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Nano-Particles Additives as a Promising Trend in Tribology: A Review on their Fundamentals and Mechanisms on Friction and Wear Reduction.

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Abstract: Nanoparticles (NPs) additives have gained notable influence in technology advancements owing to their excellent physiochemical properties with enhanced performance in application compared to previously used additives. The field of tribology has contributed significantly towards upgrading the overall engine efficiency through use of lubricating materials. The outstanding technique in achieving this is through adoption of additives derived from nanotechnology capable of preventing friction and wears during engine operations. Since only base oil cannot withstand most of the operating conditions, suitable additives are formulated and blended as to enhance the tribological properties. According to some research, nanoparticle additions have a greater impact on lubricant improvement. The ability to design NPs additives with unique qualities raises the value and demand for such class of products. The purpose of this study is to highlight the promising properties of NPs additives, mechanisms and to define the specific knowledge gaps related to the size of NPs towards friction and wear reduction. The function and mechanism performance of NPs additives during operation, such as mending, rolling, film formation, and polishing effect, is determined by their types. The excellent load carrying capability of nanoparticles during lubrication reflects their outstanding performance. The review presents an overview of the history and classifications of NPs and concisely elucidate their tribological effect when applied in lubrication. Excellent function of nano additives is the attribute of its nanoscale (1 to 100 nm) nature, thus categorized base on their size, shape, origin and composition. Certain expected characteristics of lubricating oil like friction and wear resistance, surfactant operation, load carrying capacity, extreme pressure operation, etc. were achieved through inclusion of nanoparticles additives.

Keywords: Nano particle additives, classifications, lubrications, mechanisms, friction and wear effect.

1. Introduction

In tribology, improving the tribological properties of lubricant has been a serious concern as it affects the working operation of the machine both in power output ^{1,2} and machine elements ³⁻⁵. In recent years, the field of tribology has received rapid positive attention since poor lubrication has evidently show loss of energy and waste of resources resulting from severe friction and wear ⁶⁻⁹. Solving this unwanted friction and wear during system operation, which stands as the most confronted challenges

during system working operation ¹⁰. Tribologists have come up with three possible approaches in mitigating the problems. These approaches cover all the tribological system both elements and medium involved during mechanism ^{8,11}. First was on machine element surface topography, physio-chemical aspects of solid surfaces and surface interactions as to know suitable material to be used. According to ¹², achieving smoother topography of gears to superfinishing level ,provide great reduction in friction

to about 30%. Second thought of system working design and configuration which covers sliding and rolling contact together the operating speed, size and angle of attack (area in operation)⁸⁾. The last, emphasized on the lubricant nature adaptation considering boundary, hydrodynamic, solid-film and hydrostatic conditions; thus, providing optimum performance. In addition, friction condition on elements with new improved surface coating show significant decrease of about 10-50% on boundary lubricated contacts and about 90% for dry contacts^{13,14)}. In all these, lubricant adaptation and nature yield optimal result and nano-particle technology stands as promising approach over every other mentioned approaches based on its overwhelming potentials towards friction and wear reduction^{3,15,16)}. Among all these, fast ability of nano-particles additives to build geometries or layers, withstand heat, which cannot easily achieve by any other lubricating mode, distinguished it from other counterpart^{3,17,18)}.

Engineers and researchers have focused on enhancing existing facilities and development of new techniques with formulation of novel materials for better machine operation, along with quality lubricant materials being utilized¹⁸⁻²¹⁾. In the development of lubricant additives, nanoparticles are a relatively new class as described in Fig. 1. Advanced and modified lubricants can improve productivity via energy saving and extend the life cycle of every machinery. As a result, the objective of this study was on the prevalent properties of nano-particle additives, as well as the principles that explain their superiority and dependability in the field of tribology. Understanding the lubricant, friction, and wear relationship during lubrication is important. According to²²⁾, tribological system in operation consists of four supportive elements as illustrated in Fig. 2. The two opposite elements surfaces slide against each other, represent the two bodies in operation. Another inclusive element, comprises of the environmental conditions such as humidity and temperature²²⁾. The third is the interfacial medium, termed the lubricant (liquid, solid, gas or combined state), however, the developed intermediate layer of film stands as the fourth element and highly important during lubrication¹⁹⁾. The response of these aforementioned tribological elements from their interactions and properties yield the resulting friction and wear thus not intrinsic material properties of the system.

Presence of material with good thermal properties and lubricant enhancement features, between the two sliding surfaces in contact, apparently is the possible method of mitigating the unwanted friction, thus prevent wear occurrence^{5,10,16,23)}. Furthermore, the use of a proper lubricant in both industrial and domestic applications can serve as debris removers and provide cooling to the machine through surfactant operation²⁴⁾. The inability of the lubricant (base oil and additives) to perform the designed function, lead to manifestation of friction and wear during machine operation^{15,22,25)}. Knowing that friction arises from resistance to motion of a body in

sliding or rolling contact with another body whether static or dynamic and proportional to the applied force, but independent of the rolling contact surface area^{26,27)}. It is very empirical to understand the cause of friction and wear as to ascertain the suitable lubricant and form to be applied. The following conditions should be well understood and considered. During the sliding mechanism together lubrication, friction takes place over little contact area with temperature at the sliding asperity contact increase to severe rate^{22,28)}. With this excessive heat if not controlled by lubricant, could lead to micro-structural changes to asperities with local melting. However, during sliding and lubrication, the main area of contact is influenced mainly by the applied force (load) and speed, thus with inadequate lubrication could cause plastic deformation until the area is great enough to support the load and speed²⁹⁾. Also at the asperity point with inadequate lubricant, adhesions and welding together among the two surfaces and only the frictional force required to shear the formations⁹⁾. Wear on the other hand refers to as physical loss of machine element materials resulting from poor function of interfacial fluid film of the lubricant¹⁰⁾. The wear of element materials arises from different factors like; contact fatigue, adhesion, corrosion and abrasion resulting from poor lubricant (base oil and additives)³⁰⁾. Solving most of these wear occurrences during sliding operation centered on selecting materials that do not bond together easily, increasing the lubricant film thickness and using improved materials with low shear strength additives layer^{9,31)}. Following all these aforementioned properties required, nano materials provide and stands as last resort in solving the friction and wear challenges during machine sliding operation.

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operation centered on selecting materials that do not bond together easily, increasing the lubricant film thickness and using improved materials with low shear strength additives layer^{9,31}). Problems of poor contact separation are always prevalent in lubricants without nanoparticles, however nanoparticles were discovered to have the

potential to penetrate contact areas, resulting in body to body separation. Following the requirements for all of the aforementioned features, nanomaterials provide and serve as a final resort in resolving friction and wear challenges. during machine sliding operation

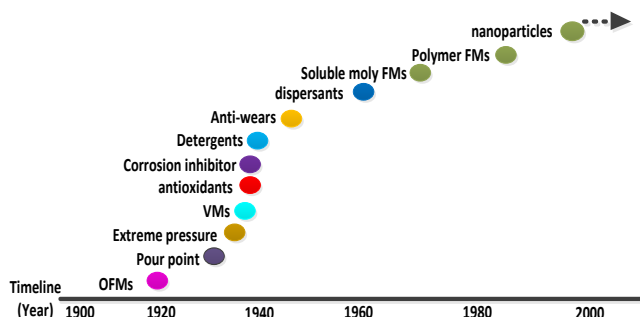


Fig. 1: Chart for development of lubricant additives³³⁾⁽¹⁶⁾

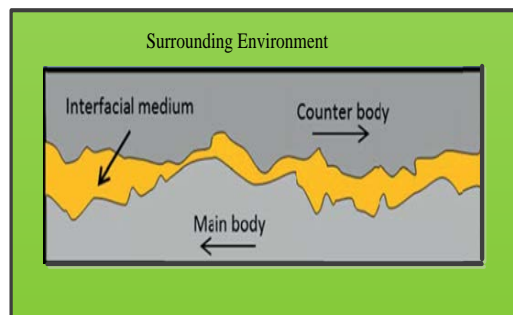


Fig. 2: Diagrammatic representation of the tribological operating system^{19,22)}

2.0 Fundamental of Nano-Particle Additives in Lubrication

Nano is a prefix, applied in dimension description of materials within the range of 1 to 100 nm. Nano as in nanotechnology was first mentioned by Nobel laureate Richard P Feynman of physics in his famous work entitled “There’s plenty of room at the bottom” in 1959 at American Physical Society meeting^{34,35}). Today nano materials has become great resources in all field of production with huge outstanding potential. Nanotechnology for the formulation of nano materials comprises of manipulation, treatment and creation of materials within the nanometer scale, which could be through scaling from unit to groups of atom or through processing by reducing the bulk size material into the desirable dimension^{15,36,37}). However, nanotechnology in the field of tribology is focused on materials in particulate substances rather than building to group and could be of 1D, 2D or 3D depending on the material type and behavior³⁸). Accordingly, 1D nanomaterials are characterized with single dimension in the nanoscale range, normally found in surface coatings and thin film nanomaterials²¹). These are usually applied in the field of tribology, chemical and biological sensors, fiber optic systems etc. In the case of

2D nanomaterials which are found in two dimensions of nanometer scale, normally of nanotubes, nanowires, dendrimers, fibers and fibrils. Also, some free particles of large aspect ratio of nanoscale dimensions are considered 2D nanomaterials during classifications. The manufacturing of 2D nanomaterials are less advanced and their behaviors together chemical properties are more complicated with less understanding. In the application, thin films due formed in various mechanisms which can be grow at a monolayer^{10,21,37}). For 3D nanomaterials, its nanoscale was observed to be in three dimensions. The materials of this form include nanocrystals or quantum dots, colloids, fullerenes, particles and precipitates^{21,39}). This class of nanomaterials are mostly found in tribology and combustion products like natural nanomaterials, titanium oxides (TiO), carbon black, and zinc oxide (ZnO). In the production of some of the products of this class like quantum dots and fullerenes gives high challenges with properties behavior are difficulty to ascertained^{5,9,40}). Different structure of various nano-additives under microscopic view are shown in Figure 3.

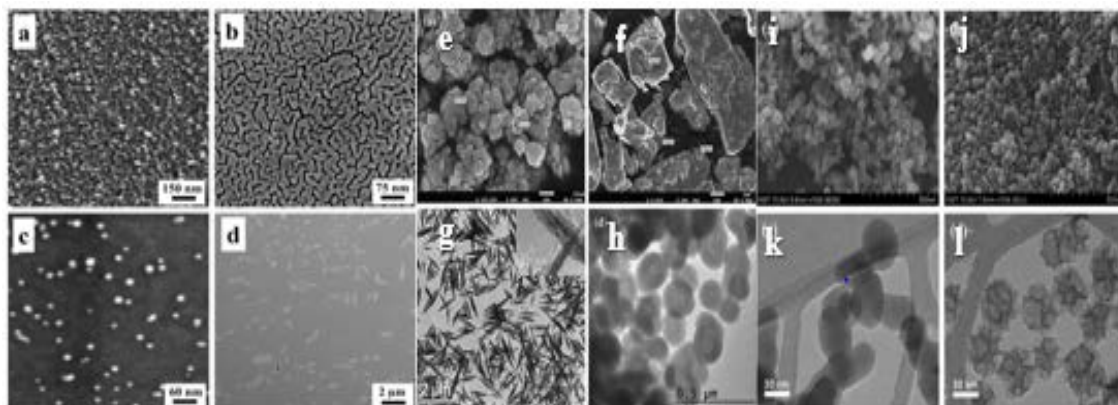


Fig. 3: Image of Scanning microscope (SEM) indicating (a) film of Ti NPs, (b) a near percolating Au film, (c) monodispersed Cu NPs and (d) Fe nano-rods ⁴¹⁾. Morphology of nanoparticles FESEM for, (e) nano CuO ⁴²⁾, (f) nano MoS₂ ⁴²⁾, HRTEM for (g) nano CuO ⁴³⁾, (h) nano MoS₂ ³¹⁾, FESEM for, (i) nonporous MA-SiO₂ NPs, (j) mesoporous MA-SiO₂ NPs, (k) MA-SiO₂ NPs and (l) mesoporous MA-SiO₂ NPs ³⁹⁾

The structure and constituents of nanomaterials are simple atoms or molecules itself but consist of different layers of three forms ²¹⁾; as shown in Fig. 4, which include; 1. The layer surface which are commonly described as Janus NPs in most literature, comprises of two types of nano-particles having common interface but of different form ³⁶⁾. The formation of this form of nano-particles tends to reduce the number of the formed bond among the elements (A and B) in the system, illustrated in Fig. 4 (i). The layer surface has potential of functionalization with other close related small molecules of organic material, inorganic material, surfactant, polymers and metal ions. 2. Mixed shell structure which are different in chemical properties from the core in every application. This can be in ordered or random form. Orderly mixed or randomly

mixed with alloys arranged in A and B atoms or solutions of solids as case may be, see Fig. 4 (ii). 3. The core-shell structure, which comprises of one type of atom say 'A' enveloped a core of different atom say 'B' as in Figure 3 (iii). This form of formation is denoted by A@B and found mostly in NPs formulations of multiple shell like onion structure ⁴⁴⁾ ⁴⁵⁾. The functionalization mostly of alternating like A-B-A shell or A-B-C arrangement when there is ternary NPs, illustrated in Fig. 4 (iv). The development of new structure of latter are normally from structure of three different elements like Fe-Ag-Si multi-shell arrangement thus formulated by adjusting the analysis selected conditions ⁴⁶⁾ ^{45,47)}. The characteristics are different from others but highly supportive during thermal activities ⁴⁷⁾.

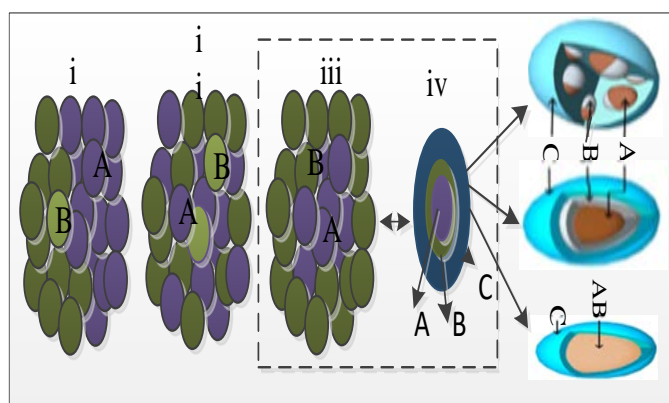


Fig. 4: Schematic description of NPs of different formation structures, (layer surface structure (i), Mixed shell structure(ii), and core-shell (iii or iv) ^{21,44,46)}

NP additives base on their source, physiochemical and formulation function are differently like anti-wear (AW), friction modifier (FM), extreme pressure (EP), surfactant and viscosity improver polymer, thus enables the base oil lubricant to withstand the working conditions ^{16,48)}. Apart from numerous physiochemical properties of NP additives, tiny size nature is another important reason of

recommending NP in tribological application because of its easy movement on the contact areas leading to good lubricating result ^{10,49,50)}. In addition, the size enables the lubricant passes through small orifices of engine components like nozzles, injectors and filter elements ^{33,50)}.

Generally, according to literatures, the microscopic

nature of the nano-materials observed to be the cardinal reason of their novel physiochemical properties that promote the wide economic benefits and applications^{10,45)}. The actualization of the prime feature of these nano-material additives were through utilization of some modern nano-technological equipment, following top

down approach as seen in Fig.5, while Fig. 6 shows the image of ball mill machine P100 as the commonest technique of formulating NPs. For good formulation of nanoparticle, appropriate step must be followed as to achieve optimal functional group of the sample^{15,37)}.

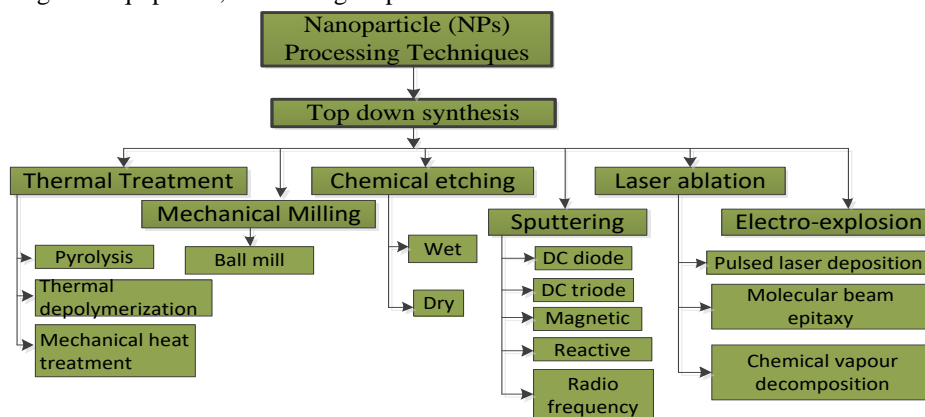


Fig. 5: Production techniques for NPs using top down synthesis approach²¹⁾.

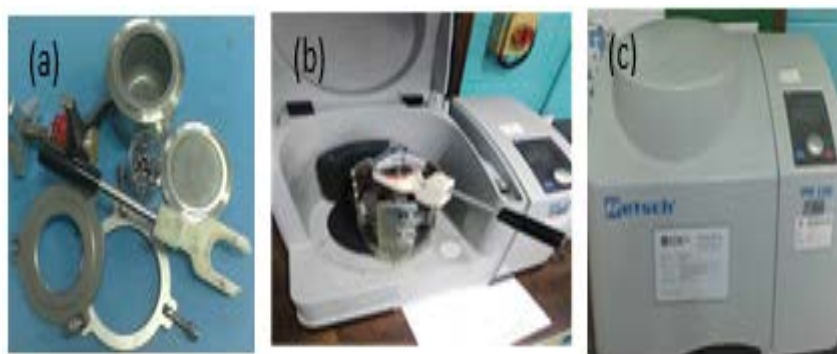


Fig. 6: Image of grinding/ball milling technique for NPs formulation, (a) machine accessories, (b) jar locking step, (c) machine in operation.

This top down approach follows destructive principle, starting from larger molecule, breaks down to smaller unit, at times, uses the generated units to formulate suitable NPs. NP additive performance in base oil depends on some serious factors ranging from morphology, compatibility with the base oil, and size⁵¹⁾ thus stands as challenge during application. Thermal treatment is a remedial approach where materials are being heated to increase the mobility and facilitate the extraction of its required contents. It can be classified into many groups; pyrolysis, thermal decomposition and mechanical heat treatment. In pyrolysis, the initiating materials are burn using heat, and may be in liquid or vapor but mostly liquid. It is being processed inside furnace t high pressure, thereby collect the formulated nanoparticles. In some operation, plasma or laser are used in place of flame to generate required temperature. In thermal decomposition, the chemical bond in the compound is broken through an endothermic process. The temperature decomposition is defined so that the element will be chemically decomposed⁵²⁾. The method of mechanical milling is a

technique best described using grinding/ball milling machine, fast de-binding and other decomposition methods⁵³⁾. The milling approach was employed as to form powders from the raw materials with the help of ceramic balls and well-known planetary mill. The overall morphology and size of the NPs are normally obtained through different characterization techniques like FESEM, TEM, SEM and particle size analyzer.

The technique of chemical etching/industrial etching or chemical milling is a manufacturing process of using baths of temperature regulated to extract materials or create a new material with unique desired shapes. The wear resistance of the honed Al-Si alloy cylinder were improved through chemical etching by applied on the surface of the Al-Si ally samples⁵⁴⁾. Sputtering method is a phenomenon of deposition of NPs through the means of ejection particles from it⁵⁵⁾. This process comprises of wet and dry, however, annealing is important for the deposition of NPs thin layer. Formulated particles are determined via some factors like; thickness of the layer, temperature, duration of annealing, and substrate^{55,56)}.

Another technique is laser ablation which is a simple method for synthesis of different NPs from different solvent, done by submerge different materials (metal) in solution by laser beam condense a plasma for the formulation of NPs^{57,58}. This technique still comprises of three other methods like; pulsed laser deposition, molecular beam epitaxy and chemical vapor decomposition⁵⁹. More importantly, during change in nano-material nature through inter-reaction with other materials, turn from bulk to molecule or macro to nano during shrinking, thus adversely affect their fundamental properties. For instance, developed nano material might display unique optical^{15,59-61} amphipathic, surfactants, and thermal behavior completely different from their bulk, macro or nano material as case may be. In all these

characteristics formations, size and materials type contribute significantly for the phenomenon⁶². Although many positive presentations on tribological improvement in lubricants with different nanoparticles, however, the issue of adequate selection of nanoparticles still occur^{15,38}. More so, several works have indicated of possibility of single nanoparticle additive saving more than one purpose as in AW, FM, and EP^{63,64}. According to^{17,33}, pointed some fundamental criterial of NP that makes them advantageous in tribology like; (a) Low reactivity with other included additives; (b), More durability, (c), Insolubility in non-polar base oil, (d). High non volatility to withstand thermal condition, (e). Easy formation of film on different material surface.

3.0 Nanoparticles Type and Classifications

Generally, NPs type and classification are organized into many different categories (composites, inorganic, organic, ceramics and carbon based nanomaterials) according to their size, nature and application^{21,41,65}. Following these features, NPs source include natural, incidental and engineered nanomaterials. Since this work focused on lubrication, the work will be limited only on engineered aspect of NPs

3.1 Composites Nanoparticles.

Composite as NPs are advance materials discovered to be the most outstanding nanoparticle for future application. The product is formulated from two or more different constituent unit material with different in physiochemical properties and when combined form one product with characteristics different from the initiated individual components.¹⁵. It consists of curing phase or a matrix and particles in nanoscale with purpose of tribological enhancement^{60,37}. The benefit of having the composite NPs materials planned in advance gives them or recommendation and the properties mainly depends on the matrix choice, curing, shape and orientation. Many presentations have it that friction and wear properties of polymeric materials can be enhanced through high stiffness, lower adhesion and strength^{33,66}. In performing the operation, suitable fillers were blended, good proportion of graphite also carefully added as to reduce the adhesion. This formulation due allows formation of friction reducing agent of film on the sliding surfaces, thus reduces the coefficient of friction (COF) in the operation⁶⁷. The use of polymer with high temperature resistance supplement (carbon nanotubes matrix) are recommended since friction generates great heat. The combination of these matrix yields composite materials with microstructure of excellent thermal and polymeric behavior. The composite blending with their expected functions are illustrated in the Fig. 7.

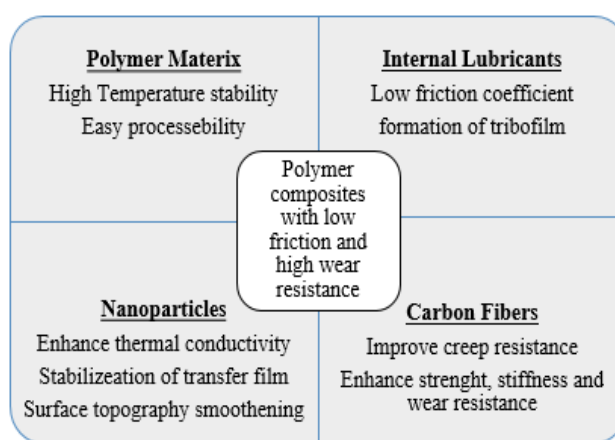


Fig. 7: Formulation of Polymer composites for optimal lubrication

3.2 Inorganic nanoparticles

This group of nanoparticles are classified into metal and metal oxide nanoparticles. Inorganic nanoparticles are mostly hydrophilic and biocompatible during lubrication and highly stable than organic nanoparticles. However, they are not mostly toxic particles due carbon products are not present in the list of inorganic nanoparticles.

3.2.1 Metal nanoparticles

Currently, metal nanoparticles are considered to the widely used additive in the field of engineering in many applications like in magnetics, semiconductors, photonic and catalysts operation^{5,68}. They are used during metallic nanoparticles synthesis through constructive or destructive approach⁵². In tribology, some interesting engineering properties found in metal nanoparticles pulls the attention of researcher towards utilization of such materials. Metal precursors are best use for formulation of pure metal nanoparticles and always exhibit unique properties because of their resonance characteristics. The persistence use of Cu nanoparticles provides excellent function especially in the area of self-repairing services together its eco-friendly attribute⁶⁹⁻⁷². On the other hand,

because of some poor physiochemical properties of these metal nanoparticles products, like weak compatibility in non-polar base oil as a result of high surface activity thus due control through further surface modification approaches⁵²⁾. In a work presentation by⁷³⁾ on the effect of metal nanoparticles of different types using mineral base oil was conducted. During the study,⁷⁴⁾ the different metals like Cu, Co and Fe were tested on their respective mixtures, with investigations to determine wear strength of the aforementioned metal materials as seen in Fig. 8.

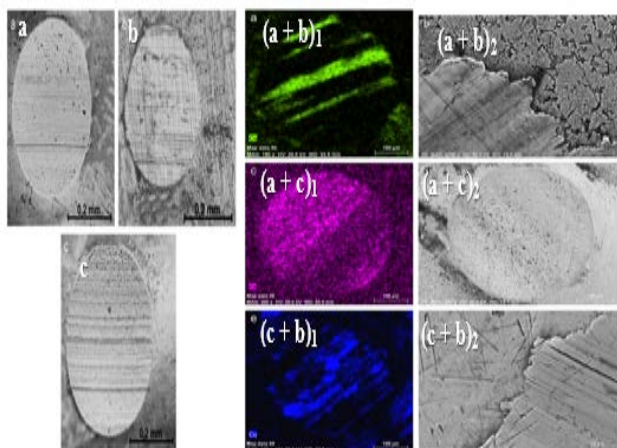


Fig.8.: Image of wear scars on balls operated with SAE 10 oil with different metallic additives: (a) Fe nanoparticles, (b) Cu nanoparticles, (c) Co nanoparticles, also with mixtures of nanoparticles: Fe+Cu (a and b), Fe+Co (a and c) and Co+Cu (c and b).⁷³⁾

Nanoparticles of Cu on its tribological behavior on diesel oil using serpentine powder was conducted⁷⁴⁾. The result shows 7.5% concentration of Cu nanoparticles gives wear reduction with maximum friction generation discovered. The ability of metal particles to exhibit all these distinctive features is because of their nanoscale (10-100nm), surface charge density, pore size, structure (amorphous and crystalline), surface area to volume, environmental factor (heat, moisture) and various shapes (tetragonal, irregular, hexagonal, spherical, cylindrical and rod). However, when the various metal constituents were mixed, surface investigation on the wear scar show improvement on the friction and wear reduction and was so because of their unique properties and resonance characteristics contributed by various metals⁷⁴⁾. After the mixture of Fe or Co with the Cu nanoparticles, the Cu nanoparticles occupied the surface and used in the medium as friction resistance⁷³⁾⁷⁴⁾. The nano nature of the additives contributes to high performance by altering the friction situation from sliding to rolling by diffusing into

the sliding contact⁷⁵⁾. Nanoparticles bear a portion of the load and form a layer between the two surfaces to avoid adhesion when the lubricant film between tribo-pairs thins and mixes, increasing anti-wear and friction reduction tribological characteristics⁷⁵⁾.

3.2.2 Metal oxide nanoparticles

This class of nanoparticles are from metal oxides and normally synthesized as to modify the property of the individual metals nanoparticles. Example is iron oxide nanoparticles developed from iron nanoparticle through oxidation. As in their function ability, the iron oxide nanoparticles reactivity is high when compared to ordinary iron nanoparticles. As to obtain high reactive and efficiency in operation, metals are synthesized like, cerium oxide, zinc oxide, magnetite, iron oxide silicon oxide and titanium oxide⁷⁶⁻⁷⁸⁾.

3.3 Ceramic nanoparticles

The class of this nanoparticles are also referred to as non-metallic solid. They are also amorphous, hollow form and porous when synthesized through successive cooling or heating⁷⁹⁾. Ceramic nanoparticles have attracted great attention, since they always successfully lubricated by water. Also discovered tribo-chemical reaction on the ceramic surface, thus found demonstrated that electric double layer of water film significantly provide super lubricity condition⁸⁰⁾.

3.4 Organic nanoparticles

Organic nanoparticles/nanomaterials are mostly product from organic matter. The materials are not toxic, biodegradable and due possess feature of like hollow sphere like in liposome/lipid and micelles, as seen in Fig. 9. Other product of organic nanomaterials includes dendrimers, and ferritin. The arrangement is by noncovalent interactions for the design of the molecules due transform the particles into the desired structures. Also, polymers are always referred to as organic nanoparticles⁸¹⁾. They are of matrix particles with overall mass almost of solid with other adsorbed molecules at the outer boundary of the spherical surface. From their functionalization, applied in lubricant as viscosity improver base on its ability of increase in strength with temperature⁸²⁾⁶⁶⁾. In the case of lipid, always of diameter from 10 to 1000 nm similar to polymer, possess core solid form of matrix contain soluble lipophilic molecules. In lubricant operation, they function as emulsion or surfactant⁸¹⁾.

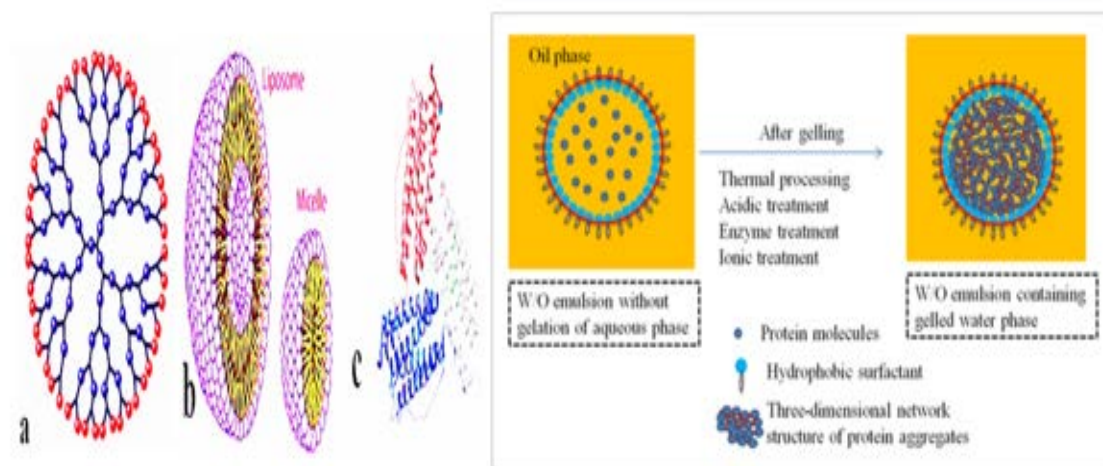


Fig. 9: Image of organic nanoparticles: (a)Dendrimers, (b) Liposomes and Micelles (c) Ferritin²¹⁾ and (d) W/O emulsions stabilized with gelled water phase^{81,83)}.

Current investigation described appealing gelling (organic) approach for formulating W/O emulsions. In the study, applied glucono- δ -lactone (GDL) to initiate the gelation of casein dispersion (3 %, w/v) instead of thermal method as shown in Fig. 10. When compared the result with W/O emulsion without inner gelation of casein

dispersion, evidently show that GDL induced exhibited great resistance to destabilization⁸³⁾. According to⁸⁴⁾ reported that Surfactant inclusion in base oil has an excellent performance, reducing the friction coefficient of water up to 80% under the experimental conditions utilized

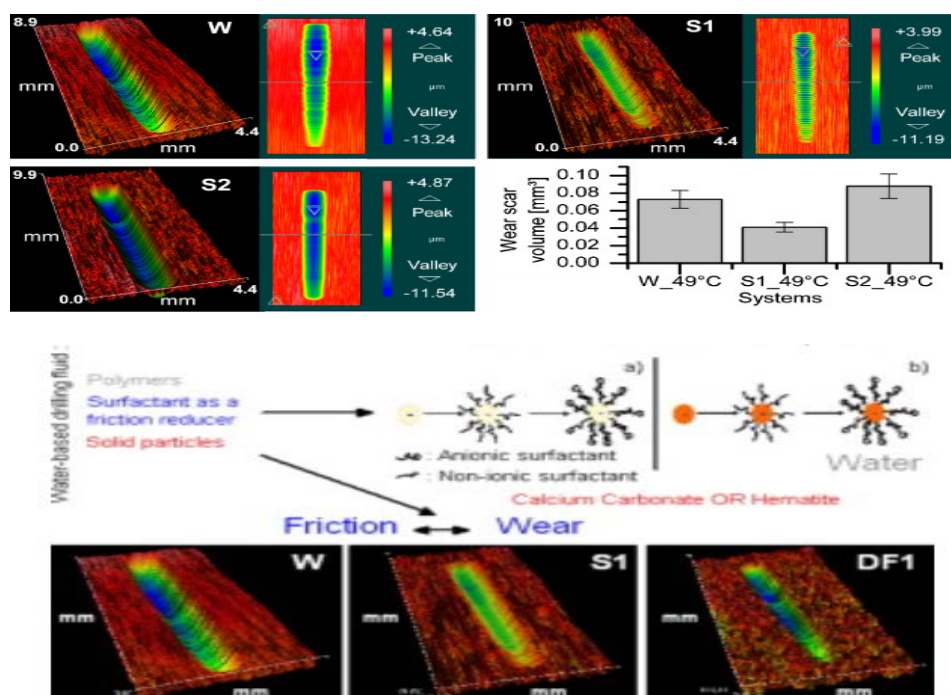


Fig. 10: Optical surface profile images of the metal block after the wear test for the surfactant operation⁸⁴⁾.

Drilling fluid formulations without inclusion of surfactant additive produce similar result of coefficient of friction compared to water. With inclusion of additive (S1) (surfactant), at 1 % (w/v), the formulation produced significant reduction in coefficient of friction (COF) by about 60 % compared water. Also, with 0.4 % (w/v) surfactant (S2), reduction turn to 30 % with wear scar

formation due to little of surfactant to adsorb by the metal material⁸⁴⁾, thus concluded that surfactant mixture in aqueous solution provide excellent anti-wear properties for reduction of friction and wear.

3.5 Carbon base nanomaterials/nanoparticles

Products of this class, completely composed of carbon

like, graphene, fullerenes, carbon nanofibers and carbon black^{15,85}, normally found in morphologies such as spheres/ellipsoids and hollow. The popular method for the production of carbon base nanomaterials were chemical vapor deposition (CVD) and laser ablation, with exception of carbon black type⁸⁶. Series of tremendous works has been presented by researcher's form carbon base nanoparticles towards lubricant properties enhancement with excellent results, owing to their potentials.

3.5.1 Carbon nanotubes (CNTs)

These are tubular cylindrical molecules, consist of rolled up sheet of single layer carbon atoms, with structure of about 1-2 nm diameter. Carbon nanotubes can be seen as semiconducting in behavior base on diameter. However, graphene is from carbon in allotropic form of hexagonal network of honeycomb carbon atom of 1 nm in thickness as see in Fig. 11. The structure of carbon nanotubes base on their rolling can be categorize as single walled (SWNTs), double Walled (DWNTs) and multi-walled (MWNTs). Also carbon nanotubes can form in three ways like; armchair, zig-zag and chiral.

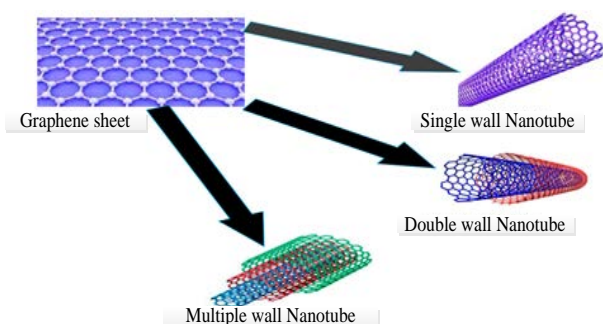


Fig. 11 : Image of graphene sheet rolled into: SWNT, DWNT and MWNT²¹⁾

As the name implies, SWNTs consist of single rolled sheet of at least 0.7 nm in diameter, DWNTs consist of double rolled sheet, while that of MWNTs is made up of multiple rolled sheet of at least 100nm in diameter. Another class of carbon base material is fullerenes, consist of nanomaterial that looks like hollow cage. This type is more conductive, versatile, electron affinity and even of high strength^{40,87}. Due to their characteristics features, CNTs easily bent and regain its normal figure without brittle. this property made them noteworthy commercially in additive formulation for lubricant application⁸⁸⁻⁹¹. In the lubricating operation on carbon nanotubes conducted by^{89,92} on its performance towards friction and wear. Result shows that coefficient of friction with and without CNTs under sliding condition as see in Fig. 12. The result evidently shows that without CNTs coefficient of friction value was 1, while that of CNTs only dropped to 0.1-0.2 at the same stable period, thus CNTs possesses the potentials to reduce friction and wear.

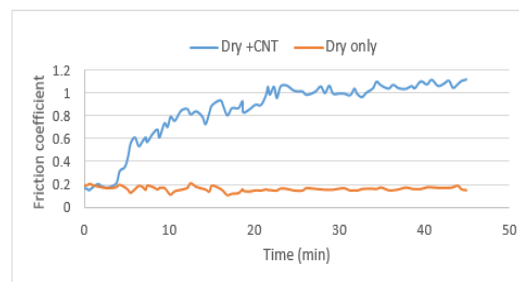


Fig. 12: Coefficient of friction under dry sliding condition with and without CNTs⁸⁹⁾

The result of this study is also in agreement with the study on Tribological Properties of Novel Multi-Walled Carbon Nanotubes and Phosphorus Containing Ionic Liquid Hybrids in Grease⁹¹⁾. Also⁴⁸⁾, reported that addition of carbon onion additive powder to PAO base oil significantly reduced the friction below 0.1 against 0.15 under pure PAO, likewise graphite exhibiting great reduction on friction as see in Fig. 13. This is because graphite powder is well known for low friction under boundary lubrication as in Fig. 13.

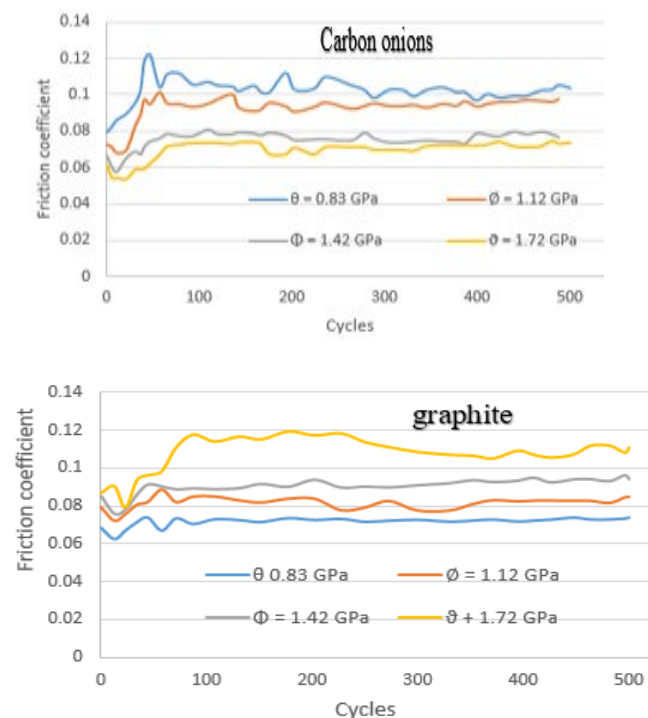


Fig. 13: Friction coefficient obtained at several contact pressure (0.83-1.72 GPa) with the dispersion of 0.1 wt % in PAO of carbon onion or graphite⁴⁸⁾

3.6 Carbon nanofiber

Carbon nanofibers (CNFs) are one of the promising materials in the field of engineering and beyond. Synthesis of carbon nanofibers are usually through chemical vapour decomposition, templating, and electrospinning. In the production, orientation of the carbon layer in CNFs seriously affect its mechanical properties^{21,93}. Again,

nano-foils with features of graphene are turn into CNFs as nanotubes, however, the nano-foils are like cone, (see Fig. 14) or cup unlike elongate cylindrical tubes of CNFs^{15,21)}.

3.7 Carbon black

These are nano-powder produced through high temperature carbonization via a carefully controlled combustion process of about 1300 ° C. Its application is verse but mostly in engineering example in ink, green technology and lubrication both coating and in base oil. The shape is mostly spherical with diameter from 20-70 nm as see in Fig. 14.to differentiate each other.

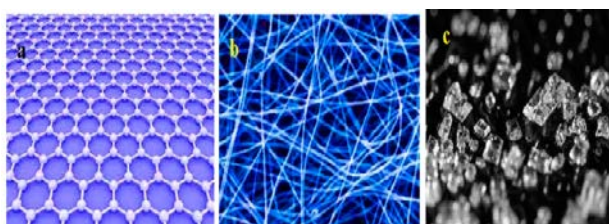


Fig. 14: Image showing (a), Graphene sheet, (b), Carbon nanofiber, (c), Carbon black²¹⁾.

According to⁹⁴⁾, carbon black provide smooth sliding with coefficient of friction range from 0.35-0.5, a volume fraction of at least 13% of soft nanoparticles. Adding that the particle clusters do not alter the smooth operation, provided the materials are completely embedded in the magnetic matrix. Some of the tribological enhancement performance from the use of some nanoparticles including categories of carbon nanotubes are listed in Table 1 and 2.

Table 1: Summary of Some Important Nanoparticle additive in lubrication

Nanoparticle	Effect	Ref.
MWCNT	MWCNT dispersed with surfactant reduces wear, low frictional force	95)
Diamond nanoparticle	Anti-wear, anti-friction, excellent load carrying capacity as a result of surface polishing from hard effect of diamond nanoparticles	96)
Titanium borate	Better (NPs) anti-wear property and friction-reducing property in base oil compared to TiO ₂ nanoparticles under the tradition process.	97)
Diamond Nanoparticle	Improvement of anti-scuffing performance significantly reduction in friction	98)
Diamond nanoparticle	Increasing the friction and wear	99)
Nano-cerium borate	Friction modifier	100)
Hexagonal boron nitride	Reducing coefficient of friction and scar diameter	101)
Diamond nanoparticle	Viscosity increasing effect, friction coefficient decrease	102)

Table. 2: Some research reports on metal matrix/nano carbon (including carbon nanotube and graphene) composites, preparation technique and properties.

Matrix	Reinforcement	Process	Properties	Ref
Aluminium	Graphene	Mixing AA2124 powder and graphene platelets and then cold compaction at 525 MPa pressure	Increasing the hardness and decreasing the relative density. On the other hand, there is an optimum point that wear rate is minimized.	103)
Copper	Nano-graphite	Powder metallurgy technique where nano graphite were dispersed in ethanol and then copper introduced to solution, after that drying the powder and using 450 MPa pressure to make composite	Copper/nano-graphite exhibit better tribological properties than copper/micro-graphite. Also increasing the volume fraction of reinforcement tends to improve tribological properties	104)
Aluminium	MWCNT	High energy ball milling, cold compaction and hot extrusion	Wear resistance and hardness of composite significantly increased while CoF decreased	105)

		were employed to synthesizing composite.	with significant decrease in wear rate	
Aluminium	CNT	Sintering the mixture of aluminium and CNTs powders in a carbon mold under 50 MPa pressure	No change in elongation while there is a significant improvement in tensile strength.	¹⁰⁶⁾

3.8 Mechanism of nanoparticles during lubrication

Since only base oil lubricant will not withstand operating condition of engine, inclusion of nano particles (additive) into an oil lubricant significantly reduces the system coefficient of friction ^{29,107)}. The reduction in the friction through the nano particles will apparently increases the load carrying capacity of the contacting components of the mechanical system ^{107,108)}. When nanoparticles are added into base lubricant oils, new solution that is stable with homogeneous feature is formed ¹⁰⁹⁾. In analyzing the nano-particles features and behavior during operation, four different mechanisms have been proposed, described the function and operation towards improvement of the lubricating oil generally ³⁰⁾. They include; rolling mechanism or bearing effect ^{110,111)}, protective or tribo film effect ^{112,113)}, mending effect ⁶⁹⁾ and polishing effect ⁹⁶⁾ as illustrated in Fig. 15.

3.8.1 Rolling Mechanism or bearing effect

In the past few years, testing of nano-particles in lubrication have emerged as the best and new choice in additive production. Many reputable works have been conducted using nan-particles, with numerous achievements from the patronize of such products towards enhancement of lubricant tribological properties ^{3,73,114)}. The mechanism of friction reduction and anti-wear of this form additives clearly believe to perform between the

rubbing material surfaces ^{10,69)}. Experimental work to demonstrate the ball bearing nanoparticle mechanism was conducted by ¹¹⁵⁾ using diamond nanoparticles with liquid paraffin. Reported that when experimentation time increased, diamond nanoparticles spherical shape turns into small rolling material like ball bearing and operate in such mechanism as see in Fig.15 and 16. Also, material with rough surface sliding due observe nanoparticles deposit in troughs on the rubbing surface, thereby serve as medium of treating the surface ^{27,116)}. Graphite nanomaterial was prepared and applied as additive in oil lubricating system, the result was so significant, but its preparation process was so challenging ^{13,112)}. The nanoscale modified graphene platelets (MGPs) if applied in lubricant operate as nano-bearings between tribo-couples ¹¹⁷⁾. According to ^{118,119)}, on synthesized Al ₂O₃/TiO₂ nanocomposite, gives good friction and wear properties on lubrication. Also observed that Al ₂O₃/TiO₂ enhanced the system tribological performance owing to the control on wear from the sliding friction to rolling effect during the operation ¹⁰⁷⁾. The mechanism theory identified two elements that contributed to the nanoparticles' outstanding performance. First, nanoparticles are normally spherical and operate as micro-bearings, preventing friction and carrying load between rubbing surfaces. As a result of the mechanism that controls lubrication, nanomaterials form tribo-film which help in preventing friction and wear ¹²⁰⁾.

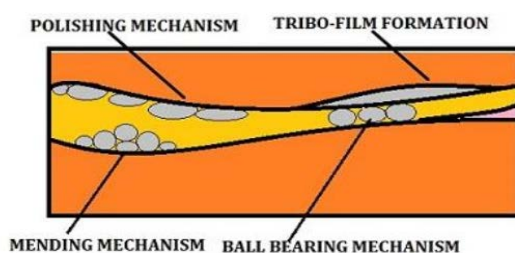


Fig. 15: Various lubrication mechanism of Nanoparticles ³⁰⁾

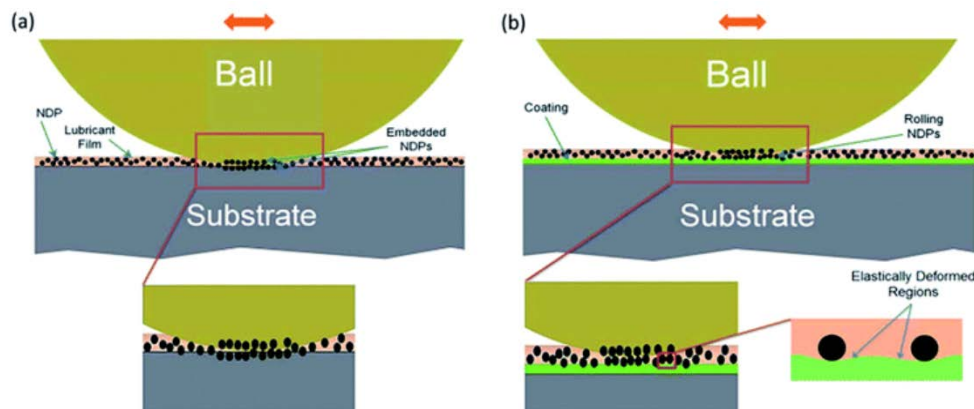


Fig. 16: Description on effects of NPs in the lubricant (a) bar and (b) coated stainless steel materials ^{121) 122)107)}

3.8.2 Protective film or Tribo film mechanism.

In this particular mechanism, layer of amorphous from nanoparticles are formed on the sliding element surface, thereby reduce the actual area of contact. NPs can form sorbent (adsorption) films on the tribo-pair body surfaces, also NPs do undergo sintering by frictional pressure and heat and fill formed wear track ^{78,107)}. NPs that dispersed in fluid well are liable to form thin film on the operating metal surface, with the film of low shear and dense separate the contact thus reduces the friction manifestation. During the system operation, the NPs in the fluid tend to move to rubbing region due system inherited phenomenon. Demonstration on protective film mechanism was carried out by ¹¹²⁾ employing magnesium borate in 500-SN base oil. Result shows that amorphous film layer was formulated between the sliding contact surfaces. Because of the new layer formation, decrease in COF was recorded with increase in the wear resistance.

According to ^{18,89)} reported that NPs in absent of oil lubricants can contribute in reducing friction and wear. Consequently, nanoparticles of nanocomposites like SiO₂ tribological performance improves via material contacting surfaces from features of hydrophilic or hydrophobic to amphipathic ^{123,124)}. During NPs additive operation, agglomeration due help in rolling and sliding on the material contact area, tribo-par material hardness, pressure on the contact and interaction pull among the nanoparticles ^{120,125)}. In addition, at contact pressure around 1GPa, the tribo-pair works in the boundary lubrication regime via mechanically sheared, however, nanoparticles developed intermediate structure, adhere on the surface of the contact elements and create gap against interference ^{17,120,126–128)} as see in Fig. 15 above. Investigation on IF-MoS₂ NPs, prove that size and crystalline shape nature alter its lubricating properties ¹²⁹⁾. The coefficient of friction on inclusion of 1 w% of IF-MoS₂ under boundary lubrication decrease compared to pure PAO oil sample as shown in Fig. 17. Moreover, the COF performance in reduction properties does not cause by contact pressure ¹⁸⁾. This tribo film mechanism is regarded as the most prevailing theory that give more

insight about friction and wear reduction behavior of lubricant. In ¹³⁰⁾ study explain that heterogeneous dropping of NPs on element wearing surfaces due lead to transition of lubricant mechanism, forming traditional mixed lubrication to modified synergistic solution that promised with asperity.

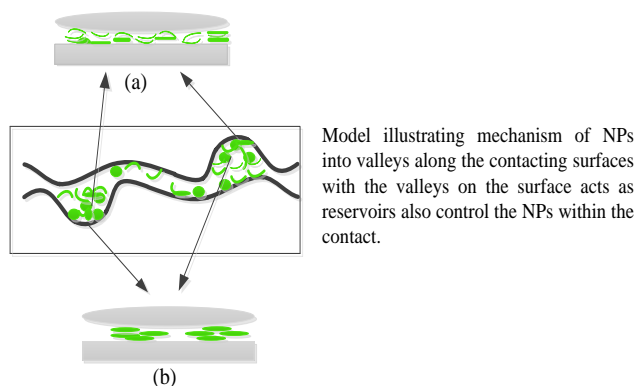


Fig. 17: Two mechanism of MoS₂ protective tribofilms, (a) Monosheet that exfoliates with thin film formed and (b) Aggregates separating interfacial contact with thick film formed ¹⁷⁾.

3.8.3 Mending or self-repairing mechanism.

When nanofluid is in operation, NPs in the lubricant tends to deposit as to fill and resurface the wounded rubbing contact through desorption and deposition and assisted in forming more smooth and flat pair ¹³¹⁾. This operation of self-repairing is mostly found with face-centered cubic soft metal. According to ⁶⁹⁾, reported that nano-copper additive in lubricant usually develop such feature thus separates the frictional materials during friction process. However, mending operation and self-repairing mechanism is not limited to accumulation of NPs on material element surfaces but also size of NPs, melting points attributes. Explaining this, at severe temperature beyond the added materials, the NPs melts or sintered in the wounded contact area, symtheniously turn to filler and repair the element working surface ^{27,69)}. Carbon nanotubes (CNTs) and Carbon nanofibers (CNFs)

observed infiltrate of small gaps between rough surfaces in contact ¹³²⁾ thus, forming self-lubricating thin films as shown in Fig. 15 above. Another experimental work on polishing mechanism using fullerene nanoparticles together with refrigeration mineral oil was conducted by ¹³³⁾. observed that using diamond particles as nanoparticle additive in paraffin oil, smooth surface was found on the rubbing contact at the starting of the tribotest compared to surface image on pure paraffin oil ^{133,134)}.

3.8.4 Polishing Mechanism or Effect.

The mechanism depend on the hardness of the nanoparticles and contributes significantly in modifying or maintaining the machine surface element smooth ⁹⁶⁾. In this, the contacting region of the tribo pairs usually increased owing to the smoother nature of the material via the mechanism and increase the load bearing capacity with decrease in coefficient of friction ^{96,107,135)}. Investigation carried out by ¹³⁶⁾, discovered that surface

morphology of aluminium alloys 6061 material lubricated separately with commercial oil and nanofluid having 0.05 % nano-diamond. Observed that the one lubricated with nanofluid decrease by 15 % if compared with ordinary commercial oil. The result on the decrease on nanofluid type was because of polishing action of diamond nanoparticles leading to element smooth surface. On this operation, working element surface with much roughness, NPs do not function appropriate as to enhance the tribological properties of the nanofluids, thus found more active in tribo pair with low surface roughness. During investigation on mending mechanism under pin (H62 bronze) on disk (20CrMnTi steel) was analysed using SEM and STM ⁶⁹⁾. The work concluded that copper particles exhibit excellent mending mechanism in the element wear areas, supported by on same analysis conducted on sliding bearing using the internal combustion engine ^{69,137)}. Table 3 concisely give some base oil/additive report on their mechanism and tribological effect during operation.

Table.3: Summary of nano-additives in base oil (lubricant), effect on lubrication, mechanism and tribological properties

Base oil/nano additive/function	Mechanism (lubricant), surface analysis methods	Tribological effect (friction and wear)	Re f.
PAO10/MoS ₂ ,BN. [FM, AW]	Tribo-film formation Raman spectroscopy	Reduction in COF, improved friction and wear reduction properties	49,138–140)
PAO/MoS ₂ . [AW, EP, FM]	Tribo-film formation SEM, EDS	Reduction in COF/Enhancement of wear properties	134)
PAO6/ ZnO, ZrO ₂ , CaO. [AW, EP]	Mending effect SEM, EDS	Improvement of Wear properties	141)
SAE 15W40/h-BN. [FM, AW]	Rolling and Polishing effect SEM	Reduction in COF/enhancement of wear properties	101)
500SN/WS ₂ [EP, AW, FM]	Mending Effect and tribo-film formation XPS and SEM	Reduction in COF/enhancement of wear properties	142)
Mineral Oil/CuO. [FM, AW]	Deposition, Polishing Effect SEM, EDS	Reduction in COF/enhancement of wear properties	143)
Palm TMP Ester/ CuO,MoS ₂ [AW, EP]	Tribo-film formation and Mending effect. SEM, Raman spectroscopy	Enhancement of wear properties	144)
Lubricating Oil ZnAl ₂ O ₄ . [FM, AW]	Mending Effect SEM and EDS	Reduction in COF/enhancement of wear properties	145)
Jatropha TMP Ester/WS ₂ [AW, EP]	Tribo-film formation. SEM	Enhancement of wear properties	146)
PAO6/ZnO, ZrO ₂ ,CaO/ [AW, EP]	Mending Effect SEM and EDS	Enhancement of wear properties	147) 141)
Mineral Oil/ ZnO. [FM, AW]	Tribo-film formation. SEM and EDS	Reduction in COF/enhancement of wear properties	143)

SE15W40/Al/Sn. [AW, EP]	Mending Effect SEM and EDS	Reduction in COF/enhancement of wear properties	148)
SAE-20W50/ Carbon Nanoball. [AW]	Mending Effect SEM	Enhancement of wear properties	149)
SAE-10/Fe, Cu, Co. [FM, AW]	Tribo-film formation. SEM and EDS	Reduction in COF/enhancement of wear properties.	73)
SAE-30/CuO. [FM, AW]	Mending and Ball bearing effect. SEM and EDS	Reduction in COF/enhancement of wear properties.	150) 151)
Liquid Paraffin/ Al/ [FM, AW]	Tribo-film formation. SEM and EDS	Reduction in COF/enhancement of wear properties.	152)
Super Gear/EP220/Gr. [FM, AW].	Polishing Effects SEM and EDS.	Reduction in COF/enhancement of wear properties.	153)
PAO6/Ca. [AW, EP]	Tribo-film formation. SEM and EDS	Enhancement of wear properties.	154)
Liquid Paraffin/ CuO. [FM, AW]	Ball bearing effect. SEM and EDS	Reduction in COF/enhancement of wear properties.	149)
TBA, Liquid Paraffin/Pb. [FM, AW]	Ball bearing, Mending effect. SEM and EDS	Reduction in COF/enhancement of wear properties.	155)
Liquid Paraffin/ MoS ₂ , TiO ₂ . [FM, AW]	Mending and tribo-film effect. XPS	Reduction in COF/enhancement of wear properties.	114)
500 SN Base Oil B ₂ Mg ₃ O ₆ . [FM, AW, EP]	Deposition, tribo-film formation SEM and XPS	Reduction in COF/enhancement of wear properties	144)
Sunflower oil/ZnO, CuO [FM and AW].	Tribo-film formation. SEM.	Reduction in COF/enhancement of wear rate.	143)
SAE10W-30/TiO ₂ [FM, AW]	Ball bearing effect. SEM and EDS	Reduction in COF/enhancement of wear properties	114) 156)
Soyabean oil/ZnO,CuO. [FM and AW]	Tribo-film formation. SEM	Reduction in COF/enhancement of wear rate.	143)
60 SN Base Oil/ ZnO. [FM, AW]	Mending effect. SEM	Reduction in COF/enhancement of wear properties	157)

4.0 Current Position of Nano-additives Globally

Nano additives (nanoparticles) or lubricants oil additives are employed globally as a way of enhancing the lubricants tribological properties as well as their performances. The application of the additive is strictly base on its property/functional requirement. Current additives (nanoparticles) offers great reduction in waste of materials and elimination of use of harmful chemicals like

cleaning solutions if compared to previously formulated additives ^{5,15)}. In order make the new technology more unique, selected additives are prepared into packages and recommended/labelled for specific base oil for a particular application. This guide is mostly found in the internal combustion engine application. The utilization of these nano-additives can also be found in other areas like gear box oil additives, machining and coolant additives, hydraulic fluid additives and in the metal working lubricants additives. following the current world

sustainability, opportunities has opened from consumption of most of neglected raw materials which turn to be of great contribution in actualizing the nano-additives products. In addition, huge achievement had been recorded from biomaterials toward nano-materials/additive formulation as they possess similar properties to fossil feedstock^{158,159}. The operations perform by these nano-additives depend on the type which include; anti-wear, detergents, extreme pressure, dispersants, viscosity index improver and oxidation inhibitors. Through provision of the various lubricant properties, nano-additives will boost production productivity by reducing friction and power losses in our industries, notably in tribology applications such as base oil, direct lubricants, and grease. Carbon base nanotubes, fullerenes, metal oxides, polymers, ceramics, and composites are the most revealing types of nano-additives. Based on current technology, most machines operate for long periods of time with high loads, necessitating the use of a sufficient additive to regulate severe pressure and prevent wear. Inorganic fullerenes (IF) possess such property and exhibits rolling mechanism, capable of

resisting the wear effects when blended with base oil and grease but the application is still limited in the field. Nano-additive anti-wear mechanism works by depositing nanoparticles in the worn surface's valley, which improves tribological characteristics by lowering boundary friction.

Metal oxides are commonly used in engine oil, metalworking fluids, and a variety of other applications due to their anti-wear and extreme pressure characteristics. They are currently being pushed higher in the global market to replace common ZDDP and Sulphur phosphorous due to their negative environmental impact. (hazardous emission)¹⁶⁰. On the other hand, products of carbon based additives and nano-diamond showcase some excellent properties during lubrication like performing almost all the four lubrication mechanism with good wear and friction resistance. Organic products (biomaterials) of nano-additives yield another opportunity in achieving sustainability goal due to their eco-friendly characteristics and good properties including thermal strength. The recorded results on nano-additives evidently provide solution to the unwanted occurrences during lubrication and stands as promising among every class of additives.

5.0 Summary and Future Scope

On account of their morphology and mechanisms, NPs have shown enormous potential that is predominantly use in lubrication. In the past decade, researchers have explored different ideas and formulations to enhance tribological properties of lubricants including material elements selection, modifications in design geometry and formulation of various forms of additives. Recently, the effect of NP additives performance in improving tribological properties of lubricating oil (base oil and additives) has attracted researcher's attention. Inability of NPs to form homogeneous mixture with lubricating oil (equilibrium of NPs in solution) due to insufficient dispersion agent could lead to agglomeration and may affect the lubricant performance. Our current state of knowledge on keeping lubricating oil (with NPs) enhanced for tribological application is briefly summarized as follows

- i. Reduction in COF and enhancement of wear properties can achieve through NPs additives with excellent thermal conductivity for stabilization of formulated film and surface topography smoothening.
- ii. Viscosity increasing effect with decrease in coefficient of friction due achieved from NPs

- additive with high temperature stability, capable to enhance strength, stiffness and resistance to wear.
- iii. Carbon nanotubes (MWCNT, DWCNT and SWCNT) dispersed with surfactant reduces wear, lower frictional force, Improve anti-scuffing performance with significant reduction in friction.

Although presence of NPs additives may facilitate film formation, enhanced wear properties and load carrying capacity, however, operation on high load for long operating hours is still facing many challenges. As lubrication is associated with various mechanisms, and difficult to understand due to many NPs with different operation for different applications. Another future studies should be directed toward organic biodegradable Nano-additives, chemically modified and hybridized for tribological application. Formulation of new modified nano additives could yield improved mechanism and performance properties which influences the tribological behavior of lubricants.

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