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Vegetable Oil-Based Nanolubricants in Machining: From Physicochemical Properties to Application

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Abstract

Cutting fluid is crucial in ensuring surface quality and machining accuracy during machining. However, traditional mineral oil-based cutting fluids no longer meet modern machining's health and environmental protection requirements. As a renewable, pollution-free alternative with excellent processing characteristics, vegetable oil has become an inevitable replacement. However, vegetable oil lacks oxidation stability, extreme pressure, and antiwear properties, which are essential for machining requirements. The physicochemical characteristics of vegetable oils and the improved methods' application mechanism are not fully understood. This study aims to investigate the effects of viscosity, surface tension, and molecular structure of vegetable oil on cooling and lubricating properties. The mechanisms of autoxidation and high-temperature oxidation based on the molecular structure of vegetable oil are also discussed. The study further investigates the application mechanism and performance of chemical modification and antioxidant additives. The study shows that the propionic ester of methyl hydroxy-oleate obtained by epoxidation has an initial oxidation temperature of 175 °C. The application mechanism and extreme pressure performance of conventional extreme pressure additives and nanoparticle additives were also investigated to solve the problem of insufficient oxidation resistance and extreme pressure performance of nanobiological lubricants. Finally, the study discusses the future prospects of vegetable oil for chemical modification and nanoparticle addition. The study provides theoretical guidance and technical support for the industrial application and scientific research of vegetable oil in the field of lubrication and cooling. It is expected to promote sustainable development in the manufacturing industry.

Keywords Cutting fluid, Vegetable oil, Chemical modification, Antioxidant, Extreme pressure additive, Minimum quality lubrication

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1 Introduction

Cutting fluids are commonly used in metal processing for turning [1], milling [2, 3], drilling, and grinding [4–6]. They provide cooling, lubrication, cleaning, and rust prevention during processing and reduce tool wear by lowering cutting temperature, extending tool service life and improving the smoothness of the metal surface [7, 8]. Cutting fluids typically have a base oil that can be mineral, synthetic, or vegetable. Approximately 85% of the cutting fluids used in the world are mineral oil-based and extracted from petroleum [9, 10].

The traditional pouring lubrication method, which is typically used in metal processing, results in large amounts of metalworking fluid being consumed during the processing process, leading to significant economic losses [11]. Moreover, the hazards posed by mineral oil-based cutting fluids to human health and the environment cannot be overlooked. The complex composition of mineral oil-based cutting fluid, coupled with the production of oil mist during pouring, makes it a breeding ground for bacteria and microorganisms that can cause irritation and allergic reactions in workshop workers, thereby posing a significant safety risk to workers [12, 13]. Long-term contact between the skin and metalworking fluid is reported to cause approximately 80% of workshop workers' occupational diseases [14, 15]. Mineral oil-based cutting fluids are also prone to chemical reactions due to high processing temperatures, leading to environmental problems such as water and soil pollution in the post-treatment process [10, 16]. However, synthetic hydrocarbons in synthetic oils and vegetable oils have poor biodegradability [17].

In recent years, modern global green manufacturing has led to an increasing awareness of the economic losses caused by the massive consumption of mineral oil-based metalworking fluids and their non-negligible harm to humans and the environment. As a result, the manufacturing industry is moving toward economic and reasonable green technology, and researchers worldwide are exploring new methods to replace traditional pouring processing. Dry cutting [18, 19], low-temperature cold air [20–24], and minimum quantity lubrication with vegetable oil [25–28] are among the technologies that scholars have explored to solve the problem of large metalworking fluid consumption caused by traditional pouring processing. Dry cutting is not restricted by cutting fluid but can lead to the deterioration of workpiece surface integrity, affecting machining quality due to its low thermal conductivity [29]. Although low-temperature air-cooling technology solves the high-temperature problem in dry machining, it still has defects in improving the lubrication performance of the machining area due to the lack of cutting fluid [30]. The minimum quantity lubrication

technology is a combination of the advantages of dry processing and traditional overflow processing [31]. It mixes and vaporizes a small amount of lubricating oil and compressed gas to form a micron-level liquid drop, which is sprayed onto the processing area in the form of aerosol to lubricate effectively during processing [32, 33]. Compared to mineral oil, vegetable oil has the advantages of renewability, low toxicity, and easy biodegradation. Therefore, vegetable oil-based minimum quantity lubrication technology has been favored by many scholars and experts. As society pursues biodegradable lubricants, scholars are increasing their research on vegetable oils, which have the most potential to replace mineral oil-based lubricants [34]. Vegetable oil has better processing performance than mineral oil, is biodegradable, and is less costly than synthetic base oil, making it an acceptable alternative for mechanical processing.

Arnsek [35] demonstrated that vegetable oil is an effective boundary lubricant due to its high polarity, which allows for strong interaction with the lubricating surface. The performance of boundary lubrication is influenced by the attraction of lubricant molecules to the surface and their reaction with the surface. Compared to mineral oil lubricants, full-formula vegetable oil lubricants exhibit a lower friction coefficient, similar scuffing bearing capacity, and better pitting resistance [36]. Rapeseed oil, in particular, has good anti-wear and antifriction properties due to the physical and chemical adsorption film formed by rapeseed oil fatty acids on the metal surface and higher viscosity index compared to mineral oil, resulting in a lower friction coefficient and temperature. However, the anti-wear properties of vegetable oils under high loads still require improvement. In a study by Belluco [37], a range of mineral and vegetable oil-based cutting fluids were evaluated for their performance in various machining operations. The vegetable oil formulation exhibited the same or better performance than the reference commercial mineral oil in all operations. Permuswan [38] evaluated the application performance of several vegetable oils on an improved engine test bench and found that these oils had reliable lubricating effects. However, over time, the viscosity of the oil gradually increased, and sediment formed in the holes [39].

Although vegetable oil has excellent properties for use in various mechanical processing fields, it is easily oxidized, which results in the loss of efficacy during processing. The oxidation of vegetable oil adversely affects its extreme-pressure performance, and the mechanism of vegetable oil oxidation remains unclear. To address these issues, current research is focused on improving the application of vegetable oil-based cutting fluids through chemical or physical modification. This study will begin by analyzing the characteristics of vegetable

oil, including the influence of viscosity, surface tension, and molecular structure (carbon chain length, unsaturated degree, and polar group) on the lubricating performance of vegetable oil. It will then analyze the mechanisms of self-oxidation and high-temperature oxidation of vegetable oil. This study will focus on the defects of insufficient antioxidation performance and extreme pressure anti-wear performance of vegetable oil. The mechanism and application effect of chemical modification and antioxidants on the antioxidation of vegetable oil will be discussed, as well as the effects of different extreme-pressure additives on the extreme

pressure and anti-wear properties of vegetable oil. The study structure is presented in Figure 1.

2 Application Characteristics of Vegetable Oil-Based Cutting Fluid

Vegetable oil is mainly composed of triglycerides, a small amount of free fatty acids, some glycerides, and phosphates. Triglycerides are the main component of vegetable oils. Triglyceride is a glycerol molecule connected to the hydroxyl groups of three long-chain fatty acids through ester bonds. Its fatty acid carbon chain length is between 14–22 carbons, but the saturation of different fatty acids varies [40]. Fatty acids in vegetable oils are mainly divided into two categories: saturated and unsaturated. Saturated fatty acids do not contain C=C. Myristic (C_{14:0}), palmitic (C_{16:0}), and stearic acids (C_{18:0}) are the most common saturated fatty acids. Unsaturated fatty acids have at least one C=C. Oleic acid (C_{18:1}), linoleic acid (C_{18:2}), and linolenic acid (C_{18:3}) are the most common unsaturated fatty acids [41]. For different vegetable oils, the type and proportion of fatty acids, carbon chain length, saturation (C=C number), and functional group type of different fatty acids are different; therefore, they have different physical and chemical properties [42].

The mechanism by which vegetable oils affect lubrication is multifaceted. The types and proportions of fatty acids contained in different vegetable oils are different; therefore, they have different physical and chemical properties during processing [43, 44]. Table 1 shows the fatty acid compositions (%) of several common vegetable oils.

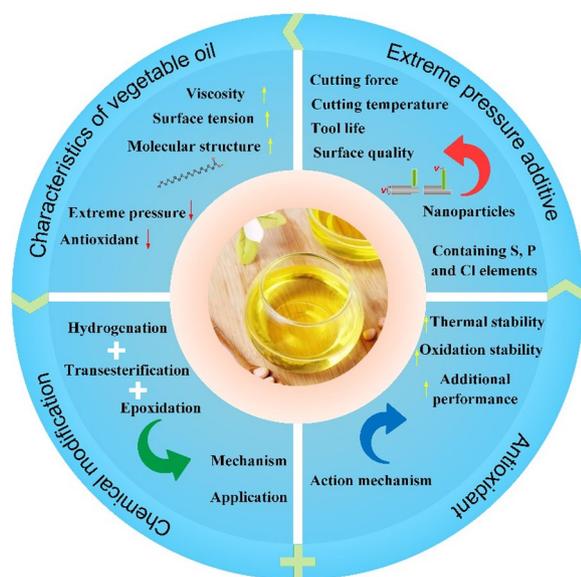


Figure 1 Article structure

Table 1 Fatty acid composition of several common vegetable oils (%)

Fatty acids	Rapeseed oil	Corn oil	Soybean oil	Peanut oil	Olive oil	Castor oil	Linseed oil
C14:0	0.06	–	0.06	0.03	–	1	0.05
C16:0	3.75	10.34	9.90	9.40	10.84	2.63	4.81
C18:0	1.87	2.04	3.94	2.65	3.59	1.51	3.03
C20:0	0.64	0.44	0.41	1.38	0.50	–	0.20
C22:0	0.35	0.31	0.48	3.14	0.15	–	–
Total saturated fatty acids	6.98	13.46	15.10	18.38	15.28	–	8.15
C16:1	0.21	–	0.08	0.06	0.02	–	–
C18:1	62.41	25.54	21.35	48.71	75.55	4.74	21.42
C18:1:OH	–	–	–	–	–	82.80	–
C20:1	1.54	0.37	0.22	1.43	0.32	–	0.40
C22:1	–	–	–	0.12	–	–	–
Total monounsaturated fatty acids	64.42	26.11	21.73	50.33	77.00	–	22.04
C18:2	20.12	59.27	56.02	31.06	7.01	8.36	15.18
C18:3	8.37	1.07	7.15	0.23	0.66	–	54.24
Total polyunsaturated fatty acids	28.60	60.43	63.17	31.29	7.72	–	69.81

2.1 Lubrication Mechanism of Vegetable Oil-Based Cutting Fluid

Different vegetable oils have different physical and chemical properties; therefore, they have different properties during the processing process. Therefore, understanding the lubricating mechanism of vegetable oil from the viscosity, surface tension, and molecular structure of vegetable oil is the key to achieving better lubricating performance of vegetable oil.

2.1.1 Viscosity

Viscosity is one of the main factors affecting the cooling and lubricating properties of vegetable oil-based cutting fluids. It is a variable exchange and adhesion caused by irregular intermolecular movement and is a method to measure the flow resistance of vegetable oil [45]. The viscosity of vegetable oil is high at room temperature, and its size is mainly affected by the composition of particles and the mutual attraction between particles. The viscosity of vegetable oil is related to the temperature, pressure, shear rate, fatty acid composition, etc. [46]. The viscosity of vegetable oil is positively correlated with the amount of monounsaturated fatty acids and negatively correlated with the content of polyunsaturated fatty acids [47]. Figure 2 shows the viscosities of several common vegetable oils at 40 °C and 100 °C.

Different cutting fluids have different lubricating effects, owing to their different viscosities. Viscosity mainly affects the heat exchange and lubrication effect of the cutting fluid at the interface between the tool and chip.

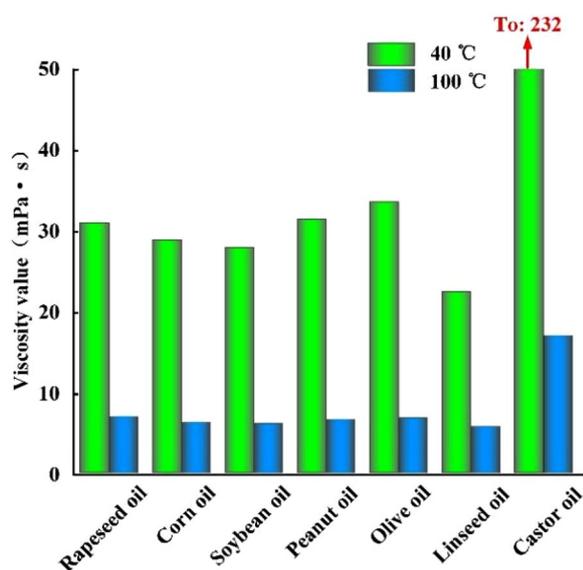


Figure 2 Viscosity of several common vegetable oils at 40 °C and 100 °C

In terms of heat exchange, cutting heat is an important physical phenomenon during the cutting process. In addition to 1%–2% of the energy consumed by cutting to form new surfaces and potential energy in the form of lattice distortion, 98%–99% of the energy is converted into heat energy. The influence of cutting fluid viscosity on the heat exchange between the tool and workpiece interface is also called the cooling effect or heat exchange performance of the cutting fluid [48, 49]. In a specific processing process, the cutting fluid enters the processing area of the tool and workpiece interface at a certain angle, forms a layer of dense lubricating oil film at the tool and workpiece interface, and flows at a certain speed along the tool feed direction. Owing to the relative motion between the tool and workpiece, the fluid layer in the lubricating oil film also exhibits relative motion [50, 51]. Therefore, the lubricating oil film formed during machining conforms to the theory of the convective heat transfer of the flowing fluid, as shown in Figure 3. The Reynolds number Re in microlubrication grinding is typically much greater than 2300. Therefore, the lubricant film formed at the interface between the tool and workpiece carries out convective heat transfer in a turbulent manner, and the size of the heat transfer coefficient is closely related to the viscosity. In a lubricating oil film with convective heat transfer in the form of turbulent flow, there is a thin layer with a significant temperature change [52]. Prandtl is known as the thermal boundary layer [53]. The thermal boundary layer of turbulent flow can be divided into a viscous bottom layer and a turbulent branch layer. According to heat transfer theory, the temperature gradient of the thermal boundary layer is the largest in the lower viscous layer and changes gently in the turbulent branch layer [54]. The heat-transfer capability of the cutting fluid depends on the viscosity of the lower layer. Therefore, the greater the viscosity of the cutting fluid, the thicker the viscous bottom layer, that is, the lower the temperature of the cutting fluid per unit time, that is, the lower the heat transfer coefficient.

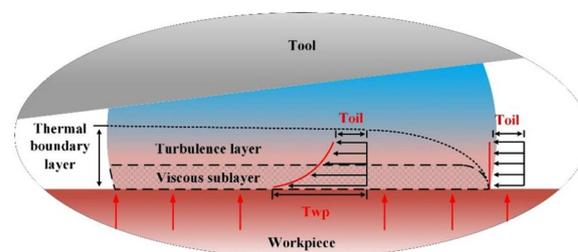


Figure 3 Convective heat transfer of fluid at the tool-workpiece interface

In lubrication, the viscosity of vegetable oil affects the permeability of the lubricating oil, and the permeability of the cutting fluid further affects its heat exchange performance during machining. As a result of inertia, vegetable oil droplets enter the interface between the workpiece and tool through the nozzle and can continue to advance for a certain distance owing to inertia. Because of the different adhesions, the running distance of vegetable oil droplets with different viscosities is different. Vegetable oil with high viscosity has a short mobility and moving distance owing to its high adhesion, and it is difficult to penetrate the gap between the workpiece and tool. Thus, low-viscosity vegetable oils can effectively penetrate these gaps. Therefore, there is a negative correlation between the viscosity and permeability of the cutting fluid.

Harpinder [55] studied the grinding effect of three vegetable oils (rapeseed, soybean, and olive oils) as lubricants when grinding Ti-6Al-4 V-ELI. Among the three oils, rapeseed oil obtained the lowest grinding force ($F_t = 4.68$ N, $F_n = 15.25$ N), friction coefficient (0.307), and specific grinding energy (17.16 J/mm³). The viscosity of rapeseed oil reached 38 mPa·s at 40 °C, while that of soybean and olive oil was 26 mPa·s and 32 mPa·s, respectively. The results show that viscosity plays a key role in improving tribological properties.

In addition, the viscosity of vegetable oil is not constant; it decreases with an increase in the cutting temperature [56]. The viscosity reduction followed the power-law model [57]. Table 2 below shows the variation trend of viscosity (mPa·s) of several vegetable oils between 35–180 °C with temperature. The viscosity value is obtained from the slope of fitting the experimental shear stress–shear rate data with Newton's viscosity law equation (the specific data are obtained from the fitting slope of the author's experimental shear

stress–shear rate data and Newton's law viscosity equation) [58].

2.1.2 Surface Tension

In minimum quality lubrication processing, the surface tension is the key factor affecting the size of the droplets, and the size of the surface tension is positively related to the size of the droplets. A cutting fluid with low surface tension has a better lubrication effect.

On the one hand, low surface tension means that the droplets formed are small in size, large in number and large in specific surface area [59]. A small droplet size implies that more droplets enter the gap between the tool and workpiece, and a higher specific surface area also represents a larger heat exchange area. On the other hand, according to the theory of convective heat transfer, droplets can be divided into thermal boundary layers and main flow areas during the process of thermal convection. The thickness of the thermal boundary layer remains unchanged. However, before absorbing sufficient heat, the grinding fluid in the mainstream area is rapidly discharged from the grinding area. In other words, the grinding fluid in the mainstream area will not provide a satisfactory heat exchange effect. When the contact angle decreases, the thermal boundary layer expands, and the proportion of abrasive fluid in the mainstream area decreases. This result explains why MQL droplets with small contact angles exhibit high cooling efficiencies [60, 61].

Different types of vegetable oils have significant effects on the viscosity of vegetable oils but do not affect the surface tension. The surface tension increases with increasing vegetable oil density and decreases linearly with increasing temperature [62]. Table 3 shows the variation trend of the surface tension of several vegetable oils at temperatures between 20 °C and 200 °C (The specific data are multiplied by the surface tension value obtained by the author based on the drop weight method by a certain correction factor ψ) [63].

2.1.3 Molecular Structure

In the process of vegetable oil used for processing lubrication, vegetable oil can form a layer of lubricating oil film on the surface of the workpiece, which can be divided into physical film formation and chemical film formation through different mechanisms of lubricating oil film formation. Physical film formation: During metal processing, owing to the strong extrusion, the vegetable oil on the workpiece surface changes from the original liquid lubrication state to the boundary lubrication state. Polar atoms contained in vegetable oils, such as S, O, N, and P, and polar groups, such as ester bonds (-COOR), carboxyl groups (-COOH), and OH, have strong metal

Table 2 Variation trend of viscosity (mPa·s) of several vegetable oils between 35–180 °C with temperature

Temperature (°C)	Viscosity (mPa·s)				
	Rapeseed oil	Corn oil	Soybean oil	Peanut oil	Olive oil
35	42.49	37.92	38.63	45.59	46.29
50	25.79	23.26	23.58	27.45	27.18
65	17.21	15.61	15.73	17.93	18.07
80	12.14	10.98	11.53	12.66	12.57
95	9.01	8.56	8.68	9.40	9.45
110	7.77	6.83	7.17	7.47	7.43
140	5.01	4.95	4.58	5.14	5.29
180	4.65	3.33	3.31	3.26	3.44

Table 3 Variation trend of surface tension (mN/m) of several vegetable oils with temperatures between 20 °C and 200 °C

Temperature (°C)	Surface tension (mN/m)				
	Rapeseed oil	Corn oil	Palm oil	Sunflower oil	Soybean oil
20	33.8	33.8	–	34.0	33.9
40	32.0	32.2	31.5	32.3	32.2
60	30.5	30.6	30.0	30.7	30.6
80	29.0	29.1	28.5	29.2	29.1
100	27.5	27.6	27.1	27.7	27.6
120	26.0	26.1	25.7	26.3	26.1
140	24.5	24.6	24.4	24.8	24.7
160	23.1	23.2	23.0	23.0	23.2
180	21.7	21.8	21.6	21.6	21.8
200	20.3	20.4	–	20.2	20.4

affinity. They can be physically adsorbed on the material surface of a metal workpiece through van der Waals forces to form a layer of molecular film, which has anti-wear and antifriction effects, thereby improving the lubrication of the cutting area [64, 65]. Due to the role of polar groups in vegetable oil, it is easier to form a high-strength lubricating oil film, which has stronger anti-wear, anti-friction, cooling, and lubrication effects in the processing process. Chemical film forming: Under the action of high temperature (100 °C) during processing, polar molecules in vegetable oil easily undergo a “metal saponification reaction” with the metal surface. During the reaction, the H atom of the carboxyl group (-COOH) in the fatty acid reacts with the metal surface atom to form a single layer of a semichemically bonded oily lubricating film. An oily lubricating film can be composed of a single molecular layer or a multimolecular layer. It is adsorbed on the surface of the workpiece material as a boundary film, thus prolonging tool life [66].

Owing to the different types of fatty acids, the saturation degree and carbon chain length of various fatty acids are different. Fatty acids with different carbon chain lengths and fatty acid saturations have different oil film formation intensities.

Regarding the relationship between the carbon chain length and lubricating performance, the cohesion between molecules is proportional to the number of carbon atoms, and the number of carbon atoms in the carbon chain has an important influence on the molecular cohesion. Therefore, the strength and lubrication performance of lubricating oil films formed using fatty acids with longer carbon chains are stronger than those formed using fatty acids with shorter carbon chains [67]. However, for saturated fatty acids, when the carbon number is greater than 16, the friction resistance and wear resistance of the formed lubricating oil film reach their

peak and remain unchanged. In this case, the lubrication effect does not change with an increase in the number of carbon atoms in the saturated fatty acids. For unsaturated fatty acids, owing to the existence of polar unsaturated bonds, the density of the adsorption film decreases under the adsorption of the olefin bond, thereby reducing the strength and lubrication performance of the lubricating oil film [68].

Regarding the relationship between different saturations and lubricating performance, the influence of different fatty acid saturations on lubricating performance can be attributed to two aspects. First, the presence of C=C makes unsaturated fatty acids easily oxidize, which leads to the degradation of vegetable oil, thereby reducing the oxidation resistance and thermal stability of vegetable oil and finally leading to the failure of the formed physically adsorbed oil film. Moreover, the higher the C=C ratio is, the worse the oxidation resistance and thermal stability, thus reducing the lubricating performance of vegetable oil. On the other hand, the attraction between fatty acid molecules is affected by the shape of the fatty acid molecules. Saturated fatty acid molecules do not have C=C, and the molecular chains are mostly straight; therefore, intermolecular binding is high [69]. However, the position where C=C exists in unsaturated fatty acids leads to bending of carbon chains, reducing the degree of intermolecular binding. In addition, because of the existence of olefin bonds in fatty acid molecules, the olefin bonds exhibit extremely strong molecular adsorption, leading to a reduction in the density of the physical adsorption film, thus reducing the strength of the lubricating oil film. Figure 4 shows the three-dimensional model of saturated and unsaturated fatty acid molecules.

Therefore, for the same carbon chain length, the lubricating oil film formed by saturated fatty acids during processing is stronger and has better lubrication

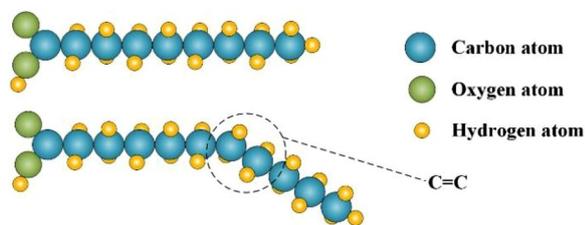


Figure 4 Three-dimensional model of saturated and unsaturated fatty acids

performance than that formed by unsaturated fatty acids during processing. Simultaneously, the workpiece surface roughness after processing is lower. At the same time, under the same carbon chain length, the lubricating oil film formed by monounsaturated fatty acids has better strength and lubrication performance than that formed by polyunsaturated fatty acids and can obtain a lower surface roughness.

Hu [70] studied the tribological properties of saturated and unsaturated long-carbon chains and short-carbon chain vegetable oil fatty acids as lubricating oil additives. The lubricating properties of two saturated fatty acids, octadecanoic acid ($C_{18}H_{36}O_2$) and eicosanoic acid ($C_{22}H_{44}O_2$), are the same. For the two unsaturated acids, the lubrication performance of long carbon chain dienoic acid ($C_{22}H_{42}O_2$) is better than that of short carbon chain octadecenoic acid ($C_{18}H_{34}O_2$). In addition, when the carbon chain length is the same, the lubricating performance of saturated fatty acids is better than that of unsaturated fatty acids.

2.2 Limitations of Vegetable Oil-Based Cutting Fluids

Vegetable oils exhibit satisfactory physicochemical properties as lubricants. However, the lack of antioxidant properties remains an unavoidable defect in vegetable oils.

2.2.1 Autoxidation

Although the specific triglyceride structure of vegetable oil determines that it has better lubricating performance than mineral oil, the structure of triglycerides also causes vegetable oil to have poorer antioxidant performance than mineral oil. Poor oxidation and thermal stability are inevitable defects in vegetable oil-based lubricating oils [71]. The higher the C=C in fatty acids, the higher the degree of unsaturation, and the more easily vegetable oil is oxidized [72]. For example, castor oil contains a large amount of ricinic acid (monounsaturated fatty acid); therefore, it has better oxidation stability than vegetable oil with a high content of polyunsaturated fatty acids [73]. Similarly, rapeseed oil (RO) contains approximately 64.42% monounsaturated fatty acids, so it has relatively high oxidation stability [74]. A study on the tribological properties of oxidized rapeseed oil showed that oxidation adversely affects its antifriction performance [75]. As shown in Figure 5.

Oxidation is the main reaction of vegetable oil when it is used as a lubricant base oil. Oxidation causes the content of unsaturated fatty acids in vegetable oil to decrease, and the contents of foam, color, viscosity, density, specific heat, free fatty acids, polar substances, and polymer compounds in vegetable oil increase with time [76]. The trend of this change is shown in Figure 6(a).

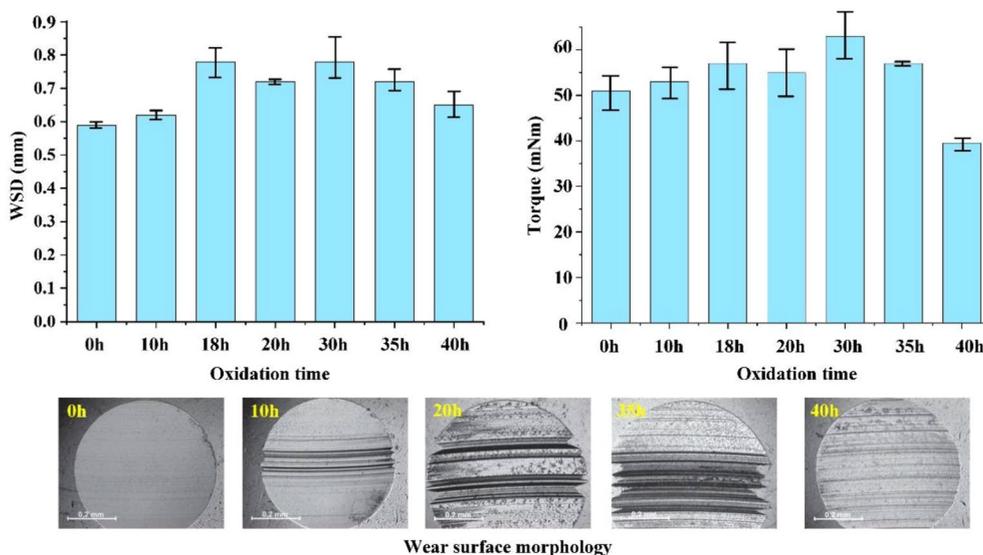


Figure 5 Wear and average torque measurement of nonoxidized and oxidized rapeseed oil

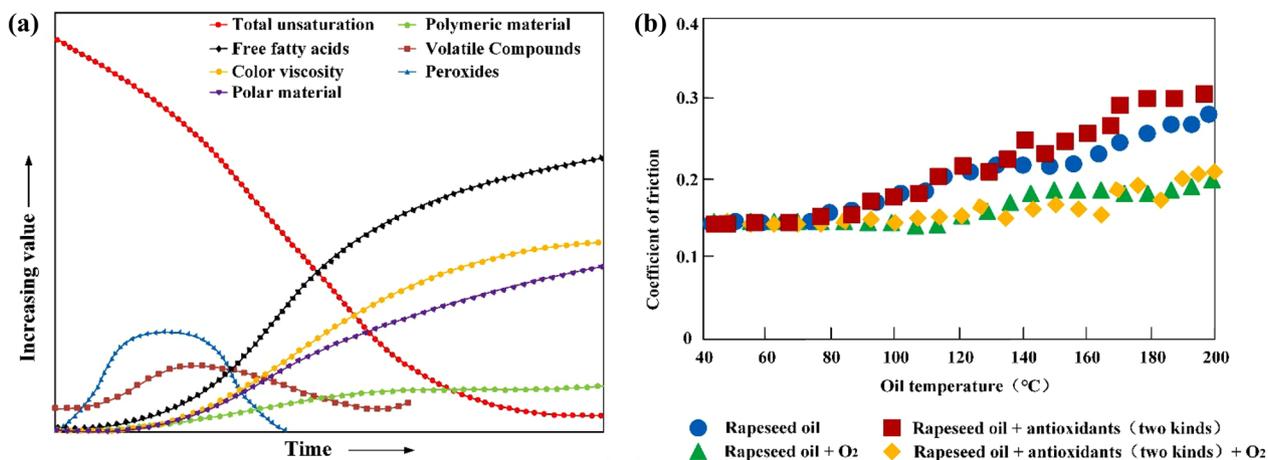


Figure 6 Change trend of the content of various substances **a** and tribological characteristics **b** under 1.38 kg in the oxidation process of vegetable oil

Because a large amount of C=C in unsaturated fatty acids can be used as the active site of the oxidation reaction, vegetable oil has very low stability. The higher the degree of unsaturation, that is, the more C=C, the more sensitive the oil is to oxidation [77]. Low oxidation stability means that, if untreated, the oil will quickly oxidize, thicken, and polymerize during use. Adverse effects on the lubrication performance. For eighteen carbon triglycerides (C-18), the relative oxidation instability of saturated, mono-, di-, and tri-unsaturated triglycerides is approximately 1:10:100:200 [78, 79].

There are four types of vegetable oil oxidation: automatic oxidation of vegetable oil, enzymatic oxidation, photooxidation, and metal oxidation. Among them, the automatic oxidation of vegetable oil is an oxidation reaction without any external influence and is the main method of vegetable oil oxidation [80, 81].

The oxidation of vegetable oils involves three stages: initiation, propagation, and termination, which are complex and involve both continuous and discontinuous reactions [82]. In vegetable oil molecules, the hydrogen atoms in methylene are more likely to detach due to the lower C-H bond strength in methylene (-CH₂-) next to C=C. The initial oxidation phase of vegetable oils involves the detachment of hydrogen atoms from the methylene next to C=C, leading to the formation of alkyl radicals (R·). These alkyl radicals react with oxygen to form alkyl peroxide radicals (ROO·). Primary antioxidants can neutralize these newly formed oxidation products, but if they are not used or cannot completely neutralize the free radicals, further oxidation stages occur [83, 84]. During the propagation phase of oxidation, alkane peroxide radicals attack the C-H bond in the remaining fatty acid molecules to obtain hydrogen atoms and new free radicals,

leading to the formation of primary oxidation compound hydroperoxide (ROOH). This process generates a series of new oxidation reactions. Hydrogen peroxide is formed in later stages, which is the rate-limiting step in the autooxidation of vegetable oils [85]. The strength of the broken carbon-hydrogen bonds determines the rate constant of the rate-limiting step. The decomposition of hydroperoxide leads to the formation of more free radicals. The newly formed alkoxy radicals (RO·) can repeatedly attack the C-H bond of fatty acids, generate more hydroperoxide, and accelerate the oxidation process [86]. The type and content of free radicals generated during the oxidation of vegetable oils depend on the composition of fatty acids, and the growth rates of free radicals vary significantly. However, not all free radicals accelerate the oxidation process. Some free radicals react with each other to terminate the oxidation process. Hydroperoxide accumulates continuously during the oxidation process and decomposes into countless volatile and nonvolatile secondary oxides when superimposed to a certain extent. Volatile organic compounds are mostly chain-broken hydrocarbons and alcohols and have little effect on the wear resistance of vegetable oils because of their low carbon chain length and low polarity. Among nonvolatile organic compounds, epoxy compounds exist longer than other nonvolatile organic compounds due to their higher oxidation stability and lower tendency to precipitate. Epoxidation has been proven to be an effective chemical modification method that has beneficial effects on the oxidation stability and viscosity of vegetable oils [87].

Certain secondary oxides can also participate in the polymerization reaction to produce macromolecular weight compounds, which eventually form precipitates on the lubricating surface of the workpiece and

Table 4 Autoxidation mechanism of vegetable oil

Initiation	$RH \rightarrow R^{\cdot} + H^{\cdot}$
Propagation	$R^{\cdot} + O_2 \rightarrow ROO^{\cdot}$ $ROO^{\cdot} + RH \rightarrow ROOH + R^{\cdot}$
Branching	$ROOH \rightarrow RO^{\cdot} + ^{\cdot}OH$ $RO^{\cdot} + RH + O_2 \rightarrow ROH + ROO^{\cdot}$ $^{\cdot}OH + RH + O_2 \rightarrow H_2O + ROO^{\cdot}$
Termination	$ROO^{\cdot} + ROO^{\cdot} \rightarrow ROOH + O_2$ $ROO^{\cdot} + R^{\cdot} \rightarrow ROOH$ $R^{\cdot} + R^{\cdot} \rightarrow R-R$
Peroxide decomposition	ROOH \rightarrow various lower molecular weight compounds
Polymerization	ROOH \rightarrow various higher molecular weight compounds

significantly improve the viscosity of vegetable oils. This represents the final stage of the vegetable oil oxidation process. The self-oxidation mechanisms of vegetable oils are summarized in Table 4.

In the early stages of vegetable oil oxidation, hydroperoxide is the main oxidation compound of vegetable oil. The presence of hydroperoxide inhibits the anti-wear property of the vegetable oil cutting fluid. With the oxidation process, the viscosity and surface tension of oxidized vegetable oil also increase with an increase in hydrogen peroxide content. Excessive viscosity and surface tension lead to poor flow performance of vegetable oil, which also reduces its lubricating performance [88]. Rounds [89] determined the effect of hydroperoxide on the wear. Under a load of 15–45 kg, an increase in the hydroperoxide concentration leads to an increase in wear. However, under a high load of 75–95 kg, the addition of hydroperoxide can reduce wear compared to pure vegetable oil, indicating that the presence of hydroperoxide has some extreme pressure properties. In addition, the addition of a zinc dialkyl dithiophosphate anti-wear additive to the oil mixture can increase the hydrogen peroxide concentration threshold and finally lead to high wear. Fox [90] showed that under boundary lubrication conditions, an increase in hydrogen peroxide content would reduce the anti-wear performance of sunflower seed oil lubricants. Habeeb [91] studied the wear effect of hydroperoxide on an engine. The wear of the cam lobe is increased by the interaction between the hydroperoxide and the metal surface, and the wear amount is proportional to the concentration of hydroperoxide. No obvious oil oxidation was detected in the experiment, indicating that the direct corrosion of the metal surface by hydroperoxide was the cause of wear.

2.2.2 High-Temperature Oxidation

The thermal stability of lubricating oil is an important parameter for its application at high temperatures, and it also affects the tribological properties of vegetable oils. The flash point is a parameter that characterizes thermal stability. When the flash point is lower than the grinding temperature, the vegetable oil is invalid. Additionally, the flash point is also an important parameter for evaluating the fire risk of flammable liquids. Combustible liquids (vegetable oils) may encounter hot surfaces (workpiece–tool interfaces), which may cause fire and explosion accidents [92]. The flash points and autoignition temperatures of several common vegetable oils are listed in Table 5.

The high temperature has sufficient energy to destroy the C-C or C-H covalent bonds in the acyl skeleton and form various free radicals. For example, alkoxy radicals are mainly found at 90 °C, and alkyl radicals are mainly found at 120 °C [93]. A free radical oxidation chain reaction occurs, which increases the oxidation rate of vegetable oils [94].

During high-temperature oxidation, the content of ROOH, the primary oxidation product of vegetable oil, increases with increasing reaction temperature. The peroxide value of vegetable oil at 60 °C is approximately twice that at room temperature [95]. However, when the temperature is higher than 150 °C, the primary oxidation product ROOH decomposes to form volatile small molecules, the decomposition rate is mostly faster than the generation rate, and the content of ROOH gradually decreases. In addition, high temperatures lead to the oxidation and polymerization of vegetable oils. The thermal decomposition of vegetable oil can be divided into three steps, which are related to the decomposition of polyunsaturated, monounsaturated, and saturated fatty acids [96]. In the first stage of the thermal decomposition of vegetable oil, the polyunsaturated fatty acids in vegetable oil begin to decompose. In the heating process, polyunsaturated fatty acids (such as linoleic acid) in vegetable oil react with heat to generate volatile compounds such

Table 5 Flash point and autoignition temperature of several common vegetable oils

Vegetable oil	Flash point (°C)	Ignition point (°C)	Autoignition temperature (°C)
Soybean oil	316	352	402
Tung oil	330	355	410
Sunflower oil	320	345	401
Rice oil	322	346	398
Peanut oil	327	351	406

as dimers, trimers, and polymers, which are continuously removed by the steam generated during the heating process. The second step of the thermal decomposition of vegetable oil corresponds to the decomposition of monounsaturated fatty acids such as oleic acid. During the reaction, the double bond of unsaturated fatty acids in vegetable oil breaks, leading to the saturation of triglyceride molecules. The third step of thermal decomposition corresponds to the thermal decomposition of saturated fatty acids such as palmitic acid [97].

The decomposition of fatty acid carbon chains caused by high temperatures is an important reason for the decline in the properties of vegetable oil. Li [98] observed that when the temperature was higher than 120 °C, linoleic acid ($C_{18:2}$), linolenic acid ($C_{18:3}$), and other polyunsaturated fatty acids began to decompose, whereas oleic acid ($C_{18:1}$) monounsaturated fatty acids decomposed slowly. Above 160 °C, the decreasing rate of monounsaturated fatty acids (oleic acid) in vegetable oil gradually increased. At 240 °C, the oleic acid content decreased by 52.33%, the linoleic acid content decreased by 58.21%, and the linolenic acid content decreased by 82.56%. This is consistent with the experimental results obtained by Jia [99]. Kim [100] studied the stability and polymerization of biodiesel through high-temperature accelerated oxidation. During the accelerated oxidation at 95 °C, the unsaturated fatty acid methyl esters in biodiesel decreased to varying degrees over time, which indicated that the oxidation led to the structural changes in unsaturated fatty acid methyl esters. In the process of high-temperature accelerated oxidation at 180 °C, oxidized unsaturated fatty acid methyl ester and unoxidized unsaturated fatty acid methyl ester are polymerized through the Diels–Alder reaction, and unsaturated fatty acid methyl ester forms a dimer and oxidized dimer and other polymerization products through the Diels–Alder reaction.

2.2.3 Extreme Pressure Performance

Pure vegetable oil is superior to mineral oil in terms of anti-wear property, friction property, bonding load capacity, and fatigue resistance under low load, but the effectiveness of vegetable oil-based cutting fluid is significantly reduced under extreme loads.

Suahrullail [101] showed that, compared with mineral oil (commercial stamping oil), the friction coefficients of *Jatropha curcas* oil (JAT), palm oil (PO) and palm fatty acid fraction (PFAD) increased by 20%, 54% and 107%, respectively, under extreme pressure conditions with a normal load of 126 kg, and the wear scar diameter increased significantly at the same time. This is because the anti-wear additives contained in mineral oils help reduce wear.

In addition, the oxidation of vegetable oil affects its extreme-pressure performance. Murakami [102] found that under 0.17 kN (average contact pressure 1.77 GPa), olive oil produced low friction at low temperature (100 °C) and increased friction at high temperature (170 °C). In contrast, rapeseed oil has a higher friction coefficient than olive oil at low temperatures, while it shows low friction above 170 °C. In contrast, olive oil contains oleic acid and a large number of saturated fatty acids, whereas rapeseed oil contains fewer saturated fatty acids than olive oil, whereas oleic acid, linoleic acid, linolenic acid, erucic acid, and other unsaturated fatty acids are high. The difference in the oxidation process of fatty acids at high temperatures leads to a difference in friction in different temperature ranges. The author then carried out an oxidation test to determine the influence of antioxidants and dissolved oxygen on the friction properties under 1.38 kN high pressure, as shown in Figure 6(b). A high oxygen content produces stable low friction and increases the bearing capacity. When the dissolved oxygen concentration is low, antioxidants reduce the carrying capacity. The authors also found that there was a friction polymer formed by oxidation around the wear scar of linolenic acid ($C_{18:3}$) with a high dissolved oxygen content, and the friction polymer played a protective role at high temperatures.

2.3 Summary of the Application Effect of Vegetable Oil

In the field of mechanical processing, the replacement of mineral oil-based lubricants with vegetable oils is a growing trend. However, the physical and chemical properties of different vegetable oils can have varying effects. Here are some key points to consider: 1) High viscosity vegetable oils have stronger heat exchange performance, reducing friction at the contact interface and lowering temperatures to improve lubrication. However, high viscosity can also mean higher adhesion, which reduces fluidity and permeability. 2) Vegetable oils with long carbon chains and low unsaturated fatty acid structures can create strong lubricating oil films that minimize wear and improve surface finish. 3) The presence of C=C is a major factor leading to the oxidation of vegetable oils. This increases the content of free fatty acids, polar substances, and polymer compounds, which weakens the lubricating oil film and anti-wear performance.

Vegetable oils with high viscosity and high percentages of monounsaturated fatty acids, such as rapeseed oil and olive oil, are popular choices for vegetable oil-based lubricants. However, all vegetable oils lack antioxidant performance, which is an unavoidable flaw.

Scholars from around the world have proposed different modification schemes to improve the performance of vegetable oils. Chemical modification and additive

modification are two methods used to enhance the antioxidant, extreme pressure, and antiwear properties of vegetable oil. The author has evaluated the effectiveness of these methods in improving vegetable oil performance.

3 Improvement of Oxidation Resistance

At present, methods to improve the antioxidant properties of vegetable oils include gene modification, chemical modification, and the addition of antioxidants. Among them, chemical modification and the addition of antioxidants are the focus of research worldwide.

3.1 Chemical Modification

The reasons for the poor stability of vegetable oil can be divided into two aspects: first, the alcohol hydroxyl group in the vegetable oil molecule β . The C-H bond in position easily decomposes at high temperature and combines with hydroxyl oxygen to form acids and olefins. Second, the presence of C=C in the triglycerides of vegetable oil

makes it extremely unstable [103]. The chemical modification of vegetable oils has attracted worldwide attention. The focus of this method is mainly to carry out chemical reactions on the carboxyl and carbon chains of unsaturated fatty acids to change the degree of unsaturation, carbon chain length, and branching of vegetable oil fatty acids to improve the thermal oxidation stability, low-temperature performance, and viscosity temperature performance of vegetable oil. The focus is on improving oxidation stability. Common modification methods include hydrogenation [104], esterification [105], epoxidation [106] and isomerization [107]. Hydrogenation, transesterification, and epoxidation are the three primary methods. The reaction process is illustrated in Figure 7. The differences in the modification methods have different effects on the molecular structure of triglycerides. The effects of fatty acid unsaturation, carbon chain length, and degree of branching on the properties of the vegetable oils after modification are shown in Table 6.

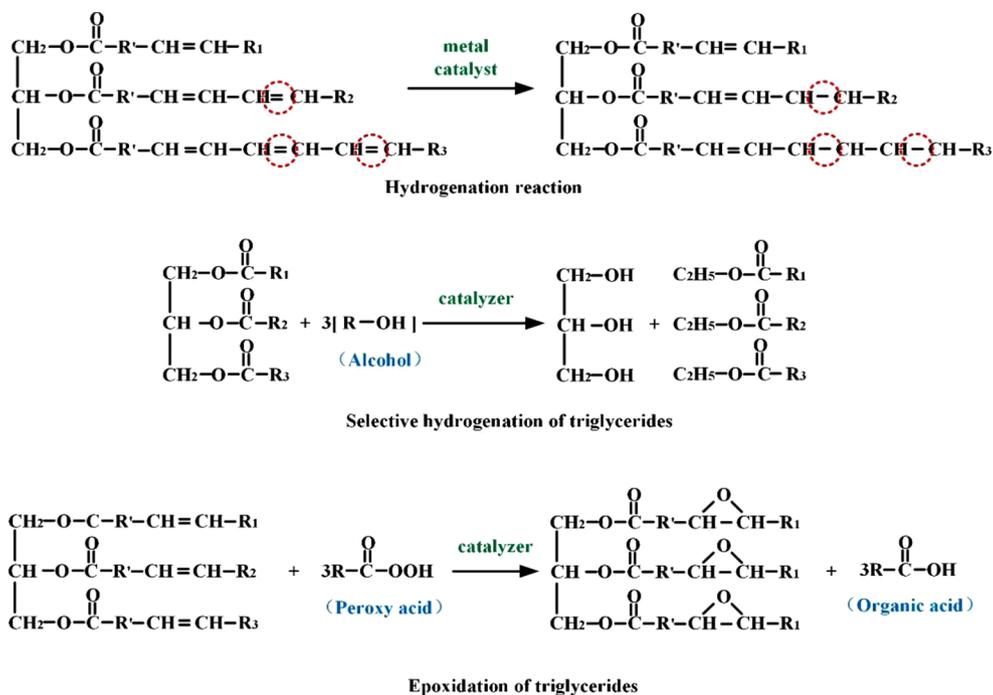


Figure 7 Hydrogenation, transesterification, and epoxidation reaction process

Table 6 Effects of unsaturation, carbon chain length and branching degree of fatty acids on the properties of vegetable oils

Project	Lubricity	Viscosity index	Low temperature performance	Oxidation performance	Volatility
Carbon chain length	++	+	-	-	+
Branching degree	--	--	+	+	-
Unsaturation	-	±	+	--	±

3.1.1 Hydrogenation

Because of the large amount of unsaturated fatty acids (oleic acid $C_{18:1}$, linoleic acid $C_{18:2}$, linolenic acid $C_{18:3}$, etc.), vegetable oil has extremely poor thermal oxidation stability and is easy to polymerize into colloidal substances at high temperatures. When used as a lubricant, sediment and corrosive substances are generated, reducing service life. Hydrogenation is the process of adding H atoms to $C=C$ in the triglycerides of vegetable oil via electrophilic addition to reduce the unsaturation of vegetable oil. The reaction process is shown in the above figure. Common hydrogenation methods include ultrasonic hydrogenation, catalytic transfer hydrogenation, magnetic field hydrogenation, electrochemical catalytic hydrogenation, etc. [108].

Paive [109] analyzed the performance of catalysts required for hydrogenation modification and summarized the effects of different reaction parameters, catalyst stability, kinetics, models, and thermodynamics. Liu [110] developed a nonsulfurized NiMoLa/ Al_2O_3 catalyst for the production of renewable C15 – C18 alkanes from the hydrogenation of *Jatropha curcas* oil. Under the conditions of adding 5.0 wt% rare earth metal La, 370 °C, 3.5 MPa and 0.9 h, the hydrocarbon yield of C15–C18 is 78%, the selectivity is 94%, and the conversion is 83%. The low-temperature fluidity of vegetable oil modified by selective hydrogenation is better than that modified by complete hydrogenation, and the thermal stability of vegetable oil is improved to a certain extent by reducing the number of double bonds in triglycerides. In terms of cost, the hydrotreatment modification method is inexpensive and can be produced on a large scale.

Although the complete hydrogenation method currently used in industry has improved the antioxidant performance of vegetable oils, the modified products are attracted to each other owing to their neat structure, which easily aggregates to form a rigid structure, affecting their low-temperature performance. The hydrogenation of vegetable oil can selectively reduce the composition of linoleic acid ($C_{18:2}$) and linolenic acid ($C_{18:3}$), thus increasing the content of oleic acid ($C_{18:1}$) in vegetable oil and avoiding an increase in the concentration of stearic acid ($C_{18:0}$). This treatment improves the oxidation stability of oil while maintaining an acceptable viscosity and low-temperature performance [111]. Belkacemi [112] used a new supported Pd catalyst for the hydrogenation of sunflower and rapeseed oils. Under mild conditions with a Pd loading of 1 wt%, compared with the commercial Ni catalyst, the iodine value (IV) is reduced, and fewer saturated fatty acids and approximately the same level of trans fatty acids are generated. Nohair [113] found that the addition of copper or lead to Pd can improve the selectivity of cis oleic acid isomers when hydrogenation of sunflower seed

oil ethyl ester was carried out with palladium (Pd), platinum (Pt) and rubidium (Ru) as catalysts. Laverdura [114] used the commercially available Lindlar catalyst to catalyze the hydrogenation of rapeseed oil and sunflower oil. The best hydrogenation effect is obtained at 180 °C and 0.4 MPa hydrogen environment, and 4 mg catalyst/ml oil. The Lindlar catalyst exhibits good selectivity for $C_{18:1}$ and can be used for sunflower seed oil with high unsaturation. However, the concentration of the trans isomers obtained from sunflower seed oil was higher than that obtained from rapeseed oil. Wang [115] designed a process to further hydrotreat vegetable oil by combining deoxidation and isomerization. On a bifunctional catalyst, vegetable oil is converted through decarboxylation, decarbonylation, hydrodeoxygenation, and isomerization to generate oxygen-free products. The acidity of the carrier plays a decisive role in the hydrodeoxygenation reaction.

The low-temperature fluidity of vegetable oil modified by selective hydrogenation is better than that modified by complete hydrogenation, and the thermal stability of vegetable oil is improved to a certain extent by reducing the number of double bonds in triglycerides. In terms of cost, the hydrotreatment modification method is inexpensive and can be produced on a large scale.

3.1.2 Esterification

In vegetable oil triglycerides, the alcohol hydroxyl group in glycerol molecule β . The C-H bond in position is easily decomposed at high temperatures [116]. This feature can lead to the partial fragmentation of beneficial molecules and the formation of unsaturated compounds, as shown in Figure 8. The formed compound can polymerize to increase the viscosity of the liquid, thereby forming precipitated particles.

As research continues to progress, the focus of transesterification research has shifted towards the synthesis of fatty acid polyol ester lubricants using polyols instead of glycerol. These synthesis pathways can be classified into two types: 1) hydrolysis of triglycerides to yield free fatty acids which are then used to generate fatty acid polyol esters, and 2) esterification of triglycerides to produce fatty acid methyl esters which are then reacted with polyols to generate fatty acid polyol esters. Polyols such as

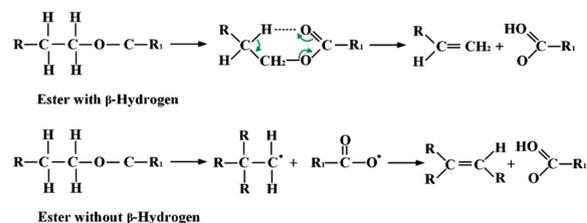


Figure 8 Thermal destruction of ester

neopentyl glycol (NPG), trimethylolpropane (TMP), and pentaerythritol (PE) are commonly used in these reactions [117].

Because the composition of vegetable oil fatty acids and the polyols reacted are different, the synthesized polyol esters have different molecular structures, so they have different physical and chemical properties. In terms of oxidation stability, its thermal oxidation stability increases with increasing number of hydroxyl (-OH) groups in polyols but decreases with increasing number of double bonds with triglycerides [118]. The free fatty acid content in vegetable oil affects the conversion efficiency of blocking transesterification. Farag [119] used pure oleic acid and H_2SO_4 as catalysts at a temperature of 60 °C, a catalyst concentration of 2.5%, and a stirring speed of 300 r/min; when the molar ratio was 6:1, the optimal conversion efficiency was 96.6%.

The transesterification of vegetable oils has been proven to improve their oxidation stability. Dodos [120] synthesized a Lunaria TMP ester using Lunaria oil. TMP esters from Lunaria have a very low oxidation rate. In the Ranchimat unit, it is easy to exceed the lower limit of 8 h, and the oxidation induction time is up to 30.5 h, which is 11.2, 11.2, 1.18, 1.9 and 5.1 times the antioxidant performance of cottonseed oil methyl ester, sunflower oil methyl ester, palm oil methyl ester, olive oil methyl ester and rapeseed oil methyl ester, respectively. Gryglewicz [121] used rapeseed and olive oils to prepare neopentyl glycol (NPG) and trimethylolpropane (TMP) esters. Olive oil-based esters have better oxidation stability than rapeseed oil-based esters because the main component of olive oil is oleic acid, which contains relatively small amounts of linoleic acid (7.2%) and approximately 14% saturated fatty acids. Compared with olive oil, rapeseed oil has a lower saturated acid content (8.3%) and a higher polyunsaturated fatty acid content (28%). The existence of a large number of polyunsaturated acids leads to a reduction in thermal oxidation stability. In addition, TMP ester had better thermal stability than NPG ester. Moreover, the NPG and TMP esters of fatty acids showed higher antioxidation activities than triglycerides. Similarly, Fernandes [122] used palm oil fatty acid distillate (PFAD) for esterification with neopentyl glycol (NPG) and trimethylolpropane (TMP) to prepare polyol esters. At high temperature (160 °C), the oxidation stability of TMP ester is about 75 % higher than that of NPG ester. However, both TMP ester and NPG ester have relatively high pour points, which are 12 °C and 18 °C, respectively. This can be achieved by the addition of anticoagulants. Arumugam [123] synthesized rapeseed oil pentaerythritol ester (PE) and compared its thermal stability with synthetic compressor oil (SAE30). PE demonstrated similar thermal stability as SAE30 with an initial oxidation

temperature of 240 °C and 236 °C, respectively. As the temperature increased, PE and SAE30 showed two-stage and single-stage decomposition, respectively. The decomposition peaks of PE ester were detected at 339 °C and 424 °C, corresponding to the oxidation of saturated and unsaturated fatty acids, respectively. On the other hand, SAE30 exhibited a decomposition peak at 376 °C. Thus, PE ester exhibited better thermal stability than SAE30. Padmaja [124] synthesized pentaerythritol ester of 10-undecenoic acid (UDA) by reacting it with three polyols (trimethylolpropane (TMP), neopentyl glycol (NPG), and pentaerythritol (PE)). The thermal stability of a lubricating oil base material is directly proportional to its decomposition starting temperature. The pentaerythritol ester of UDA exhibited excellent thermal stability with an initial temperature of 390 °C, followed by TMP ester at 336 °C and NPG at 264 °C. Reeves [125] demonstrated that the thermal and oxidation stability of vegetable oils depends on the fatty acid composition. Natural oils with a high percentage of monounsaturated fatty acids, such as oleic acid, exhibit superior thermal oxidation stability in high-temperature environments.

In terms of tribological properties, modified vegetable oil is competitive with the corresponding part of petroleum. Dodos [120] used lunaria oil to synthesize Lunaria TMP ester, which exhibited excellent lubricating performance. Compared with conventional GpI base oil, the friction coefficient (CoF) was reduced by 52.4%, and the wear scar diameter was reduced by 21.8%. Arumugam [123] observed a reduction in the average friction torque between rapeseed oil pentaerythritol ester (PE) and synthetic compressor oil (SAE30). The friction torque of SAE30 compressor oil was approximately 0.18 N·m, whereas that of biodegradable compressor oil was approximately 0.15 N·m, a decrease of approximately 16.7%. The friction torques of the SAE30 and PE mixed compressor oil samples were significantly reduced. Compared with SAE 30, the friction torques of PE50 and PE75 were reduced by approximately 27% and 38%, respectively. This reduction can be attributed to the presence of fatty acids such as oleic acid, linoleic acid, linolenic acid, palmitic acid, and erucic acid in rapeseed oil and their related polar groups, which can adsorb onto the friction pair surface, leading to the formation of a carbon structure layer. This layer ultimately reduces friction by avoiding metal-to-metal contact of the friction pair.

Moreover, transesterification reaction can improve the low-temperature performance of vegetable oils by improving their branching degree and antioxidant performance. Vegetable oil can solidify at -15 °C generally, so when used as the base oil of cutting fluid, it must have excellent low-temperature performance. Robiah [126] synthesized trimethylolpropane (TMP) ester based on

palm and palm kernel oil. In the TMP ester of high oleic acid palm oil, the pour point was increased to $-33\text{ }^{\circ}\text{C}$, demonstrating excellent low-temperature performance. Additionally, it exhibited good thermal stability and lubricity.

3.1.3 Epoxidation

As a very convenient method, epoxidation modification is one of the most commonly used methods to improve the thermal stability of vegetable oils. Relevant research shows that the epoxidation reaction can remove unsaturated components in the triglyceride molecules of vegetable oil, thus significantly improving the thermal oxidation stability of vegetable oil and improving the high-temperature application performance of lubricants [127]. During the reaction, the double bonds in triglycerides react with H_2O_2 , peroxyformic acid, or peracetic acid to form epoxides. The commonly used catalysts are zeolite, No. 120 phenol formaldehyde ion-exchange resin, anhydrous Na_2CO_3 , $(\text{NH}_4)_2\text{SO}_4$ and SnC_{12} .

Due to the elimination of multiple unsaturations, the oxidation induction time of epoxidized soybean oil is still six times higher than that of unmodified soybean oil at $150\text{ }^{\circ}\text{C}$ even at $195\text{ }^{\circ}\text{C}$. Because the metal surface of soybean oil modified by epoxidation can form a stable polymer film and has stronger stability, it has excellent anti-wear and antifriction properties. A higher level of performance has been achieved in various applications [128]. Compared with the original oil, epoxidized oil has a good molecular weight, a higher polar structure, and stronger interactions between molecules. Wu [129] proposed that epoxidized vegetable oil can form a friction polymer film on a metal surface, thus having better anti-friction and extreme pressure properties than the original oil. Chaurasia [130] modified Sal oil via epoxidation, and the friction coefficient of the modified Sal oil decreased by approximately 23.5%.

Furthermore, the three atoms in the epoxy group formed during epoxidation modification are co-planar, with the oxygen atom bond angle at approximately 60° , which is about 49.5° lower than the normal oxygen atom bond angle. This significant deviation in the two chemical bond angles of the oxygen atom in the epoxy group indicates that the tension of the C-O bond in the ternary ring is much higher than that of the conventional C-O bond, as shown in Figure 9. As a result, the fatty acid molecular chain can be selectively added to side chains of different chain lengths after the ring-opening reaction through epoxidation, effectively improving the low-temperature performance of vegetable oils and their tribological properties [131]. Additionally, the oxidation stability of modified vegetable oils decreases as the carbon chain length of the branched group increases.

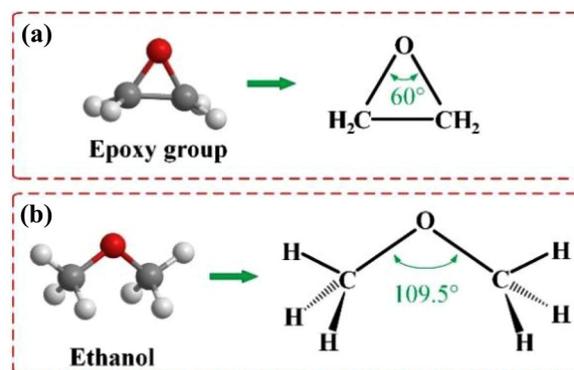


Figure 9 Epoxy key a and normal C-O key b

Brajendra [132] used acetic anhydride to modify the structure of epoxidized soybean oil (ESBO) and obtained the reaction product ACE-SBO. ACE-SBO has similar thermal stability to ESBO at high temperatures ($300\text{ }^{\circ}\text{C}$). Butyric acid, isobutyric acid, and caproic anhydride were used simultaneously to generate the diester derivatives But-SBO, Isobut-SBO, and Hex SBO, respectively. However, ACE-SBO has better oxidation stability than unmodified soybean oil (SBO) and dihydroxy derivatives. The initial oxidation temperature (OT) decreases with increasing carbon chain length of the branched group. The OT value of Ace SBO was the highest ($174\text{ }^{\circ}\text{C}$), whereas that of Hex SBO was the lowest ($161\text{ }^{\circ}\text{C}$). Similarly, Brajendra [133] used different anhydrides and soybean oils for epoxidation ring-opening reactions. Acid anhydrides, such as acetic acid, propionic acid, butyric acid, valeric acid, caproic acid, and heptanic acid, produced the corresponding diester derivatives ASO, PSO, BSO, VSO, HxSO, and HpSO, respectively. The reaction process is illustrated in Figure 10. The properties of these diester derivatives have also been studied [134]. With the addition of side chains, the low-temperature properties of modified soybean oil significantly increase. For example, compared with unmodified soybean oil ($-9\text{ }^{\circ}\text{C}$), the pour points of HxSO and HpSO can reach $-21\text{ }^{\circ}\text{C}$. In another study, the pour point of soybean oil isopropyl ester reached as low as $-50\text{ }^{\circ}\text{C}$ [135]. In addition, the antioxidant property of modified soybean oil is increased by 5.1%–7.4% compared with unmodified soybean oil. The author added the modified soybean oil to cetane at a concentration of 5%. At a high load of 400 pounds (181.44 kg) and a low speed of 5 r/min, all CMSO products showed excellent antifriction performance at a concentration of 5% (w/w), and the coefficient of friction (COF) was within the range of 0.11–0.12. Compared with pure cetane, the COF decreased by 75.6%.

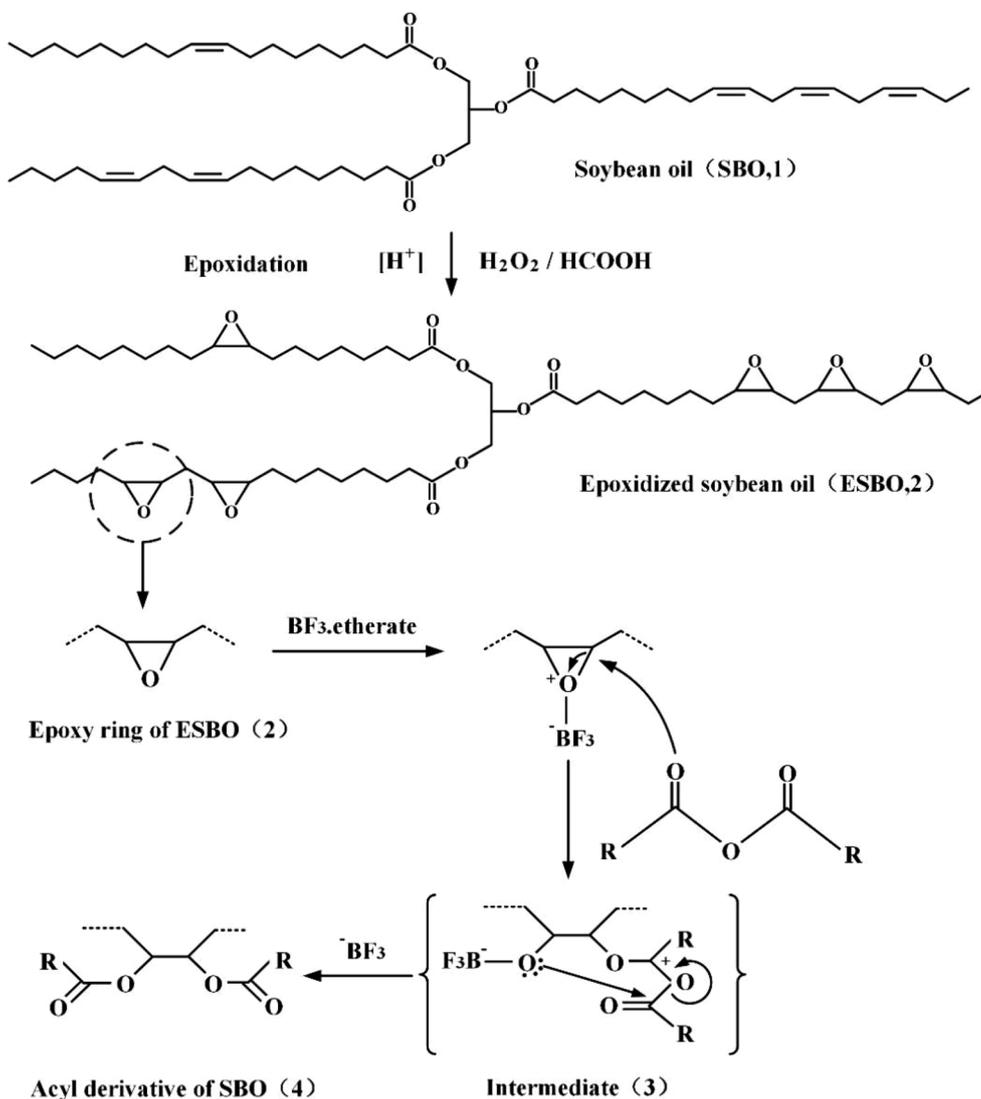


Figure 10 Epoxidation ring opening reaction of soybean oil

In addition to the epoxidation ring-opening reaction, the epoxidation ring-opening reaction of vegetable oil fatty acid methyl esters is also an important method for improving the oxidation stability of vegetable oil. Sharma [136] synthesized a series of branched-chain fatty esters from commercially available methyl oleate and carboxylic acids. These are the propionic ester of methyl hydroxy-oleate (PMO), the levulinic ester of methyl hydroxy-oleate (LMO), the hexanoic ester of methyl hydroxy-oleate (HMO), the octanoic ester of methyl hydroxy-oleate (OMO), and the 2-ethylhexyl ester of methyl hydroxy-oleate (EHMO). PMO had the highest oxidation initial temperature (OT) (175 °C), followed by EHMO (166 °C), LMO (162 °C), and OMO

(160 °C). The authors also showed that the oxidation stability decreased with an increase in ester side chain length. This may be because the longer side chains have oxidation sites that are easier to reach, making them easier to split than shorter side chains.

3.1.4 Summary of the Application Effect of Chemical Modification

Different chemical modification methods can have different effects on the physicochemical properties of vegetable oils (such as viscosity, pour point, tribological properties, and oxidation stability). In conclusion, among the three modification methods mentioned above, (i)

hydrogenation modification reduces the unsaturation of vegetable oil to some extent; however, the relatively neat molecular structure leads to poor low-temperature performance, and the problem of easy decomposition of the β -H atom at high temperatures cannot be solved. (ii) The transesterification reaction improves the tribological properties of the base oil. However, double bonds are still present in the reaction products. To a certain extent, the transesterification reaction depends on the type of base oil. Vegetable oil with a high percentage of monounsaturated fatty acids is the best choice for improving the oxidation stability and thermal stability. The initial oxidation temperature of rapeseed oil pentaerythritol ester reached 236 °C. (iii) The epoxidation reaction eliminates the double-bond structure by generating epoxy groups and improves the oxidation stability of the modified products from the molecular structure. Subsequently, the side chain structure and polar groups were added via the ring-opening reaction. It can produce a higher-strength lubricating oil film and improve the anti-wear performance. In addition, with the addition of a side chain, the low-temperature flow performance of the base oil improved.

In summary, ester exchange and epoxidation ring-opening reactions eliminate β -H, $C=C$ increases the antioxidant properties of vegetable oil, and the oxidation induction temperature increases accordingly, which can be used as a potential source of vegetable oil-based high-temperature lubricants. The epoxidation ring-opening reaction can improve the physical and chemical properties of base oil in many ways by adding side chains with different structures and can be used as an effective means for the development of vegetable oil-based lubricants in many ways.

3.2 Antioxidants

Antioxidants are substances that prevent the oxidation reaction between oxygen and vegetable oil and can help capture and neutralize free radicals to eliminate a class of substances in which free radicals cause the oxidation of vegetable oil [137]. After the base oil is modified, the oil quality can be further improved, and the production cost can be reduced by adding antioxidants. After chemical modification of vegetable oil, Wu [129] epoxidized rapeseed oil to obtain epoxy rapeseed oil with an epoxy value of 4.13%. After adding 1% antioxidant to epoxidized rapeseed oil, the oxidation induction time is about 4.9 times higher than that of epoxidized rapeseed oil without addition and 13 times higher than that of pure rapeseed oil.

3.2.1 Mechanism

The degradation of vegetable oil after oxidation significantly affects its lubricating performance and increases wear. Antioxidation additives can delay or resist the

oxidation process by protecting the cutting fluid from oxidative degradation. Antioxidants generally refer to any type of chemical reagent that inhibits the supply of oxygen or ozone. When applied to vegetable oils, antioxidants inhibit the oxidation process by inhibiting the formation of free radicals in the initial steps or blocking the propagation of free-radical chains [138]. Antioxidants can be divided into free-radical scavengers (primary antioxidants) and peroxide decomposers (secondary antioxidants) according to their mechanism of action.

Free radical scavengers are the primary antioxidants. It preferentially reacts with fat-free radicals in vegetable oil to generate stable free radicals that do not react quickly with oxygen. Free radicals have stronger thermodynamic stability. Common free-radical scavengers include butyl hydroxyanisole (BHA), butyl hydroxytoluene (BHT), monotert butyl hydroquinone (TBHQ), propyl gallate (PG), and naturally produced tocopherol (vitamin E). Hydroxyphenol compounds with various ring substitutions are the most popular free-radical scavengers [139]. In the study of phenolic antioxidants, it was found that their antioxidant capacity was related to the number of hierarchical patterns occupying 1,2 or 1,4 positions in the aromatic rings and the volume and electronic properties of the existing ring substituents. In contrast to the action mechanism of free radical scavengers, peroxide decomposers react with hydroperoxides to decompose them, generate stable compounds at the same time, accelerate the actual speed of the chain reaction, and thus achieve antioxidation effects. In addition, some antioxidants, such as dialkyl dithiocarbamate compounds and butyl hydroxytoluene (BHT), also act as free-radical scavengers and peroxide decomposers [140].

The mechanisms of action of the free radical scavenger and peroxide decomposer are shown in Figure 11. The free radical scavenger contains unstable hydrogen, which is quickly supplied to hydrogen peroxide and interferes with the lipid oxidation process, as shown in Figure 11a. Figure 11b shows the competition between the peroxide decomposing agent and chain propagation reaction.

3.2.2 Application Effect

Antioxidants can significantly improve the antioxidant properties of vegetable oils. Lampi [141] tested the effect of two tocopherols on the antioxidant activities of

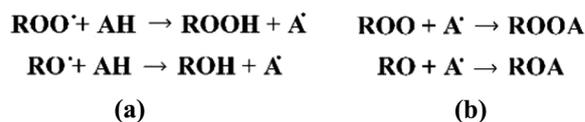


Figure 11 Action mechanism of **a** free radical scavenger and **b** peroxide decomposing agent

α - and γ -antioxidants. Tocopherol can inhibit autoxidation of rapeseed oil. As long as tocopherol is present, the stability of hydroperoxide will be greatly improved, and no volatile aldehyde will be formed. Compared with the two, at a low level ($\leq 50 \mu\text{g/g}$), the α -tocopherol ratio γ -tocopherol is more stable. When the α -tocopherol level is high ($>100 \mu\text{g/g}$), the formation of hydrogen peroxide α - The consumption of tocopherol shows a relatively increasing trend, while γ -tocopherol shows no such trend. Therefore, above $100 \mu\text{g/g}$, the γ -tocopherol ratio α -tocopherol is a better antioxidant. Hamblin [142] introduced the application of ashless antioxidants to control the oxidative degradation of several types of biodegradable vegetable oils. The effects of naturally occurring antioxidants (e.g., vitamin E) were also evaluated. Ash-free antioxidants can improve the oxidation performance of natural esters (such as rapeseed oil), whether alone or in combination. The most effective phenolic antioxidants for maintaining the stability of rapeseed oil are mixed phenols. Vitamin E showed moderate activity in short-term experiments, but this requires further study. Brajendra [143] studied the oxidation resistance of zinc dialkyl dithiocarbamate (ZDDC), alkylated diphenylamine (ADPA), butyl hydroxytoluene (BHT), and an alkylated phenol, dithiophosphate, and diphenylamine mixture (APDD) in soybean oil (SO). The initial oxidation temperature of soybean oil with 2% ZDDC increased from 171°C to 207°C . ADPA exhibited the worst antioxidation performance. The low reaction temperature (150°C) is an important reason for its poor antioxidation performance. It showed good antioxidation performance at high temperatures (175°C). Diphenylamine has been reported to have different inhibition mechanisms at low and high temperatures.

Antioxidants also affect the thermal degradation temperature of vegetable oils, and the adaptability of different vegetable oils to antioxidants varies. Quinchia [144] evaluated the effects of four antioxidants, (+)- α -tocopherol (TCP), propyl gallate (PG), L-ascorbic acid 6-palmitate (AP) and 4,4'-methylenebis (2,6-di-tert-butylphenol) (MBP), on the antioxidant properties of sunflower oil (HOSO) and castor oil (CO). The initial temperature of thermal decomposition of the HOSO/antioxidant mixture is usually low, and only PG, an antioxidant, can improve the thermal stability of HOSO. The thermal degradation temperature increased from 331°C to 347°C . In contrast, the three antioxidants (PG, MBP, and AP) delayed the thermal degradation of CO. The authors explained that the affinity of most antioxidants with CO is higher than that with HOSO because of the existence of functional polar groups (-OH). Therefore, although the initial temperature of the antioxidant thermal decomposition is lower than that of the pure oil studied, the

interaction of the -OH group of castor oil prevents the volatilization of antioxidant molecules.

Xu [145] synthesized an environmentally friendly antioxidant (pentaerythritol rosin ester (PRE)) by esterification of rosin acid and pentaerythritol. Compared with rapeseed oil (RO) and soybean oil (SO) without additives, when the concentration of PRE was 20 wt%, the oxidation induction times (OIT) of RO and SO increased by 305% and 124%, respectively. In addition, at 425°C , the weight loss rates of RO and SO decreased by 35.8% and 25.5%, respectively, and with an increase in PRE concentration, the thermal stability of lubricant samples increased gradually. This is because the thermal stability of the lubricating oil depends on its composition and chemical structure. Owing to the high hydrophenyl content and similar aromatic groups in PRE, this stable molecular structure can absorb a large amount of chemical energy and delay the occurrence of free radical reactions, resulting in good thermal stability [146].

Feng [147] utilized an intramolecular synergistic strategy to generate highly effective oxidation inhibitors by combining biobased phenol and aromatic amines. The Mannich condensation of hydrocyanhydrin (HC) with aromatic amines and polyformaldehyde was employed to produce three phenolic amine antioxidants (BAs). Due to the presence of the diphenylamine group, the thermal stability of the three types of BAs is much higher than that of the commercial antioxidants BHT and DPA. BA1 had an initial degradation temperature approximately 100°C higher than that of DPA. The higher molecular weight, more aromatic rings, and more closely aligned long alkyl chains (C15H31) of BAs, as well as the difference in thermal stability between phenol and aniline molecules, contribute to their high thermal stability. Similarly, Zhao [148] synthesized three types of biobased multifunctional additives, BMA1, BMA2, and BMA3, by combining biological phenol and aromatic amine in one molecule. The preparation process of the three antioxidants is shown in Figure 12. The chemical bond between biological phenol and aromatic amine increases the molecular weight and electron transfer rate, making BMAs very effective as antioxidants, regardless of the saturation of the vegetable oil. The oxidation induction time (OIT) of rapeseed oil (RO), coconut oil (CO), and epoxidized soybean oil (ESO) increased by 2.2, 14.0, and 32.0 times, respectively. The antioxidant activity of BMAs in epoxidized soybean oil was 2 and 12 times that of the commercial antioxidants BHT and DPA, respectively. Furthermore, the three BMAs have better thermal stability than the commercial antioxidants BHT and DPA, with BMA1 exhibiting the best thermal stability. The temperature at 5% weight loss (T5%) is as high as 296°C , much higher than that of BHT (103°C) and DPA (136°C). The excellent thermal stability

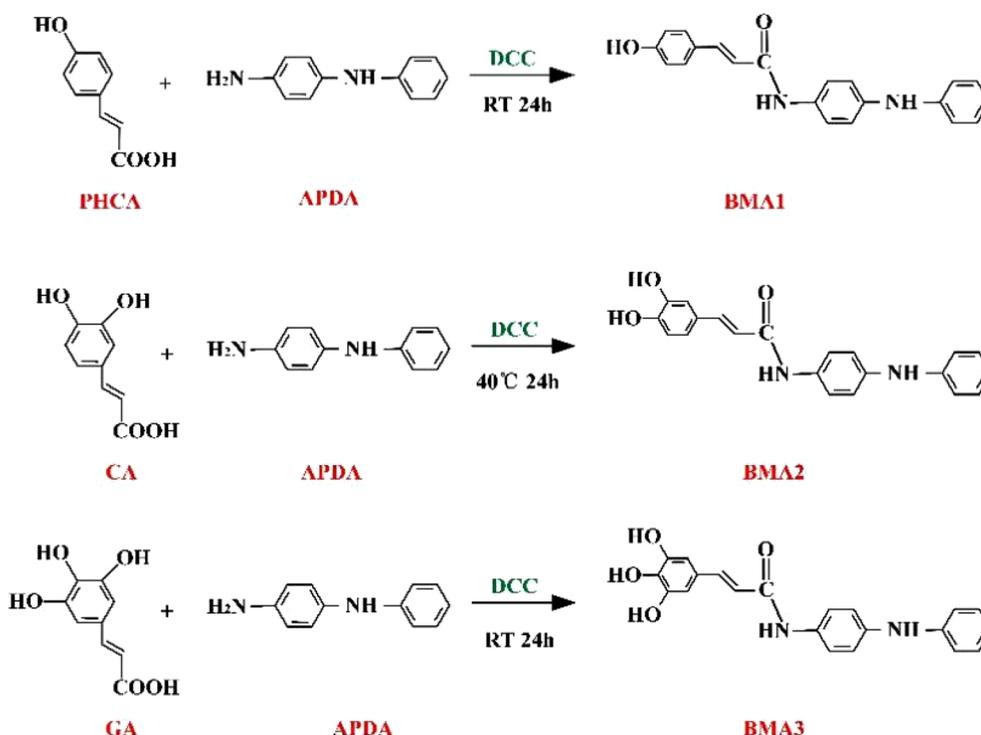


Figure 12 Three biological multifunctional antioxidants 1, 2 and 3 (BMA1, BMA2, BMA3) prepared with gallic acid (GA), caffeic acid (CA), hydroxycinnamic acid (PHCA), 4-aminodiphenylamine (APDA) and dicyclohexyl-3 carbodiimide (DCC) as catalysts

of BMA can be attributed to its high molecular weight and possible intermolecular hydrogen bonds. Concerning free radical scavenging activity, the ability of BMA3 to scavenge free radicals can exceed 90%. The scavenging activity of BMA3 and BMA2 on free radicals was superior to that of the existing commercial antioxidants BHT and DPA. The order of radical scavenging activity of the three BMAs was BMA3 > BMA2 > BMA1, with the scavenging activity increasing significantly with an increase in the number of hydroxyl groups (-OH) due to the radical scavenging activity caused by hydroxyl groups.

Another successful approach to developing high-temperature stable antioxidants is to link single phenol molecules to prepare polyphenol compounds. Jin [149] synthesized two polyphenol antioxidants, THA and PTP, which showed excellent thermal stability. Compared to THA, the PTP antioxidant has better thermal stability due to the instability of the C-S bond in the THA antioxidant at 310 °C, while the polyphenol antioxidant (PTP) bridged by ester has a more stable molecular structure at high temperatures. The authors tested the antioxidant performance of the two compounds by adding them to ester lubricants and measuring the oxidation initiation temperature (OOT) and oxidation induction time (OIT) of the oils. Both compounds showed higher oxidation resistance compared to the non-added base oil, but PTP

had better oxidation resistance than THA. The OOT of PTP (199.4 °C) was higher than that of THA (180.7 °C), and the OIT was 12.3 min longer than that of the THA antioxidant. This shows that PTP has excellent oxidation resistance in ester lubricants compared to THA. The difference in antioxidant capacity is related to the functional groups of the compounds. At room temperature, the free radical scavenging activity of the functional groups follows the order O->NH-, and the free radical scavenging activity increases with an increase in the number of hydroxyl (-OH) groups. Therefore, PTP, which has more hydroxyl groups, has a stronger antioxidant effect than THA.

3.2.3 Additional Properties

Some antioxidants can improve the oxidation stability of vegetable oils and meet the requirements of the industry.

Guo [150] found that conventional additives such as dispersants, detergents, antioxidants, friction modifiers, and viscosity index modifiers can reduce the friction coefficient of diisooctyl acetate (DIOS). Especially for AO-1 (T534, amine antioxidant), the average friction coefficient decreased by 28.93%. The friction coefficient of DIOS oil with added AO-2 (RF1135, phenolic antioxidant) slightly decreased. It is worth noting that the friction coefficient of Cu NPs added to DIOS with

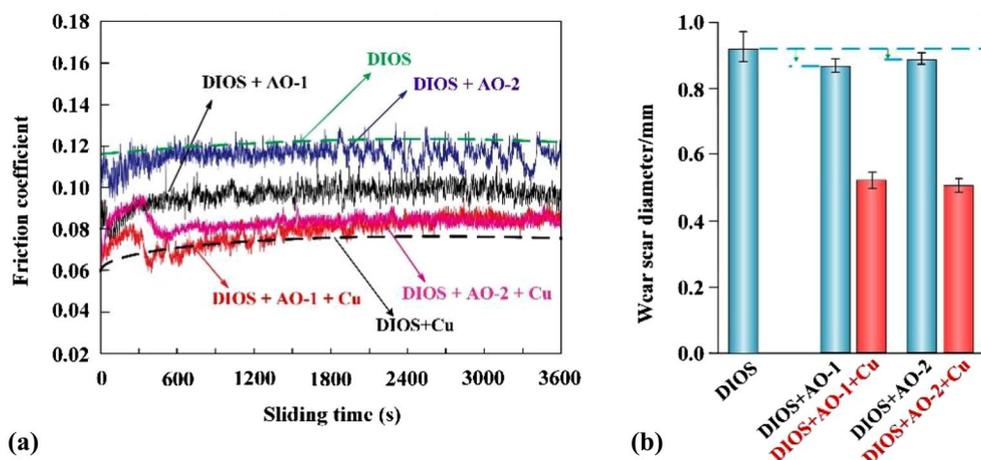


Figure 13 Friction coefficient and wear scar diameter with AO-1, AO-2 and Cu NPs added

antioxidants is larger than that of DIOS+Cu, as shown in Figure 13a. This means that AO-1 and AO-2 are antagonistic to Cu NPs in improving the lubrication effect of the DIOS oil. This was mainly due to the performance and structure of the additives used. However, the addition of AO-1 and AO-2 optimized the wear resistance of pure DIOS oil, and the wear scar diameter decreased slightly. The friction coefficients and wear scar diameters of AO-1, AO-2, and Cu NPs are shown in Figure 13b.

Jin [149] investigated the tribological properties of two polyphenol antioxidants (THA and PTP) and examined their structural changes under high-temperature oxidation friction. The lubricating oil's friction coefficient order was base oil+THA>base oil>base oil+PTP. The addition of THA did not enhance the base oil's lubricating performance but adversely affected the boundary film formation. On the other hand, the addition of PTP antioxidant improved the base oil's lubricating performance under high-temperature oxidation conditions and reduced the wear scar diameter by 22.91% compared to the base oil. The ester group of the PTP antioxidant can be adsorbed on the friction surface, reducing friction and wear. Therefore, PTP can be used as a multifunctional additive for ester lubrication at high temperatures. Xu [151] synthesized an environmentally friendly multifunctional additive called pentaerythritol rosin ester (PRE) by esterifying picric acid and pentaerythritol. Compared to rapeseed oil (RO) and soybean oil (SO), the dynamic viscosities of RO and SO increased by 136.11% and 179.3%, respectively, improving their lubricating properties and expanding their application range [152]. Zhao [148] synthesized three new BMA antioxidants by amidation reaction of biological phenol and 4-16 aminodiphenylamine. BMA2 and BMA3 were found to improve the

antioxidation and lubrication performance of vegetable oil. BMA3 exhibited the best tribological performance, reducing wear and friction by 18% and 25%, respectively, in the four-ball tests of RO and CO.

The synergistic effect of antioxidants and anti-wear additives is also an embodiment of the additional performance of antioxidants. It was previously reported that the antiwear effect of the antiwear additive ZDTP decreased with an increase in the peroxide content. Minami [153] studied the antiwear effect of ZDTP in antioxidant oxidized oils. Among them, free radical scavengers, such as DBPC and MBDBC, can prevent oxidation intermediates from converting into peroxides by invalidating carbon free radicals during autoxidation, thus preventing peroxides from reacting with ZDTP. The anti-wear properties of ZDTP remain unchanged.

Wang [154] investigated the impact of four Schiff base-bridged phenolic diphenylamine antioxidants (SSPDs) on the multifunctional properties of the traditional additive zinc dialkyl dithiophosphate (ZDDP). The addition of any SSPD with ZDDP resulted in enhanced EP performance of the base oil, with the PB value increasing by 18%–22% compared to the single addition of ZDDP. This improved performance can be attributed to the higher reaction rate of the mixture with the metal surface, leading to the formation of a harder EP film. The synergistic effect of the two additives was due to the decomposition of the cage structure of ZDDP by SSPD, which then formed complexes through coordination of the imine nitrogen and zinc atoms of ZDDP. The complexes were more easily adsorbed on the metal surface through various active centers, such as the sulfur atom of ZDDP, the amino group of SSPD2b, and the phenyl group, thus accelerating the formation of the friction film during the friction process. Additionally, excessive SSPD2b reduced the consumption of ZDDP by reacting with free radicals

and retaining the active ingredients for other functions. Figure 14 illustrates the synergistic mechanisms of antioxidant SSPDs and ZDDP, and Table 7 summarizes the molecular structures and effects of the antioxidants.

3.2.4 Summary of the Application Effect of Antioxidants

Antioxidants inhibit the oxidation process by preferentially reacting with free radicals to generate free radicals that do not rapidly react with oxygen. In practical applications, (i) owing to the different antioxidant activities of different functional groups, an antioxidant with more hydroxyl (-OH) causes an increase in free radical scavenging activity, so it has a stronger oxidation inhibition effect. (ii) In high-temperature environments, the preparation of polyphenol antioxidants through molecular connections is a successful means of high-temperature applications. The stability of intermolecular chemical bonds is a key factor for improving high-temperature resistance. (iii) The use of antioxidants can optimize the viscosity and anti-wear performance of the base oil and can be used as multifunctional additives in addition to simple antioxidants.

4 Improvement of Extreme Pressure Performance

The oil-based lubricant can form a dense adsorption film at the friction interface, thereby reducing the friction between the tool and workpiece interface. However, with an increase in the pressure and temperature of the friction interface, the intermolecular activity of the adsorbed film also increases, and the strength of the adsorption film decreases. The oily agent in the lubricant becomes

invalid, and the oil film will be damaged by the basic friction interface, thus losing the lubrication effect [155]. The addition of an extreme-pressure additive is a necessary condition for realizing high-temperature and high-pressure boundary lubrication of lubricating oil. Its main role is to improve the bearing capacity of the lubricating oil and reduce the wear of the workpiece and tool during machining. The EP-enhanced oil film could withstand greater loads and provide stronger anti-friction properties. Therefore, the ideal properties of extreme pressure lubricants include providing wear resistance under mild friction conditions and preventing adhesive wear under harsh extreme pressure conditions [156].

4.1 Conventional EP Additives

Traditional extreme-pressure additives containing sulfur [157, 158], phosphorus [159], and chlorine [160] belong to the category of active extreme-pressure agents. In the application process, molecules containing polar groups in the additives are first adsorbed onto the metal surface. Under the load of high temperature and high pressure, elements such as sulfur, phosphorus, and chlorine in the molecules react with the metal surface by friction chemistry, forming a boundary lubricating film with good adhesion and easy shearing, thus achieving anti-wear and antifriction properties [161, 162]. Extreme pressure additives based on sulfur, phosphorus, and chloride are activated by reaction with metal surfaces within a specific temperature range [163]. The extreme pressure additive containing chlorine can react with metal at 200–300 °C, withstand a high temperature of 600 °C, and exhibit

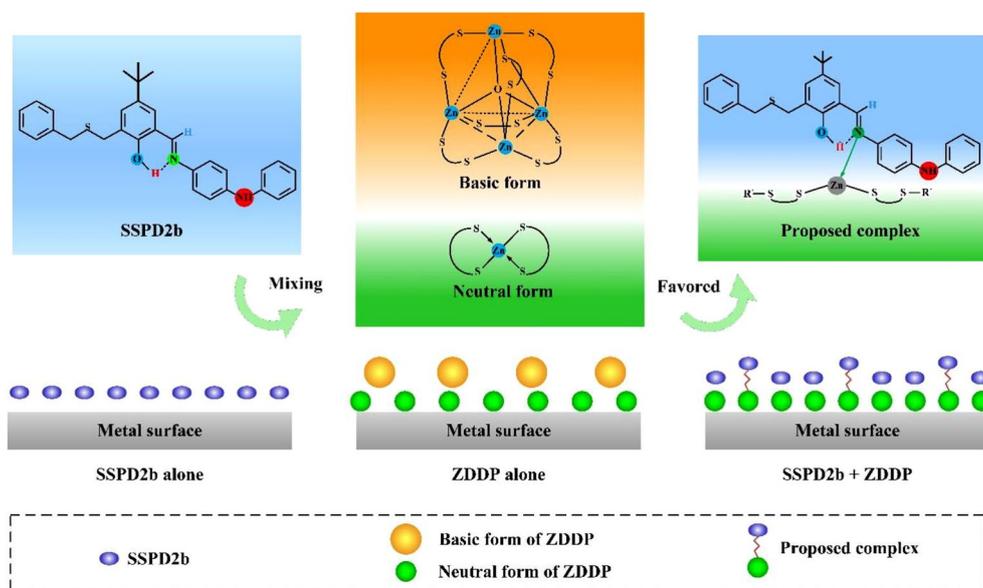


Figure 14 Synergistic mechanism of antioxidant SSPDs and ZDDP

Table 7 Main types and effects of antioxidants

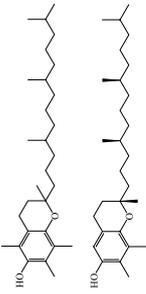
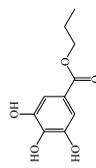
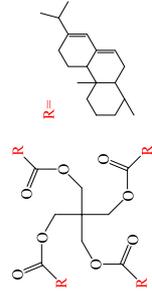
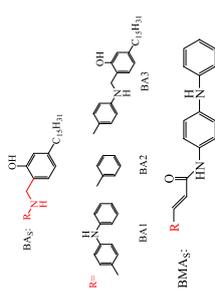
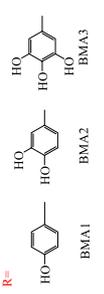
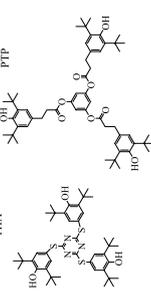
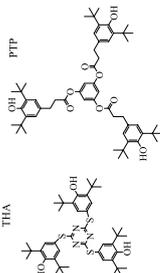
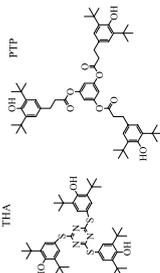
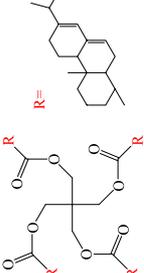
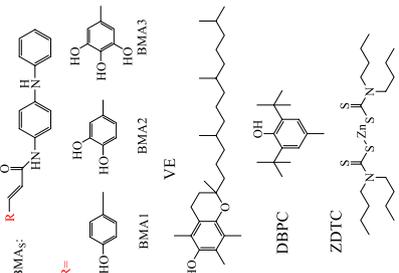
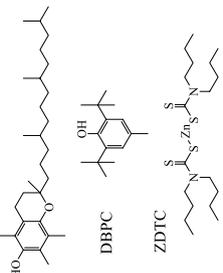
Ref.No.	Main additives	Molecular structure	Basic fluid	Application effect
[141]	Antioxidant effect α -tocopherols, γ -tocopherols		Rapeseed oil triacylglycerols	$\leq 50 \mu\text{g/g}$, α -tocopherols $>$ γ -tocopherols $> 100 \mu\text{g/g}$, γ -tocopherols $>$ α -tocopherols.
[143]	ADPA, BHT, APDD, ZDDC		Soybean oil	ZDDC $>$ BHT $>$ APDD $>$ ADPA Too low reaction temperature (150 °C) is an important reason for poor oxidation resistance of ADPA. PG increases the thermal degradation temperature of vegetable oil from 331 °C to 347 °C.
[144]	Propyl gallate (PG)		High-oleic sunflower, Castor oils	
[145]	Pentaerythritol ester (PRE)		Rapeseed oil, Soybean oil	When the concentration of PRE was 20 wt%, the oxidation induction time (OIT) of RO and SO increased by 305% and 124%, respectively.
[147]	Biobased antioxidants (BAs)		Polyol ester oil	The initial degradation temperature of BAI is nearly 100 °C higher than that of diphenylamine (DPA).
[148]	Biobased multifunctional additives (BMAs)		Rapeseed oil, Coconut oil, Epoxidized soybean oil	No matter how saturated the vegetable oil is, BMAs are very effective as antioxidants. The oxidation induction time (OIT) of rapeseed oil, coconut oil and epoxidized soybean oil increased by 2.2, 14.0 and 32.0 times.
[149]	Two multiphenol antioxidants, THA and PTP		Ester lubricant	The oxidation initiation temperature (199.4 °C) of PTP antioxidant was higher than that of THA (180.7 °C), and the oxidation induction time was 12.3 min longer than that of THA antioxidant.

Table 7 (continued)

Ref.No.	Main additives	Molecular structure	Basic fluid	Application effect
[150]	Additional effects AO-1 (T534, amine antioxidant), AO-2 (RF1135, phenolic antioxidant)		Diisooctylsebacate (DIOS)	The addition of AO-1 and AO-2 improved the lubrication effect of antiwear performance of DIOS, and AO-1 reduced the friction coefficient of DIOS by 18.93%.
[149]	Two multiphenol antioxidants, THA and PTP		Ester lubricant	The addition of PTP antioxidant reduced the wear scar diameter by 22.91% compared with the base oil.
[145]	Pentaerythritol rosin ester (PRE)		Soybean oil, Rapeseed oil	When the concentration of PRE was 20 wt%, the dynamic viscosity of RO and SO increased by 136.11% and 179.3%, respectively.
[148]	Biobased multifunctional additives (BMAs)		Rapeseed oil, Coconut oil, Epoxidized soybean oil	The wear and friction of vegetable oil added with BMA3 decreased by 18% and 25%, respectively.
[153]	alpha-Tocopherol(VE), 2,6-dibutyl-4-methylphenol (DBPC), (O,O-diisopropyl-dithiophosphate) (ZDTP)		Rapeseed oil	Phenolic antioxidants and zinc dialkyldithiophosphate exhibited good results with regard to preventing peroxide generation. ZDTP has synergistic effect with antioxidant additives.
[154]	Schiff base bridged phenolic diphenylamine antioxidants (SSPDs), zinc dialkyldithiophosphate (ZDTP)		Base oil	Compared with the single addition of ZDDP, when any SSPD is added at the same time, the EP performance of base oil is enhanced, and the PB value increases by 18%–22%.

good lubricating performance. The reaction temperature between the extreme-pressure additive containing phosphorus and metal is 350–600 °C. The reaction temperature between the sulfur extreme pressure additive and metal surface is 600–900 °C, and the formation of a vulcanization film can maintain the lubricating performance at a high temperature of approximately 1000 °C. The use of these additives also has a synergistic effect and can satisfy a wide range of cutting temperatures. Figure 15 shows the operating range of the three extreme-pressure additives.

4.1.1 Action Mechanism

During the application of additives, their interaction with the environment, such as oxygen, water, and carriers, can generate active substances. These active substances can be adsorbed onto the metal surface and further interact with other active molecules, forming a polymer film

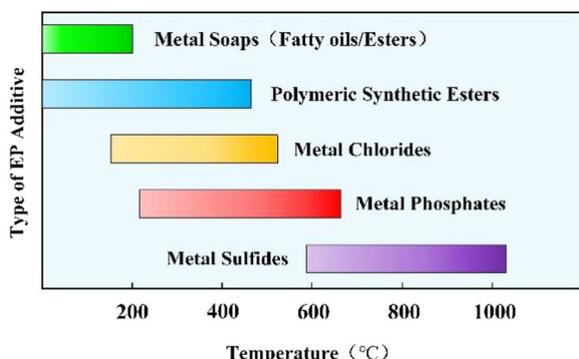


Figure 15 The operating range of the three extreme pressure additive types

under high temperature and pressure. As the processing progresses, the polymer film undergoes wear due to sliding and chemical dissolution or enters the entire lubricant because of the extrusion contact point, which changes the lubricant’s rheological properties. The reaction product is then recycled through the above steps. The adsorption of additives onto the workpiece surface is critical in the above reaction. Higher adsorption concentration of the additive on the workpiece surface leads to a higher reaction rate [164]. The adsorption layer can be formed through basic methods, and the formation process is illustrated in Figure 16.

When additives containing Cl, S, and P collide with the metal surface, spontaneous formation of the polymer film is detected, accompanied by the formation of iron compounds and participating products. Ferric chloride, ferric sulfide, and ferric phosphate are important products of reactions with additives containing Cl, S, and P.

The mechanism of action of the extreme pressure agent is similar to that shown in Figure 16. In the application process, a chlorine-based extreme-pressure agent is first adsorbed on the friction surface. With an increase in pressure and temperature, chlorine reacts with the friction metal surface to form a ferric chloride lubricating oil film with a layered structure, which has extreme pressure and anti-wear effects. Chlorinated paraffin, which has good chemical stability and flame retardancy, is commonly used as an extreme pressure agent. Other commercial types include chlorinated olefins, fatty acids, and esters [165].

The mechanism of action of the sulfur extreme-pressure agent can be described starting from physical adsorption, chemical adsorption, and sulfur cracking and

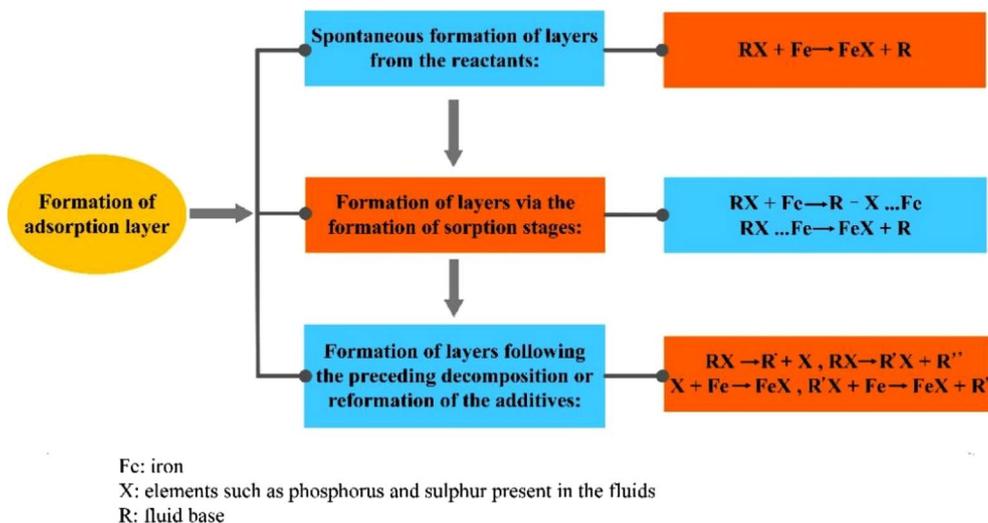


Figure 16 Formation process of adsorption layer of EP additive on workpiece surface

its reaction with the metal surface [166]. Sulfur-based extreme-pressure agents first adsorb on the metal surface to form a physical fluid adsorption film, which plays a role in fluid lubrication. With an increase in pressure and temperature, the S–S bond breaks, and the generated sulfur reacts with the metal surface via friction chemistry, forming a chemical adsorption film of iron mercaptan. As the pressure and temperature rise again (above 800 °C), the C–S bond breaks and forms a solid film of iron sulfide, which effectively inhibits sintering. The common sulfur extreme pressure agents are vulcanized esters, vulcanized lubricating grease, vulcanized hydrocarbons, and polysulfides. According to different processes, these additives are either dark or light [167].

The phosphorus extreme-pressure agent mainly refers to the phosphate ester extreme-pressure additive. Phosphate esters are mixtures of monoesters and diesters. The EP performance is optimized by adjusting the ratio of monoesters and diesters. Under mild conditions, phosphate ester is first adsorbed on the friction pair surface. When water is present in the medium, the phosphorus extreme-pressure agent is first hydrolyzed into phosphoric acid, and then phosphoric acid and metal form iron phosphate. With an increase in load and temperature, phosphate esters react with metal oxides to form phosphate or polyphosphate under boundary lubrication conditions, which have extreme pressure and anti-wear effects [168].

4.1.2 Application Effect

Wang [169] investigated the tribological properties of chlorinated paraffins as extreme pressure additives for vegetable oil (rapeseed oil). The chlorinated paraffin additive significantly improved the bearing capacity and wear resistance of rapeseed oil, and the optimal mass fraction of the additive was 0.5%. However, the effect of the chlorinated paraffin additive on improving the bearing capacity is not obvious in high-load (>600 N) processing. When the concentration of the additives was too high, the lubricating effect declined. The tribological properties of sulfurized isobutylene and sulfurized cottonseed oil as extrusion anti-wear additives for rapeseed oil were also determined. Sulfurized isobutylene can significantly improve the bearing capacity and anti-wear properties of rapeseed oil. Compared with sulfurized cottonseed oil, the maximum bite free load (PB) and wear scar diameter (WSD) of rapeseed oil with sulfurized isobutylene decreased by 32% and 30.6%, respectively. It is worth noting that the higher the content of additives, the better; otherwise, the WSD value increases. Asadauskas [170] tested the effect of chlorinated paraffin on the boundary lubrication properties of soybean oil (SO) and mineral oil (150 N oil). In the 4-Ball EP test, the weld points of both

oils were 120 kgf without the EP additive. The boundary lubrication performances of the two oils were significantly improved by adding 5% EP. The weld point of SO increased to 350 kgf. This value is much higher than the 160 kgf value of 150 N oil. The 150 N oil weld point with 20% EP additive was only 300 kgf. Therefore, the EP additive is more effective in soybean oil than in 150 N. The melting point of the 5% EP additive mixture in soybean oil is equal to or higher than that of the 20% EP additive mixture in 150 N oil. In addition, the wear damage of the 5% EP additive mixture in soybean oil and the 20% EP additive mixture in 150 N oil during 8 s of TCT operation is similar.

Li [171] evaluated the tribological properties of three phosphate ester additives in rapeseed oil (RSO). The synthesized phosphate esters exhibited good bearing capacity and anti-wear performance when used as rapeseed oil additives. Addition of the phosphate ester additives to RSO significantly reduced the diameter of the wear scar. Similarly, Li [172] prepared three phosphate esters containing benzotriazole groups as RSO additives and tested their influence using a standard four-ball tester, finding that they exhibited good wear resistance and load-bearing capacity. However, the adaptability of different additives to different base oils can vary, and the effect of additives depends on the chemical properties of the base oils and additives, as well as the performance of the material surfaces and the wear state. Fan [173] synthesized a new extreme pressure additive (PN) derived from ammonium benzotriazole phosphites, which exhibited excellent anti-friction and anti-wear performance under extreme pressure conditions. The friction coefficient (COF) of 1 wt% PN was approximately 22.2% and 12.5% lower than that of common additives ZDDP and T304 under a load of 392 N. The synthetic PN additive showed better anti-friction effects than ZDDP and T304 under low load and high speed conditions, and its COF remained relatively stable with increasing load, as shown in Figure 17. Johnson [174] reviewed the application of phosphate esters and thiophosphate esters as anti-wear or extreme pressure additives and discussed their mechanisms of action. While the details and film-forming conditions of the film vary for different additives, they all form an effective coating that reduces friction and wear on the surface.

Wang [175] compared the tribological properties of two different vegetable oil additives, sulfurized isobutylene and sulfurized cottonseed oil, when added to rapeseed oil. The results showed that the PB value (the maximum load-bearing capacity before the oil film breaks down) of rapeseed oil supplemented with sulfurized isobutylene was 66.7% higher than that of sulfurized cottonseed oil, and the wear scar diameter was 48% lower than that of sulfurized cottonseed oil. Thus, sulfurized

isobutylene exhibited better extreme pressure and anti-wear properties in rapeseed oil than sulfurized cottonseed oil. Additionally, increasing the amount of additives used improved performance, with the optimal amount being 2%. Anand [176] developed a biodegradable and nontoxic energy-saving EP gear oil by using a sulfur-containing vegetable oil additive based on vegetable oil, phosphothio capric acid methyl ester, and phosphothio hydrogenated cashew nut amine borate. In comparison to traditional sulfated olefins and alkyl aryl thiophosphates, sulfated and phosphorous sulfated vegetable oils and hydrogenated cardanols provide hydrocarbon-based and ester-based lubricants with excellent anti-wear and anti-wear properties. Moreover, the hydrocarbon oil formula based on vegetable oil ester and cashew nut shell liquid is non-toxic, whereas the dialkyl formula based on sulfurized olefin and alkyl aryl thiophosphate, molybdenum thiocarbamate, and thiocarbonate belongs to the slightly toxic/harmful category.

The effectiveness of additives can be influenced by the different unsaturated fatty acids present in vegetable oils. Waleska [177] studied the effects of organic sulfur and phosphorus anti-wear additives (A), phosphate diesters (B), and amine phosphates (C) in three types of vegetable oils: normal refined soybean oil (containing approximately 25% oleic acid side chain), high oleic soybean oil (containing approximately 85% oleic acid), and epoxidized soybean oil (in which all double bonds have been chemically modified by adding oxygen to form an epoxy ring). Among the three vegetable oils without additives, the epoxidized vegetable oil demonstrated the best wear resistance. After adding 1 wt% of A, B, and C additives, A and B additives showed good anti-wear properties in two unsaturated vegetable oils (refined soybean oil and high oleic soybean oil), reducing wear by 54.5%–77.8% compared to vegetable oil without additives. However, the wear of A and B additives increased in the saturated epoxidized vegetable oil. Additive C showed a lower wear rate in the three vegetable oils with different saturations, reducing wear by approximately 40%–53.5%.

Traditional extreme-pressure additives have a satisfactory effect on improving the extreme pressure and anti-wear properties of vegetable oils. However, the environmental pollution caused by traditional extreme pressure antiwear agents containing sulfur, phosphorus, and chlorine cannot be ignored. Sulfur pollutes the environment, and phosphorus causes rich oxidation of water. Chlorine-containing additives have been restricted in many countries owing to their toxicity and corrosivity [178]. Therefore, research on new additives in various countries is urgently needed. Researchers worldwide have conducted various studies on green nanoparticle additives.

4.2 Nanoparticle Additives

Nanoparticles generally refer to particles with a size of 1–100 nm, which are in the transition area between atomic clusters and macroscopic objects. Nanoparticles are nonvolatile, stable at high temperatures, small in size, high in surface energy, and exhibit good heat transfer [179, 180]. A nanofluid refers to a new fluid formed when nonmetal or metal nanoparticle additives with sizes less than 100 nm are dispersed into the base fluid. Nanofluids exhibit better extreme pressure performance, anti-wear and antifriction performance, heat transfer performance, and bearing capacity than basic fluids [181]. The lubrication and heat transfer performance of nanofluids can be controlled by changing the concentration of nano additives. Due to the anti-wear, friction reduction and cooling characteristics of nanoparticles, when processing difficult-to-process materials, adding nanoparticles to vegetable oil lubricants can better solve the high temperature generated during processing [29, 182]. In addition, nanoparticles do not pollute the environment; therefore, they are an effective substitute for traditional extreme pressure antiwear agents [183]. Common nanoparticles are shown in Table 8.

Mello [184] compared the tribological properties of conventional EP additives and nanoparticles under boundary-lubrication conditions. ZnO nanoparticles

Table 8 Common nanoparticles

Classification	Nanoparticles
Carbon and its derivatives	Graphene, Diamond, Single wall carbon nanotube, Multi wall carbon nanotube, Single-layer carbon nanotube, Multilayer carbon nanotube
Nano metal element	Sn, Fe, Bi, Cu, Ag, Ti, Ni, Co, Pd, Au
Nano oxide	TiO ₂ , Al ₂ O ₃ , ZnO, CuO, AlOOH, Fe ₃ O ₄
Nano disulfide	WS ₂ , CuS, MoS ₂ , NiMoO ₂ S ₂
Nanometer rare earth compounds	LaF ₃ , CeO ₂ , La(OH) ₃ , Y ₂ O ₃ , CeBO ₃
Nanocomposites	Cu/SiO ₂ , Cu/graphene oxide, Al ₂ O ₃ /SiO ₂ , Serpentine/La(OH) ₃ , Al ₂ O ₃ /TiO ₂
Other	CaCO ₃ , ZnAl ₂ O ₄ , Zeolite, ZrP, SiO ₂ , PTFE, Hydroxide, BN, Serpentine

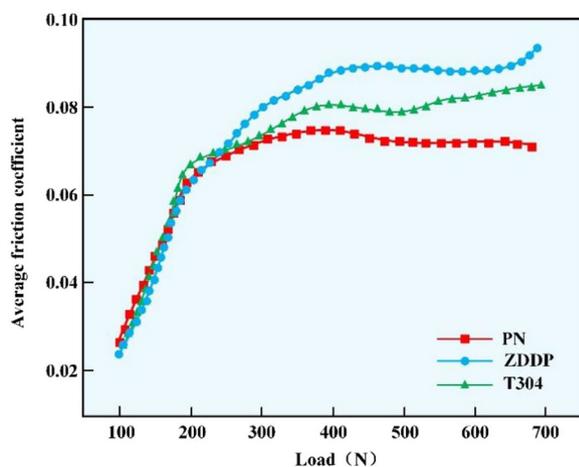


Figure 17 COF curve of three different additives

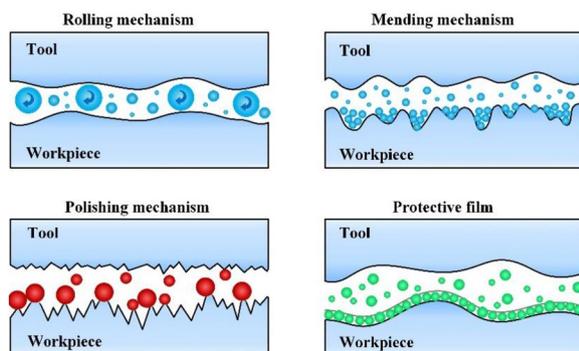


Figure 18 Different mechanisms by which nanoparticles improve tribological properties

showed a stronger affinity for mineral oil and showed similar anti-wear properties as ZnDDP and low friction properties as sulfur additives. This indicates that the deposition of nanoparticles on the surface may form a physical friction film, which is adsorbed on the friction surface and forms a protective film. The SEM images showed a uniform surface comparable to that of the other additives. Therefore, it can be used as a good anti-wear and antifriction additive.

4.2.1 Action Mechanism

Nanoparticles reduce friction and wear mainly through rolling, repair, polishing, and protective film formation [185]. The specific process is illustrated in Figure 18.

Nanoparticles can be divided into layered, granular, onion, tubular, and spherical structures. The addition of nanoparticles with different structures results in better extreme pressure performance, anti-wear and antifriction performance, and bearing capacity. Due to

the “ball effect” and “filling effect” of nanoparticles, the lubricating performance of the cutting fluid has also been greatly improved with the addition of nanoparticles. Owing to the differences in the physical properties and shape characteristics of nanoparticles with different structures, there are also differences in lubrication mechanisms; therefore, they have different lubrication effects [186]. Layered, spherical, and onion-shaped nanoparticles show superior tribological properties compared with other material structures [187, 188].

Because of their specific crystal structure, layered nanoparticles are connected by van der Waals forces between layers, and slip easily forms between surfaces. After the molecules break along the molecular layer, slip surfaces are formed to form an extended physical film to isolate the two friction surfaces and reduce friction and wear [189, 190]. Sliding of the layered nanoparticles is shown in Figure 19a. On the other hand, because the boundary of layered nanoparticles often has a stronger surface energy, under extreme conditions such as high temperature and high pressure during processing, nanoparticles adsorbed on the metal surface are prone to oxidation reactions, forming dense oxide films, which have excellent anti-wear and antifriction effects. It exhibits good lubrication performance in the field of machining lubrication. However, the heat transfer coefficient of these nanoparticles is low, and the heat transfer capacity is not ideal. Some layered nanoparticles, such as molybdenum disulfide, fail owing to their easy carbonization, leading to a decline in the lubricating performance. Common nanoparticles with layered structures include graphite and Al_2O_3 nanoparticles.

The surface energy of the spherical nanoparticles is uniform in all directions. During processing, spherical nanoparticles form a layer of physically deposited film at the interface between the tool and workpiece. Moreover, because of the structural characteristics of spherical nanoparticles, they can be regarded as one after another “small balls”, which play a similar role as “micro bearings” on the surface of friction pairs [191]. Thus, the sliding friction between the tool and workpiece interface is changed to rolling friction, and the friction coefficient is reduced. In the machining process, the peaks of some microconvex bodies on the machined surface break through the oil film and cause collision and fracture, accompanied by new wear marks. Under the action of a high friction shear force and high normal load at the interface between the tool and workpiece, spherical nanoparticles will deform owing to extrusion, and some of them will peel off. With the generation of new wear marks, the detached nanoparticles fill in new scratches, repair the friction surface, make the machined surface smooth and flat, and further

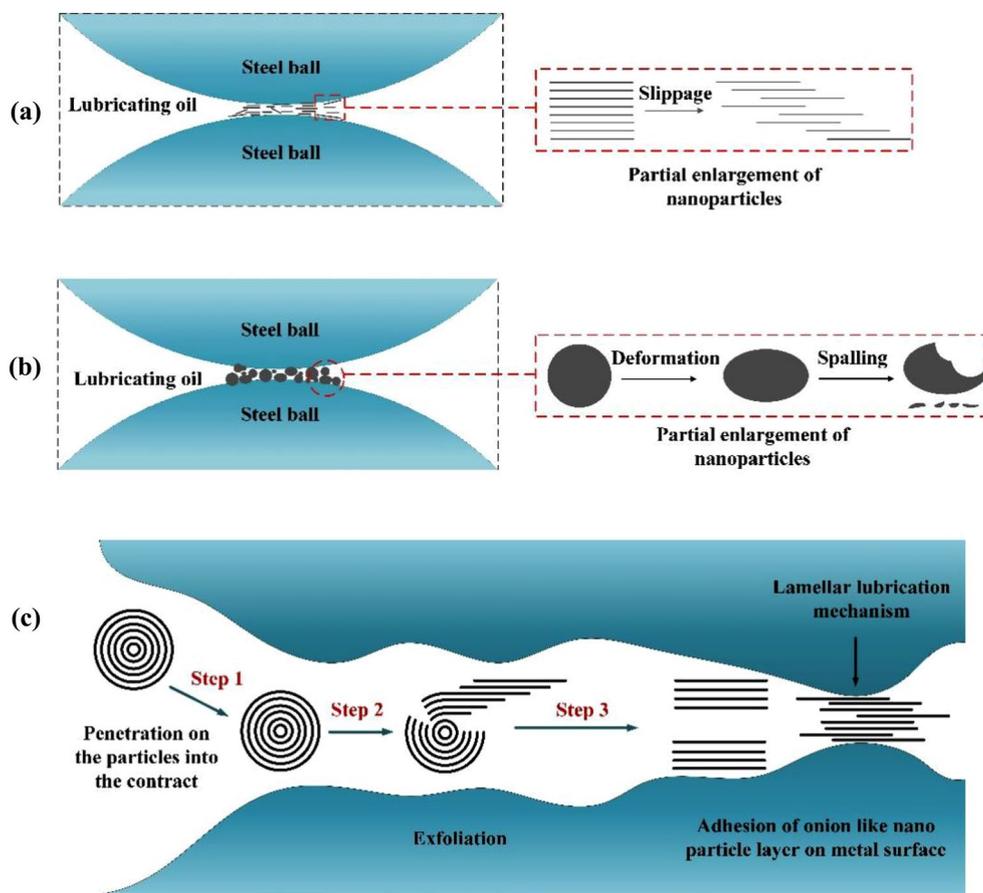


Figure 19 Anti-wear mechanism of **a** layered, **b** spherical and **c** onion nanoparticles

reduce wear [192]. The deformation and spalling of the spherical nanoparticles are shown in Figure 19b.

Similar to spherical nanoparticles, onion-like nanoparticles are spherical. However, it is layered in the interior, with no hanging bonds inside, resulting in less interaction between the particles and surfaces. Similar to the lubrication mechanism of spherical nanoparticles, onion-like nanoparticles can also enter the machining surfaces of tools and workpieces due to their small size, sliding or rolling in the contact area, playing the role of “micro bearing”. With the increase in load, under anisotropic processing pressure, the layered structure of onion-like nanoparticles splits, and the external flakes peel off and adhere to the friction. The formed nanoparticles, similar to the layered structure, form a layer of physical film with anti-wear and antifriction effects on the friction surface [193]. Onion nanoparticles combine the advantages of spherical nanoparticles and layered nano ions, so their lubrication ability is more prominent than that of spherical nanoparticles and layered nanoparticles, among which common nanoparticles such as molybdenum disulfide and onion graphene belong to this structure.

The anti-wear mechanism of onion-like nanoparticles is shown in Figure 19c.

4.2.2 Application Effect

In view of the shortcomings of traditional casting processing, such as the large consumption of cutting fluid, difficult post-processing, and harm to the environment and human body, minimum quality lubrication technology has become an effective alternative [194–196]. Minimum quality lubrication (MQL) processing, also known as quasidry or semidry processing, is a processing method that mixes and vaporizes a small amount of lubricating oil with compressed gases, such as air, nitrogen, and carbon dioxide, to form micrometer-sized droplets, which are sprayed to the processing area in the form of aerosols to effectively lubricate during processing [197–199]. As a green processing method that matches environmentally friendly vegetable oil-based cutting fluids, it has become the focus of scholars all over the world to explore how to improve the performance of nanoparticles as extreme-pressure additives in vegetable oil-based minimum quality lubrication processing.

(1) Cutting Force

The addition of nanoparticles to vegetable oils can significantly reduce the cutting force. Singh [55] conducted an in-depth study of the tribological properties of graphene nanosheets. Compared with traditional casting machining with synthetic cutting fluid, the MQL of rapeseed oil with 1.5 wt% graphene reduced F_n and F_t by 22.1% and 33.8%, respectively. The stress situation is shown in Figure 20a. This is because the smooth surface and unique two-dimensional (2D) structure of graphene help reduce friction. In addition, the number of graphene layers determines tribological properties. Because sliding occurs in the layers when a tangential load is applied, the friction decreases with an increase in the number of layers. However, a small number of layers can easily produce a fold effect.

Gao [200] conducted a study on the influence of milling on milling force under four lubrication conditions: dry machining, flood machining, minimum quantity lubrication (MQL) with cottonseed oil, and MQL with alumina (Al_2O_3) nanoparticles. The stress on the milled aluminum alloy under the four different lubrication modes is shown in Figure 20b. The cutting force in dry machining without lubrication was significantly higher in the x , y , and z directions (234.486 N) compared to the other lubrication conditions. The cutting force in MQL with cottonseed oil was 19.2% lower than that in dry machining, while the cutting force in MQL with vegetable oil-based lubricant after adding alumina nanoparticles was further reduced. The study found that adding nanoparticles can change the chip removal from sliding mode to rolling mode during cutting, which improves lubricity and reduces friction.

Zhang [201] conducted a study on the lubricating properties of MoS_2/CNT mixed nanoparticles and single nanoparticles, and found that the mixed nanoparticles

exhibited better cooling and lubricating properties than single nanoparticles. The optimal mixing ratio of MoS_2/CNT and nanofluid concentration were 2:1 and 6 wt%, respectively. Mix (2:1) showed higher grinding force than pure MoS_2 , while the grinding forces F_n and F_t were 16.29% and 29.12% lower than those of pure CNT, respectively. The lowest friction coefficients were 15.32% and 8.79% lower than those of pure MoS_2 and pure CNT, respectively. Mixing two types of nanoparticles with different structures can improve the lubrication state of the grinding area through physical synergy, and significantly reduce the grinding force. However, different forms of nanoparticles have varying effects on the cooling and lubricating properties. Spherical and layered nanoparticles have good antifriction and antiwear properties, but low thermal conductivities. Tubular CNTs have higher thermal conductivity and heat transfer capacity but are not conducive to lubrication.

On the other hand, not all nanoparticles can increase the processability of vegetable oil-based lubricants. Alves [202] investigated the effects of adding nanoparticles to vegetable oil-based lubricants, and found that adding CuO nanoparticles to sunflower oil and soybean oil increased the friction coefficients by approximately 20% and 7.5%, respectively. The study indicated that the addition of nano-oxide particles may deteriorate film formation in vegetable-based lubricants. The author explained that this may be related to the third-body behavior of these oxides, which increases friction and reduces the conductivity (less ECR) between sample surfaces [203].

(2) Cutting temperature

The addition of different nanoparticles to vegetable oil also affects the cutting temperature. According to the theory of enhanced heat transfer, the heat transfer capacity of a solid is better than that of a liquid. Therefore, an appropriate amount of solid nanoparticles

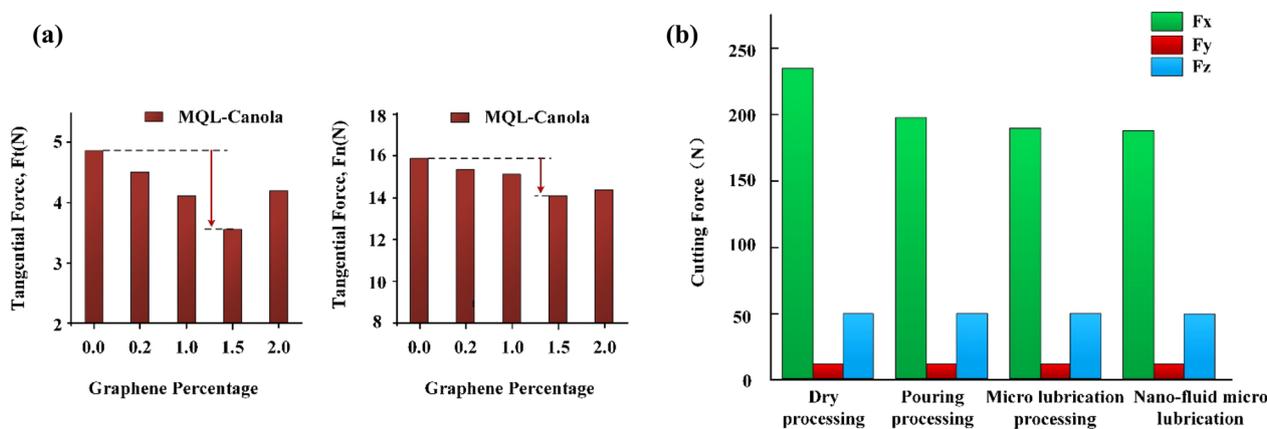


Figure 20 Effect of graphene a and Al_2O_3 b nanoparticles on cutting force reduction in MQL processing of vegetable oil

should be added to the vegetable oil base oil to form a nanofluid. During the minimum quality lubrication process, the nanofluid is sprayed onto the flooded area in the form of high-pressure gas atomization, thereby enhancing the heat exchange capacity of the liquid in the grinding area and improving the cooling effect. Li [204] studied the influence of a variety of nanofluid vegetable oil (palm oil)-based minimum quality lubrication on the surface grinding temperature, including six types of nanoparticles: molybdenum disulfide (MoS_2), zirconia (ZrO_2), carbon nanotubes (CNT), polycrystalline diamond, aluminum oxide (Al_2O_3), and silicon dioxide (SiO_2). The grinding temperatures of the six nanofluids are shown in Figure 21. The CNT nanofluids exhibits the best cooling performance. Adding nanoparticles to the base oil can significantly improve the heat transfer performance of the base oil, and nanoparticles with high thermal conductivity have better heat transfer performance, while CNT nanoparticles have the highest thermal conductivity ($3000 \text{ W}/(\text{m}\cdot\text{K})$) among the six nanoparticles. From the perspective of increasing the viscosity of nanoparticles, the author analyzed the convective heat transfer effect of nanofluid minimum quality lubrication by using boundary layer theory.

Li [205] studied the effect of palm oil-based nanofluids with carbon nanotubes on the heat transfer performance of three different workpiece materials (45 steel, nickel-based alloy, and ductile iron) during grinding. A numerical simulation heat-transfer model for the MQLC grinding temperature was established. In the process of minimum quality lubrication grinding with 2% palm oil-based carbon nanotubes, the highest grinding temperature ($363.9 \text{ }^\circ\text{C}$) was obtained for 45 steel, the lowest grinding temperature ($143.2 \text{ }^\circ\text{C}$) was obtained for ductile iron, and the grinding temperature of the nickel-based alloy was $245.7 \text{ }^\circ\text{C}$. This indicates

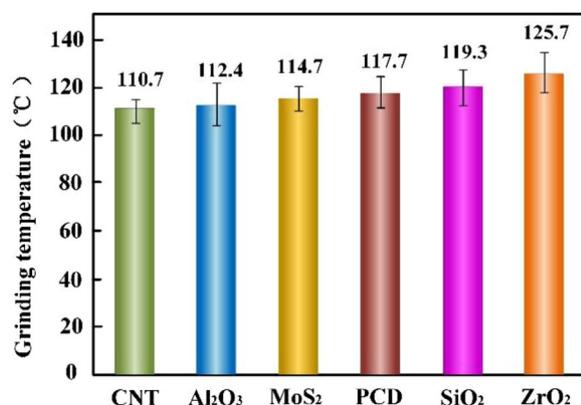


Figure 21 Grinding temperature of six nanofluids

that under the same working conditions, the type of workpiece material has a significant impact on the grinding temperature.

Krishna [206] studied the change in cutting temperature of AISI 1040 steel using cemented carbide tools during turning. The study was carried out in a nanoboric acid suspension in the environment of SAE-40 and coconut oil. Compared with SAE-40, the cutting temperature of coconut oil was lower. Under all lubrication conditions, the cutting temperature increased with increasing feed speed. Compared with 0.5% SAE-40 lubricant, the cutting temperature of the solid lubricant containing 0.5% nano boric acid particles in coconut oil decreased by approximately 21% under the combined action of the solid lubricant. This is because of the oily nature of vegetable oil molecules, that is, the chemical affinity between long and heavy dipoles and metal surfaces. In lubricants containing boric acid nanoparticles, a solid lubricant softens and forms a film at high temperatures. These nanosolid lubricant particles can increase the heat transfer performance of lubricants. The combined effect of coconut oil and nanosolid particles reduced the cutting tool temperature. At the same time, compared with the base oil, the grinding temperature decreased significantly after the addition of boric acid nanoparticles, which is related to the reduction in friction caused by the nano boric acid suspension.

(3) Tool wear

Tool wear is also a key factor that affects tool life. Krishna [206] studied the tool wear performance in the turning process of AISI 1040 steel using a nanoboric acid suspension. Compared to the base oil, the lubricant containing boric acid nanoparticles can significantly reduce the side wear of the tool. This is because under a high processing temperature, the solid lubricant forms a thin lubricating oil film at the interface between the workpiece and the prop; at the same time, the solid lubricant particles flow at the interface of the oil, reducing the plastic contact and side wear. Among them, the 0.5% nano boric acid particle suspension in coconut oil had the least side wear. The author explains that, compared with 0.5%, 0.25% boric acid cannot provide a sufficient lubrication effect, and 1% inclusion may reduce the fluidity of the lubricant and prevent it from entering the cutting area, thus reducing its effectiveness.

Yildirim [207] studied the influence of hBN nanoparticles on tool life in the MQL processing of ester lubricating oil. Compared with pure MQL and dry cutting, the tool life with 1% hBN nanoparticle MQL increased by 24% and 105.9%, respectively. At the same time, when the cutting speed, feed speed and cutting depth are kept constant at 40 m/min, 0.1 mm/rev and 0.8 mm, respectively, the tool wear of the tool with 1% hBN nanoparticles

MQL is reduced by approximately 20% and 36% compared with that of pure MQL and dry cutting, respectively. This is because nano additives retain oil particles and prevent the immediate release of cutting oil from the cutting area. At the same time, the high thermal conductivity of the nanofluids also reduces the cutting zone temperature. Therefore, it exhibits a good lubrication effect.

Adding nanoparticles in the minimum quality lubrication process of vegetable oil can reduce the wear of the rear tool surface and further improve tool life. The existence of a nanosheet reduces the microcutting and tool fracture caused by the impact effect in intermittent machining. During minimum quality lubrication, MQL containing a nanoplate solid lubricant is superior to conventional MQL in reducing the maximum side wear width and steady side wear.

(4) Surface quality

Different nanoparticles have different effects on the anti-wear and friction reduction during processing. Zhang [208] studied the influence of MoS₂, carbon nanotubes (CNTs), and ZrO₂ nanoparticles on the grinding surface quality during the microlubricating processing of rapeseed oil. The three types of nanoparticles clearly improved the surface quality, and the MoS₂ nanoparticles had the best lubrication performance. Tangential friction force (0.23) and specific grinding energy (32.7 J/mm³) are significantly lower than those of the CNT and ZrO₂ nanoparticle jets (0.247/0.258) and specific grinding energy (35.63 J/mm³, 36.49 J/mm³) and have the best surface roughness. The ground surface of MoS₂ nanoparticles can form a MoO₃ oxide film through oxidation, thus producing friction resistance. MoS₂ exhibited the best surface quality. Some substances accumulate in carbon nanotubes (CNTs) and ZrO₂. In addition, the surface texture gap and wave crest height were large. Hosseini

[209] studied the grinding of vegetable oil (sunflower oil)-based and mineral oil-based lubricants with different concentrations of MoS₂, graphite, and Al₂O₃ nanoparticles. The surface roughness along the grinding and transverse grinding directions is shown in Figure 22. In the two lubrication modes of pure minimum quality lubrication and nanoparticle minimum quality lubrication, the surface roughness of vegetable oil-based lubricants along the grinding direction and transverse grinding direction is always better than that of mineral oil-based lubricants. For different nanoparticles, MoS₂ and graphite nanoparticles can reduce the roughness values in two cutting directions (along the grinding direction and the transverse grinding direction). This is because MoS₂ and graphite nanoparticles are solid lubricants that can reduce friction and thus reduce surface roughness. However, the addition of vegetable oil and mineral oil base to Al₂O₃ increased the roughness values in all grinding directions. The authors speculated that this may be due to the high wear resistance of Al₂O₃ nanoparticles and the wear scratches formed on the workpiece surface during the grinding process.

Li [210] used graphene-dispersed vegetable oil-based cutting fluid MQL in the milling of TC₄ alloy to improve the milling characteristics of TC₄ alloy and evaluated and analyzed the milling force, milling temperature, surface microhardness, and surface roughness of TC₄ alloy. When graphene-dispersed vegetable oil-based cutting fluid is used for the MQL milling of TC₄ alloy, the concentration of the cutting fluid is a more important MQL parameter affecting the milling characteristics. The cutting fluid concentration, cutting fluid flow rate, and gas pressure of the nanoparticles in vegetable oil have a significant impact on the surface roughness. The most important minimum quality lubrication parameter is the

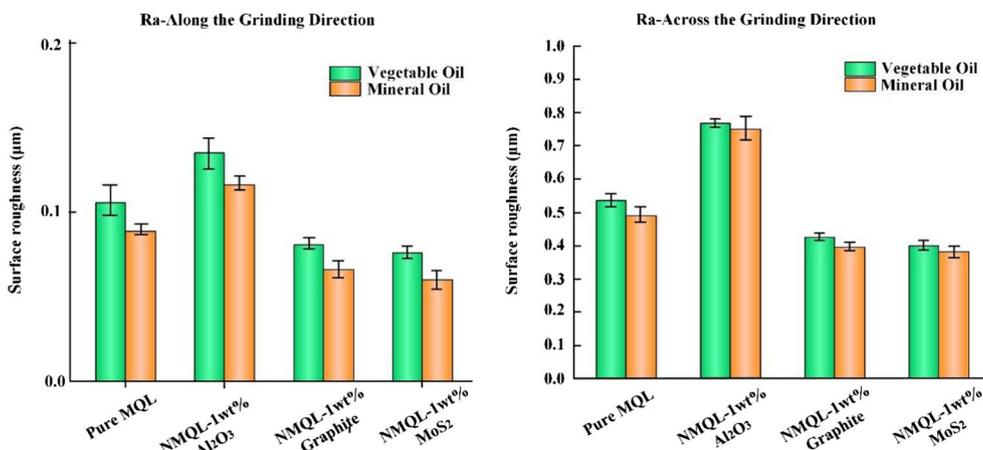


Figure 22 Surface roughness along the grinding direction and transverse grinding direction

cutting fluid concentration. This is because a proper concentration of nanoparticles can improve the lubricating and cooling properties of the oil film to improve the surface quality and reduce the surface roughness.

Zhang [211] studied the lubrication performance of MoS₂ nanoparticle palm oil-based nanofluids under MQL conditions. When 2% nanoparticle jet MQL grinding was carried out with paraffin and palm oil liquid, the corresponding R_a values perpendicular to the direction of grinding particles were 50.0% and 61.1% lower than those of dry grinding, respectively, and R_z values were 41.1% and 48.2% lower. Compared to the MQL of paraffin oil and palm oil liquid without nanoparticles, R_a decreased by 25% and 30%, and R_z decreased by 15.4% and 23.7%, respectively. This is because MoS₂ has a strong absorption capacity and high film-forming strength, which makes it difficult to break the oil film. The MoS₂ nanofibers have little influence on the abrasive resistance of the nanofiber because of their main abrasion on the friction pair surface to form a lubricating film.

Table 9 summarizes the application effects of various nanoparticles in different vegetable oils.

5 Conclusions

This paper summarizes the application mechanism of vegetable oil-based cutting fluids. The principle and application effect of chemical and additive modification methods are summarized in view of the problems of easy oxidation of vegetable oil and insufficient extreme-pressure performance. The main conclusions are as follows:

(1) The mechanism of autoxidation of vegetable oils was analyzed. The main reason for the increase in wear is the destruction of the fatty acid structure of the vegetable oil and the increase in peroxide content at high temperatures. Although peroxide has certain extreme pressure performance, adding 2.5% hydroperoxide can reduce wear by approximately 40% compared with pure vegetable oil under a 75–95 kg high load. However, under extreme pressure (126 kg), compared with mineral oil, the friction coefficient of vegetable oil increases by approximately 20%–107%. However, the oxidation stability and extreme pressure characteristics of vegetable oil-based lubricants still cannot meet the processing requirements.

(2) The mechanism of the antioxidation effect of vegetable oil was studied. Transesterification is an effective way to improve the oxidation stability of vegetable oils and reduce their pour point. For example, the initial oxidation temperature of rapeseed oil pentaerythritol ester is as high as 236 °C, and the pour point of the high-oleic palm oil TMP ester is –33 °C. The transesterification reaction depends on the type of vegetable oil used. Vegetable oils with a high percentage of monounsaturated fatty acids (such as rapeseed oil) are the best choice for

the development of high-temperature lubricants. The epoxidation ring-opening reaction increases the side chain through isomerization, which improves the anti-oxidation performance of vegetable oil, reduces the pour point, and improves its anti-wear performance. The oxidation stability decreases with an increase in side chain length. For example, the oxidation starting temperature of methyl hydroxy oleate propionate is 175 °C. Adding 5% epoxidized soybean oil to hexadecane reduces the COF by approximately 75.6%. The pour point of soybean oil isopropyl ester can reach –50 °C. The epoxidized ring-opening modification can be used as an effective means for the development of vegetable oil-based lubricants in many aspects.

(3) Antioxidants have varying compatibility with different vegetable oils. For example, owing to the presence of a polar group (-OH) in castor oil, most antioxidants have a higher affinity for castor oil. Antioxidants with high molecular weights and long alkyl chains have higher thermal stability, and their antioxidant capacity is related to the antioxidant activity of their functional groups. The application of molecular binding synergy is an effective means to develop high-temperature antioxidants. For example, when the temperature is approximately 300 °C, the weight loss of BMA1 is only 5%, whereas the ability of BMA3 to remove free radicals can exceed 90%. Antioxidants can be used as multifunctional additives. For example, the wear scar diameter of vegetable oil with added PTP decreases by approximately 18%–22.91%, the friction force of vegetable oil with added BMA3 decreases by approximately 25%, and the viscosity of vegetable oil with added PRE increases by approximately 136%–179.3%.

(4) The improvement mechanism of the extreme-pressure performance of vegetable oils was studied. Compared with the harmful effects of sulfur, phosphorus, and chlorine extreme pressure additives on the environment, nanoparticle additives can be used as an effective substitute for traditional extreme-pressure additives owing to their environmental protection characteristics. Compared with pure oil, the advantages of nanoadditives are reflected by the cutting force, cutting temperature, tool life, and surface quality. The cutting force, cutting temperature, tool wear, and surface quality of vegetable oil with nanoparticles decrease by 19.2%–33.8%, 21%, 20%–36% and 25%–30%, respectively.

In recent years, significant achievements have been made in research on methods to improve the resistance of vegetable oil-based cutting fluids, but there are still some limitations. According to a previous systematic review, future research directions may focus on the following topics.

(1) The antioxidant and physicochemical properties of different vegetable oils are different. Scholars worldwide

Table 9 Main types and effects of nanoparticles

Ref. No.	Lubricant	Nanoenh-ancer	Workpiece	Evaluation parameters	Conclusion
[55]	Rapeseed oil	Graphene nanosheets	Ti-6Al-4 V-ELI	Cutting force	Compared with the traditional casting machining with synthetic cutting fluid, the MQL of rapeseed oil with 1.5 wt% graphene reduced F_n and F_t by 22.1% and 33.8%, respectively.
[200]	Cottonseed oil	Al ₂ O ₃	Aviation aluminum 7075	Cutting force	The cutting force of cottonseed oil minimum quality lubrication machining is 19.2% lower than that of dry machining, while the cutting force of vegetable oil minimum quality lubrication machining with nanoparticles is further reduced.
[201]	Vegetable oil	MoS ₂ , CNT	GH4169 Nickel-based alloy	Cutting force	The best MoS ₂ /CNT mixing ratio and nanofluid concentration were 2:1 and 6 wt%, respectively. The friction coefficient is 15.32% and 8.79% lower than that of pure MoS ₂ and pure CNT, respectively.
[204]	Palm oil	MoS ₂ , ZrO ₂ , CNT, Polycrystalline diamond, Al ₂ O ₃ , SiO ₂	GH4169 Nickel-based alloy	Cutting temperature	CNT nanoparticles have the highest thermal conductivity (3000 W/(m·K)) among the six kinds of nanoparticles, and show the best cooling performance.
[205]	Palm oil	CNT	45 steel, nickel base alloy, ductile iron	Cutting temperature	The grinding temperature of 45 steel is the highest (363.9 °C), while that of ductile iron is the lowest (143.2 °C), and that of nickel base alloy is 245.7 °C.
[206]	Coconut oil, SAE-40	Nano boric acid suspension	AISI 1040	Cutting temperature, tool wear	Compared with SAE-40 lubricant, the cutting temperature of the solid lubricant containing 0.5% nano boric acid particles in coconut oil decreases by approximately 21% under the combined action of the solid lubricant. In addition, the 0.5% nano boric acid particle suspension in coconut oil has the least side wear.
[207]	Ester lubricating oil	hBN nanoparticles	Inconel 625	Tool wear	Compared with pure MQL, the tool wear and tool life with 1% hBN nanoparticles MQL are improved by approximately 20% and 24%, respectively.
[208]	Rapeseed oil	MoS ₂ , ZrO ₂ , CNT	Hardened Steel 45	Surface quality	It is observed that MoS ₂ has the best surface quality. Some substances are accumulated in carbon nanotubes (CNT) and ZrO ₂ . In addition, the surface texture gap and wave crest height are large.

Table 9 (continued)

Ref. No.	Lubricant	Nanoenh-ancer	Workpiece	Evaluation parameters	Conclusion
[209]	Paraffin oil, sunflower oil	MoS ₂ , graphite, Al ₂ O ₃	Tungsten carbide (WC) grade YG8	Surface quality	MoS ₂ and graphite nanoparticles can reduce the roughness values in both cutting directions. However, the addition of Al ₂ O ₃ will increase the roughness values in all grinding directions.
[211]	Palm oil, liquid paraffin	MoS ₂	45 steel	Surface quality	When paraffin and palm oil liquids were milled with 2% nano particle jet MQL, compared with the MQL of paraffin oil and palm oil liquids without nano particles, Ra decreased by 25% and 30%, Rz decreased by 15.4% and 23.7%, respectively.

have studied several methods to improve the performance of a single vegetable oil. However, the complementary effect of the physical and chemical properties of different vegetable oils is also a method for improving the antioxidant and extreme pressure properties of vegetable oils. However, the ratio of vegetable oils with different physical and chemical properties remains unclear.

(2) Both the modification of vegetable oils and the addition of antioxidants can improve the oxidation stability of vegetable oils. The synergistic mechanism of the two is an effective method for further improving the oxidation stability of vegetable oils.

(3) Nanofluids can significantly improve the lubrication and cooling effects of vegetable oil-based cutting fluids, but their physical and chemical properties and multifield mapping laws of process parameters under various working conditions are still unclear.

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Authors' Contributions

CL and BL were responsible for the theoretical guidance; XZ wrote the manuscript; ZZ was responsible for technical support. All authors read and approved the final manuscript.

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Availability of Data and Materials

Not applicable.

Declarations

Ethics Approval and Consent to Participate

We declare that the papers we submitted are my research work under the guidance of the instructor and research results we have obtained. We confirm that this article has not been published previously and is not being submitted for publication elsewhere. We have not considered elsewhere except *Chinese Journal of Mechanical Engineering*. We confirm that this article has had the full consent of all authors. If this article was accepted, we confirm that it will not be published elsewhere in the same form, in English or in any other language, without the written consent of the publisher. The authors declare that they participated in this paper willingly.

Consent for Publication

The authors declare to consent to the publication of this paper.

Competing Interests

The authors declare no competing financial interests.

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