*lubricants*

*Review*

# $MoS_2$ Nanomaterials as Lubricant Additives: A Review

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**Abstract:** Improving the lubricating properties of base oils through additives is a crucial objective of tribological research, as it helps to reduce friction and wear of materials. Molybdenum disulfide ($MoS_2$) is a 2D nanomaterial with excellent tribological properties that is often used as a lubricant additive. Several studies have been conducted on the preparation and utilization of $MoS_2$ and its nanocomposites as lubricant additives. This paper reviews the research progress on $MoS_2$ nanomaterials as lubricant additives. It firstly introduces various synthesis methods of $MoS_2$ nanomaterials while focusing on the preparation of nano-$MoS_2$ as lubricant additives. It then summarizes the dispersion stability of nano-$MoS_2$ in lubricating oils which has been paid extensive attention. Moreover, this paper reviews and discusses the tribological properties of nano-$MoS_2$ and its various composites as lubricant additives. The possible anti-wear and friction reduction mechanisms of nano-$MoS_2$ and its composites are also discussed. Finally, this paper presents the challenges faced by nano-$MoS_2$ additives in the field of lubrication and the prospects for future research in view of previous studies.

**Keywords:** nano-$MoS_2$; dispersion stability; anti-wear; friction reduction; lubrication mechanism



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## 1. Introduction

Friction and wear have always been the top issues in different industrial fields. While modern industry is developing rapidly, global energy consumption also maintains rapid development. In mechanical systems, energy waste due to friction and wear accounts for about 1/3–1/2 [1,2], and friction and wear of machineries and equipment will seriously affect the normal operation and life cycle of its components [3] and may even produce more dangerous consequences under the process of operation [3]. Mechanical systems in industrial production are usually lubricated with lubricants, which can not only significantly reduce friction and wear and improve lubrication efficiency, but also have important significance for energy saving and environmental protection [4]. Currently, lubricants commonly used in various industries and research experiments are composed of base oils and various lubricant additives [5]. Traditional anti-wear and extreme pressure additives such as molybdenum dialkyl dithiocarbamate (MoDTC) and zinc dialkyl dithiophosphate (ZDDP) can greatly reduce friction and wear by generating beneficial products in situ from the reaction between the additive and the metal friction pair surface [6,7]. However, traditional lubrication additives are facing serious challenges with the development of the industry. In order to fulfill the needs of different fields such as ultra-low friction, wear, and extreme conditions, new requirements are placed on lubricants such as resistance to high temperature and pressure [8]. Lubricant additives can give new properties to the lubricant and make up for the shortcomings of the base oil, thus greatly improving the tribological properties of the lubricant.

The utilization of nanomaterials as additives in lubricants presents a promising avenue to enhance the performance of mechanical systems, surpassing that of the traditional lubricant additives. Nanomaterials are tiny particles, with at least one dimension ranging between 1–100 nanometers, which allows them to easily penetrate the contact zone [9]. Due to their high surface activity, they can strongly adsorb onto the friction surface and form a stable protective film that prevents direct contact between solid surfaces, effectively reducing friction. These nanomaterials include metals [10], metal oxides [11], metal sulfides [12], carbon-based materials [13], nitrides [14], and their composites. During friction processes, nanomaterials can be physically or chemically adsorbed onto the contact surfaces of friction pairs, thus improving lubricant tribological properties, a subject that has been extensively researched in the field of tribology [15–17]. Transition metal dichalcogenides (TMDs), due to their structural similarity to graphene, have attracted significant attention from global scholars in recent years for their ability to significantly enhance lubrication and anti-wear properties in lubricants.

The study of using transition metal disulfides as nano-lubricating additives is still under research and development processes. $MoS_2$ is a typical representative of transition metal disulfides and is used as an anti-wear and friction-reducing additive in solid lubricants, lubricants, and greases, or as a lubricating component in coatings [18]. $MoS_2$ is an excellent solid lubricant and has gained the reputation of "king of lubrication" because of its good lubrication performance even under special working conditions like ultra-high temperature and ultra-vacuum. $MoS_2$ also has certain problems, such as humidity and oxygen enrichment that can degrade its performance [19–22]. Low humidity and low oxygen environments need to be constructed for them to perform. However, $MoS_2$ can overcome the effects of moisture and oxygen by developing specific structures or morphologies. Although some progress has been made in improving $MoS_2$'s resistance to moisture and oxidation [20], there is still much work to be undertaken.

$MoS_2$ was initially used in the mid-19th century during the California Gold Rush as a lubricant for horse-drawn carriage bearings. $MoS_2$ as a lubricant has been the subject of a great deal of scientific research, with reviews and books on the subject published as early as the 1960s [23,24]. Although its use as a dry film lubricant was established in the mid-20th century, its effective use in oils took longer to perfect. Research to understand and improve the lubricating properties of $MoS_2$ is still active. Meanwhile, compounding various nanomaterials provides an alternative approach to creating new materials with excellent tribological properties. Recent studies have focused on integrating $MoS_2$ with other nanomaterials (e.g., graphene) into nanocomposites and improving their lubrication properties through controlled doping [25–28].

This paper aims to provide guidance for researching on lubricant additives, with specific focus on $MoS_2$ as a lubricant additive. It reviews the progress of tribological research on $MoS_2$ nanomaterials in recent years and summarizes the current understanding of lubricant additives. This study does not address applications of $MoS_2$ as a solid lubricant or in coatings. Section 2 provides details on the structure and synthesis of $MoS_2$, while Section 3 focuses on methods to improve the dispersion stability of $MoS_2$ in base oils by surface modification and preparation of composites. Section 4 discusses the tribological properties and lubrication mechanisms of $MoS_2$ and its complexes. Finally, Section 5 summarizes the challenges that may arise with the use of $MoS_2$ materials as lubricant additives, as well as new directions for future development.

## 2. Structure and Synthesis

### *2.1. Structure of $MoS_2$*

A single layer of $MoS_2$ is composed of three atomic layers. It consists of a molybdenum layer sandwiched between two sulfur atom layers, forming a sandwich-like structure. $MoS_2$ belongs to the hexagonal or rhombohedral crystal system, with six sulfur atoms distributed around each molybdenum atom and three molybdenum atoms oiled around each sulfur atom, Mo and S are bonded to each other by covalent bonding, whereas the triatomic layer of S-Mo-S is bonded to each other by van der Waals forces, with a layer-to-layer spacing of about 0.615 nm (PDF#37-1492). In addition, due to the difference in the relative positions between Mo and S atoms, three crystal structures are formed as shown in Figure 1 [29]: the 1T (lattice parameters a = 5.60 Å, c = 5.99 Å9), 2H (lattice parameters a = 3.15 Å, c = 12.30 Å), and 3R (lattice parameters a = 3.17 Å, c = 18.38 Å) crystal structures [30]. The 1T-$MoS_2$ structure has an octahedral coordination, which is metallic, and belongs to the substable structure; the 2H-$MoS_2$ crystal form contains two Mo-S units, which belongs to the stable state structure; and the 3R-$MoS_2$ crystal form has one Mo-S unit more than the 2H-$MoS_2$ crystal form which has one more Mo-S unit, i.e., it contains three Mo-S units and belongs to the substable structure. Two of these substrates, 1T-$MoS_2$ and 3R-$MoS_2$, can be transformed into the stable 2H-$MoS_2$ form at high temperatures [31,32]. Based on the fact that $MoS_2$ layers are connected by van der Waals forces, the layers slide very easily, which confers excellent lubrication properties to $MoS_2$.

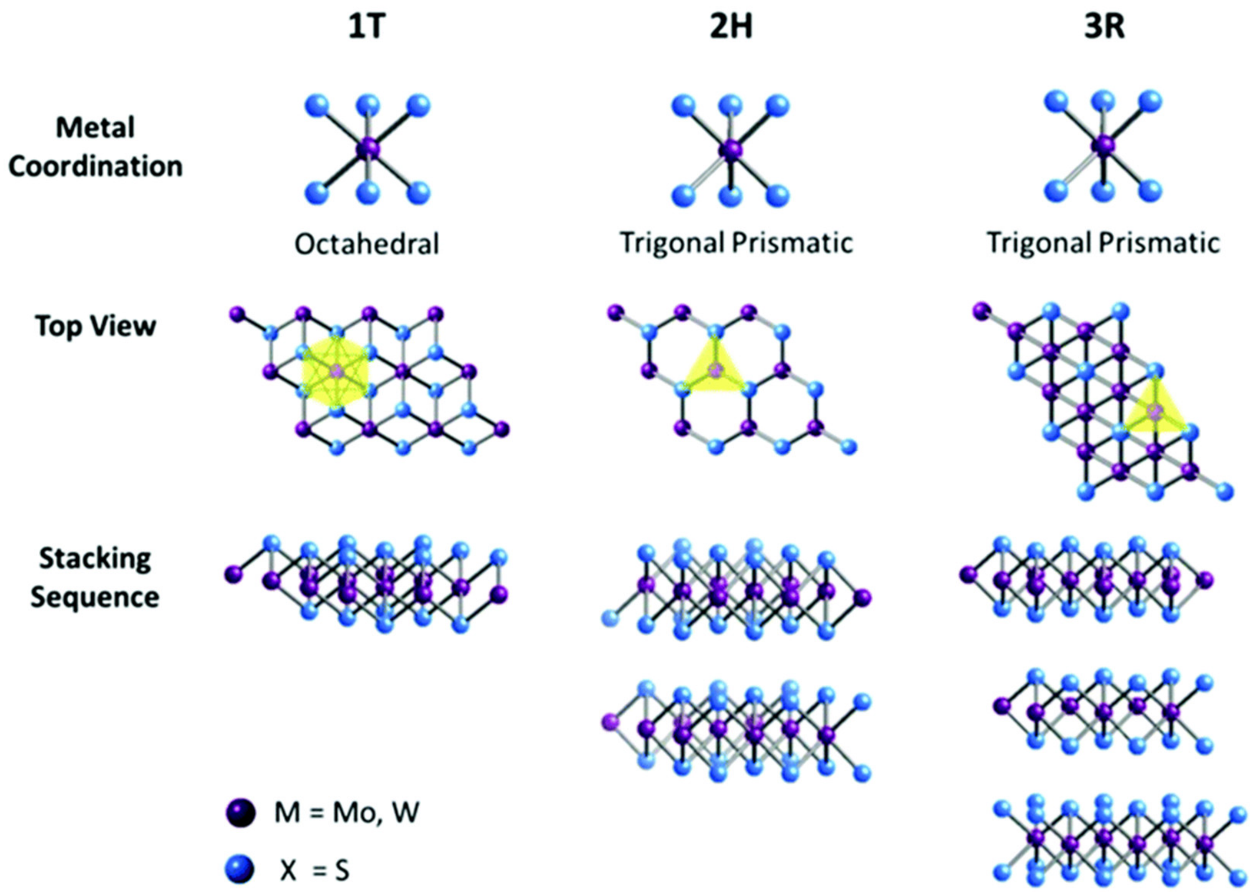


**Figure 1.** 1T, 2H, and 3R phases of $MoS_2$ (Reproduced from Ref. [29] with permission from the Royal Society of Chemistry).

### *2.2. Synthesis Method of Nano-$MoS_2$ Nanomaterials*

$MoS_2$ is a substance with excellent lubricating properties and has gained widespread attention in the field of tribology. The last few years have seen rapid advancements in the preparation technology and processes of $MoS_2$, which have laid a solid foundation for its basic research and application and significantly promoted its development. The synthesis method of $MoS_2$ is the cornerstone for its development and application and holds significant scientific and economic significance. Similar to graphene, the preparation method for two-dimensional layered $MoS_2$ starts with the mechanical stripping method, and several other preparation methods have been developed through continuous research and exploration. The preparation methods of $MoS_2$ include mechanical exfoliation, hydrothermal, solvothermal, liquid-phase precipitation method, and more.

### 2.2.1. Mechanical Exfoliation

The earliest method used to prepare $MoS_2$ was micromechanical exfoliation, i.e., the use of adhesive tape to strip thin sheets of $MoS_2$ from a block of $MoS_2$. This method was first proposed by Frindt et al. [33] in 1965, and several tens of layers of $MoS_2$ were successfully obtained by this method. Although the $MoS_2$ obtained by this method has a high degree of crystallinity, the preparation is inefficient and reproducible, which makes it difficult to realize large-scale applications. For this reason, researchers have explored the ultrasound-assisted exfoliation method. Liu et al. [34] prepared graphene-like $MoS_2$ materials by ultrasound exfoliation with the assistance of 1-Dodecanethiol (Figure 2A). Dual-solvent ultrasonic exfoliation was achieved by introducing chloroform and the highest concentration of graphene-like $MoS_2$ was prepared when the volume ratio of 1-dodecanethiol to trichloromethane was 1:1 and the ultrasonication time was 12 h. Yao et al. [35] used chitosan/acetic acid (CS/HAc) solution to sonicate the native phase $MoS_2$ to obtain oligomerized $MoS_2$, the (002) crystalline surface stacking of the treated $MoS_2$ samples was reduced, and the crystalline phase was transformed from a polycrystalline to a monocrystalline structure.

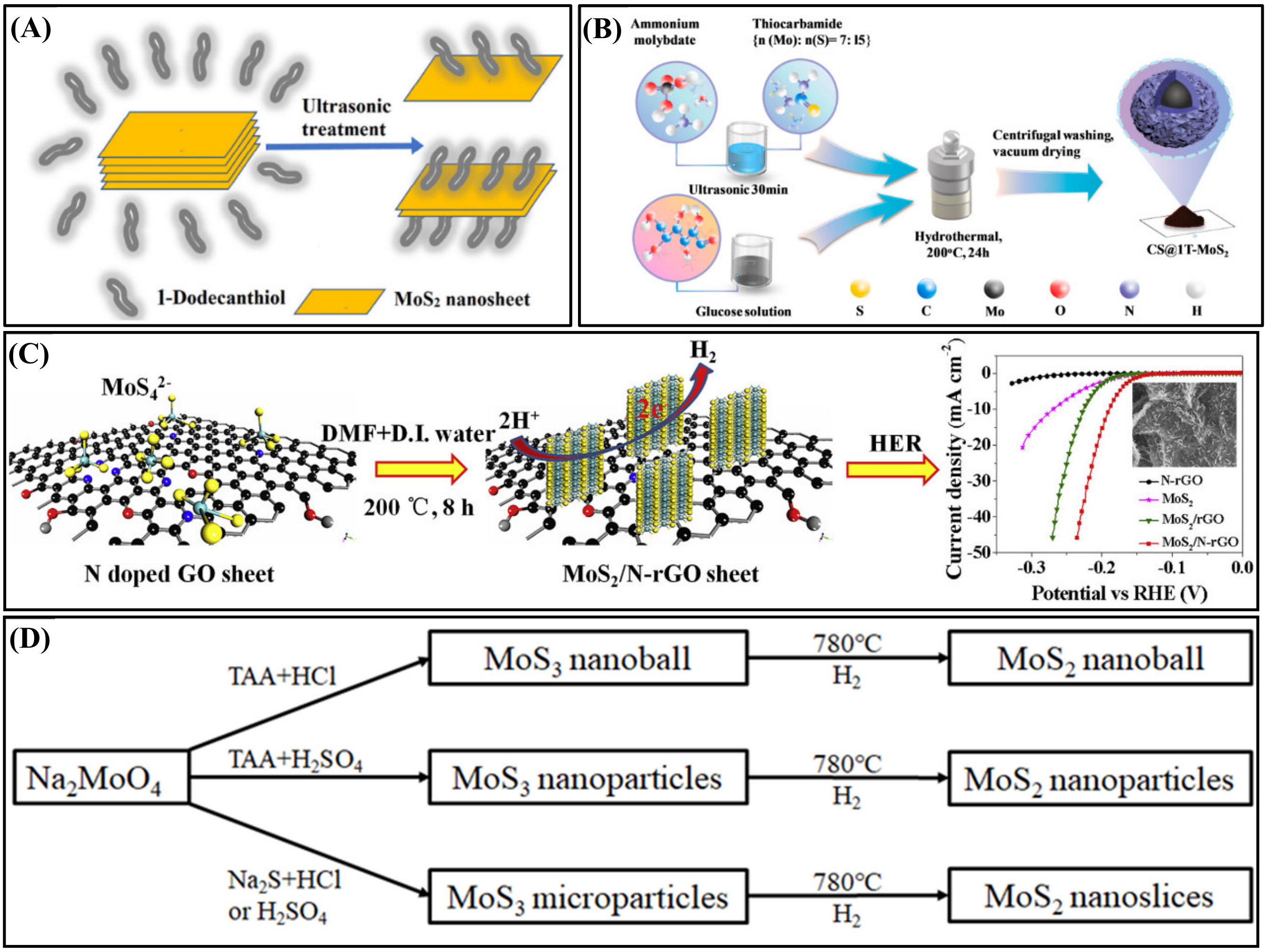


**Figure 2.** Schematic diagram of $MoS_2$ preparation. (**A**) Ultrasonic stripping of $MoS_2$ nanosheets (Reproduced from Ref. [34]), (**B**) Hydrothermal synthesis of CS@1T−$MoS_2$ nanocomposites (Reproduced from Ref. [36]), (**C**) Solvothermal preparation of $MoS_2$/N−rGO nanocomposites (Reproduced from Ref. [37]), and (**D**) Liquid−phase precipitation method for the preparation of $MoS_2$ nanoparticles (Reproduced from Ref. [38]).

#### 2.2.2. Hydrothermal

The hydrothermal is a method to complete the material synthesis and preparation by heating and pressurizing the reaction system (or self-generated steam pressure) in a specially designed closed reactor (e.g., autoclave) with water as the reaction medium to create a relatively high-temperature and high-pressure reaction condition. The advantage of this method is that the synthesized products are characterized by complete grain development, small and uniform particle size distribution, and light particle agglomeration, and it is one of the most commonly used methods for the preparation of two-dimensional materials, including $MoS_2$ and its nanocomposites [39,40]. Tang et al. [41] successfully obtained flower-like microspheres consisting of $MoS_2$ nanosheets by the surfactant-assisted hydrothermal method at 180 °C for 24 h using a certain proportion of $(NH_4)_2MoO_4$, $NH_2OH$-HCl and $CH_4N_2S$ as the initial reactants. Li et al. [42] obtained $MoS_3$ precursors by adding an appropriate amount of [EMIM]Br to an acidic aqueous solution of sodium molybdate and thioacetamide and reacting at 200 °C for 24 h, followed by high-temperature desulfurization to obtain hollow core/shell $MoS_2$ microspheres. Vijaya et al. [43] mixed ammonium hexamolybdate tetrahydrate ($(NH_4)_6Mo_7O_{24}\cdot 4H_2O$) and $H_2NCSNH_2$ with 30 mL of deionized water and hydrothermally reacted at a temperature of 200 °C for 24 h to obtain $MoS_2$ nanosheets. Xie et al. [41] dissolved ammonium molybdate, thiourea, and glucose in deionized water and placed the reaction at 200 °C for 24 h to obtain CS@1T-$MoS_2$ (Figure 2B).

It can be seen that the hydrothermal method usually involves pre-preparing an aqueous solution containing the raw material, placing it in a sealed autoclave reactor reacting it for some time at a temperature of about 200 °C, and then cooling it naturally to room temperature. The precipitate after the reaction is collected for some additional post-treatment (e.g., calcination) to optimize the product. However, the hydrothermal method requires high-temperature and high-pressure reaction conditions, high dependence on equipment, expensive autoclaves for fabrication, safety issues during the reaction process, and the inability to observe the reaction while synthesizing the nanomaterials, which makes it difficult to realistically control the parameters [44], thus limiting the development of the method to some extent.

#### 2.2.3. Solvothermal

Solvothermal is a high-pressure growth method that does not require any catalyst. It is developed from the hydrothermal method, which is similar but differs in using organic solvents or non-aqueous solvents (such as N, N-dimethylformamide (DMF) [37,45], ethylene glycol [46]) instead of water as the reaction medium. This method is used to prepare materials that cannot grow in aqueous solution and are easily oxidized, hydrolyzed, or sensitive to water, such as III-V semiconductor compounds, nitrides, and sulfur compounds.

Li et al. [45] used ammonium tetrathiomolybdate ($(NH_4)_2MoS_4$) as a precursor for Mo and S and reacted it in DMF at 210 °C for 36 h to obtain $MoS_2$ nanosheets. In some cases it is also necessary to use DMF and water as a mixed solvent (Figure 2C). Zhao et al. [37] reported that the proper DMF/water ratio is very important for the construction of $MoS_2$ nanocomposites, because the use of DMF as a solvent does not allow for the self-assembly of the hybrid material into the desired structure, and the use of pure deionized water does not result in homogeneous $MoS_2$ nanoparticles. Li et al. [46] conducted an experiment where $CoC_4H_6O_4\cdot 4H_2O$ and $Na_2MoO_4\cdot 2H_2O$ were dissolved in ethylene glycol and stirred with sublimated sulfur and GO to form a homogeneous solution, which was placed in an autoclave and kept at 200 °C for 24 h to obtain the target product. In addition, various other solvents such as N-methyl pyrrolidone (NMP) [47], ethanol [48,49], polyethylene glycol [50], and isopropanol [51] are favorable for solvothermal methods.

Solvothermal has some limitations. Although they are usually less demanding than hydrothermal, they still require high temperature and pressure conditions in most cases. Additionally, the synthesis process usually lasts longer [52–54] and consumes more energy. Comparing the two methods, the simple hydrothermal method usually has no negative impact on the material properties, whereas solvothermal methods usually have a greater impact on material properties due to differences in solvents. However, the solvothermal method is more likely to result in a better morphology than the hydrothermal method.

#### 2.2.4. Liquid-Phase Precipitation

Liquid-phase precipitation is a process in which a solution containing the desired reactants is generated by controlling the acidity and temperature of the reaction. By adjusting these factors, a large number of particles can be generated, and the use of surfactants prevents agglomeration of the particles, resulting in uniformly dispersed nanoparticles. This method is low-cost, easy to operate, and has a mild reaction process, making it suitable for industrial production.

Hou et al. [55] prepared $MoS_2$ nanoparticles using a simple method. They dissolved $Na_2MoO_4 \cdot H_2O$ and $CH_3CSNH_2$ in deionized water, added a surfactant, and adjusted the pH to 1.0 using $H_2SO_4$. The mixture was stirred for 5 min at 90 °C, resulting in the formation of $MoS_3$. The product was washed, dried, and desulphurized at 800 °C under a hydrogen atmosphere, which led to the formation of $MoS_2$ nanoparticles with a size range of 150–300 nm. Hu et al. [56,57] utilized thioacetamide ($CH_3CSNH_2$, TAA) as the sulfur source and dissolved it in distilled water along with $Na_2MoO_4 \cdot 2H_2O$. The resulting solution was heated to 82 °C for 5 min, then HCl or $H_2SO_4$ was rapidly poured into the reaction system and the reaction was continued for 5 min. The product was then washed and dried to give the desired $MoS_3$ precursor. Hu and colleagues [58,59] utilized $Na_2S \cdot 9H_2O$ as a source of sulfur, which was then dissolved in deionized water along with $Na_2MoO_4 \cdot 2H_2O$. While stirring the mixture at room temperature, they added HCl or $H_2SO_4$ dropwise to the reaction system. The obtained precipitate was washed and dried to obtain the desired $MoS_3$ precursor. The $MoS_3$ precursors obtained under both conditions were subjected to calcination under a high-purity hydrogen atmosphere at a selected temperature for 50 min to obtain $MoS_2$ nanoparticles. It was also suggested that the sulfur source has a significant effect on the morphology and size of $MoS_3$ precursors [38], and the $MoS_3$ precursors prepared by the TAA sulfur source consisted of hollow nanorods and solid nanoparticles, while those prepared by the $Na_2S$ sulfur source consisted of micrometer-sized particles (Figure 2D). The $MoS_3$ hollow nanorods and solid nanoparticles obtained by calcination at 780 °C under $H_2$ yielded ~150 nm $MoS_2$ hollow nanospheres and ~40 nm solid nanoparticles, respectively, and the $MoS_3$ micrometer-sized particles prepared by calcination at 780 °C produced ~30 nm $MoS_2$ nanosheets.

Xu et al. [60] were able to successfully synthesize spherical nano-$MoS_2$ particles using an improved method based on the original synthesis method in the literature [55]. The improvements made included optimizing the ratios of reactants, lowering the calcination temperature to 480 °C, and substituting $H_2$ with $N_2$. This resulted in the formation of spherical nano-$MoS_2$ particles with a solid structure, as opposed to hollow ones.

## 3. Stability of $MoS_2$ Nanomaterial Dispersion

In recent years, $MoS_2$ nanoparticles have received increasing attention as lubricant additives for their excellent tribological properties. In practice, poor dispersion of the additive causes agglomeration and precipitation, which can adversely affect the friction system. However, due to the small particle size of $MoS_2$ nanoparticles with high surface energy, such nanoparticles have a strong tendency to agglomerate and are prone to aggregate and form agglomerates with larger sizes, thus making it difficult to form a stable lubricant suspension, which has become a challenging problem for the application of nano-$MoS_2$ as a lubricant additive [61–63], seriously hindering the application of nano-$MoS_2$ materials in practice. Therefore, optimizing the dispersion stability of $MoS_2$ in oil solutions and slowing the tendency of particle agglomeration in suspensions can be regarded as prerequisites for studying other properties. Currently, methods for improving the dispersion stability of $MoS_2$ nanomaterials in base oils are divided into two main categories: physical and chemical methods.

### 3.1. Physical Methods

Physical dispersion of nanomaterials in the base lubrication medium can be achieved by mechanical techniques such as mechanical agitation, ultrasonic treatment [64–67], ball milling [68,69], and high-pressure homogenization [70], with ultrasonic treatment being one of the most commonly used physical methods.

Ultrasonic dispersion is a relatively mature technology, which is used in textile, printing and dyeing, chemical, biological, pharmaceutical, and many other industries and fields. The mechanism of ultrasonic dispersion occurs when ultrasonic waves propagate in a dispersing medium, wherein the pressure by the speed of sound oscillatory changes, which produces a series of rapid compressions of the liquid so that the suspended particles are instantly broken and dispersed. However, overheated ultrasonic mixing should also be avoided, as the chance of particle collision increases with the increase in mechanical energy in the thermal phase, leading instead to further agglomeration. It was mentioned earlier that ultrasonic treatment helps in stripping the bulk material into nanoscale products without any other reactants [33,34] and also helps in homogeneous dispersion of $MoS_2$ material in solution or solvent [71,72].

Alazemi et al. [73] dispersed CS-$MoS_2$ particles in reference oil at different weight percent concentrations using a 750 W piezoelectric ultrasonic probe at a power amplitude of 20% for 120 s. The particles were then dispersed in a reference oil. Visual inspection of the results showed that using the ultrasound-mediated technique, CS-$MoS_2$ particles could remain suspended in the reference oil for a long period (about 2 weeks) even without the addition of surfactant. Xu et al. [74] dispersed graphene and $MoS_2$ in esterified bio-oil (EBO) with different mass ratios and treated them with ultrasonic oscillation at room temperature for 20 min. The dispersion stability of EBO containing 0.5 wt% of the copolymer was better and remained stable for at least 240 h after ultrasonic dispersion. Usually, the combination of the two methods gives better results. Wang et al. [75] homogeneously dispersed modified reduced graphene oxide and modified $MoS_2$ in base oil (PAO6) by ultrasonic treatment and microwave-assisted ball milling (Figure 3).

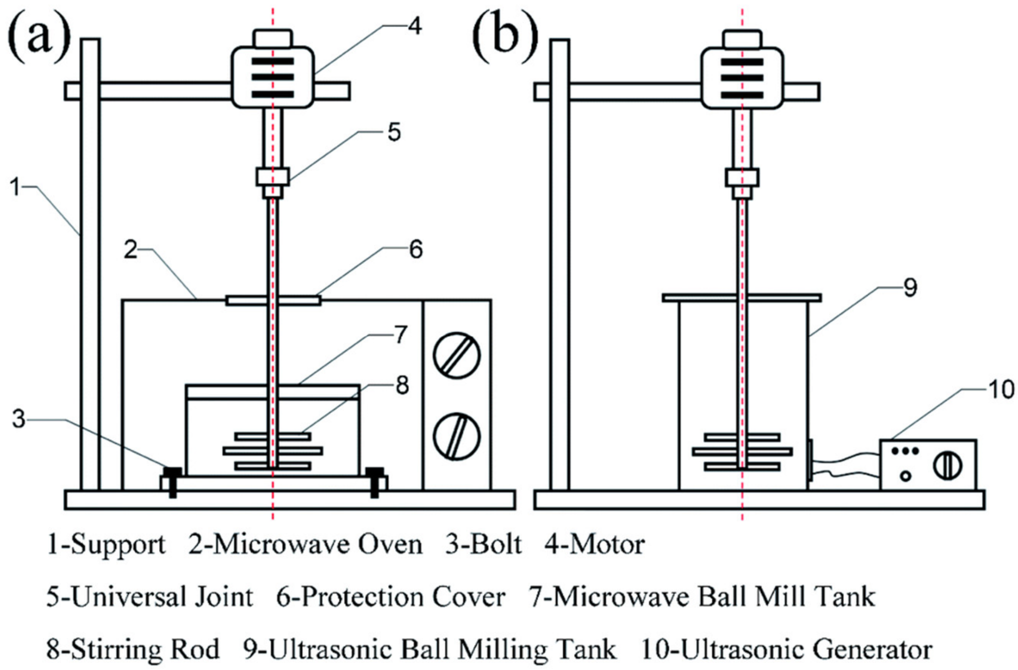


**Figure 3.** (**a**) Microwave ball milling device and (**b**) ultrasonic ball milling device (Reproduced from Ref. [75]).

Ultrasonic dispersion technology is a simple and efficient dispersion method with easy operation, fast speed, and no pollution, but insufficient ultrasonic time cannot completely break the inter-particle agglomeration, while too long ultrasonic time will produce re-agglomeration among nanoparticles, thus weakening the dispersion performance of the solution, and the high cost of ultrasonic equipment is prone to cause a drastic increase in temperature in the process of use, which in turn leads to the dispersion of the particles for further agglomeration. In addition, the difficulty of acoustic energy transfer and heat dissipation in large-scale operations affects its industrial application.

### *3.2. Chemical Methods*

Graphene-like topic nanomaterials cannot be dissolved in most solvents due to the strong $\pi$-$\pi$ bonds, and neither direct dispersion nor simple ultrasonic dispersion can make $MoS_2$ stably dispersed in lubricating oils. Currently, surface modification or composite materials with other materials are commonly used to improve the dispersion of $MoS_2$ in lubricating oils.

Adding surfactants as dispersants is a relatively easy method. Organic dispersants are adsorbed by these particles suitably to create a state of repulsion between the particles, thus counteracting the natural attraction between the particles [76]. There are many common dispersants such as Span 80, tween 80, oleic acid (OA), sodium dodecyl sulfate sodium dodecyl benzene sulfonate, etc. Hou et al. [77] investigated the effect of different dispersants (OA and Span 80) on the dispersion stability of $MoS_2$ nanomaterials in PAO6 to provide a theoretical basis for the stability of the $MoS_2$ nano-lubricant. Oleic acid and Span 80 successfully modified the $MoS_2$ nanomaterials, and the $MoS_2$ nano-lubricant was able to remain somewhat stable at room temperature for a period of 7 days. The paper also points out that concentration, sonication time, and morphology (spherical and sheet) all have a certain effect on the dispersion stability of $MoS_2$ in base oils as well. Gulzar et al. [78] examined the effect of $MoS_2$ particles on the dispersion stability and anti-wear properties of PAO10 and bio-based base oils and found that poorly dispersed nanoparticles led to a partial loss of wear protection. Surfactants contributed to the suspension of $MoS_2$ particles in both base oils and the corresponding suspensions were found to give more favorable anti-wear. Physical adsorption of surfactants on the particle surface reduces the surface tension of the particles, thus preventing the formation of aggregates [79]. In addition, surfactant-coated additives can overcome the van der Waals gravity between them through electrostatic/spatial repulsion [80]. However, surfactant molecules are subject to shedding, decomposition, and degradation during friction due to shear stresses and localized high temperatures at the contact interface, resulting in unstable friction properties [81].

Surface modification is a commonly used method for $MoS_2$ surface modification. Through surface grafting modification, specific functional groups in surfactants are grafted on the surface of nanoparticles to improve their oil solubility, so that the nanoparticles can be stably dispersed in lubricating oils, which has become an important direction in the research on the practicalization of nano-lubricant additives [82–84]. OA is an unsaturated fatty acid, which is often used as a chemical modifier for $MoS_2$ [85,86]. Wu et al. [85] successfully obtained OA-$MoS_2$ by one-step modification of homemade five-layer-thickness $MoS_2$ nanosheets with oleic acid (Figure 4A). The OA molecules chemically confined the $MoS_2$ nanosheets through C-S bonding, which significantly improved the dispersion and stability of the $MoS_2$ nanosheets in the base oil. The results of four-sphere machine tests showed that the addition of OA-$MoS_2$ nanosheets significantly improved the friction reduction, anti-wear, and extreme pressure capabilities of the base oil. Kumari et al. [87] chemically functionalized $MoS_2$ using octadecanethiol (ODT). Structural defects and empty sulfur sites on the Mo atoms of $MoS_2$ nanosheets were the targets for the grafting of ODT via Mo-S coordination bonds (Figure 4B). Van der Waals interactions between the octadecyl chains of $MoS_2$-ODT and the oleate chains of polyol ester lubricants facilitated $MoS_2$-ODT nanosheet dispersion (Figure 4C).

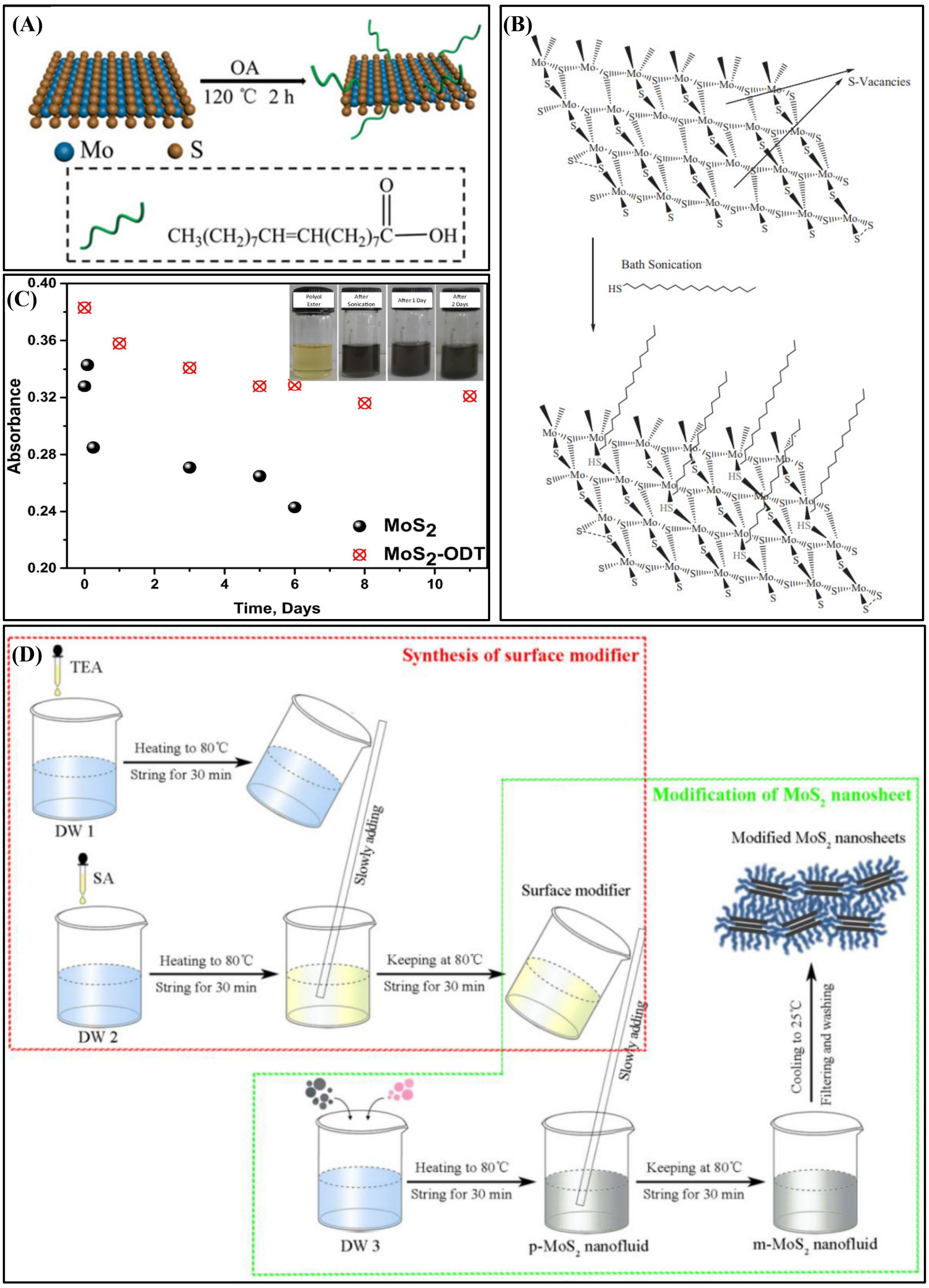


**Figure 4.** Chemical modification and dispersion stability of $MoS_2$ (**A**) Synthesis route of OA-$MoS_2$ nanosheets (Reproduced from Ref. [85]), (**B**) Schematic diagram of grafting ODT molecules onto sulfur vacancies in $MoS_2$ nanosheets (Reproduced from Ref. [87]), (**C**) Dispersion stability of pristine $MoS_2$ and $MoS_2$-ODT nanosheets in polyol ester lubricant base oils (Reproduced from Ref. [87]), (**D**) Surface modifiers for the synthesis and flow chart of the modification of $MoS_2$ nanosheets (Reproduced from Ref. [88]).

Meng et al. [88], in order to mitigate the agglomeration and deposition of $MoS_2$ nanofluid, conducted an experiment wherein surface modification of $MoS_2$ nanosheets was carried out by using homemade surface modifiers based on triethanolamine (TEA) and stearic acid (SA), and modified $MoS_2$ nanosheets with better dispersion stability were obtained (Figure 4D). The effects of the surface modification on the tribological properties of the modified $MoS_2$ nanofluids were investigated using a four-ball friction and wear tester and molecular dynamics simulation. The diffusion rate, interlayer spacing, interlayer adsorption energy, and shear stress of $MoS_2$ nanosheets before and after modification were calculated. The interlayer spacing of the modified $MoS_2$ nanosheets increased due to the interaction between the $MoS_2$ nanosheets and the surface modifier. Especially at atmospheric pressure, the interlayer adsorption energy and interlayer shear stress were decreased. The modified $MoS_2$ nanosheets not only have better dispersion stability but also have excellent tribological properties.

Due to the strong attraction between metal atoms and polar functional groups (-OH, -O-, -$NH_2$, etc.) in $MoS_2$, it helps to graft surfactant molecules containing such groups onto the nanoparticles [89–91], which not only reduces the agglomeration of nanoparticles but also avoids the separation of the nanoparticles and grafted functional groups due to frictional stress, which effectively improves the dispersive stabilization and lubrication properties of the nanoparticles. With the deepening of the research, a single decentralized method can no longer meet the demand, and the combination of methods has become the choice of many researchers.

The composite material realizes the result of "1 + 1 > 2" for two or more materials. The composite of $MoS_2$ with a dibasic material to improve its dispersion in the base oil and then improve its lubrication performance is also one of the research hotspots of $MoS_2$ in recent years.

The hexagonal boron nitride (*h*-BN) nanosheets prepared by Kumari et al. [92] via alkali-assisted hydrothermal stripping were used for the growth of a $MoS_2$ sheet by chemical reduction in the presence of cetyltrimethylammonium bromide (CTAB). The CTA molecules on the surface of the composite nanomaterials avoided their re-stacking through the spatial site resistance of the long alkyl chains, and furthermore, the alkyl chains of the CTA molecules in the *h*-BN-$MoS_2$-CTAB had van der Waals interactions with the hydrocarbons in the 5W30 engine oil, resulting in the formation of a heterogeneous structure of *h*-BN-$MoS_2$-CTAB. Thus, the dispersion of *h*-BN-$MoS_2$-CTAB in 5W30 engine oil was stable for one month (Figure 5A). Farsadi et al. [93] complexed functionalized reduced graphene oxide (FrGO) with $MoS_2$ to improve its dispersion stability in base oils. $MoS_2$-loaded FrGO was better dispersed compared to other materials and remained uniformly distributed over a month. Guan et al. [94] synthesized magnesium silicate hydroxide (MSH)-enhanced $MoS_2$ hybrid nanomaterials using a hydrothermal method, and MSH-$MoS_2$ showed good dispersion stability due to its abundant active anchor sites. Chouhan et al. [95] synthesized ZnO-modified reduced graphene oxide/$MoS_2$ (Gr-MS-Zn) nanosheets and the obtained Gr-MS-Zn was formed by three to seven molecular layers of $MoS_2$ nanosheets fully distributed on the graphene backbone through weak interfacial interactions. The curved and bent structure of $MoS_2$ nanosheets grown on graphene sheets subsidized the cohesive interactions, and these $MoS_2$ nanosheets wrinkled on graphene nanosheets to promote dispersion by minimizing the contact or adhesion interactions between the nanocomposites, thus reducing the interactions between the Gr-MS-Zn components, which was important for inhibiting the formation of agglomerates and maintaining their good dispersion in the fully formulated 10W40 engine oil, which is in turn important to obtain a good dispersion of Gr-MS-Zn in the final stable dispersion of the fully formulated engine oil (Figure 5B).

Therefore, although physical methods are one of the simplest and most economical ways to disperse $MoS_2$ into lubricants, the nanoparticles are prone to re-agglomerate due to degradation problems under frictional conditions. Surfactant-modified $MoS_2$ has better dispersion properties, but the presence of other atoms or functional groups may lead to significant degradation of its intrinsic properties. The composites obtained by compositing with other two-dimensional nanomaterials not only obtain good stability of the products, but also can choose the composite two-dimensional materials according to the need for their own defects to make up for the corresponding defects, and at the same time, the composite nanoparticles are able to realize self-dispersion in lubricating oils, which is a very promising method.

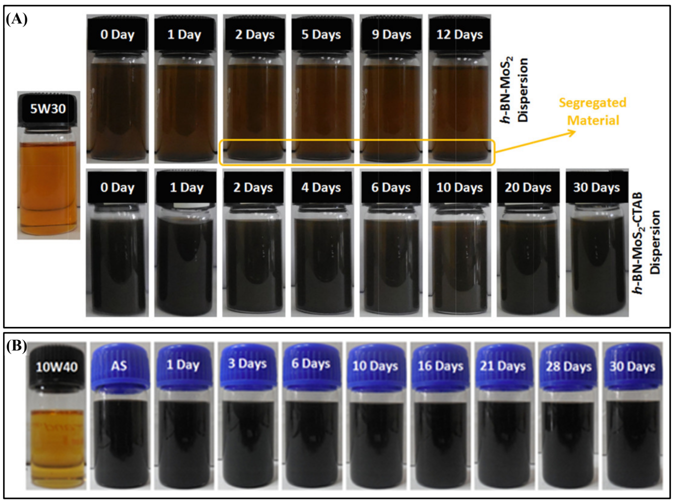


**Figure 5.** (**A**) Dispersions of 5W30 engine oil, h-BN-$MoS_2$, and h-BN-$MoS_2$-CTAB in 5W30 engine oil (Reproduced from Ref. [92]), (**B**) Dispersion of Gr-MS-Zn in 10W40 engine oil with different resting times (Reproduced from Ref. [95]).

## 4. Tribological Behavior and Lubrication Mechanism of $MoS_2$ Nanomaterials

$MoS_2$ has excellent mechanical and physical properties, remarkable chemical stability, and a unique layered structure, showing its great potential for application as a lubricant additive [96–98]. $MoS_2$ nanoparticles and their composites were prepared and dispersed into lubricating oils, and their tribological behavior was investigated on a friction tester. In the course of this study, the lubrication mechanism was illustrated by a number of modern surface analysis tools.

### *4.1. Tribological Properties of Nano-$MoS_2$*

It is widely recognized that the tribological properties of 2D nanomaterials are closely related to their size and morphology [99–102]. In recent years, with the continuous development of the synthesis process, researchers have prepared $MoS_2$ particles with various morphologies, such as nanoflower [103], nanospherical [104], hollow core-shell [105], nanorod [106], and nanowire [107].

Hu et al. [99] investigated the effect of nanospheres, nanosheets, and bulk 2H-$MoS_2$ additives on the tribological properties of liquid paraffin (LP) using a four-ball friction and wear tester. The results showed that all the $MoS_2$ additives used could improve the tribological properties of LP, and the lubrication effect of nano-$MoS_2$ particles in LP was better than that of micrometer $MoS_2$ particles. The LP containing nanospheres had the best friction reduction and anti-wear properties when the $MoS_2$ content was 1.5 wt%. This was attributed to the chemical stability of the layer-closed spherical structure of the nanospheres. This study further deepens the understanding of the relationship between the tribological properties and morphology of $MoS_2$. Tontini et al. [103] synthesized nanoflower-like structured $MoS_2$ particles with an average diameter of 250 nm and evaluated their suitability as lubricant additives. In order to ensure good stability of the nano-oils, the particles were dehydrated and lipophilized by solvent heat treatment with anhydrous ethanol. Tribological characterization was carried out by using a pin-disc tribometer with a reciprocating motion under impregnation. The results showed that the coefficient of friction of polyolester nano oil, on the other hand, decreased by 86%. Luo et al. [104] developed a new green laser-assisted solution growth of ideally spherical fullerene-like $MoS_2$, where the extreme non-equilibrium conditions generated by laser irradiation can reconfigure $MoS_2$ nanosheets into perfect solid nanorods with layer-closed structures to release the high surface tension energy of the nanosheets. Such $MoS_2$ nanospheres are effective as additives to paraffinic fluids to reduce the coefficient of friction (~47% reduction) and enhance the extreme pressure performance (>2.24 GPa). This excellent lubrication performance may be attributed to the molecular bearing-like rolling effect of the fullerene-like $MoS_2$ nanospheres and the formation of friction film between mechanical contact surfaces (Figure 6A). This study is important for the advancement of nano microspheres as additives for internal combustion engine oils in preserving mechanical energy and reducing mechanical failures due to wear. Xu et al. [105] prepared hollow core-shell $MoS_2$ nanoparticles with flower-like surfaces using a two-step solvent-thermal method (Figure 6B), and the friction and wear performance of the prepared hollow core-shell $MoS_2$ nanoparticles in oil was investigated by a ball-and-disk friction and wear tester. The results show that the prepared $MoS_2$ nanoparticles can significantly improve the friction reduction and anti-wear performance of lubricating oils. The friction coefficient was reduced by 43.80% and the wear was reduced by eight times after the addition of hollow core $MoS_2$ to the lubricating oil.

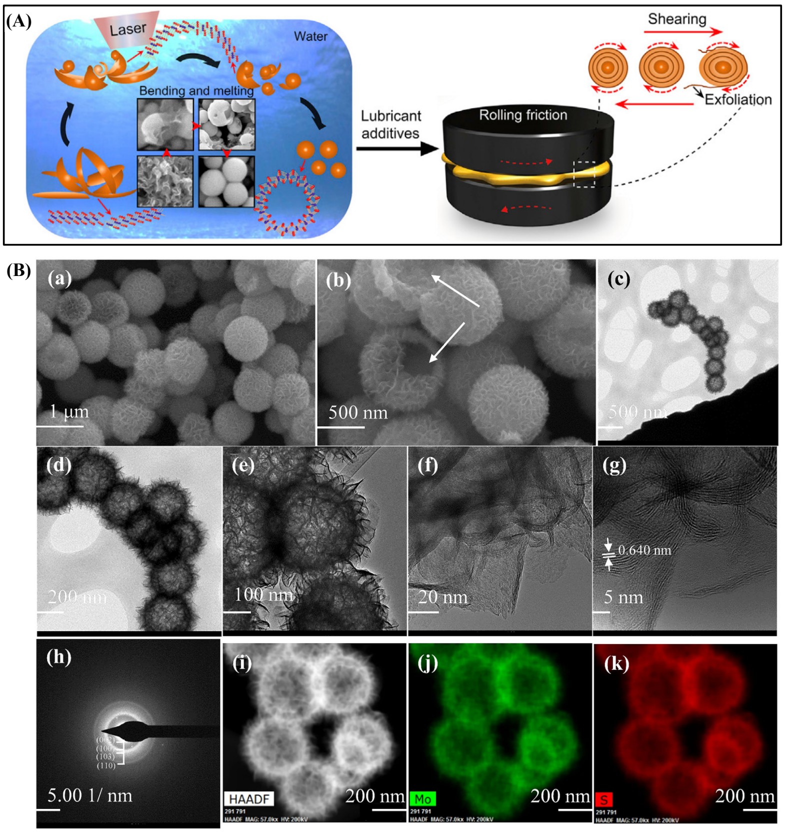


**Figure 6.** (**A**) Schematic of the growth of fullerene-like $MoS_2$ nanospheres and their lubrication mechanism as lubricant additives: Under laser irradiation-induced instantaneous ultrahigh temperature and ultrahigh pressure, MoS2 nanoflakes will bend and melt in water at ambient conditions. The following quenching process of surrounding water solidifies the liquid MoS2 droplet into ideally fullerene-like nanospheres. (Reproduced from Ref. [104]), (**B**) SEM and TEM images of hollow core-shell $MoS_2$ nanoparticles: (**a**,**b**) SEM and (**c**–**k**) TEM images of the hollow core-shell $MoS_2$ nanoparticles. Moreover, the broken hole in Figure 2B (directed by arrows) illustrates a hollow structure (Reproduced from Ref. [105]).

### *4.2. Tribological Properties of $MoS_2$ Composites*

When single $MoS_2$ particles are used as lubrication additives, the $MoS_2$-based friction film adsorbed on the contact surface of the friction pair due to the experimental process is easily destroyed under certain extreme friction conditions and easily removed from the friction interface. A feasible way to improve the tribological performance of pure $MoS_2$ additives is to develop $MoS_2$ nanocomposites, which, as mentioned in the previous section, can not only enhance the dispersion stability of the material in the base oil but also realize the synergistic enhancement effect during the friction process.

Carbon-based solid lubrication materials have been one of the focuses of research in the field of new materials for friction reduction and anti-wear, with broad application value and prospects. Scholars at home and abroad have shown strong interest in the research of carbon materials and $MoS_2$ composites, especially graphene. Xu et al. [108] outlined the development of graphene/$MoS_2$ nanocomposites in recent years, and discussed the synthesis method, dispersion behavior, tribological properties, and lubrication mechanism of the composites, and put forward the challenges that graphene/$MoS_2$ nanocomposites will face in the field of tribology. It is also pointed out that modified graphene/$MoS_2$ nanocomposites have good prospects in the field of tribology. Hu et al. [109] prepared rice husk charcoal/$MoS_2$ (RHC/$MoS_2$) nanoparticles by the precipitation method and modified the synthesized products with surface modifiers. The tribological properties of the nanoparticles in polyethylene glycol (PEG) before and after modification were tested by a ball-and-disc friction and wear tester. When the experimental tests were conducted at 80 N and 300 rpm for 30 min, the nanoparticle additions before and after modification were 0.5 wt%, and the wear rates were reduced by 88.6% and 97.8%, respectively, compared with pure PEG. The modified RHC/$MoS_2$ particles are more effective in improving the anti-wear and friction reduction performance of PEG. Gong et al. [110] synthesized $MoS_2$ nanoparticles grown on three carbon materials, i.e., carbon nanotubes ($MoS_2$@CNT), graphene ($MoS_2$@Gr), and fullerene C60 ($MoS_2$@C60), using the solvothermal method. Incorporation of the three composites as lubricant additives into polyalkylene glycol (PAG) base oils showed significantly improved friction reduction and anti-wear behavior compared to PAG containing CNT, Gr, C60, and $MoS_2$ as well as pure PAG (Figure 7). Xie et al. [41] prepared a novel carbon sphere (CS)@metallographic $MoS_2$ (1 T-$MoS_2$) hybrid using a one-step hydrothermal method. They investigated the effect of this complex on the friction and wear performance of PEG 200 in the MM-W1A friction and wear tester to investigate the effect of the complex on the tribological properties of PEG 200. When 0.3125 wt% CS@ 1T-$MoS_2$ was added, the coefficient of friction was reduced by 43% compared to pure PEG 200. The wear diameter was significantly reduced from 1860 μm in pure base oil to 880 μm.

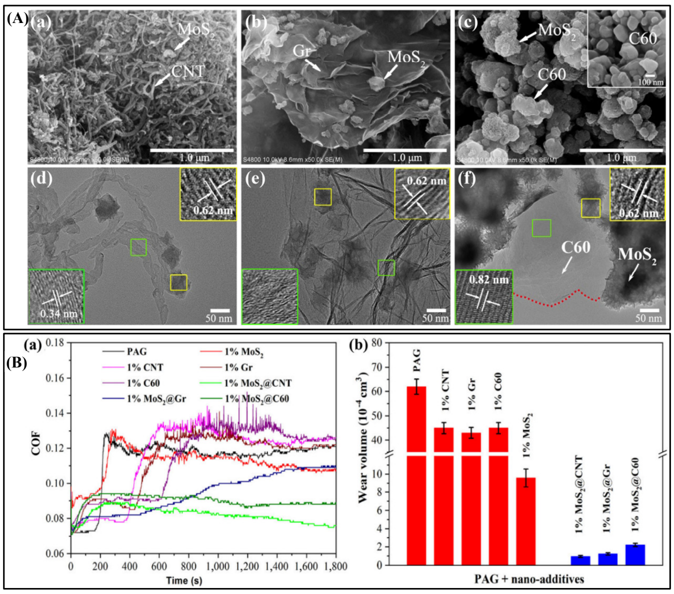


**Figure 7.** (**A**) Electron microscopy characterizations of $MoS_2$@CNMs. FE-SEM images of (**a**) $MoS_2$@CNT, (**b**) $MoS_2$@Gr, and (**c**) $MoS_2$@C60. Inset in (**c**) is the SEM image of pure C60. TEM images of (**d**) $MoS_2$@CNT, (**e**) $MoS_2$@Gr, and (**f**) $MoS_2$@C60; (**B**) (**a**) COF and (**b**) wear volumes of steel discs lubricated by PAG base oil and PAG addicted with 1 wt% CNT, 1 wt% Gr, 1 wt% C60, 1 wt% $MoS_2$, 1 wt% $MoS_2$@CNT, 1 wt% $MoS_2$@Gr, and 1 wt% $MoS_2$@C60 at 100 °C (SRV conditions: load, 100 N; stroke, 1 mm; frequency, 25 Hz; duration, 30 min) (Reproduced from Ref. [110]).

Combinations with other nanomaterials are still remarkable, such as *h*-BN [92,111], graphitic phase carbon nitride (g-$C_3N_4$) [112], and MSH [94,113]. Kumari et al. [92] used the design idea of generating ultra-low friction at the interface of two asymmetrically stacked 2D/2D heterostructures with different lattices to prepare *h*-BN-$MoS_2$-CTAB by alkali-assisted hydrothermal stripping. *h*-BN-$MoS_2$-CTAB, which was added to the engine oil (5W30) as a 30 ppm compound, resulted in a reduction of friction and wear of a steel friction pair by 44% and 96%, respectively (Figure 8A). The 2D/2D heterostructure in this study could be a revolutionary material for the development of a new generation of lubricants. Min et al. [112] successfully synthesized covalently bonded g-$C_3N_4$/$MoS_2$ nanocomposites using a one-step hydrothermal method. g-$C_3N_4$/$MoS_2$ nanocomposites showed an average coefficient of friction and wear rate of 27.86% and 70.87% lower than that of the pure oil at a concentration of 0.20 wt%. Guan et al. [94] synthesized MSH-reinforced $MoS_2$ hybrid nanomaterials into PAO by a hydrothermal method at 220 °C. Tribological properties were tested by a four-ball machine, and the anti-friction performance, wear resistance, and final non-seize load of the friction pair under this lubricant were increased by 44.8%, 41.2%, and 116.6%, respectively. Guan et al. [113] also evaluated the tribological properties and wear surface repair performance of MSH-$MoS_2$ nanocomposites as lubricant additives in PAO by a reciprocating ball-and-disk tester. The results of the tribological tests showed that the anti-friction performance and wear resistance of the friction pair were improved by 27.7% and 37.4%, respectively, with the addition of 1.0 wt% MSH-$MoS_2$ in PAO (Figure 8C,D). What is more, the wear of the friction disk could be repaired by 33.2% after 5 h of sliding under the lubrication of PAO and MSH-$MoS_2$.

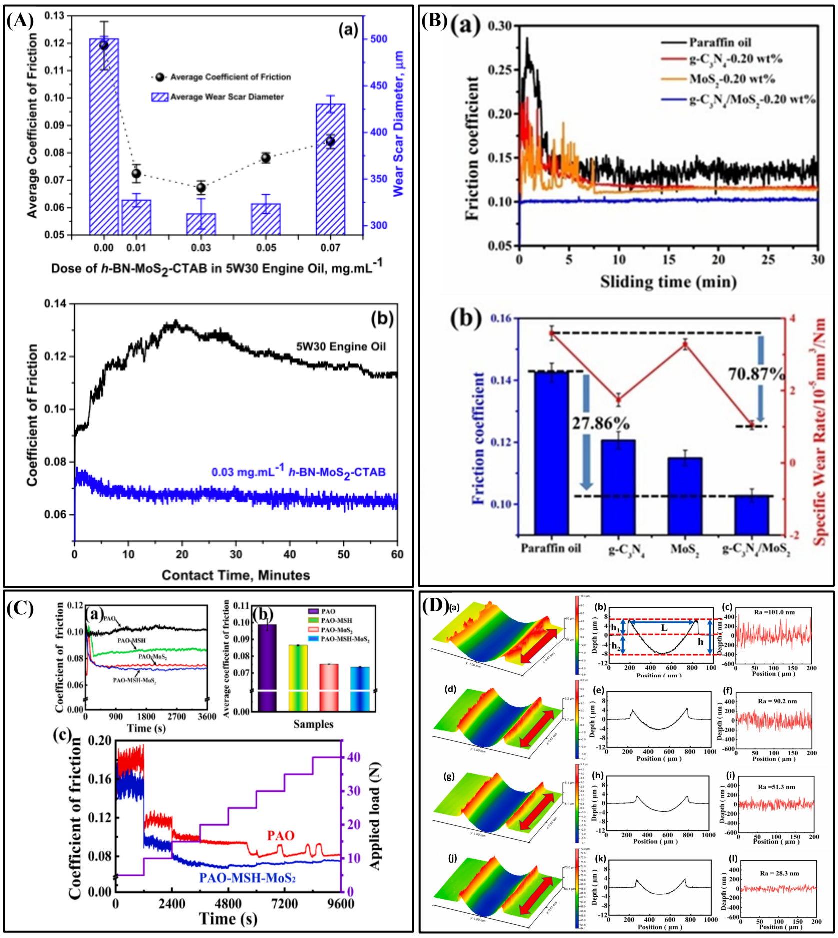


**Figure 8.** (**A**) (**a**) Average friction coefficient and WSD for steel tribe-pair using 5W30 fully formulated engine oil and its blend with variable doses of the *h*-BN-$MoS_2$-CTAB heterostructure, (**b**) Friction profiles

of steel tribe-pair using 5W30 fully formulated engine oil and its blend with *h*-BN-$MoS_2$-CTAB (Reproduced from Ref. [92]). (**B**) (**a**) The curves in regard to friction coefficient of oil with g-$C_3N_4$ nanosheets, spherical $MoS_2$, and g-$C_3N_4$/$MoS_2$ nanocomposites at 0.20% concentration or without additive; (**b**) the parallel average friction coefficient and wear rate of steel sheets after being lubricated by these samples in sequence (Reproduced from Ref. [112]). (**C**) (**a**) COF curves as a function of sliding time, (**b**) average values for steel/steel under the lubrication of four kinds of oil samples, and (**c**) COF curves with the lubrication of PAO and PAO-MSH-$MoS_2$ under different loads. (**D**) 3D topographies, wear scars profiles perpendicular to sliding directions, and surface height profiles along with the sliding directions in GCr15 discs under the lubrication of PAO (**a**–**c**), PAO-MSH (**d**–**f**), PAO-$MoS_2$ (**g**–**i**), and PAO-MSH-$MoS_2$ (**j**–**l**) (Reproduced from Ref. [113]).

### *4.3. Lubrication Mechanism*

Understanding the lubrication mechanism of $MoS_2$ additives is crucial for enhancing the preparation of these additives and for their application in the field of lubrication. There has been a considerable amount of research conducted by scholars from all over the world on the lubrication mechanism of $MoS_2$ particles, but a unified conclusion has not yet been reached. Based on the previous studies of the tribological properties of $MoS_2$ additives, the most widely accepted lubrication mechanism can be summarized into four aspects: rolling mechanism, shear slip, formation of friction film, and synergistic lubrication.

#### 4.3.1. Rolling Mechanism

According to researchers, spherical nanoparticles can roll between two sliding surfaces and help reduce friction and wear. Stabilized spherical nanoparticles can also improve extreme pressure performance and lubricant load-carrying capacity [114–116]. The rolling effect of spherical nanoparticles depends on the thickness of the film. When the nanoparticle diameter is close to the thickness of the membrane, the shape of the nanoparticles will remain unchanged and the ball-bearing mechanism will dominate [117,118].

When the nanoparticles are spherical in shape, they can act as a kind of ball-bearing during friction [119–121], thus improving the lubrication performance. Alazemi et al. [73] synthesized CS-$MoS_2$ particles and added them to engine oil (SAE 5W30) and studied the tribological properties of the lubricant blend using a friction and wear meter. When 1 wt % CS-$MoS_2$ particles were kindly added to the engine, there was a significant reduction in friction and wear (15–35%) compared to the original reference oil at various disk speeds. Raman spectroscopic investigations of wear scars after tribological tests showed that the CS-$MoS_2$ particles were highly chemically stable. The improved tribological performance of the CS-$MoS_2$ and engine oil blend lubricant was attributed to the fact that the CS-$MoS_2$ particles prevented direct contact between the sliding surfaces and acted as particulate ball bearings on the nanoscale. Meng et al. [122] prepared $MoS_2$ quantum dots stably dispersed in paraffin oil, and the spherical $MoS_2$ quantum dots have a proper ball-bearing lubrication effect during friction. Luo et al. [123] prepared a lamellar composite structure consisting of ultra-smooth $MoS_2$ submicroscopic spheres embedded in a multilayer graphene. The ultra-smooth $MoS_2$ spheres within this layered structure can significantly reduce friction by transforming sliding friction into rolling friction under the strong shear force generated by the moving contact surface.

#### 4.3.2. Shear Slip

$MoS_2$ has good lubricating properties due to its unique crystal structure. Within each molecular layer of $MoS_2$, sulfur atoms are strongly bonded to molybdenum atoms through covalent bonds, while sulfur atoms between the layers are connected to molybdenum atoms through weak van der Waals forces. As a result, a low-shear plane is formed. When a small amount of shear force is applied between the molecules, the molecular layer breaks easily and a slip plane is created. For instance, a $MoS_2$ surface film with a thickness of 0.5 μm contains 800 molecular layers and 799 slip planes. The numerous slip surfaces transform the original relative slip of the two metal surfaces in direct contact into the relative slip of the $MoS_2$ molecular layer, which reduces the friction factor and wear.

Xie et al. [124] pointed out that the tribological mechanism of lamellar nanocomposites is different from that of spherical nanocomposites. They concluded that the lamellar nanocomposites were able to provide lower COF and lower wear rate due to the shearing effect of weak van der Waals bonds between molecular layers. Compared to spherical nanocomposites, sheet nanocomposites are less likely to roll on the friction surface. Meng et al. [125] analyzed the lubrication mechanism using molecular dynamics simulations, which showed that an increase in normal pressure promotes interlayer slip and reduces the friction coefficient between the $MoS_2$ nanosheets. Wang et al. [126] used alkylamine-grafted $MoS_2$ oxide nanosheets ($MoS_2$-O-OLA) dispersed in 15# industrial white oil for friction experiments, confirming that $MoS_2$-O-OLA nanosheets exhibited better friction and wear resistance, resulting in a significant reduction of friction (36.2%) and wear (22.4%) by 0.02 wt% $MoS_2$-O-OLA nanosheets as compared to pure 15# industrial white oil. $MoS_2$-O-OLA nanosheets played an important role in improving tribological properties by generating interlayer slip at the steel ball contact interface, resulting in surface protection and a uniform oil film.

For rGO/$MoS_2$ nanocomposites [127], when their concentration is low and their size is smaller than the oil film thickness, they can form interlayer slips between the friction interfaces and provide friction reduction. However, once the concentration is too high, the excess nanocomposites will agglomerate in the base oil, and this interlayer slip to reduce friction is more difficult to realize.

#### 4.3.3. Tribofilm Formation

Numerous studies have shown that nanoparticles tend to migrate toward the friction surface during the friction process due to two factors. One is that the localized high temperature caused by friction may cause the undulation of nanoparticles on the friction surface to be greater than that of nanoparticles in the nanofluid. Due to this undulation, random migration is more active and ultimately increases the chances of nanoparticles migrating to the friction surface. Secondly, the acceleration of escaping electrons generated during friction enhances the electric field at the friction surface, the presence of which may lead to the aggregation of nanoparticles at the friction surface [128]. Nanoparticles that flow to the friction contact surface during oil lubrication are highly susceptible to friction chemical reactions under stress and friction heat, forming a friction-responsive film in the friction region [67,129,130].

Wu et al. [131] investigated the effect of different percentages of polyisobutylene amine succinimide (PIBS) on the tribological properties of $MoS_2$ nanosheets (Figure 9A). At low percentages of PIBS and without any PIBS, the nanosheet aggregates gathered in front of the contact area tend to enter the contact area and form a uniformly distributed friction film in the contact area, effectively reducing the coefficient of friction (COF) and the amount of wear. When lubricated with a high percentage of PIBS, the discrete particles tend to move and spread around/away from the contact region due to lateral flow, and thus the particles cannot enter the contact region resulting in poor lubrication performance. To investigate the friction film source of $MoS_2$ nanoparticles as lubricant additives, Wu et al. [132] investigated the effect of $MoS_2$ nanoparticles on the oil film thickness of PAO4 and 0W20 engine oils by examining the effect of $MoS_2$ nanoparticles on the oil film thickness of PAO4 and 0W20 engine oils as well as by observing the particle flow pattern (Figure 9B). The expected increase in oil film thickness, from 30 nm to 60 nm in 15 min, was obtained with the addition of 0.1 wt% $MoS_2$ to PAO4 base oil, whereas during the test period using 0.1 wt% $MoS_2$ blended with 3 wt% PIBS as a dispersant in PAO4 base oil and 0.75 wt% $MoS_2$ in 0W20 engine oil, no increase in oil film thickness and particle aggregation were observed. The results suggest that the nanoparticles responsible for friction film formation originate from particle aggregation rather than well-dispersed nanoparticles in point contact. This understanding should contribute to advances in the design of novel lubricant additives.

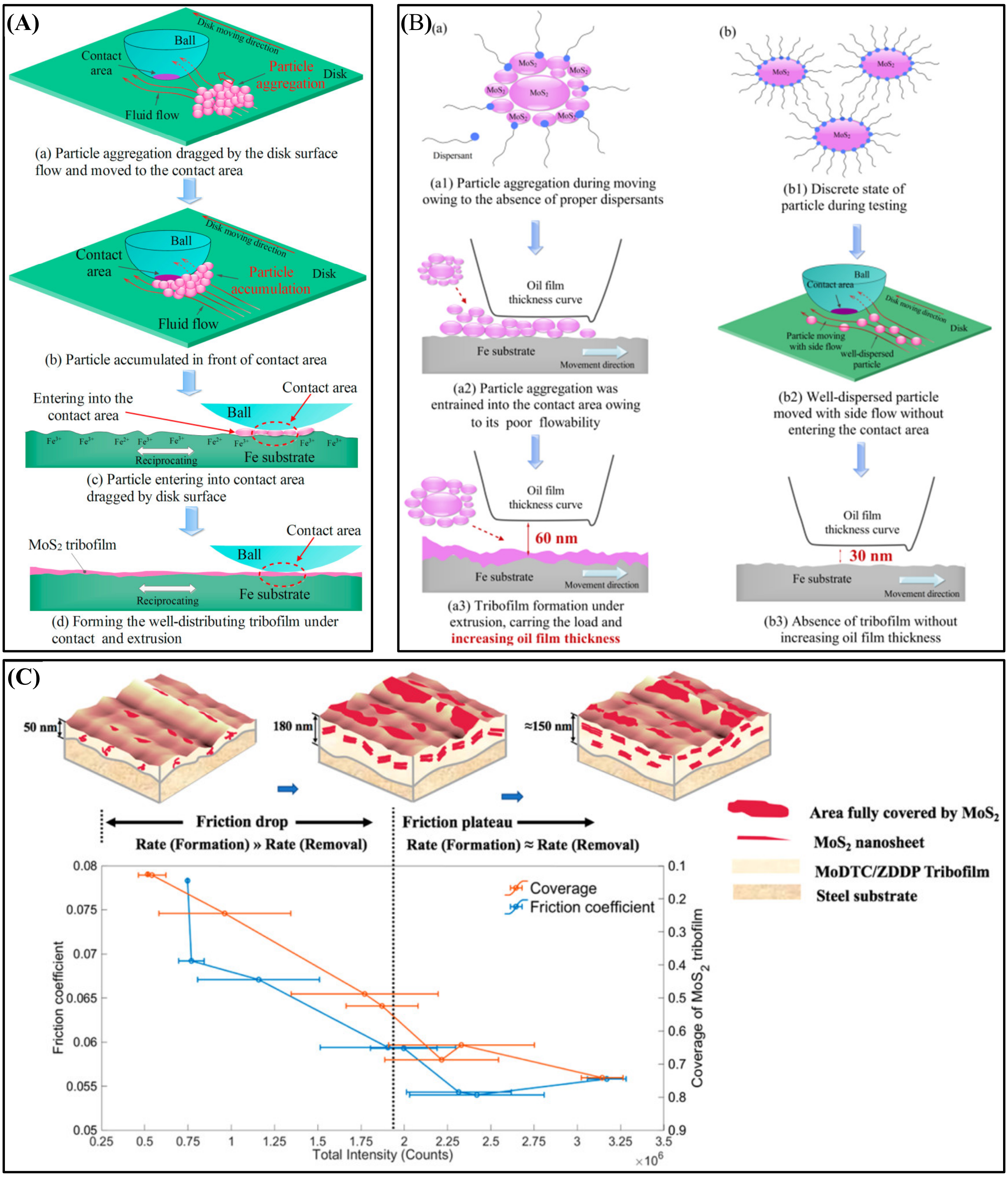


**Figure 9.** (**A**) Proposed lubrication mechanism for influence of dispersants on oil film thickness (Reproduced from Ref. [131]). (**B**) Proposed lubrication mechanism for influence of dispersants on oil film thickness. (**a**) In PAO4 base oil without proper dispersants and (**b**) in the presence of rich dispersants (Reproduced from Ref. [132]). (**C**) Schematic diagram of the MoDTC-derived tribofilm evolution (Reproduced from Ref. [133]).

Xu et al. [133] used Raman microscopy (Raman) and atomic force microscopy (AFM) to characterize the friction behavior and friction film formation and removal of MoDTC/ZDDP (Figure 9C). Friction tests were combined with a collection of non-in situ Raman intensity maps to analyze $MoS_2$ friction film accumulation. The rates of $MoS_2$ removal from friction films obtained at different temperatures indicate that $MoS_2$ friction films are more readily removed from friction contacts compared to anti-wear ZDDP friction films. In addition, new insights into the link between $MoS_2$ film formation and friction properties are provided: the formation of $MoS_2$ films in localized friction contacts leads to an immediate reduction of microscopic friction, which is terminated as soon as the amount of $MoS_2$ reaches a so-called "saturation value". In the "saturation zone", a balance between the formation and removal of $MoS_2$ is achieved. This is the first study to relate $MoS_2$ content and coverage to friction behavior and provides a basis for the development of numerical models that can predict friction by taking into account friction chemical processes.

Tribofilm formation is currently the most popular theory to explain the anti-wear and friction reduction properties of nanocomposites. Most researchers attribute the excellent lubrication properties of nanoparticle lubricants to the fact that a friction film is formed on the friction surface during the friction process.

#### 4.3.4. Cooperative Lubrication

The lubrication ability of nano $MoS_2$ can be enhanced by utilizing the synergistic effect of nano $MoS_2$ with other materials [124,134,135]. MSH-reinforced $MoS_2$ hybrid nanocomposites synthesized by Guan et al. [94] as lubricant additives produced a strong two-layer friction film on the friction surface during sliding due to the synergistic effect of MSH and $MoS_2$ (shown in Figure 10); the upper layer was rich in $MoS_2$, which had an excellent friction-reducing effect, and the neighboring layer, which consisted of the reaction products of the MSH and the lubricant additives with the substrate, strongly ensured the stability of the first layer, which resulted in the reduction of friction and wear. Hu et al. [136] found that composite $MoS_2$ additives with different morphologies ($MoS_2$ nanorods and nanosheets) improved the wear resistance and friction reduction of liquid paraffin waxes more than those used individually due to the presence of synergistic lubrication between the two different morphologies of $MoS_2$ particles. Hu's team also investigated the synergistic lubrication of a series of material-loaded $MoS_2$ complexes during friction, including sericite (DHSM/SM) [137], montmorillonite ($MoS_2$/K10) [138], and fly ash ($MoS_2$/FA) [139], in which $MoS_2$/FA lubricant, in addition to offering strong lubrication performance, featured complexes that contain about 71% of low-cost FA, and it is of great environmental significance to obtain high-value-added products from FA and thus to turn waste into treasure.

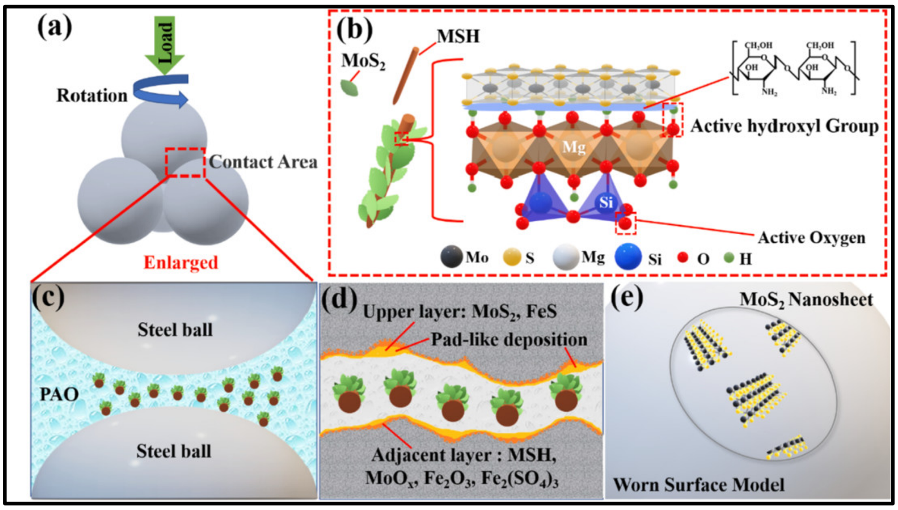


**Figure 10.** Schematic diagrams of the lubrication mechanism of MSH-$MoS_2$ nanocomposites (**a**) four-ball tester contact model, (**b**) MSH-$MoS_2$'s crystal structure, (**c**) an enlarged contact area shows the entry and adhesion of MSH-$MoS_2$ nanocomposites on the sliding surfaces, (**d**) the formation of pad-like tribofilms on the worn interface, (**e**) the formation of pad-like tribofilms on the worn surface (Reproduced from Ref. [94]).

#### 4.3.5. Other Mechanisms

Apart from the aforementioned mechanisms, other mechanisms have also been demonstrated such as repair and inlay, elastic deformation, and stripping. During the friction process, nanoparticles fill and repair the microcracks on the surface by desorption or deposition, which eventually results in a smoother and more even friction surface. It is worth noting that the "self-healing" mechanism is not just about the accumulation of nanoparticles on the friction surface. As the size of the nanoparticles reduces, their melting point also decreases drastically. Under the high temperature of the friction surface, these nanoparticles can easily melt or sinter in the microcracks in the contact zone, forming ordered nanoparticles and closely bonding with the friction surface [128]. Yi et al. [140] used a molecular dynamics approach to construct rough surfaces and multilevel dimpled structures to simulate oil-poor lubrication before oil film rupture, and the simulation results showed that multilayered $MoS_2$ acted as a load-bearing agent at light loads or low speeds and went into the grooves to repair the surfaces at heavy loads or high speeds. Yi et al. [141] used a solvent/hydrothermal method to prepare $MoS_2$ particles in three morphologies: flower-like, microspheres, and nanosheets. The tribological behavior of the synthesized $MoS_2$ particles as liquid paraffin lubrication additives was investigated using a ball and disc friction tester. It is noted in the paper that under heavy loads, $MoS_2$ nanosheets exhibit superior friction and wear reduction behavior compared to the other two types of $MoS_2$ particles. The reason for the optimal lubrication performance of nanosheet $MoS_2$ is discussed based on their exfoliation into ultrathin nanosheets and nanofragments in friction tests.

#### 4.3.6. Diversification of Lubrication Mechanism

It is worth noting that in most practical friction processes, nanoparticles do not act through a single lubrication mechanism, but rather two or more mechanisms act simultaneously [128]. The combination of different mechanisms leads to some extent to the complexity of mechanism studies. Kumari et al. [87] showed that when fully dispersed $MoS_2$-ODT was used as an additive in polyol lubricant base oils, the uninterrupted supply of $MoS_2$-ODT nanosheets on the friction contact surfaces resulted in the formation of $MoS_2$ films at the friction interface, and the easy shear driven by weak van der Waals interactions between the $MoS_2$ lamellae and the high mechanical strength of $MoS_2$ combined to improve friction performance. Zhang et al. [142] prepared carbon ball surface-coated $MoS_2$ (C@$MoS_2$) by a hydrothermal method to realize the unification of the lubrication characteristics of carbon balls (CS) with the high load carrying capacity and low shear friction performance of $MoS_2$. Then, 0.5 wt% of C@$MoS_2$ particles were dispersed into PAO40 to enhance the anti-wear and friction reduction capability. The excellent tribological properties of C@$MoS_2$ particles were attributed to the filling effect of CS and the synergistic lubrication between bearings as well as the friction reduction capability of the $MoS_2$ shell layer. In this study, the synergistic lubrication mechanism of C@$MoS_2$ can be categorized into different stages (shown in Figure 11): (1) shell layer fragmentation stage, which corresponds to the rolling bearing effect of C@$MoS_2$ particles at the friction interface; (2) core/shell separation stage, which corresponds to the rolling bearing effect and the self-repairing effect of the C@$MoS_2$ particles; and (3) core/shell composite stage, which corresponds to the formation of the protective film and the enhancement in which the friction and wear performance is substantially improved.

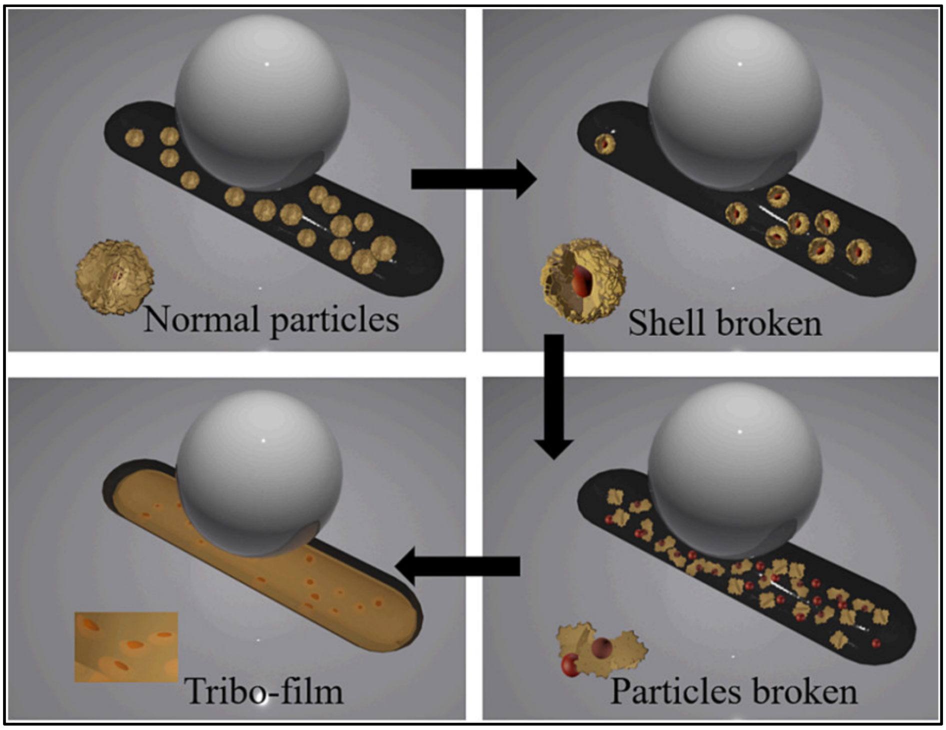


**Figure 11.** The staged action mechanism of C@$MoS_2$ particles in the lubrication process (Reproduced from Ref. [142]).

## 5. Summary and Outlook

This paper reviews the notable research progress in the field of $MoS_2$ nanomaterials as lubricant additives in recent years, focusing on the synthesis strategy, structure, dispersion stability, tribological performance evaluation, and lubrication mechanism of $MoS_2$ nanomaterials. Dispersion stability is the key to the research and application of graphene lubricant additives. There are physical and chemical methods to improve the dispersion stability of $MoS_2$ nanomaterials in lubricants, in which the dispersion, stability, and tribological properties can be easily adjusted and improved by combining $MoS_2$ with other materials, further expanding the application of $MoS_2$ nanomaterials in tribology. Various lubrication mechanisms such as interlayer sliding, ball action, formation of friction film, and synergistic action, which are currently recognized, are outlined, and the excellent tribological properties of $MoS_2$ nanomaterials are explained in depth.

Although significant breakthroughs and developments have been made in the research on $MoS_2$ nanomaterials as lubricant additives, there are still some difficulties and challenges to overcome as follows.

(1) The dispersion stability of $MoS_2$ nanomaterials in lubricating oils has not been adequately addressed. During the friction process, the organic modifier is prone to degradation due to the heat generated by friction, which leads to the re-colonization of $MoS_2$ nanoparticles in the lubricating oil. Therefore, research on the long-term dispersion stability of $MoS_2$ nanomaterials is still necessary.

(2) In terms of lubrication mechanism, there are fewer systematic descriptions of the lubrication mechanism of $MoS_2$ nanomaterials with different morphologies in lubricants. The influence of each component on the tribological performance is still unclear, such as the interaction between additive materials and lubricants, the synergistic lubrication of each component in composite nanomaterials, and the interaction between nanomaterials and modifiers. It is necessary to further elucidate the relevant mechanisms by combining advanced characterization techniques, molecular dynamics simulations, and theoretical calculations.

(3) Most reported additives for $MoS_2$-based nanomaterials have been studied under laboratory conditions. In the future, there is a need to develop new additives with good tribological properties under extreme conditions or multiple environments. In addition, low-cost, large-scale preparation routes and evaluation of tribological properties in practical applications are essential for the practical application of these additives.

To promote the use of nanomaterials as lubricant additives, the following aspects should be considered for future research, taking into account future developments and trends in $MoS_2$ lubricant additives:

First, it is important to prepare high-quality $MoS_2$ through a green, economical, and scaled-up production method. Combined with modern computer simulation methods, it can not only reduce the unnecessary waste of resources but also optimize the design of $MoS_2$ nanoparticles with good structure and obtain better performance. Secondly, dispersion stability is very important as a prerequisite for practical applications. However, there are still difficulties in improving the dispersion stability of nanomaterials for practical industrial applications. For example, simple physics is susceptible to reaggregation. Chemical modifications may mask the original properties and chemical modifiers tend to degrade during friction. Therefore, further research is needed to improve dispersion methods. The self-dispersion method does not require any modifier and has good dispersion stability. The stability of the prepared $MoS_2$ and $MoS_2$-based composite nanomaterials dispersed in the lubricant is crucial for long-term enhancement of tribological properties. Finally, with the rapid development of advanced industrial equipment, many advanced mechanical devices need to meet extreme working conditions such as high load, high temperature, and high speed. It is important to study new lubricant additives for extreme friction conditions. Many highly efficient lubricant additives decompose at 150 °C and have a flash point not exceeding 100 °C. The application of lubricant additives is still limited to temperatures of 1000 °C and loads of 3000 N. Therefore, it is of great significance to further improve the performance of $MoS_2$ and $MoS_2$-based composites under extreme conditions to overcome the bottleneck of friction and wear inefficiency.

In future research, it is believed that with the efforts of a large number of researchers, $MoS_2$ nanomaterials will make surprising breakthroughs and progress and realize the expected concepts.

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## List of Abbreviations

| | |
|---|---|
| MoDTC | Molybdenum dialkyl dithiocarbamate |
| ZDDP | Zinc dialkyl dithiophosphate |
| TMDs | Transition metal dichalcogenides |
| CS/HAc | chitosan/acetic acid |
| DMF | N, N-dimethylformamide |
| NMP | N-methylpyrrolidone |

| | |
|---|---|
| TAA | Thioacetamide |
| OA | Oleic acid |
| ODT | Octadecanethiol |
| TEA | Triethanolamine |
| SA | Stearic acid |
| *h*-BN | hexagonal boron nitride |
| CTAB | Cetyltrimethylammonium bromide |
| FrGO | Functionalized reduced graphene oxide |
| MSH | Magnesium silicate hydroxide |
| Gr-MS-Zn | ZnO-modified reduced graphene oxide/$MoS_2$ |
| PAO | Polyalphaolefin |
| LP | Liquid paraffin |
| SEM | Scanning electron microscope |
| TEM | Transmission electron microscopy |
| RHC/$MoS_2$ | Rice husk charcoal/$MoS_2$ |
| PEG | Polyethylene glycol |
| CNT | Carbon nanotubes |
| PAG | Polyalkylene glycol |
| CS @ 1 T-$MoS_2$ | carbon sphere @ metallographic $MoS_2$ |
| g-$C_3N_4$ | graphitic phase carbon nitride |
| $MoS_2$-O-OLA | Alkylamine-grafted $MoS_2$ oxide |
| PIBS | Polyisobutylene amine succinimide |
| COF | Coefficient of friction |
| Raman | Raman microscopy |
| AFM | Atomic force microscopy |
| K10 | Montmorillonite K10 |
| FA | Fly ash |

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