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A Review on Tribological Study of DLC Coatings in Combination with Bio Based Lubricants

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Abstract: In past few years DLC coatings and bio based lubricants have gained significant attraction due to their excellent tribological properties. Biolubricants showed synergetic behavior with contact surfaces, when used as a lubricant. Reason behind the attraction towards biolubricants is that they are renewable and biodegradable source of energy. The dominant properties of biolubricants are high flash point, less coefficient of friction, good wear resistance, high viscosity index, lower toxicity and high biodegradability. On the other hand, diamond like carbon (DLC) coatings have also gained attraction due to their excellent tribological properties which enables them to reduce the COF and wear of contact surfaces. In literature, many experimental studies have been carried out by researchers on DLC coating and biolubricants to analyze their interaction. Although biolubricants are not much applying in practical tribology fields but their properties are significant as compared to conventional synthetic lubricants. In this review paper, data from past few years published papers have been arranged in an organized manner to study the interaction of DLC coatings with biolubricants. Most widely used DLC coatings (W-DLC, a-C:H DLC, ta-C DLC) and biolubricants (palm oil, coconut oil, canola oil, sunflower oil, jatropha oil and rapeseed oil) were considered for this study. Tribological performance of symmetric (DLC) contacts and asymmetric (DLC and steel) contacts with biolubricants have been analyzed by comparing the average values of coefficient of friction and coefficient of wear. Synergetic behavior was obtained when biolubricants were used with symmetric DLC coated contact while tribological results were not much effective in case of asymmetrical contact of DLC coatings and steel.

Keywords: DLC Coatings, Bio Lubricants, Friction, Wear, Tribofilm, Biodegradability

1. Introduction

The steady depletion of crude oil and the adverse environmental impacts of its use have pushed researchers to look for its alternatives [1]. One area of application is automobile lubrication. In this regard, vegetable oil based lubricants are most suitable. Biolubricants have low shear property, which can improve engine efficiency [2]. They also have good anti-wear character, high biodegradability, high viscosity index, lower toxicity, excellent lubricating property, high fire point, high value of flash point and less coefficient of friction [3, 4]. Vegetable oil-based lubricants are becoming more popular not only because of their environment friendliness but also due to their brilliant lubricating properties as compared to mineral based oils [5]. Reduction

in greenhouse gases, less dependence on non-renewable sources and increase in agricultural market have attracted many countries toward biolubricants [6].

Vegetable oils major consist of triglycerides, which are fatty-acid tri-esters of glycerol [7, 8]. Vegetable oils are amphiphilic as a molecule contains both, polar and non-polar groups. Due to their amphiphilic nature they are suitable to use in all lubrication system whether they are thin layer or thick layer lubrication condition [9-11]. The reduction in friction caused by the use of biolubricant directly translates to better fuel economy in an automobile. About 25% of the world's total energy consumed is needed to counter friction [12, 13]. In an automobile engine, much of the fuel's energy is utilized in running auxiliaries and to overcome friction. Main areas in an engine where these losses occur are piston

and cylinder liner assembly, crankshaft bearings, vehicle tires and brake system of vehicles [7]...

There are three types of lubrication regimes which differ due to the thickness of lubricant layer. In boundary lubrication regime, asperities on the surface are in direct contact and applied load is fully shared. However, in mixed lubrication regime load is partially or fully shared by lubricant film. It is observed that lubricant layer shear strength is directly proportional to the friction between the contacting surfaces [16]. Experimental analysis also showed that surface hardness is directly proportional to the wear resistance of contacting bodies. [17]. Therefore, we can add additives in the lubricants to improve their COF and wear resistant property. The main additives in this matter are Zinc dialkylthiophosphate (ZnDDP), Molybdenum dithiocarbamate (MoDTC) and glyceryl monooleate (GMO). These are frequently used and are compatible with most of lubricants and improve their properties to dominant extent. One of the best solutions for improving the surface roughness is surface coating. There are many surface coatings available but the diamond like carbon (DLC) coating are optimal in improving friction and wear response of a system. The most effective DLC coating having both anti-friction and ant-wear properties, when used under specific operating conditions, is ta-C DLC coating [18]. Diamond, the hardest known material has excellent tribological properties due to a phenomenon known as self-lubrication. Experimentation showed that tribological characteristics of diamond film depends on the environmental and working conditions of the system, and has the ability to retain COF against itself or other materials. COF by researchers for diamond is about (0.03) in 1960s [19]. In past few years, a lot of studies have been conducted for understanding the tribological behavior of diamond film which suggests that in sliding contact condition, diamond film is a suitable candidate to control the wear and friction of system [20-24]. The deposition method of coating non-crystalline carbon and defects in grain boundaries of carbon may cause degradation of tribological properties [25].

In this review, tribological properties of biolubricants are studied when the contact surfaces are coated with DLC. Besides this, average values of COF reported in previous works are also analyzed and discussed. Several researches have been conducted to analyze the effect of DLC coatings with biolubricants [5, 9, 26-34].

1.1. Biolubricants

Vegetable oils are extracted from plants and they can be used in most applications directly. However, in most modern applications, such as an engine, vegetable oil are not suitable because of their low oxidative and thermal stability. Chemical modification of these vegetable oil becomes necessary for modern applications. Chemical modification can be done via transesterification reaction, Estolide formation or epoxidation, but transesterification is most common, in which the glycerol backbone of fatty acid is replaced with another, more stable alcohol. The main feedstock oil for chemical modification include castor oil, soybean oil, rapeseed oil, coconut oil, karanja oil, jatropha oil, sunflower oil and palm oil [35]. They are less toxic and highly biodegradable as compared to petroleum-based oils. Type of feedstock used mainly depends upon the geographical location and varies from country to country. There are many countries of world which are known for their vegetable oil crops [3].

1.1.1. Nature of Biolubricants

The polar nature of fatty acids makes biolubricants a suitable candidate to use in boundary lubrication conditions. High flashpoint, high viscosity, good adhesive property and high lubricity of biolubricants is also due to its amphiphilic nature [6]. Long polar chain makes them stick to metal and metal oxide surface layer in boundary lubrication regime [43-45]. Table 2 shows a comparison between mineral oils from American Petroleum Institute (API) group 1 to 3 with the vegetable oils. Properties like viscosity, density, solubility, flash point, pour point, oxidation stability etc. are compared in this table. Although vegetable oils have many desirable properties as lubricant but have poor oxidative and thermal stability, and they do not perform well in low temperature conditions.

Table 1. Most widely used VEGETABLE OILS.

S. No.	Edible Vegetable oils	VOLUME% OF OIL	Non-edible Vegetable oils	VOLUME% OF OIL
1	Peanut	45–55	Moringa	20–36
2	Coconut	63–65	Jatropha	40–60
3	Rapeseed	38–46	Linseed	35–45
4	Corn	48	Neem	30–50
5	Palm	30–60b	Mahua	35–50
6	Olive	45-70	Karanja	30–50
7	-	-	Castor	45–60

1.1.2. Biolubricants Classification

Classification of biolubricants can be done in many ways like source of oil, use of oil, chemical properties of oil etc. Vegetable oils are mainly classified on the basis of toxicity. These oils are extracted by placing plant seeds under pressure which squeezes the oil present in it [6]. Depending on the toxicity they are classified as edible and non-edible. Table 1 shows the vegetable oils as two main categories: edible and non-edible oils [36-42].. Countries import edible oils for their domestic use and the biggest importer of edible oils is India [6]. To use these oils as lubricants is not advisable. Edible oils cannot fulfill the lubrication as well as food demand simultaneously. However, non-edible oils being less expansive and readily available can be used as lubricants.

Table 2. *Properties of vegetable oils.*

| Properties	Vegetable oils	Mineral oils
Viscosity index	100…200	100
Cloud flow behaviour	Poor	Good
Sludge forming tendency	Poor	Good
Solubility in water	Not miscible	Not miscible
Density @20°C (kg/m^3)	940	880
Shear stability	Good	Good
Pour point °C	-20…+10	-15
Miscibility with mineral oils	Good	–
Seal swelling tendency	Slight	Slight
Oxidation stability	Moderate	Good
Hydrolytic stability	Poor	Good
Sludge forming tendency	Poor	Good
Hydrolytic stability	Poor	Good
Oxidation stability	Moderate	Good
Seal swelling tendency	Slight	Slight

1.2. Diamond-Like Carbon (DLC) Coating

In last few decades DLC coatings have been used in many applications and have gained attention due to their excellent antifriction properties. Their main properties include high optical permeability, low specific inducting capacity, low surface energy, high rigidity, low COF and good biocompatibility. Diamond, carbon, and graphite coatings all three have the same chemical structure. Concentration of sp^3 bonding in DLC is about 35-85% and due to this high percentage it forms tetrahedral structure that is similar to internal bonding of diamond [49]. Properties of a structure can be determined by sp^3/sp^2 hybrid bonding ratio. Deposition techniques of DLC coating also effect its tribological properties. The most widely used DLC deposition techniques are physical vapor deposition (PVD) and chemical vapor deposition CVD [18]. These two types have further classifications. Common method which fall in PVD category are filtered cathodic arc deposition (FCVAD) and ion beam assisted deposition (IBAD) while common method under CVD are hot filament chemical vapor deposition (HFCVD) and direct photochemical vapor deposition (DPCVD) [18, 50-52].

1.2.1. DLC Coatings Classification

Main classifications of DLC coating are non-doped DLC coatings and doped DLC coatings. There are further more classifications of both types. Doped DLCs have two main categories i.e. metal doped (W, Cu, Ag, Ti) DLC coatings and non-metal doped DLC coatings. Similarly, non-doped (F, Si, N) DLC coatings have two main branches as hydrogenated coatings and non-hydrogenated coatings. Figure 1 represents DLC coating classifications. The main difference in both types is the percentage of hydrogen present in them. Amorphous carbon coating which have hydrogen content more than 20% are known as hydrogenated (ta-C:H and a-C:H) coatings [53], and coatings less than 20% hydrogen are known as non- hydrogenated (a-C and ta-C)[25, 54, 55]. DLC coatings have significant attraction due to its synergetic properties with lubricants and adhesion with contact surfaces. Reason behind the low COF is its specific properties such as resistance to internal stresses, chemically inertness, high thermal conductivity and excellent adhesion with the substrate material [52, 56-58]. Carbide and nitride coating are also well known for their synergetic tribological properties in oil-lubricated sliding conditions [24, 59, 60]. DLC coating is much harder and stiffer than these coatings [61]. Advantage of DLC coating over these carbide and nitride coatings is its excellent tribological performance even in dry lubricated conditions [62-72]. This is due to their inherent property of self-lubrication.
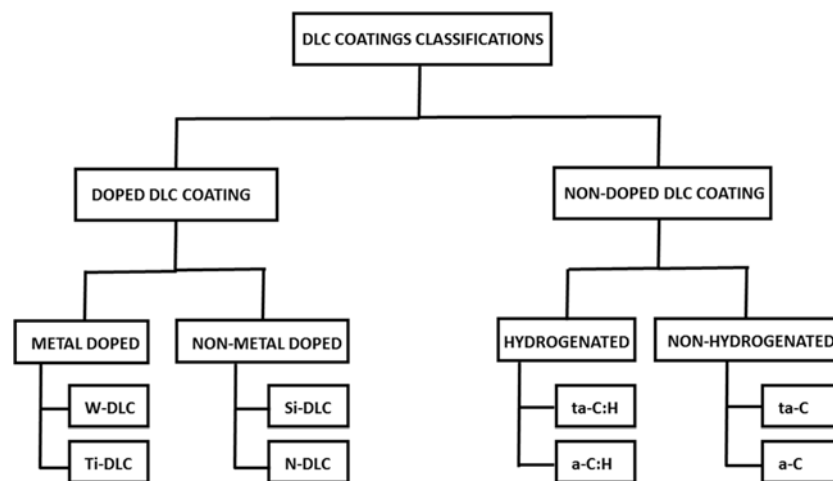

Figure 1. *Major classification of DLC coatings.*

1.2.2. Industrial Applications of DLC Coatings

Now-a-days DLC coating are employed mainly at extreme operating conditions like high contact pressure, high operating temperature and high applied loads. In these conditions, DLC coatings have been most suitable. If the lubricant film between mating parts prevails, then no surface contact occur and result will be low friction and wear values. DLC coating also have self-lubrication property which enables it to be useful in such extreme conditions where the lubricant film can bebroken. In engine components like camshaft, tappet, inlet and exhaust valves, piston of cylinder and cylinder liner are coated with

DLC to improve wear and reduce friction to increase power output.
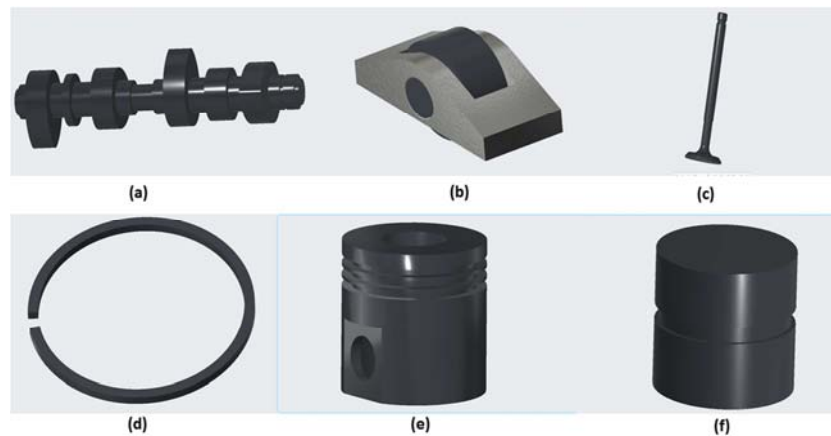
Figure 2. Industrial use of DLC coatings (a) Camshaft (b) Finger Roller Follower (c) Inlet/Exhaust Valve (d) Piston ring (e) Engine Piston (f) Tappet.

Besides friction, DLC coatings also protect the material from corrosion and abrasive wear. Materials with a magnetic property are especially coated with DLC [18, 73, 74]. Many researchers have conducted experimentation by depositing DLC coating on engine parts like and engine assemblies, piston liner, valve train, bearing and gear assembly [75-88]. DLC coated engine parts are shown in Figure 2. All these parts perform in extreme working conditions and DLC coatings have worked extremely well as compared to un-coated parts. In machining of parts, tools are coated with DLC to increase the tool life and to reduce frictional and wear losses. Based on DLC coating's inherent properties their applications are widely spread. Due to its hardness they are coated on surgical blades. Due to its anti-sticking property, they are used in injection molding machine to obtain fine surface finish of product. Due to its anti-wear property, they are used in eye lenses to protect them from scratches and wear [89]. DLC coating cannot withstand the machining of hard materials like mild steel due to the high temperature and absence of lubricant, so for soft metalwork, cutting tools with DLC coatings are used [73].

2. Tribological Study Review: DLC Coatings with Biolubricants

In past few years, researchers conducted tribological experiments on DLC coated contacts with biolubricants. Different experiments with different test parameters have been conducted by researchers which will be discussed later in this review.
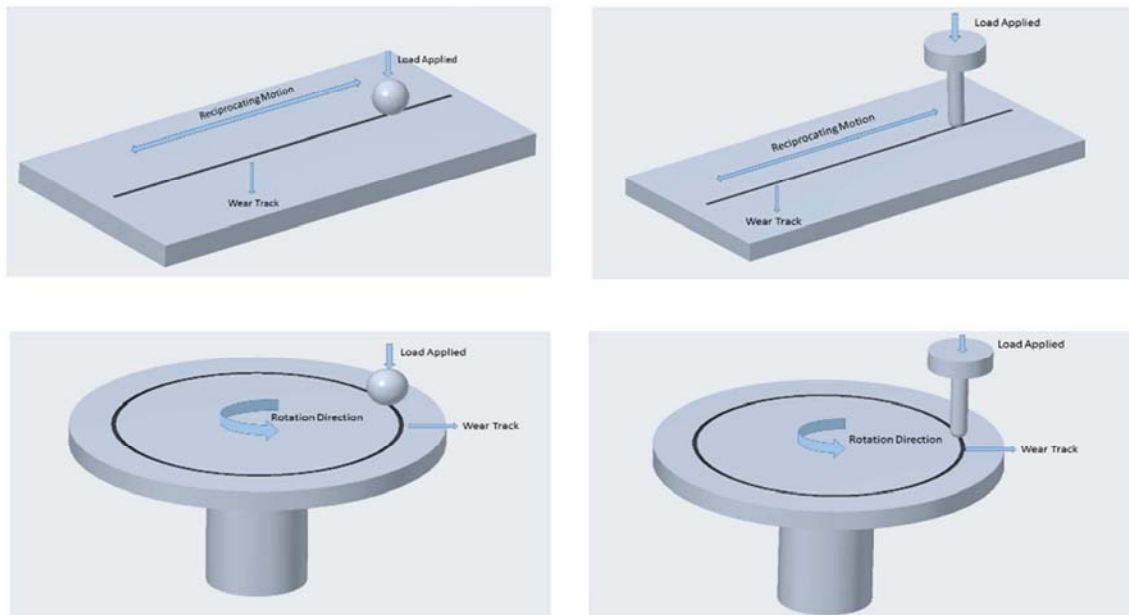
Figure 3. Most widely used tribological test setups (a) ball-on-plate (b) pin-on-plate (c) ball-on-disc (d) pin-on-disc.

2.1. Experimental Setup

To evaluate the tribological properties of DLC coatings with biolubricants, researchers have conducted tribological experiments with many different configurations. Different tribological testing configurations are shown in the Figure 3.

The purpose of these experiments is to examine the friction and wear response of DLC coatings with the biolubricants. Most widely used tribological testing schemes are ball-on-disc, ball-on-plate, pin-on-plate and pin-on-disc. Figure 3 shows the layout of these four types of experimental setup. The testing conditions for experiments vary from each other. Extreme temperature conditions are used to examine the DLC chemical inertness property with biolubricants [90-92]. Each tribotestor has its own specification, based on the direction of motion of substrate or counter body. Some testing configurations are unidirectional i.e. ball-on-disc or pin-on-disc, while others are bidirectional testing rigs i.e. ball-on-plate or pin-on-plate. There are many other testing geometric configurations but these are most widely used.

2.2. Substrate and Counter Body Material

Mostly, substrate and counter body are made up of ferrous and non-ferrous materials. Depending upon the conditions of application, their material is selected. In symmetric contact, substrate and counter body are made up of same type material and in asymmetric contact both have different materials. In asymmetric testing, counter body is made up of nonferrous material like silicon nitride, germanium and aluminum oxide [93-95]. Ferrous materials are also used for substrate and counter body material due to its good hardness property which enables its anti-wear properties. Table 3 shows most common materials used for substrate and counter body literature [5, 9, 7, 24, 26, 30, 32-34, 47, 48, 96-105].

Table 3. Substrate and counter body materials.

S. No.	Reference	Ferrous	Reference	Non Ferrous
1	[34, 93]	AISI 440C	[47, 48]	Silicon nitride (Si_3N_4)
2	[5, 9, 32, 33]	AISI 316	[94]	Aluminium (6061)
3	[26]	High speed steel	[24]	Silicon Carbide (SiC)
4	[7, 30]	AISI 52100 Chrome Steel	[95-98]	Silicon (Si 100)
5	[99]	M2 steel	-	-
6	[100]	Cast iron	-	-
7	[101, 102]	High carbon steel	-	-

2.3. Biolubricants and Synthetic Lubricants

In literature biolubricants gain importance few years back and in recent years many researchers include biolubricants in the tribological investigation. Most widely used biolubricants are shown in Table 1 and their properties are also shown in Table 2. DLC coatings were used with biolubricants by many researchers to investigate their combined effect on wear and friction. Some biolubricants perform very well with respect to tribological outcomes and some do not. The reason behind this is the compatibility of lubricant with the DLC coatings. Synthetic lubricants are also used with DLC coatings and outcomes of the use of synthetic and bio lubrican was examined by many researchers. Synthetic oils were mostly used with additives like zinc dialkylthiophosphate (ZnDDP), glycerol monooleate (GMO) and molybdenum dithiocarbamate (MoDTC) to enhance the tribological properties.

2.4. Characteristics of DLC Coatings

Characteristics of DLC coating can be measured with many techniques which may be pre-process or post process. Many researchers used different techniques to evaluate the properties of DLC before and after tribological testing. Hardness is the most important property of DLC coating and it is measured with nano-indentation hardness testing technique [29]. These techniques help in finding the optimum results of wear, friction, adhesive property and roughness etc. AFM technique is employed to find the surface roughness of DLC coated surface [106]. NMR spectroscopy technique is used to determine the hardness as well as elastic modulus of DLC coating [96, 103]. Scratch testing technique is used to find the adhesion of DLC coating, Feld emission scanning electron microscopy (FESEM), x-ray diffraction and scanning electron microscopy (SEM) are post process techniques and used to find the wear profile and wear scar diameter [7, 26, 96, 97, 104, 107-109].

Table 4. Most widely used DLC coating Characterization Techniques.

S. No.	Reference	Techniques Used	Purpose
1	[97, 106]	Atomic force microscopy (AFM)	Surface roughness
2	[93, 96]	Nuclear magnetic resonance (NMR)	Hardness and Elastic modulus
3	[5, 34]	Scanning electron microscopy (SEM)	Wear and Friction analysis
4	[94, 99]	Field Emission Scanning Electron Microscopy (FESEM)	Wear analysis
5	[95]	Filtered cathodic vacuum arc (FCVA)	Wear Rate
6	[98, 110]	Feld emission electron microscopy (FEI)	Particles observation
7	[94, 99]	Field Emission Scanning Electron Microscopy (FESEM)	Wear analysis
8	[29]	Nano-hardness tester (NHT)	Nano Hardness measurement
9	[9, 101]	Raman spectroscopy	Extent of graphitization
10	[96]	Transmission electron microscopy (TEM)	Nano structure analysis
11	[111]	Elastic recoil detection analysis (ERDA)	Depth profiles analysis
12	[102]	Optical Microscopy	Wear analysis

Raman spectroscopy is used to determine the structural properties after tribotesting. Raman spectroscopy finds the extent of graphite and diamond present in the DLC structure. Due to the chemical interactions, diamond coatings are

converted to graphite. This phenomenon is known as graphitization. This technique is also used to determine the percentage of sp^3 and sp^2 content present [5, 9, 26, 30, 33, 34, 99, 107]. Techniques used in the study under consideration and the purpose of these techniques are shown in Table 4 [5, 9, 29, 34, 96, 97, 99, 100-106, 110, 111].

2.5. Tribological Performance of DLC Coatings

In this study DLC coatings are evaluated with respect to

tribological performance. Biolubricants are used with DLC coatings to further improve the tribological results. DLC coatings are doped with metals and non-metals to increase their chemical reactivity, which results in low value of COF and increases wear resistance of DLC coatings. Un-doped DLC coatings are also analyzed and discussed in this review. Table 5 shows the DLC coatings and type of contacts that are used in the literature that is considered in this review.

Table 5. DLC coating and Contact type used in the study under consideration.

Reference	DLC Coating	Substrate Material	Contact Type
[107]	W-DLC	Hardened steel	W-DLC/Steel
[108]	ta-c DLC	Cast iron	ta-C/Cast Iron
[26]	ta-C DLC	440C stainless steel	ta-C/Steel
[97]	a-C:H DLC	Silicon	a-C:H/Steel
[27]	Metal Doped	Ductile iron	Metal doped DLC/Cast Iron
[29]	a-C:H DLC	-	Steel/Steel
	TiAlN DLC	-	-
[30]	W-DLC	AISI 52100	Steel/Steel
	-	-	Steel/Cat Iron
	-	-	W-DLC/W-DLC
	-	-	W-DLC/Cast Iron
[110]	ta-C DLC	-	Steel/Cast Iron
	-	-	Cast Iron/Cast Iron
	-	-	ta-C/Cast Iron
[31]	Si-DLC	-	Steel/Si-DLC
[32]	ta-C DLC	AISI316 stainless steel	ta-C/Steel
[33]	a-C:H DLC	AISI316 stainless steel	a-C:H/Steel
	ta-C DLC	-	ta-C/Steel
[9]	a-C:H DLC	440C stainless steel	Steel/Steel
	-	-	a-C:H/Steel
	-	-	a-C:H/a-C:H
[34]	a-C:H DLC	440C stainless steel	Steel/Steel
	-	-	a-C:H/Steel
	-	-	a-C:H/a-C:H
[5]	a-C:H DLC	AISI316 stainless steel	a-C:H/Steel
	ta-C DLC		ta-C/Steel

In this review comparison between DLC coatings with biolubricants and DLC coatings with synthetic or mineral base oils is also consider. Doped DLC coating and un-doped DLC coatings that are used are shown in Table 5. Substrate material that is mostly used in tribotesting is AISI 52100 stainless steel. Beside these other materials were also used by researchers and are also shown in Table 5 [5, 9, 26, 27, 29-34, 104, 107, 108, 110,]. In past few years, many researchers have worked on biolubricants and synthetic oil with DLC coating mated parts. In some cases, biolubricants perform well and in some cases synthetic and mineral oils perform well, depending on the testing conditions, contact type and DLC coating type.

Experimental data from published papers have been considered for this study to examine the compatibility of biolubricants with DLC coatings. For friction value average COF values are considered and compared with each other in

similar contact conditions. Lubricants with additive are not included in this review. For wear calculations, data was available in different configurations. Some researchers discuss wear coefficient values; some discuss wear scar diameter while others discuss wear volume. Beside this some discussed wear values for composite of substrate and counter body and some discus for only substrate or counter body. To address this issue average values are taken and represented in the form coefficient of wear or wear coefficient. A list of biolubricants, synthetic oils and mineral base oils that are used in the works under consideration are shown in Table 6 [5, 7, 9, 26, 27, 29-34, 97, 101, 104, 106, 108, 110, 112]. Testing parameters like temperature, test duration, tribological setup used, calibration standard, type of motion and load applied are shown in Table 7 [2, 5, 7, 9, 26-34, 96-116].

Table 6. List of Lubricants which are used in current study.

Reference	Bio Lubricants	Synthetic Lubricants	Fully Formulated
[108]	-	-	-
[7]	Karanja Oil	-	-
[26]	Palm Oil	PAO	-

Reference	Bio Lubricants	Synthetic Lubricants	Fully Formulated
[106]	Palm Oil	-	-
[99]	-	PAO	-
[97]	-	PAO	-
[27]	Bio Fuel (ph 2-3)	-	-
[94]	Waste Cooking oil	-	SAE 40
[29]	Bio Degradable Polymers	-	-
[30]	Palm Oil	PAO	-
[110]	-	-	SAE 5w-30
[31]	Rapeseed oil	-	-
	Soybean oil	-	-
[112]	Pongamia oil	-	-
[32]	Sunflower oil	-	-
	Palm oil	-	-
	Coconut oil	-	-
[33]	Jatropha oil	-	-
[9]	Jatropha oil	-	SAE 40
[34]	Canola oil	-	-
[5]	Jatropha oil	-	-

***Table 7.** Testing Parameters of study under consideration.*

Reference	Calibration Standard	Normal Load (N)	Test Temperature (°C)	Test Duration (min)	Setup Configuration	Motion Type
[107]	ASTM D4172	15, 25 and 35	28–30	60	Ball-on-plate	Reciprocating
[108]	-	100 and 1000	50	180 and 66	Block-on-ring tests	Rotational
					Pin-on-disc	Rotational
[7]	ASTM D4172-94	150 and 400	75	60	Four-ball Tribotestor	Rotational
[95]	-	2	-	15, 30 and 45	Ball on disc	Rotational
[109]	ASTM G99-05	20 to 100	2	-	Ball on disc	Rotational
[111]	-	2, 3 and 4	25	-	-	-
[26]	-	100	40–125	120	Ball-on-plate	Reciprocating
[93]	-	10	23	-	Pin-on-disc	Rotational
[106]	-	1-10 nN	15–100	-	-	-
[96]	-	1	25	60	Pin-on-disc	Rotational
[100]	-	150	25	30	-	-
[99]	ASTM D-2783	400-1600	40 and 90	10 sec	Four-ball Tribotestor	Rotational
[97]	-	1	25	60, 120, 180 and 240	Ball on disc	Rotational
[101]	-	-	55 - 970	3	-	-
[27]	-	140	85	60	Cylinder-Liner Tribotestor	Reciprocating
[2]	-	20	25	3	Pin-on-disc	Rotational
[98]	ASTM E384–11	5	25	-	-	-
	ASTM C1327–15				-	-
[28]	-	40	100	60	Ball-on-plate	Reciprocating
[116]	-	20	25	-	Pin-on-disc	Rotational
[94]	-	20 to 90	70	10	Cylinder-Liner Tribotestor	Reciprocating

***Table 7.** Continued.*

Reference	Calibration Standard	Normal Load (N)	Test Temperature (°C)	Test Duration (min)	Setup Configuration	Motion Type
[113]	ASTM G-99a	-	23 to 150	-	Pin-on-disc	Rotational
[114]	-	5	40 and 100	60	Ball on disc	Rotational
[102]	ASTM 4172.	392	75	60	Four-ball Tribotestor	Rotational
[29]	ISO 14577	50	23 ±1	-	Ball on disc	Rotational
	ASTM E25				-	-
[30]	ASTM D-2783	40 and 615	90	120	Four-ball Tribotestor	Rotational
[110]	-	5	25	-	Ball on disc	Rotational
[31]	DIN51834	10	25 and 150	120	Ball on disc	Rotational
[112]	-	15	-	30	Pin-on-disc	Rotational
[115]	ASTM D2596	400 and 800	30, 45, 60 and 75	5	Four-ball Tribotestor	Rotational
	ASTM D2783				-	-
[32]	-	390	50	60	Ball on disc	Rotational
[33]	-	392	50, 100 and 150	120	Ball-on-plate	Reciprocating
[9]	ASTM D4172	392.4	100	60	Four-ball Tribotestor	Rotational
[34]	-	10	80	-	Ball-on-plate	Reciprocating
[5]	-	392	50, 100 and 150	60	Ball-on-plate	Reciprocating

3. Friction Performance

3.1. W-DLC/W-DLC Contact

In recent years Tungsten DLC coatings (W-DLC) have emerged as potential source to reduce the friction and wear of the mating surfaces. It has been found that when these coatings are used on mating surfaces they interact with the lubricants in positive manner by forming tribofilms which help in improvement of tribological parameters. Wear behavior has been investigated using different lubricants. Rapeseed Oil, PolyalphaOlefin (PAO) and Di-ester oil were used by Karzan et. al. It was found that in presence of rapeseed oil COF of W-DLC symmetric contact was reduced to minimum while highest friction was encountered in case PAO lubricant. This shows that W-DLC coatings have synergetic relation with rapeseed oil which helped to reduce the friction of the mating surfaces [117]. In another study friction properties of symmetrical tungsten doped DLC coatings were studied in API (American Petroleum Institute) group III base oil. These coatings also performed well with mentioned base oil. The positive behavior of these coatings with base oil was attributed to the capability of these coatings to form tribofilms which help in sliding of the mating surfaces which, in turn, reduces metal to metal contact [118].

In few experimental studies these coatings were tested using different experimental techniques with paraffinic mineral oil. Several tests were conducted with and without anti-wear and extreme pressure additives in the base oil. In all of the tests these coatings performed extremely well with mineral oil [119, 120]. W-DLC symmetrical contacts were also tested with TMP (Trimethylolpropane). It was found that synergetic relation exists between these two. As COF was found lower as compared to dry lubrication condition. This positive effect on COF was attributed to the capability of these metal doped DLCs to form tribofilms by interacting with different lubricants [30]. Figure 4 shows the comparison of average COF when different lubricants were tested with W-DLC/W-DLC symmetrical contacts under different operating conditions. It can be seen that paraffinic mineral oil was not a suitable lubricant to be used with W-DLC coated contacts. Although rapeseed oil performed well and resulted in low COF, the average COF for API group III oil was found to be lowest. This shows that a synergetic relation exists between tungsten doped DLC coating and API group III oil. This positive behavior of group III oil on the average COF associated with tungsten doped DLC coatings can be attributed to the surface graphitization or formation of oxygen containing tribofilm [118].
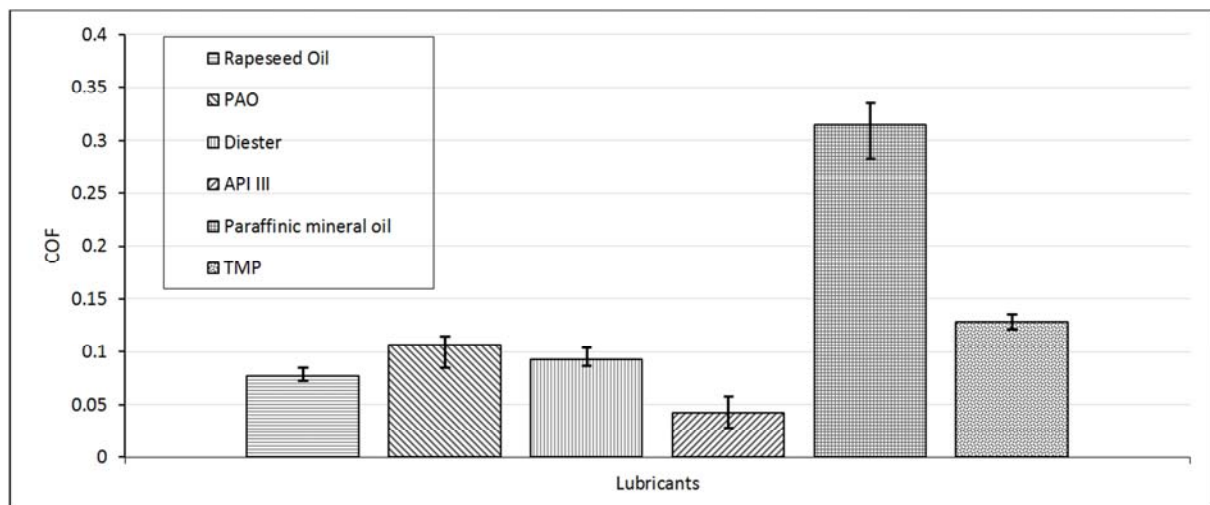

Figure 4. Effect on average COF for W-DLC/W-DLC contact for different lubricants.

3.2. a-C:H DLC / a-C:H DLC Contact

In recent years hydrogenated amorphous carbon is the most widely tested DLC coating. In a number of experimental studies have been conducted to check the suitability of these coatings for different lubricants. In these studies, COF was found to be reduced as compared to dry sliding conditions. Few studies have been conducted in which PAO was used with these coatings to enhance the tribological properties of the mating surfaces. PAO performed well with these coatings and average COF was found to be 0.0776. This low COF associated with use of these coatings with PAO was attributed to formation of weakly adhered tribofilms which helped in

increased lubricity resulting in reduced friction and wear [121, 122]. It has been found that these coatings are capable to perform extremely well with biolubricants. In one of the study these coatings were used in presence of Jatropha oil which is a biolubricants. Result of the experiment showed that these coatings have a potential to reduce the COF and wear of the mating surfaces. In the same study these coatings were also tested with SAE 40 oil. The results of the tribological test showed that average COF associated with use of these coatings with the mentioned lubricant was similar to jatropha oil. This low COF associated with use of jatropha oil can be attributed to high polar nature of the oil and presence of large unsaturated molecules in the biolubricants [9]. Mineral oil was not able to

perform well with a-C:H DLC symmetrical contact and resulted in higher COF as compared to COF associated with use of other lubricants. This negative effect on COF associated with use these coatings with mineral oil can be attributed to the extremely week tribofilm formed. The make and break of this tribofilm resulted in high COF [118]. Friction performance of these coatings were considered with API group III oil. Experiments were carried out under different operating conditions to find most optimum operating conditions. Results of the tribological tests showed that COF was greatly reduced by using API group III oil. This positive behavior on COF was attributed to synergetic relation between hydrogenated amorphous carbon DLC and API group III oil. Surface

graphitization and formation of tribofilm were the main reasons for this positive behavior COF [118, 123]. Canola oil performed extremely well with neat API group III oil and COF was found to be lowest as compared to other lubricants considered for comparison [34]. Figure 5 shows the variation in COF when hydrogenated amorphous carbon is used as a coating in different tribological tests in presence of different lubricants. It can be clearly seen that canola oil exhibited best performance and lowest COF was reported in this case. This extremely well behavior of canola oil can be attributed to large amount of polar component of oil that promotes lubricity by forming a lubricating film with a-C:H coatings and graphitization of these coatings [34].
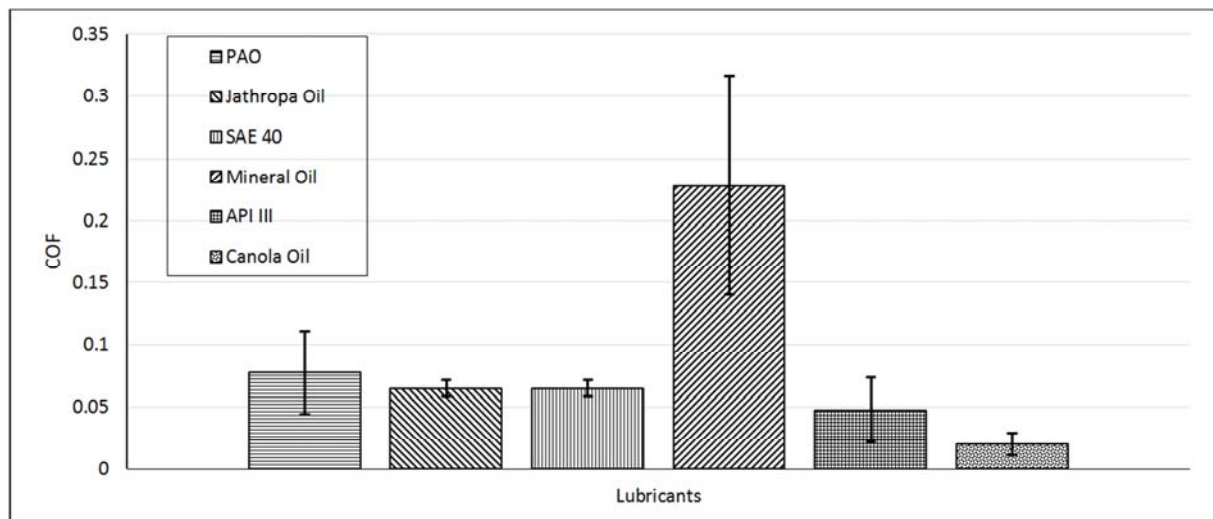

Figure 5. Effect on average COF for a-C:H/a-C:H contact for different lubricants.

3.3. ta-C DLC / ta-C DLC Contact

A lot of experimental work has been conducted in recent years to check the suitability of non-hydrogenated tetrahedral carbon DLC coatings with different lubricants. But no considerable work is conducted to check the suitability of biolubricants with these coatings. In an experimental study, friction performance of these DLCs was investigated with mineral oil. Friction tests were conducted using a Mini Traction Machine (MTM), where DLC coated ball was rubbed against DLC coated plates at different operating conditions. Results showed that COF was reduced by using DLC coated mating surfaces as compared to the interacting surfaces without DLC coatings. While the presence of lubricant further reduced average COF. Average COF was found minimum when balls and plates were coated with DLC and tests were conducted in the presence of mineral oil. This reduction in COF was attributed to graphitization and formation of oxygen or parent material based tribofilm which improved the tribological characteristics of the interacting surfaces [118]. In another study tribological suitability of these DLC coatings was investigated with API group III oil. When compared with effect of these coating on tribological characteristics associated with mineral oil it was found that this coating has more potential to enhance the tribological properties of API group

III oil. This reduction in COF associated with use of API group III as a lubricant can be explained by the formation of highly efficient tribofilm and positive interaction between the lubricant and non-hydrogenated tetrahedral carbon DLC coatings [123]. Among the considered lubricants used in past few years to improve the tribological properties of different mating surfaces PAO has performed extremely well to enhance the friction resistance of the mating surfaces when used with ta-C DLC having symmetrical contacts [124]. Figure 6 shows the comparison of the effect on average COF associated with these coatings when tested in presence of different lubricants. It can be clearly seen that average COF was much lower in case of PAO as compared to other oils. Which highlights the suitability of this coating with PAO and shows that a synergetic relation exists between the lubricant oil and applied coating. Although a number of studies have been conducted in order to check the suitability of different lubricants with these DLC coatings. But in recent years no considerable study is found where these coatings are tested with biolubricants. Which shows that there is a need to study the effect of different biolubricants on friction and wear performance associated with these coatings. So that a lubricant can be suggested for this DLC coating with which it can show a synergetic relation and help to reduce the average COF and wear to lowest possible levels.
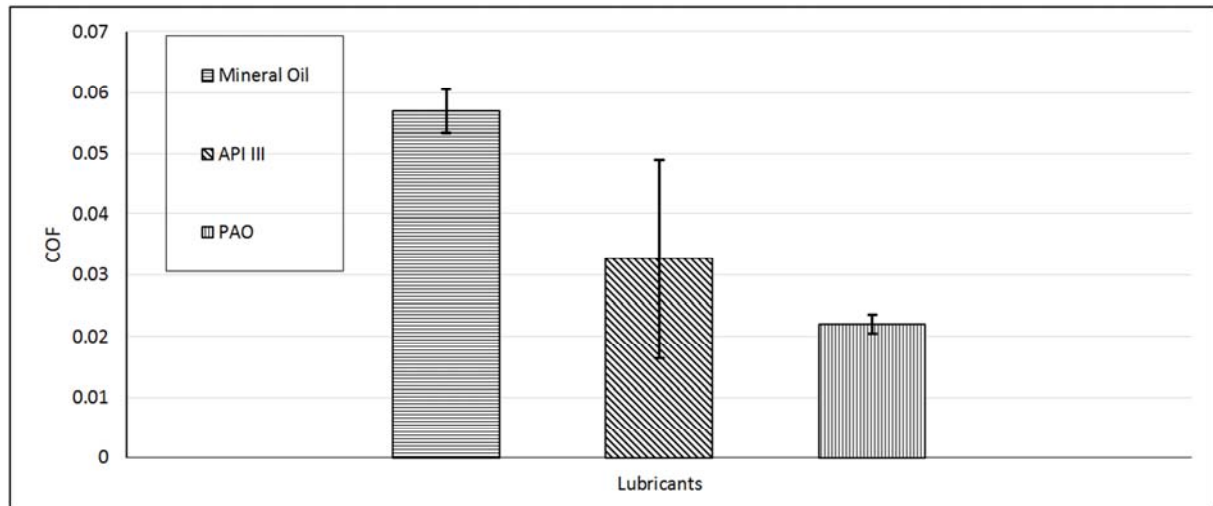
Figure 6. Effect on average COF for ta-C/ta-C DLC contact for different lubricants.
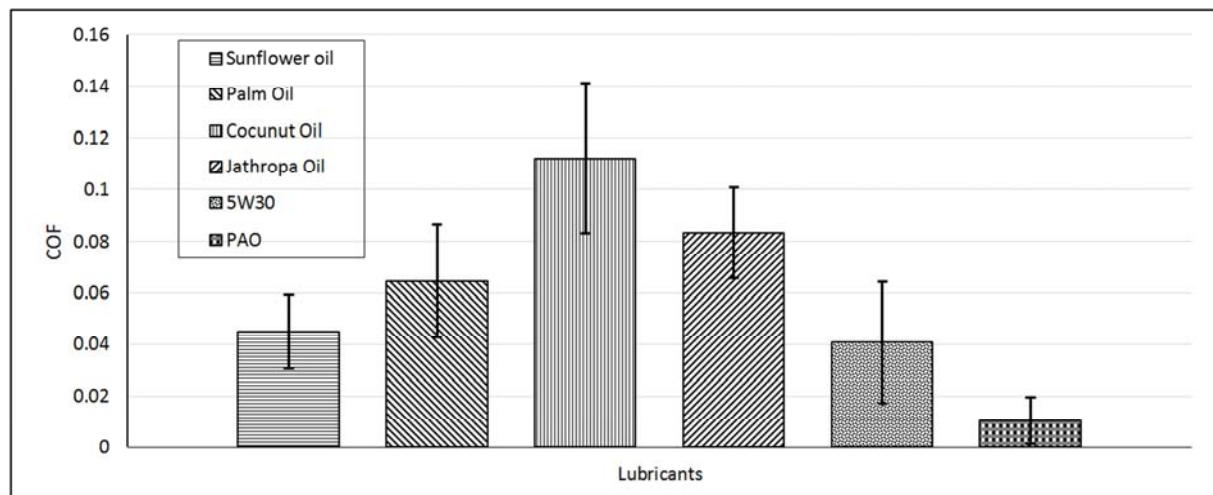

Figure 7. Effect on average COF for ta-C/Steel contact for different lubricants.

3.4. ta-C DLC / Steel Contact

A number of experimental studies have been conducted in order to find the best lubricant to be used with ta-C/Steel contact. A few biolubricants are considered in this regard. In one study, sunflower oil was used as a lubricant in a tribological test in order to find its suitability with ta-C/ Steel contact. Friction and wear performance of the mating surfaces was highly improved with the presence of sunflower oil in the interface formed between ta-C DLC and steel contact. This positive effect on tribological properties was attributed to the formation of tribofilm and carbon transfer layer. In the same study palm oil was also used as a test lubricant in order to check its performance with asymmetrical ta-C/steel contact. Palm oil also exhibited good tribological performance but average COF found in this case was higher as compared to sunflower oil. Although a tribofilm was formed in this case too but it was inefficient. Like sunflower oil, carbon transfer layer was also found in palm oil which helped to reduce the wear of the mating surfaces. Coconut oil was not able to perform efficiently with the asymmetrical

DLC contact and the COF resulting from the tribological test was found higher as compared to other lubricants. It was reported that coconut oil was not able to show synergetic relation with the ta-C/steel contact and was unable to form tribofilm in this case [32]. When in the same contact type jatropha oil was used, it showed better performance with respect to average values of COF. Jatropha oil showed slightly good behavior with this interface as compared to coconut oil and reduced COF of the mating surfaces. This positive effect on COF due to the use of jatropha oil was attributed to the chemical interaction of this oil with the coating which helped in formation of tribofilm. With the use of jatropha oil, COF decreased first and then began to increase. The reason behind this behavior of jatropha oil is attributed to the graphitization of DLC coating [33]. Like most of the previously discussed contacts, synthetic lubricants also showed better performance in this case. COF was greatly reduced when ta-C/Steel contact was lubricated with SAE 5w-30 oil. The positive behavior of these coatings with such fully formulated oils can be attributed to the presence of certain friction and wear modifying agents present in the package. Which interacts chemically with the DLC coatings and

helps in formation of efficient tribofilms thus reducing friction and wear associated with the interacting surfaces [86]. Among the lubricants tested with this interface, PAO performed best and showed lowest COF as compared to other lubricants. It was found that this ultralow friction coefficient associated with use of PAO in a sliding contact is approximately equal to friction coefficient of the roller bearing. Figure 7 shows the comparison of COF when different lubricants were used in ta-C/Steel contact. It can be clearly seen that among the lubricants used, PAO performed extremely well and had lowest COF as compared to other lubricants. Among the biolubricants, Coconut oil showed highest COF and was unable to form efficient tribofilms at the interface while sunflower oil was able to reduce the friction coefficient. This effect in not prominent on COF of the interacting surfaces for other biolubricants. This shows that only PAO has a synergetic relation with the DLC by chemically interacting with the DLC and forming tribofilm.

3.5. a-C:H / Steel

A lot of work has been done on the study of a-C:H/Steel contact with different lubricants. Some studies have also focused on the use of biolubricants with this interface. In one of the study PAO was used as a lubricant in order to modulate the friction performance of the interacting surfaces. It was found that friction coefficient was gradually reduced with use of PAO as compared to dry lubrication condition. While the addition of anti-wear and antifriction additives also enhanced the friction performance [104]. In another study SAE 40 oil was used as lubricant to improve the friction performance of the interacting surfaces. COF was reduced with use of this fully formulated lubricant. It has been found that DLC coatings perform well with the most of the fully formulated lubricants due to the presence of anti-wear and

extreme pressure additives in them [9]. Canola oil also exhibited better performance with the a-CH/Steel contact and modulated the friction coefficient in positive manner. The reason behind the low friction associated with a-CH DLC coating in the presence of canola oil is the presence of extremely large amount of polar components in canola oil which helps in increasing the lubricity between the interacting surfaces by forming a lubricating film with a-CH coating. The canola oil also has potential to promote graphitization of the a-CH coating which ultimately reduces the friction coefficient and increases friction resistance of the interacting surfaces [34]. Presence of jatropha oil at a-CH/steel contact modulated the friction performance of the interacting bodies. Jatropha oil helped to decrease the variation of the friction coefficient with the increase in temperature and helped to maintain a steady friction coefficient [33]. Like the asymmetrical contact discussed in the case of ta-C DLC, asymmetrical contact in this case showed the good performance with the SAE 5w-30 oil, too [125]. Among the considered lubricants for asymmetrical contact of this category of DLC least COF is found when this DLC is used with SAE 5w-30. Figure 8 shows the comparison of friction performance of a-CH/Steel contact when different lubricants are used as an additive. It can be clearly seen that lowest COF is reported for the case of SAE 5w-30 oil as compared to other lubricants. This positive behavior associated with use of SAE 5w-30 can be attributed to the presence of antifriction and anti-wear additives present in the package which helps to attain a lower COF as compared to other lubricants. Although canola oil performed well with asymmetrical a-CH DLC contact, the positive effect is much less as compared to fully formulated oils.
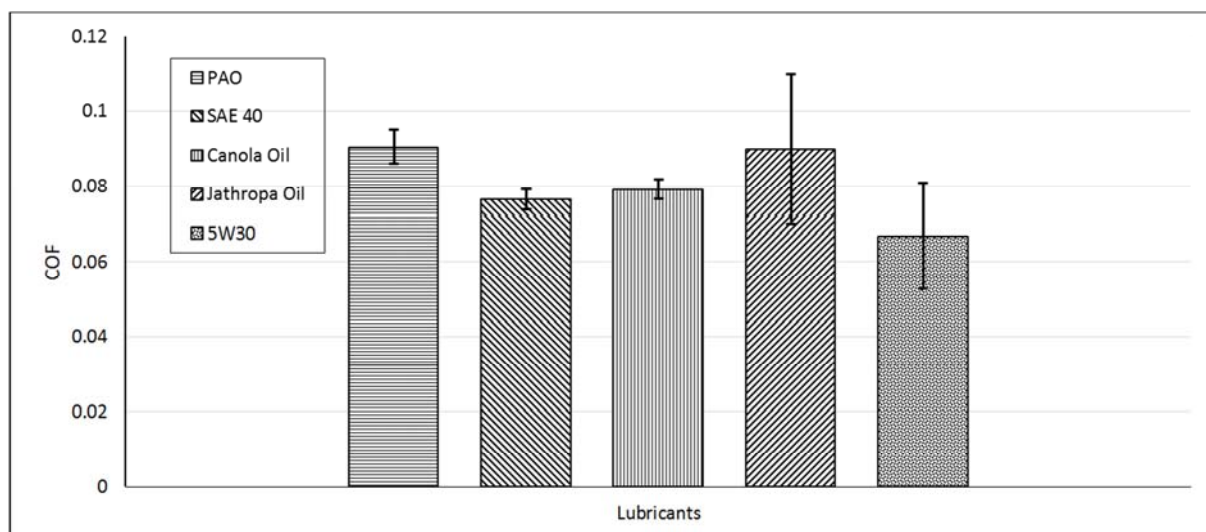

Figure 8. Effect on average COF for a-CH/Steel contact for different lubricants.

4. Wear Performance

Wear between two mating parts exist when they have

boundary lubrication between them i.e. direct contact between surfaces. Wear can be classified in four major categories: adhesive wear, abrasive wear, third-body wear and fatigue wear. Adhesive wear occurs due to the localized

bonding between the contacting surfaces. Abrasive wear occurs due to the removal of material from soft body when surface asperities strike with each other. Third-body wear is exhibited by soft body, when hard particles from a third body are present between mating surfaces ploughing one of the two mating bodies. Fatigue wear occurs due to the formation of debris when a process is cyclic. All these types of wear are illustrated in Figure 9.
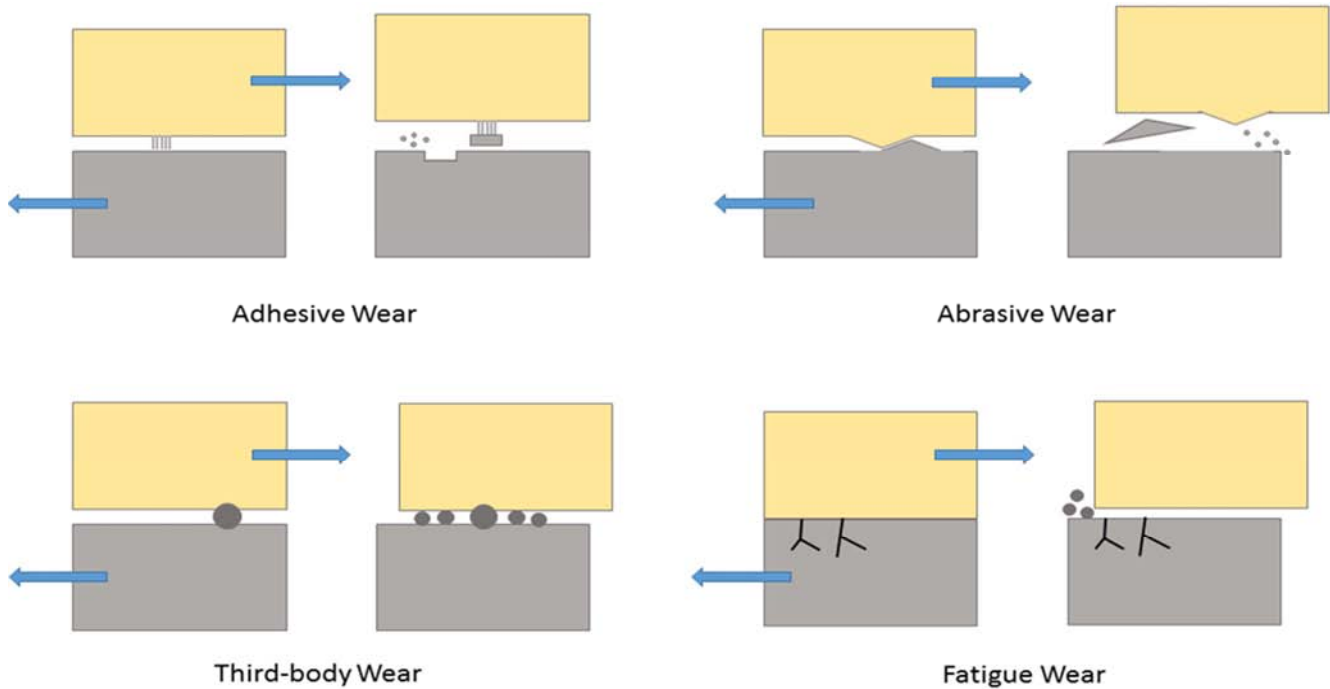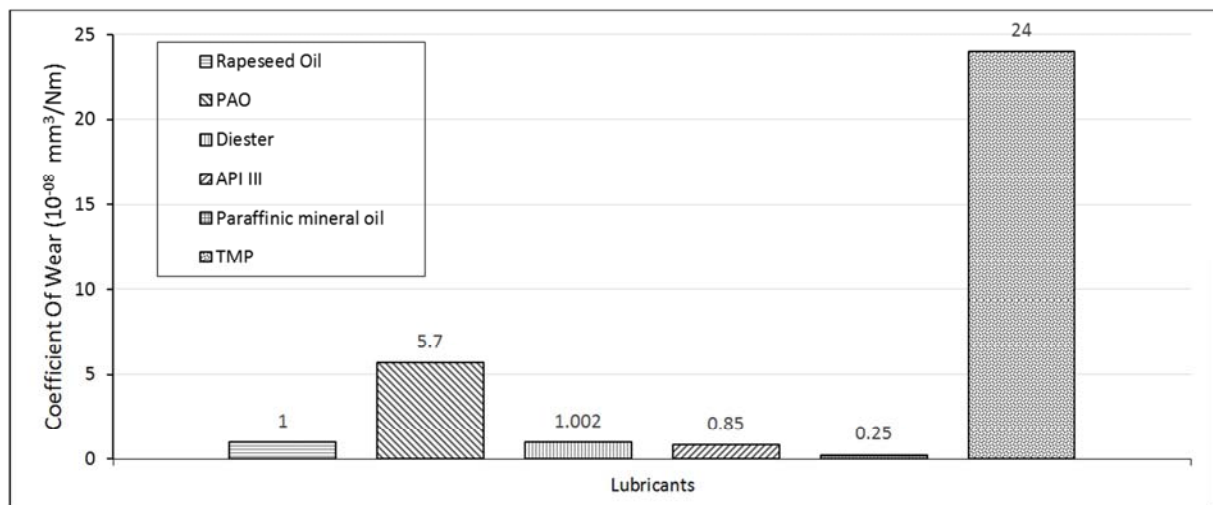

Figure 9. *Types of wear.*

Figure 10. *Effect on wear coefficient for W-DLC/W-DLC contact for different lubricants.*

4.1. W-DLC / W-DLC Contact

Symmetric contact between metal doped DLC coatings shows excellent performance in wear and COF reduction. This is attributed to the self-lubrication property of DLC coatings. In this study, lubricants are evaluated with respect to their synergetic behavior with DLC coatings. In past few years, many studies have been conducted to evaluate the tribological properties of biolubricants and synthetic lubricants. When rapeseed oil is tested with W-DLC/W-DLC contact it showed synergetic behavior and increased the wear resistance of DLC coating [117]. This is due to the presence of thick film of lubricant, which resist to form boundary lubrication regime. As a result, wear rate is reduced. The polar functional group in triglyceride structure of rapeseed oil provide resistance in wear rate. With the same testing conditions and parameters, a synthetic lubricant, PAO, is used and in this case wear rate is very higher. This is due to the absence of effective protective layer of lubricant on the DLC coating surface. When a diester is used with same configuration then its wear rate is almost the same as in case of rapeseed oil. This is due to the saturated chain structure of fatty acid which is present in diester [117].

Here it can be accomplished that the polarity of ester effects its tribological properties. In case of API III mineral oil, the wear rate is slightly low than rapeseed oil. This is due to the surface graphitization of W-DLC coating [118]. In case of paraffinic mineral oil, due to the graphitization phenomenon, the wear rate is the lowest with respect to all others. This shows the synergetic behavior of coating and lubricant [119]. In case of TMP ester lubricant, which is a synthetic biolubricant. Wear rate is very high which might be due to the formation of graphite layer of DLC coating. This soft graphite layer greatly effects the wear rate of contact [30]. Figure 10 shows a comparison between the lubricants which are used for W-DLC/W-DLC contact type. It can be seen that the minimum wear occur in case of paraffinic mineral oil and highest wear rate is obtained when we use additive free TMP ester as lubricant. Bio lubricants also show good behavior in wear resistance and are supposed to behave more synergetic when anti-wear or anti-friction additives are present in them.

4.2. a-C:H / a-C:H Contact

Symmetric a-C:H coated contact have been studied by many researchers in past few years. a-C:H coatings have been proved to be optimum in wear and friction reduction. In current study our aim is to analyze the symmetric a-C:H DLC contact with both biolubricants and synthetic lubricants. When PAO is used as lubricant with this contact, it behaves better as compared to uncoated steel contact but significant wear can be observed. While if additives were added in lubricants, then their tribological properties were improved [121]. In another study

jatropha oil is used with same contact and results were analyzed. Jatropha oil did not perform well with a-C:H symmetric contact. This poor behavior is because of the formation of transfer layer on the counter surface. This layer contains nano graphite particles which cause third body wear phenomenon and graphitization of DLC coating [9]. When SAE 40 oil, fully formulated synthetic oil, is used with amorphous hydrogenated contact type as discussed above, wear results were not satisfactory as compared to PAO. This is due to the DLC coating graphitization and thermal stresses which were produced at the wear track region [9, 126]. Mineral oils were tested with same contact type and results suggest the synergetic behavior of mineral oils with DLC coating. The reason behind this low wear is the local adhesion of the lubricant with DLC surface [118]. Mineral oil performed well as compared to both jatropha oil and PAO. The best results of wear were obtained when canola oil is used. Canola oil performs with synergetic behavior with a-C:H coating due to its polar components which form the lubricant film on the coated surface [34]. A comparison between all lubricants which were discussed earlier is shown in Figure 11. Least wear was obtained with canola oil and maximum wear was obtained in the case of fully formulated lubricant SAE 40. Mineral oil performed well as compared to synthetic oil. Positive behavior of canola oil is attributed towards the formation of lubricant film layer on the coated surface and due to the presence of polar components in the canola oil. Reason behind the poor performance of SAE 30 is the graphitization of DLC coating and development of thermal stresses and strains at wear track area.
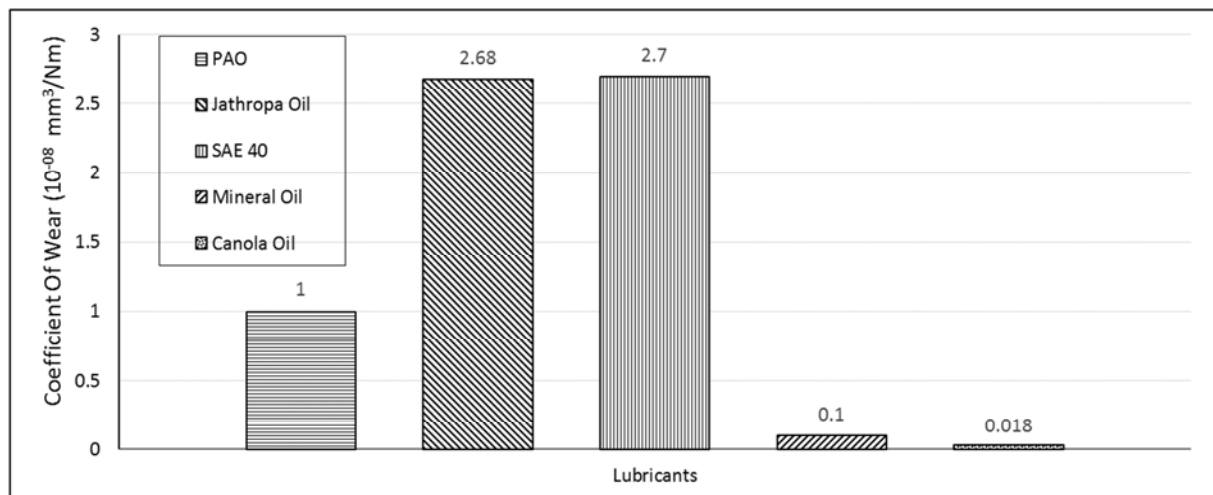

Figure 11. Effect on wear coefficient for a-C:H DLC / a-C:H DLC contact for different lubricants.

5. Discussion

In this study, interaction of DLC coatings with biolubricants was investigated by going analyzing previous works. The tribological behaviors of lubricants with symmetric DLC contacts and asymmetric DLC coated and steel contact is studied. It was found that in most of the cases, biolubricants show synergetic behaviors with DLC coatings

while results of few studies was not in the favor of biolubricants. Reason behind this is the formation of transfer layer on the counter body which contains nano graphite particle causing third body wear between contacting surfaces. When W-DLC symmetric contact was analyzed, API III group oils perform the best in average COF value and biolubricants were at second place but they have also shown good behavior in tribological respect. Paraffinic mineral oil was not suitable to use with W-DLC coatings as it has the

worst results in comparison with other oils. With wear characteristics, TMP ester which is a synthetic oil show poor results while paraffinic mineral oil and rapeseed oil show synergetic behavior with respect to wear. It can be stated that API III and rapeseed oil will be the good choice to use with W-DLC symmetrical contact as they both performed well in wear and friction case.

While studying the hydrogenated (a-C:H) DLC symmetric contact. It was found canola oil biolubricant performed well in terms of reduced friction and wear. The reason behind the good performance of biolubricants may be attributed toward their polar nature. Polar components help in improving lubricity and also the graphitization process. While with the same DLC coating i.e. a-C:H DLC, mineral oil did not perform well and have the worst results of COF with respect to all other lubricants. This shows that with this DLC coating, mineral oil will not be an appealing candidate due to the formation of weak tribofilms which can be easily broken with even less contact pressure.

In the past, no work has been done on ta-C symmetric contact with biolubricants and there is a need to carry out research in this area. Although investigation of other lubricants was done for this contact and found that PAO oil performed well. With asymmetric contact of DLC coating and steel it was found that no biolubricants under study was able to show its dominant behavior but PAO oil show very good result with the DLC coatings and making the tribofilm with the coating which reduces the COF. Like this case, when asymmetric contact between a-C:H and steel was studied by average values then results were also identical to ta-C and steel contact. SAE 5W-30 fully formulated lubricant performed the best in all test lubricnats. The reason behind this was the presence of anti-wear and anti-friction particles in the SAE 5w-30 lubricant. Although in biolubricants canola oil performed well but not as significant as of SAE 5W-30 oil.

6. Conclusions

Review and comparison of the available data on DLC coatings and biolubricants the following conclusions can be drawn.

Due to the self-lubricating property of DLC coatings, DLC symmetrical contact were much effective as compared with asymmetrical contact with respect to friction and wear. Due to the presence of polar components in the structure of biolubricants, their tribological behavior with DLC coating is synergetic. Biolubricants show synergetic behavior with symmetric DLC coated contact but their tribological properties are not much significant with asymmetrical contact of DLC coating and steel. In asymmetrical DLC and steel contact, fully formulated oil (SAE 5W-30) have strong interaction with contact surfaces and its wear results were better due to the presence of anti-wear particles in lubricant. Graphitization occur when lubricant interact with the coating and this form a graphite layer on the coating which promotes the lubrication of the contact and reduces the COF and wear

values. In some cases, a transfer layer is formed which contains nano particles of graphite. These particles case severe damage of the DLC coating and act as debris, due to their presence third body wear phenomenon occur and surface starts to deteriorate. Based on the above discussed all observations it can be concluded that biolubricants show synergetic behavior with symmetrical DLC contacts due to the presence of polar components in its structure.

7. Recommendations

The study put a comprehensive light on literature of DLC coating and biolubricants use. Though there are many areas in this topic range which need more work on it and a vast range of research can be conducted on these areas. This will help the automobile industry to optimize the tribological part of an automobile's engine. These is no work done on an actual automobile engine with DLC coated parts contact. Future work would be needed on this portion in different ways.

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