient of friction were obtained with 5 wt% graphite, while the lowest coefficient of friction was obtained with 20 wt% graphite. It can be concluded that the graphite/UHMWPE biocomposites displayed a remarkable combination of enhanced mechanical properties such as hardness, wear resistance, impact strength, and coefficient of friction making the composites attractive potential candidates as a tibia insert for artificial joints in the human body.

Keywords: Ultra-high molecular weight polyethylene (UHMWPE), graphite, biocomposites, mechanical properties

Introduction

Ultra-high molecular weight polyethylene (UHMWPE) is one of the best engineering thermoplastics that possesses high impact strength, low friction coefficiency, good biocompatibility, high chemical inertness, and the highest wear resistance as compared with other thermoplastics due to its long chain entanglement (Budinski, 1997). For many years, it has been an established material for application as a component of artificial joint replacement in prosthetic joints that can replace human joints degraded by severe arthritis or injuries. Despite its exceptional properties, wear problems that occur after certain service periods still remain as the main challenge. The production of wear debris will cause adverse effects to the human body's system which subsequently leads to osteolysis and aseptic

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loosening of the implant. On this subject matter, it is imperative for research to be carried out in polymer tribology in order to reduce the wear rate of UHMWPE components in artificial joint replacements. There are several methods that have been attempted and studied by researchers in order to improve the tribological properties of UHMWPE. To enhance the performance of UHMWPE in terms of reducing its wear rate and wear particle generation, attempts have been made to improve the lifespan of the component. For example, the Hymaler, a high crystalline UHMWPE (with 73.2% crystallinity) was identified as a potential material for arthroplasty application, since this material presents a high resistance to fatigue and creep propagation. However, it is susceptible to oxidation related degradation, which affects its clinical performance (Baena *et al.*, 2015). This material was replaced by Maraton crosslinked UHMWPE in 1997 (Shi *et al.*, 2004). In regard to a UHMWPE composite, it also has been considered as a potential alternative to improve the wear performance of artificial joints. The UHMWPE reinforced with carbon fibers (CFR-UHMWPE), named Poly II, was used in orthopedic implants in the 1970s. This composite was discontinued due to evidence of reduced crack resistance, rupture of the fibers on the surface, and other issues (Pearle *et al.*, 2005). Lahiri *et al.*, (2014) evaluated the evolution of the wear resistance at the nano-scale by scratching the UHMWPE-GNP composite at different graphene nanoplatelet (GNP) concentrations (0.1, 0.5, and 1.0 wt%) and using 3 different loads (100, 200, and 300 µN) (Lahiri *et al.*, 2014; Baena *et al.*, 2015).

Recently, graphite-filled polymeric materials have been discussed in tribological research in many published articles (Wang *et al.*, 2010; Zouari *et al.*, 2010). The use of graphite as a filler material is known to ameliorate the tribological behavior of polymer matrix composites (Suresha *et al.*, 2007). Difallah *et al.* (2012) evaluated the evolution of the addition of a small amount of graphite which improves the friction behavior and the anti-wear abilities of the acrylonitrile butadiene styrene (ABS) polymer. Graphite strengthens the wear resistance of ABS composites and effectively reduces the adhesive and plowing wear and enhances the formation of a third body with better quality on the sliding stripe. A composite filled with 7.5 wt% graphite corresponds to the best friction and wear abilities (Difallah *et al.*, 2012).

The present study focuses on the improvement of the mechanical properties of UHMWPE such as wear resistance, the coefficient of friction, hardness, and impact strength by the addition of graphite as a filler. Graphite has solid lubrication, biocompatibility, high temperature stability, a low coefficient of friction under high loads, excellent thermal shock resistance, and high chemical resistance.

Materials and Methods

Commercially available UHMWPE powder with a mean diameter of 170 μm (IRPC Public Company Limited, Rayong, Thailand) (0.94 g/cm^3 density) was used as the matrix. Graphite powder with a mean diameter of 80 µm was used as the filler. The graphite particles were mixed in the UHMWPE with 5 to 40 wt% with a dry powder mixer. Composites were formed by using a hot compression molding machine with the pressure at 10 MPa at a temperature of 250°C for 30 minutes. The hardness of the composite samples was measured by means of a hardness tester (Heinrich Bareiss Prüfgerätebau GmbH, Oberdischingen, Germany) with Shore hardness type-D. The study of the friction behavior was performed using a Param MXD-02 coefficient of friction tester (Labthink Instruments Co., Ltd., Jinan, China). Rectangle-shaped specimens were 64 mm long, 4 mm thick, and 39 mm wide, and the contact area was 39 mm × 64 mm in the abrasion wear test. The specimens were slid onto a SiC paper P800 (Ra ~120 nm) and the abrasive was of a median diameter of 25.8 microns. The normal load was 14.71 N for 5 minutes/sample. The study of the impact strength was measured using the Izod mode with an Instron CEAST 9050 (Illinois Tool Works Inc., Norwood, MD, USA) at room temperature using the impact pendulum with an impact energy of 22 joules for the notched specimen. The microstructure of the composite samples was observed by means of a JEOL-JSM-6010 LV scanning electron microscope (SEM) (JEOL Ltd., Tokyo, Japan). The section of the samples was cut under liquid nitrogen as a coolant.

Results and Discussion

The work reports the results of systemic studies of the mechanics and characteristics of the coefficient of friction, impact strength, hardness, and wear resistance of the graphite/UHMWPE composites.

Figure 1 shows the hardness of the graphite/UHMWPE composites with different amounts of graphite. The highest hardness was obtained with 5 wt% of graphite filler, an increase of 0.16% in comparison with ordinary UHMWPE. At the higher graphite content (> 5 to 30 wt%), there was a decrease in hardness due to the decreasing of the UHMWPE matrix as the binding phase. Nevertheless, with a large amount of 40 wt%, the graphite content could not be tested due to the brittleness of the sample.

Figure 2 presents the coefficient of friction of the UHMWPE composites with different amounts of graphite. It can be seen that the increased graphite content reduced the friction coefficient. The 20 wt% of graphite showed the lowest friction coefficient due to the self-lubricating ability of the graphite which easily causes interlayer shearing, whereas its high in-plane strength resists its total disintegration and thus keeps the lubrication long-lasting and

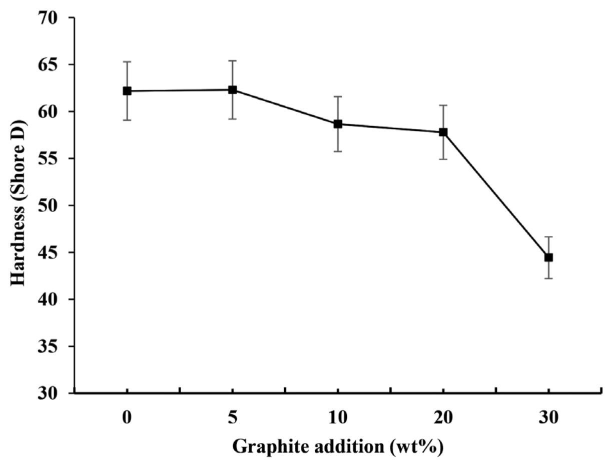

Figure 1. Variations of hardness as a function of graphite addition

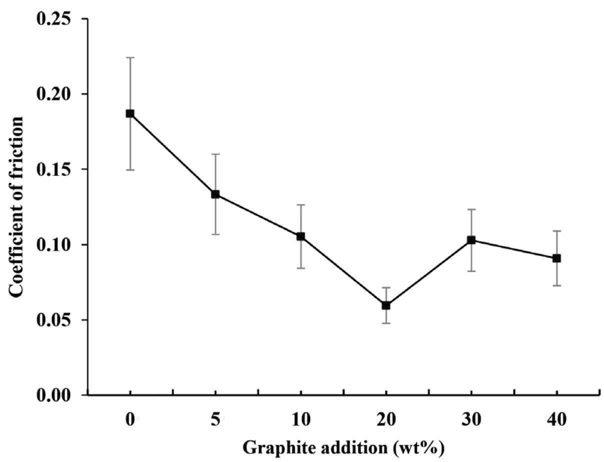

Figure 2. Variations of the coefficient of friction as a function of graphite addition

effective. Corresponding to the work of Difallah *et al.* (2012), the addition of graphite in the ABS matrix exhibits a lower coefficient of friction and the best coefficient of friction was obtained with 7.5 wt% of graphite.

Figure 3 presents the weight loss of the UHMWPE composites with different amounts of graphite. The composite with a 5 wt% of the graphite showed the lowest weight loss because the 5 wt% of graphite addition could reduce the transmission of shear stresses throughout the bulk sample. However, by increasing the graphite content over 5 wt%, the weight loss was increased as compared to pure UHMWPE due to the fact that the crosslinking reduces the distance between the folds caused by decreased polymer chain mobility and, compared with the behavior of UHMWPE reinforced with a graphene nanoplatelet (GNP) content from 0.1 to 1 wt%, it also increases the wear resistance (Lahiri *et.al.*, 2014). Finally, the 40 wt% of the graphite content could not be tested due to the brittleness of the sample.

Figure 4 presents the impact strength of the UHMWPE composites with different amounts of graphite. The highest impact strength, obtained with 5 wt% graphite addition, is 16.4% higher than that of the UHMWPE. However, in increasing the graphite content over 5 wt%, cracks were able to propagate through weak points within the large particle agglomerations, while decreasing the polymer matrix in the composites leads to the

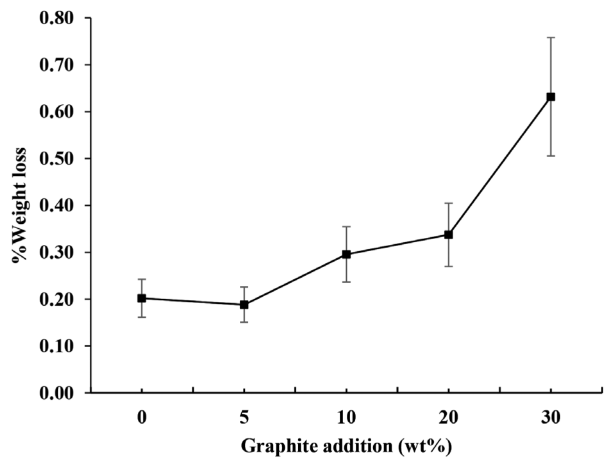

Figure 3. Variations of %weight loss as a function of graphite addition

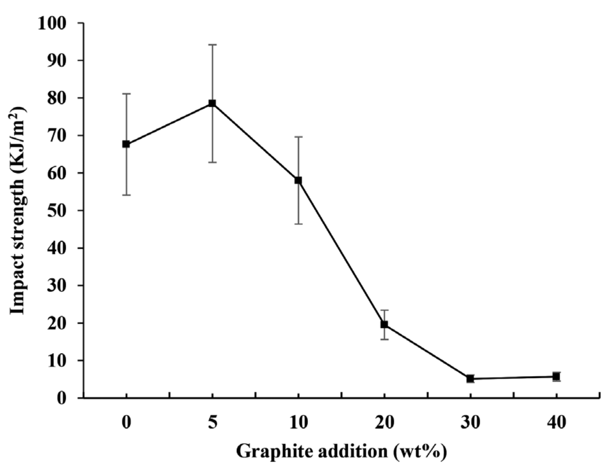

Figure 4. Variations of impact strength as a function of graphite addition

brittleness of the sample and also the poor dispersion of particles in the composites; this creates a path of weak regions within the polymer matrix. When the polymer is cyclically loaded, fatigue cracks can quickly propagate around the boundaries of the graphite particles, creating separated regions which can deform more freely and independently of each other (Plumlee, 2008).

Characterization of the SEM

Scanning electron images were compared to reveal the distribution of graphite particles within the matrix, as seen in Figures 5 and 6. The images revealed that the graphite particles accumulated in long veins that ran through the entirety of the samples. This can be explained by the difference in particle size between the graphite and the UHMWPE during the mixing state. This causes the graphite particles of 80 μm to gather in the empty regions between the larger UHMWPE particles of 170 μm while the groupings were caused by the particle geometries; the graphite particles did not appear to naturally agglomerate together even when in close proximity, suggesting that dispersion could be improved by simply altering the initial particle size of the UHMWPE powder, with a diameter ratio of 1:1, resulting in the most uniform distribution (Plumlee, 2008).

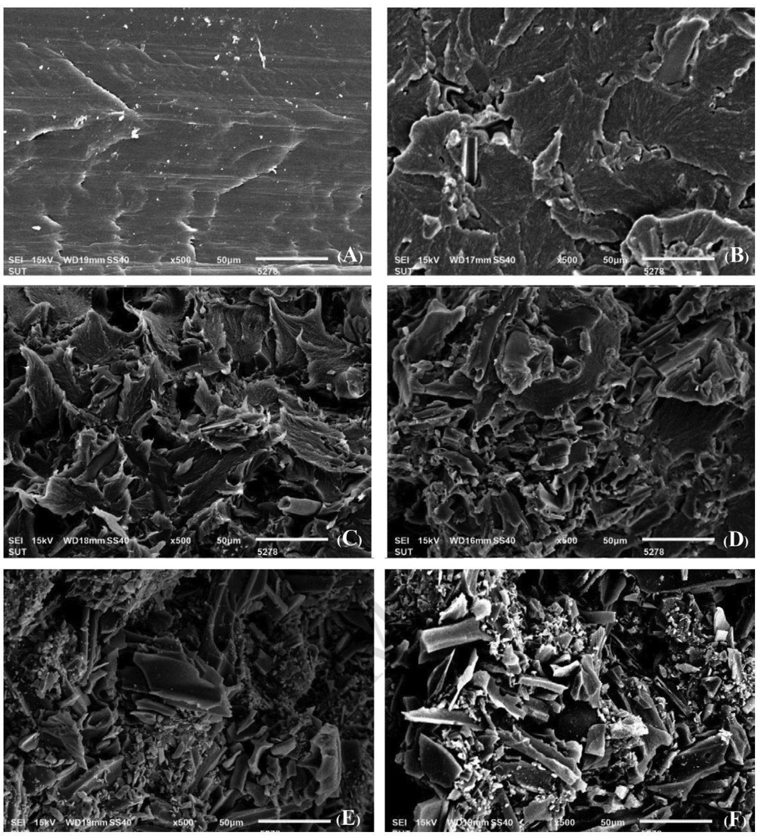

Figure 5. SEM images (magnification = 500x, bar = 50 μm) of the surfaces of unfilled and filled graphite composites:
(A) UHMWPE
(B) 5% Graphite
(C) 10% Graphite
(D) 20% Graphite
(E) 30% Graphite
(F) 40% Graphite

The 5 wt% of the graphite/UHMWPE composites, as seen in Figures 5(b) and 6(h), showed that individual graphite particles appear to be fully encompassed by the UHMWPE. It confirmed that the melted UHMWPE was able to flow around the graphite particles during the molding process leading to 0.16% of hardness and 16.14% of impact strength increasing in comparison with the UHMWPE.

At the higher graphite content of over 5 wt%, the results in Figures 5 and 6 showed that, in the regions between the UHMWPE powder particles, many graphite particles accumulated. The extremely small crevices between these closely packed particles, along with the high melt viscosity of the UHMWPE, lead to voids where the melted polymer could not penetrate (Plumlee, 2008).

Conclusions

The addition of graphite could reduce the coefficient of friction of the UHMWPE. The 20 wt% of the graphite exhibited the lowest coefficient of friction as compared to the UHMWPE. A decrease in the coefficient of friction is possibly due to the decrease in shear strength. 5 wt% of the graphite/UHMWPE composites exhibited the lowest %weight loss, creating an increase of 0.16% of hardness and

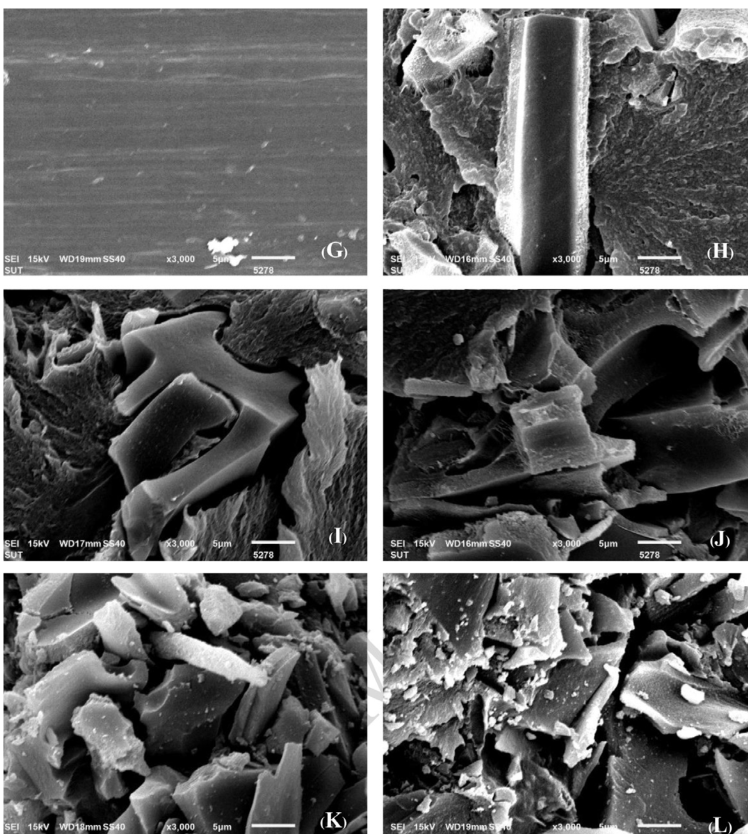

Figure 6. SEM images (magnification = 3000x, bar = 5 µm) of the surfaces of unfilled and filled graphite composites:
(G) UHMWPE
(H) 5% Graphite
(I) 10% Graphite
(J) 20% Graphite
(K) 30% Graphite
(L) 40% Graphite

16.14% of impact strength in comparison with the UHMWPE. For the most uniform dispersion, the UHMWPE particle size should be equal to the graphite particle size, and the results showed that the addition of graphite could improve the friction behavior and the anti-wear ability of the UHMWPE to yield longer performance of a tibia insert.

Acknowledgments

The authors would like to thank IRPC Public Company Limited for support with the ultra-high molecular weight polyethylene powder. This research work was financially supported by Suranaree University of Technology (SUT).

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