TRIBOLOGICAL BEHAVIOR OF ULTRA-HIGH MOLECULAR WEIGHT POLYETHYLENE NANOCOMPOSITES

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Graphical Abstract

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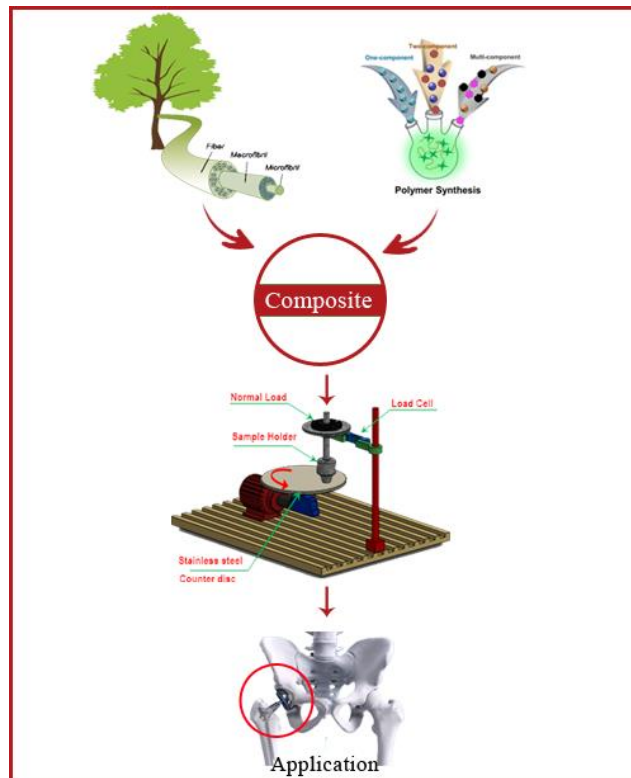
ABSTRACT
Thermoplastic polyethylene (PE), such as ultra-high molecular weight polyethylene (UHMWPE), is commonly used in industrial applications as bearing materials. UHMWPE has also been used effectively as a main ingredient in artificial knee and hip joint replacements due to its superior wear resistance and low friction surface properties. However, the wear debris produced during joint motions can cause osteolysis and implant loosening, contributing to the primary reason for joint revision. Many attempts have been made to increase the wear resistance of PE materials in order to extend the service life of the material. Carbon nanofibers, carbon nanotubes, nano-alumina, and nano-montmorillonite have all been used as suitable reinforcing materials to alter the tribological properties of PE. This article summarizes the current state of research in tribological studies of UHMWPE-based nanocomposites, focusing on UHMWPE nanocomposites, including nano-filler forms, fabrication methods, and tribological characterization approaches. The dependence of the tribological properties on nano-filler concentrations and modifications, as well as other tribological parameters, is summarized.

KEYWORDS
Ultra-High Molecular Weight Polyethene (UHMWPE); Tribology; Artificial Joint.

INTRODUCTION
Despite the fact that tribological phenomena are normal in daily life, the term tribology remains unknown to most people. Consequently, it is customary to describe tribology in its literal sense before discussing it with non-tribologists. Tribology is characterised as the science of rubbing surfaces and is derived from the Greek words tribos, which means rubbing/friction, and logos, which means science. In other words, it's the science of interacting surfaces in relative motion, and it covers a broad variety of topics including friction, wear, and lubrication. [1]. The chair on the floor, the blinking of the eye, the tyre rolling over the roadnd so on are all examples of tribology. When communicating surfaces are found on the animal or human body, such as artificial joints, the term biotribology is commonly used.

Total joint arthroplasty is a procedure that replaces the joint in the body. This technique entails removing the natural joint and replacing it with an artificial joint in order to regain the patient's functions and alleviate discomfort for a long time, [2]. As many as 400,000 total knee and hip replacements are done per year in the medicare community, this is projected to rise by 160% within the next 20 years, [3 – 5]. In clinical practice, the significance of tribological efficiency of an Artificial Joint Replacement (TJRs) is well understood. Despite the fact that joint arthroplasty is regarded as one of orthopaedic surgery's major achievements, implant wear remains a critical problem that restricts the implant's lifetime to 10-15 years, [6]. Lifesaving joints are in demand for more than one purpose, such as sports injuries and because of the ageing population This is supported by the fact that the general population is now living longer, and demand for prostheses with a longer life expectancy is on the rise, [7].

In artificial substitution, polymer materials have become the preferred alternative due to their advantages such as Light weight, self-lubrication, corrosion resistance, and processing ease are just a few of the advantages. This 1960, Charnley preferred polytetrafluoreether (PTFE) for plastic acetate for the flexible cup because of its lower coefficient of friction (COF), [8]. It was then decided to use high-density polyethylene (HDPE) for arthroplast, as it has lower friction and wear, [9 – 12].

Since 1962, ultra-high molecular weight polyethylene (UHMWPE) has acted as the bearing material for tribology applications. where wear and friction are issue because it is a special polymer that outperforms all other polymers in terms of wear resistance. Because of its wear-resistant properties, hardness, low friction, and biocompatibility are some of the characteristics of this material, UHMWPE has become one of the most modified polymers in the manufacture, replacing existing traditional polymers. So, Biomedical materials for TJRs, [13 – 15], engineering bearings, [16 – 17], automotive, valves, [18], are only a few of the many uses for UHMWPE. It has thus become the preferred substrate for bearing surfaces in TJRs, especially in the last 30 years,[16, 19 - 21].
Polymer composites have increasingly been used to replace metal materials in engineering applications entailing tribology (wear and friction) in recent years. Nano cellulose, which is highly mechanical superior and has strong compatibility with most polymers in the world is one of the most abundant bio-nanofiller. It was used therefore in very broad applications, in particular biomedically.

There are still large numbers of total hip and total knee replacements that use a combination of metal and other biomaterials today, but the most commonly used of these is UHMWPE. But, still wear debris was created in spite of the excellent anti-wear properties of UHMWPE, causing osteolysis, necessitating removal, and replacement of the implant, [22, 23]. Because of UHMWPE's viscoelasticity, this material often had some inherent disadvantages, such as poor creep and fatigue strength in comparison with the metal and cortical bone, [24]. As several attempts have been made to enhance wear resistance and anti-creep properties of PE, people have looked to discover how to prolong the existence of man-made joints. Ageing the cross-linked density to XLPE, known as the "cross-link polyethylene" (the improved wear resistance of UHMWPE) has been viewed as a promising strategy at first, [25]. Some approaches used crosslinking of the UHMWPE including of chemical and ionizing radiation techniques used, such as using peroxides or silane treatments, [26 – 28]. Of in hip replacements, the crosslinked UHMWPE showed superior wear-resistance in joint simulators and has thus been widely used in the United States. While the increase in material wear resistance that the cross-linking treatment provided was a good, the ductility and yield strength were adversely affected as well. And decreased toughness, [29, 30], which increased the risk of acetabular cup wear and which could have eventually lead to the cup's catastrophic failure, [31]. Still, other attempts were made to improve the crystallinity, [32], as well as the use of different fillers , and to strengthen the PE by itself, [33, 34]. Polymer composites have increasingly been used to replace metal materials in engineering applications entailing tribology (wear and friction) in recent years. Polymers. The argument that one bearing material is superior to all others is still awaits definitive proof.

Reinforcement with polymers is an effective method for improving the properties of UHMWPE, [35]. Many reinforcing materials have been used to increase the wear resistance of UHMWPE, including zinc oxide particles, glass, carbon nanoparticles, and others, [36 – 38]. Organic inorganic Particles like alumina, [39], silica, [40], and hydroxyapatite, [41], have been confirmed to improve bone health. Mechanical efficiency and wear. UHMWPE fibers are also considered as high-performance fibers for various applications due to their high mechanical performance with low density, [42]. The word "natural fibers" referred to fibers derived from natural sources. Extraction from natural sources is a speculative concept that various scholars interpret differently. They do, however, share an overarching concern about 'extraction,' which includes organic, regenerative, naturally abundant, cultivable, biodegradation, environmentally sustainable, causing little or minimal harm to the ecosystem, green, and so on. Plants, animals, minerals, and other natural sources are examples. However, the focus of natural fibers is skewed more toward plant sources than animal and mineral sources. This is because a higher percentage of natural fibers are derived from plant sources (80 to 85 percent of global natural fiber production), [43 – 45].

Although natural fibers can be extracted from animal and mineral sources, they cause more environmental damage than natural fiber extracted from plant sources. The size, texture, and form of natural fibers, as well as the type of thermoset, all influence how natural fibers reinforced thermoset polymers are processed. Fibers are usually short, long, or mat-like. Thermosets, also known as resins, are liquids in which short fibers are randomly impregnated or resin-saturated long continuous fibers are deposited layer by layer in unidirectional, bidirectional woven mat shape and allow for curing at ambient temperature or elevated temperature with or without pressure. Polymerization of resins fully solidifies the fiber-impregnated resin to composite. Hand lay-up, spray lay-up, and hot compression molding are all low-cost and widely used techniques for producing unsaturated polyester and epoxy composites. For higher quality epoxy or phenol formaldehyde composite materials, however, more complicated and costly techniques such as resin transfer molding (RTM), pultrusion, and vacuum bagging are used.

Since thermoplastic resins are solid, dispersion and distribution of short fibers in plastic matrix or wetting of long continuous fiber by thermoplastic are critical issues in composite fabrication. Short fibers are combined with plastic resins in a cyclone vortex or internal mixer, where both the fibers and the thermoplastic are premixed and transformed to pellet form. These pellets are then used in injection molding or hot compression molding. For long or mat fibers, hot compression molding or thermoforming processes with different time and temperature interval stages are used to ensure proper thermoplastic wetting of the fibers. Natural fiber thermoplastic composites typically have high manufacturing costs due to the use of costly machinery such as extrusion, injection molding, or hot compression molding machines, however since natural fibers are less abrasive, the equipment maintenance costs are low in composite production. Typically, engineering thermoplastics need high temperatures in the range of 200–350 °C, which is incompatible with natural fibers, which degrade at or above 180 °C. As a result, the use of engineering thermoplastics is restricted, whereas PE, PP, and PU, which have low processing temperatures, are used for lignocellulosic fibers. Some popular natural fiber

compatible thermoset and thermoplastic products generated using various processing techniques, [46].

Carbon nanoparticles such as carbon nanotubes (CNT), carbon nanofibers (CNF), graphene, and nanodiamonds are used as reinforcement materials to improve the mechanical properties of UHMWPE and to extend the lifespan of implants. CNTs' unique optical, electrical, mechanical, and thermal properties, as well as their use in composites, have received a lot of attention, [47]. CNT is an effective additive for polymer composites to increase wear resistance, [48]. The dispersion of CNT into polymer materials, as well as the interaction between the CNT and macromolecular chains, are critical factors in transferring CNT properties to polymeric matrix, [49]. To investigate the wear of UHMWPE under lubricated conditions, Liu, [50], mixed three forms of CNTs and nacre into UHMWPE coated with Perfluoropolyether (PFPE). UHMWPE coated with PFPE and strengthened with nacre wore out faster than plain UHMWPE. The dispersion of all forms of CNTs into UHMWPE increases the wear rate of the material. The explanation for the increased wear rate is that non-covalent stresses and merely shear interactions between UHMWPE molecules and CNT, as well as CNT dispersion into UHMWPE, cannot increase the necessary energy dissipation to decrease the buildup of plastic strain. The hardness value was reduced by reinforcing PFPE with UHMWPE, most likely due to softening of the UHMWPE surface and decreased resistance between the material and indenter surface. Because of the inclusion of hard nacre particles, the dispersion of nacre into UHMWPE and PFPE coatings increases hardness value slightly. Figure 1 depicts the data.

Under water-lubricated sliding conditions, Golchin (51) investigated the tribological efficiency of nanodiamond reinforcing particles in UHMWPE. Nanodiamond dispersion in UHMWPE greatly decreased friction and wear properties. Nanodiamond, with its nanoscale dimensions and semi-spherical morphology, functions as bearing balls between the tribo surfaces, allowing detached wear debris to roll. This increases direct asperity-asperity interactions while decreasing friction between the mating surfaces. Because of the decreased friction force, the shear stress acting on the UHMWPE chains is reduced, resulting in a decrease in the wear rate of the tribo-pairs.

CNF, like CNT and nanodiamond, has received a lot of attention as a polymer reinforcement due to its promising intrinsic properties and strong compatibility (52, 53). Galetz (54) studied the mechanical properties of CNF dispersed in extruded UHMWPE. The yield stress and modulus were increased while the ductility was maintained. Furthermore, the hardness of UHMWPE was increased due to the smoothness of the surface given by nanoscale particle reinforcement. Sui (55)incorporated CNF into UHMWPE. The incorporation of CNF increased the tensile modulus and tensile strength of the UHMWPE/HDPE, whereas it decreased at higher material. The effect of inter-ply stacking positions on the mechanical efficiency of hard ballistic UHMWPE/carbon fibre composites was investigated by Zulkifli(56). By varying the slight change in orientation of carbon fibres, major variations in back-face signature, flexural yield intensity, and ballistic impact were observed. The results show that a strategic sequence of carbon fibres in UHMWPE can improve ballistic efficiency.
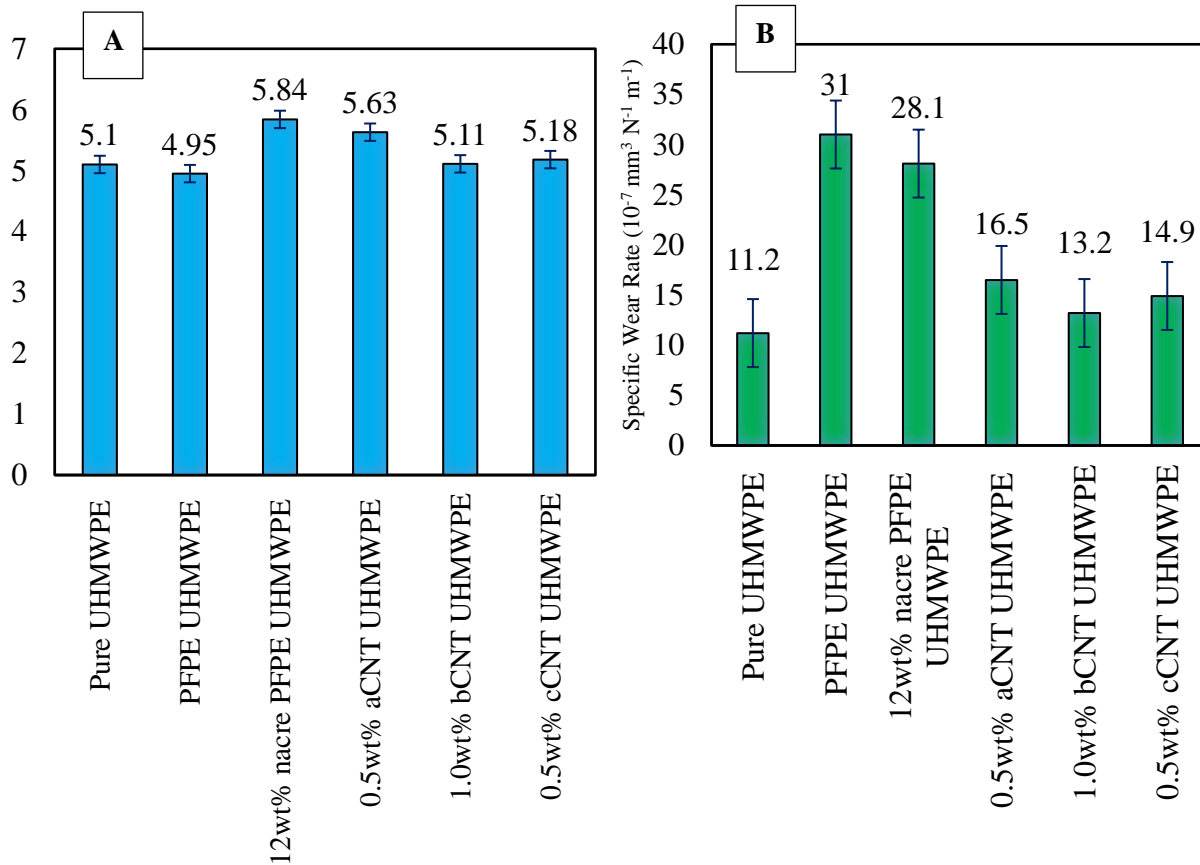
Fig. 1 (a) Vickers hardness values for different UHMWPE composites, (b) Wear rate values for different UHMWPE composites. Reprinted from, [50].

Because of its excellent thermal, mechanical, and electrical properties, graphene has received a lot of publicity as a filler material in UHMWPE, [57]. According to Puertolas [58], the use of graphene as a filler in UHMWPE could result in an improvement in mechanical properties. The filler content of graphene was discovered to be very important for the strengthening of UHMWPE, since fillers improve mechanical efficiency at low concentrations; however, after this preliminary point, the material showed a decrease in performance with increasing filler content. Mechanical properties in nanocomposites achieve their best values at an ideal filler concentration, which is not always the same for the various mechanical parameters, regardless of the graphene product used. Aliu, [59], created composites of graphene nanoplatelets (GNPs) and UHMWPE to improve tribological properties. When 0.25 wt. % GNPs particles were added to UHMWPE, the wear rate was reduced by 31% when compared to pure UHMWPE. Figure 2 depicts the friction response of GNPs/UHMWPE composites measured at 0.1 m/s velocity and 8 MPa pressure. With the addition of GNPs, the COF increased to 0.24. This is due to GNPs anchoring the UHMWPE links, stopping them from slipping over each other. Alam (*60*) investigated the effect of GNPs in UHMWPE. A substantial increase in electrical properties at 3–10 wt. % GNP concentration of GNP/UHMWPE composite demonstrates that such composites are useful for smart biomedical implants as crystallinity and elastic

properties increase. Furthermore, the addition of graphene to UHMWPE improved thermal stability as compared to pure UHMWPE, [61].

Carbon nanoparticle concentration, orientation, and distribution state are critical factors in the reinforcement of UHMWPE, [62, 63]. Because of its high melt viscosity, dispersion of carbon nanoparticles within UHMWPE is difficult. Baena, [64], investigated the effect of multi-walled CNT dispersion state in UHMWPE on tribological and mechanical efficiency. The variance in the content of multi-walled CNTs improved tribological performance; however, this improvement was less apparent due to defects caused by multi-walled CNT agglomeration.
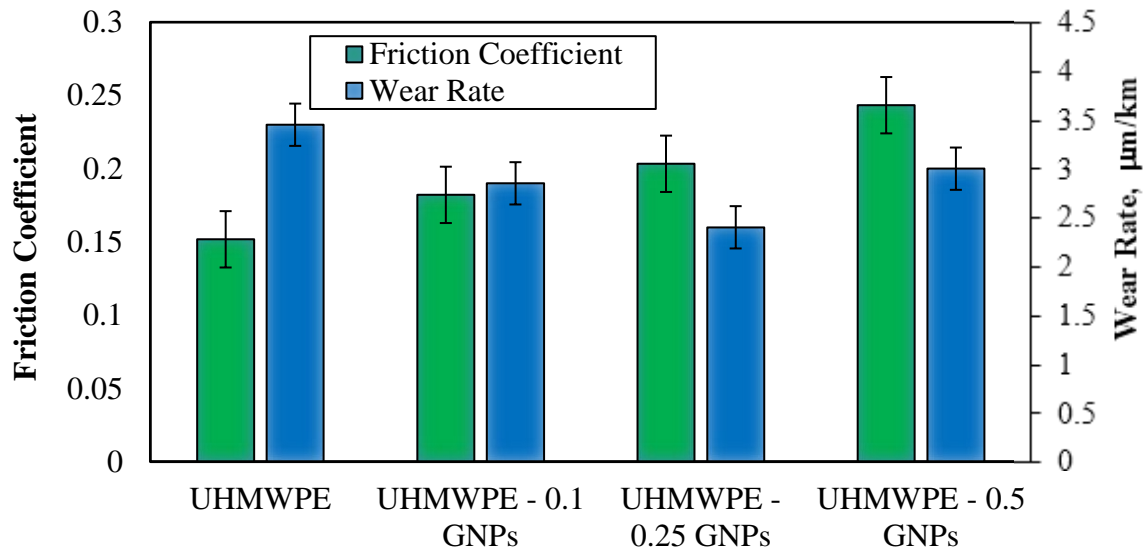

Fig. 2 COF and wear rate of pure UHMWPE and UHMWPE graphene nanocomposites, Reprinted from [59].

The addition of soft particles improves the viscoelastic behaviour of UHMWPE, while the addition of hard particles improves the load-carrying capacity and wear resistance of UHMWPE. It is critical to remember that the form, size, and type of the filler, as well as the filler–matrix interaction, filler loadings, and filler dispersion into UHMWPE, all play a role in determining the wear behavior of UHMWPE composites. Table 1 shows the effect of particle reinforcement on crystallinity, tribological and mechanical efficiency as stated in previous studies.

Table 1 The effect of particle reinforcement on crystallinity, tribological and mechanical efficiency.

Ref.	Reinforced Particle	Concentration/Size	Crystallinity	Tribological Results	Mechanical Results
[65]	Polyimide	10–90 wt.% Optimum-50% wt.%	Increase in crystallinity and stability	COF 65–75% Wear rate-15%	-----

[51]	Nano-diamond	0.5, 1 & 2 wt.% 30–40 nm Optimum-1 wt.%	97.8%	COF-76% Wear rate-28%	Yield Stress-No change Micro Hardness-97.6%
[66]	Carbon Nanotubes	0.1, 0.45 & 0.5 wt.%, Optimum-0.1 wt.%	3% decrease in melting peak.	Wear rate-118%	Micro Hardness-100.2%
[67]	Zeolite	10wt.% 20wt.% Optimum-10 wt.%	-----	COF-approx. 80–90% Volume loss-approx. 80–85%	Tensile Strength-89% Impact Strength-125% Modulus-131% Elongation-89.2%
[66]	Nacre coated with PFPE	12 wt.%	12% reduction in melting peak	Wear rate-251%	Micro Hardness-114%
[56]	Carbon Fibers	Variations in no. of layers Optimum-CF/UF/CF-2/12/2			Flexural Strength-509% Flexural Modulus-284% Ballistic Limit-91%
[68]	Nanoclay	0.5, 1.5 & 3 wt.% Optimum-1.5 wt.%		Wear Life-greater than 10,000 cycles	Hardness-134%
[64]	Multi-walled carbon nanotubes	0.1, 0.5 & 1 wt.% Optimum-1 wt.%		COF-approx. same Wear Rate-74%	Hardness-105%
[69]	Polyethylene glycol (PEG)	Best UHMWPE/PEG ratio 60/4		Shear viscosity-33.3% Storage Modulus-25.5% Loss of modulus-68%	Flexural strength-79.8% Flexural Modulus-77.5%
[60]	Graphene nanoplatelets (GNP)	0.1 wt.% to 10 wt.% Optimum-0.5 wt.%	Crystallinity %-103%		Elastic modulus-130% Yield strength-113% Tensile strength-75% Toughness-76%
[70]	Aramid	2, 3 & 5 wt.%		Roughness-172% Specific wear-60% COF-107%	Hardness-700%
[70]	Poly-tetra-fluoro-ethylene	2, 3 & 5 wt.% Average		Roughness-159% Specific wear-83% COF-91%	Hardness-500%

[47]	High density polyethylene (HDPE)	20, 40, 50, 60, 80 wt.% Optimum- 50 wt.%			Tensile yield stress. 86.3% - Tensile strength-69.5% Strain at the break. 380%
[71]	SiO$_2$ nano-spheres	0.5, 1, 2, 4 wt.% Optimum-1 wt.%	Degree of crystallization %-96%	COF-50% Volume wear rate-29.4% Mass wear rate-90%	
[72]	Alendronate sodium (ALN)	1.0 wt.%		COF-approx. 90% Specific wear rate- approx. 110%	Young's Modulus-97.5% Micro-hardness-96.8% Tensile strength-84.4%
[73]	Carbon Fibers (CF)	5–30 wt.% Optimum-20 wt.%		COF-139% & 220% Wear Volume-20% & 35%	Hardness-140%
[74]	Nano ZnO	5–20 wt.% Optimum-10 wt.% Size-<100 nm		Wight loss (mg)-58.5% COF-100%	
[75]	Hydroxyapatite (HA)	4.7–22. wt.% Optimum-22.8 wt.%			Modulus-888% Yield strength-104% Elongation at break-74%
[18]	kaolin	Size-10 µm 11–26.5 wt.% Optimu-20 wt.%		COF-87% Wear rate-56%	
[61]	Graphene	0.5–3 wt.% Optimum-0.7 wt.%	Degree of crystallization (%)-101%		Linear weight loss temperature-102 % Micro-hardness-110% Toughness-55%
[76]	Talc	10 & 20 wt.% Optimum-20 wt.%	Degree of crystallization (%)-108%		COF-55% Wear rate-50%

Chang, [67], investigated the tribological and mechanical efficiency of UHMWPE with zeolite, particles. By varying the concentrations of zeolite particles, elongation at break and tensile strength were reduced while modulus was increased. Furthermore, the COF was reduced with the addition of zeolite. Furthermore, zeolite reinforcement resulted in smoother surfaces and shallower grooves. The counter surface of UHMWPE was rough, partially coated, and discontinuous for lubricant film transfer, while the counter surface of zeolite/UHMWPE was smooth, covered, and continuous. Overall, the incorporation of zeolite into UHMWPE improved tribological efficiency significantly.

Chen, [65], investigated polyimide/UHMWPE composites for the creation of matched sliding materials. The friction efficiency of the mating pair decreased as the polyimide concentration increased, while surface roughness and wear rate increased. In terms of

surface roughness and wear rate, the optimum polyimide concentration was 50 wt. Because of their strong barrier and mechanical properties, particle size, morphology, abundance, and low cost, nanoclays are also a very promising filler material. The addition of 1.5 wt. % nanoclay greatly improved the wear life and mechanical properties of UHMWPE, [68]. Gurgen, [70], discovered that UHMWPE interacts well with aramid additives, resulting in lower wear rates for aramid/UHMWPE composites. Because of the increased moulding pressure during the manufacturing stage, microstructural consolidation is increased, resulting in a wear resistant PTFE/UHMWPE composite. Because of its lower COF, PTFE has a lower frictional interaction in sliding conditions. Shi, [77], discovered that SiO_2 nanospheres (SNS)/UHMWPE composites have significantly better tribological properties than pure UHMWPE. Huang, [72], published his findings for Alendronate sodium (ALN)/UHMWPE composites. Low COFs were observed at lower loads, while COFs at higher loads were higher than those of UHMWPE in deionized water and saline.

Chang, [74], claimed that incorporating micro and Nano-ZnO with different filler loadings in UHMWPE matrix can improve composite wear behaviour. Under sliding speeds of 0.033 and 0.368 m/s, UHMWPE composites with 5–10 wt. percent micro-ZnO fillers exhibit the lowest weight loss, while Nano-ZnO/UHMWPE composites with 10 wt. percent exhibit the lowest weight loss. Weight loss increases as applied loads and sliding speeds increase for both micro and Nano-ZnO/UHMWPE. Prasad, [78], investigated the impact of micro and nano ZnO material in UHMWPE and discovered that UHMWPE with 5% nano ZnO had the lowest wear rate. When nano-ZnO/UHMWPE composites were compared to micro-ZnO/UHMWPE composites, smooth worn-out surfaces were observed.

UHMWPE has low processability due to high melt viscosity caused by high inter-chain entanglement density, [69] . The most popular approach for improving processability is to blend UHMWPE with a viscosity polymer. Many researchers combined UHMWPE with polyethylene, polypropylene [79, 80], polysilane and paraffin oil, [81, 82], or organic clay, kaolin, organic montmorillonite, and other materials (*83, 84*) Polyethylene is the most desirable of these due to its better miscibility and structural similarity. Khashoggi combined UHMWPE and HDPE and discovered that increasing the HDPE material reduced viscosity, storage modulus, and loss modulus. The mechanical strength, [83, 84], was also reduced as the HDPE content was increased. Li, [69] found that when UHMWPE was blended with PEG, the chain entanglement level of the blended UHMWPE was lower. The addition of HDPE to the UHMWPE/PEG composites improved their processability and mechanical properties. When compared to UHMWPE/PEG (100/4), the flexural modulus, flexural strength, and tensile strength of UHMWPE/HDPE/PEG (60/40/4) were increased by 32.5 percent, 25.7 percent, and 13.8 percent, respectively.

CONCLUSIONS
The development of UHMWPE-based nanocomposites for tribological applications is a novel and exciting area. The addition of nano-fillers has been shown to be an efficient method of improving the wear resistance of UHMWPE-based polymer matrix without losing other mechanical properties. Many nano-fillers, including ceramic and clay nano-

fillers, metal-based nano-fillers, and carbon-based nano-fillers, have been tried to reinforce UHMWPE matrices. Due to Van der Waals forces and high surface energy, nanofillers are extremely difficult to disperse uniformly in polymer matrices. The agglomeration of nano-fillers has been identified as a common issue that impedes the achievement of desired properties in the development of high-performance polymer nanocomposites. To improve the wear efficiency of the nanocomposite, it is critical to ensure a good dispersion of nano-fillers in the UHMWPE matrix.

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