

## Article

# Preparation and Tribological Properties of Wind Turbine Lubricating Oil Modified by Nano-ZnMgAl Layered Double Hydroxides

Hao Xiao <sup>1,2</sup>, Yihao Zhang <sup>1,2</sup>, Xueqiang Ding <sup>1</sup>, Mingmin Zheng <sup>1</sup>, Qiuya Tu <sup>1</sup>, Zongde Liu <sup>2</sup> , Jingbin Han <sup>3</sup> , Xin Zhang <sup>1,\*</sup> and Yuan Xu <sup>4</sup>

<sup>1</sup> New Energy and Future Technologies Research Institute, State Power Investment Corporation Research Institute Co., Ltd., Beijing 102209, China; xhhbldx@163.com (H.X.); zhangyihao0310@sina.com (Y.Z.); dingxueqiang@spic.com.cn (X.D.); zhengmingmin@spic.com.cn (M.Z.); tuqiuya@spic.com.cn (Q.T.)

<sup>2</sup> School of Energy Power and Mechanical Engineering, North China Electric Power University, Beijing 102206, China; lzd@ncepu.edu.cn

<sup>3</sup> College of Chemistry, Beijing University of Chemical Technology, Beijing 100029, China; hanjb@mail.buct.edu.cn

<sup>4</sup> Ordos Power Supply Company, Inner Mongolia Electric Power (Group) Co., Ltd., Ordos 017000, China; xuxinyuan1987@126.com

\* Correspondence: kmzx201@163.com

## Abstract

ZnMgAl layered double hydroxides (LDHs) were synthesised via coprecipitation, and oleic acid and stearic acid were grafted onto their surfaces via dehydration condensation to obtain two nano-lubricant additives, OA-ZnMgAl LDH and SA-ZnMgAl LDH. These surface modifications significantly improved the dispersion stability of ZnMgAl LDH in lubricating oil. Tribological tests showed that, at their respective optimal concentrations for friction reduction or wear resistance, ZnMgAl LDH, OA-ZnMgAl LDH, and SA-ZnMgAl LDH reduced the coefficient of friction by 3%, 20%, and 16%, and decreased the wear scar diameter by 7%, 9%, and 14%, respectively, compared with the base oil (XMP-Mobil 320). To clarify the lubrication mechanism, the wear morphology and chemical composition were analysed using 3D optical profilometry, X-ray photoelectron spectroscopy, scanning electron microscopy, and FIB-SEM. The results indicate that LDHs react with the steel surface under load and shear to form a multilayer protective film consisting of an inner oxide layer and an outer graphite layer, preventing direct contact between friction pairs. In addition, the rolling and filling effects of partially unreacted LDHs further reduce friction and wear.

**Keywords:** LDHs; surface modification; lubricant additives; anti-friction and anti-wear



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## 1. Introduction

Energy constitutes the core driver of economic growth. The traditional global energy mix has been overwhelmingly dominated by fossil fuels (coal, oil, and natural gas), whose combustion releases substantial greenhouse gases, contributing to global warming and frequent extreme weather events. Moreover, the uneven distribution of fossil fuel resources, coupled with geopolitical instability, readily triggers energy supply crises. To address these challenges, numerous nations worldwide have set carbon neutrality targets, driving the transition towards low-carbon, clean, and reliable energy consumption [1]. This necessitates shifting from a fossil fuel-centric energy structure to a new system underpinned

by renewable energy. In 2023, renewable energy accounted for 14.6% of global primary energy consumption, an increase of 0.4% year-on-year, with renewable electricity generation rising by 13% [2]. Among renewable sources, wind power plays a pivotal role. Since 2014, global wind turbine capacity has nearly tripled, surpassing 1 terawatt for the first time in 2023 [3]. With the continuous increase in installed wind turbine capacity and the progressive scaling up of wind turbine units, operational and maintenance management issues for wind generators have consequently emerged. Statistics indicate that lubrication failures account for over 34% of transmission system faults. The main gearbox constitutes a core component within wind turbine units, featuring a high speed ratio, high transmission efficiency, substantial start-up loads, frequent start–stop cycles, and complex operating environments. Consequently, it imposes stringent requirements on gear oils. Should gearbox failure occur, the intricate repair process necessitates prolonged downtime for the wind turbine, significantly impacting both operational safety and economic viability. Conventional lubricants increasingly fail to meet the demands of larger turbine designs, rendering the need for composite lubricants with superior physicochemical properties and lubricating performance ever more pressing.

To enhance the overall performance of lubricating oils, various types of additives must be incorporated to meet the diverse requirements of mechanical components under specific operating conditions. Commonly used additives include anti-wear agents, antioxidants, extreme pressure additives, dispersants, and corrosion and rust inhibitors [4]. Layered double hydroxides (LDHs) are a class of inorganic compounds with a layered structure, composed of divalent metal ions, trivalent metal ions, and anions. Researchers have summarised the common properties of LDHs and proposed their structural formula,  $[M_{1-x}^{2+}M_x^{3+}(\text{OH})_2](\text{A}^{n-})_{x/n} \cdot m\text{H}_2\text{O}$ , where  $M^{2+}$  and  $M^{3+}$  denote divalent and trivalent metal ions, respectively. Common divalent cations include  $\text{Mg}^{2+}$ ,  $\text{Zn}^{2+}$ ,  $\text{Co}^{2+}$ ,  $\text{Mn}^{2+}$ , etc., while trivalent cations encompass  $\text{Ni}^{3+}$ ,  $\text{Al}^{3+}$ ,  $\text{Cr}^{3+}$ ,  $\text{Fe}^{3+}$ , etc. [5]. In the structural formula,  $\text{A}^{n-}$  denotes an interlayer anion bearing  $n$  negative charges, including  $\text{Cl}^-$ ,  $\text{CO}_3^{2-}$ ,  $\text{NO}_3^-$ , and others.  $x$  denotes the molar fraction of trivalent metal cations within the total cation content, typically ranging from 0.20 to 0.33.  $m$  indicates the number of crystallisation water molecules, generally falling within the range of 0 to 6 [6–8]. Due to their unique structure, shape memory effect, and chemically tunable composition, hydrotalcite minerals have been extensively studied across diverse fields, including catalysis, adsorption, flame retardancy, and medicine [9–12].

In 2012, Bai et al. pioneered the use of cobalt–aluminium-based LDH powder with a particle size of 150 nm as a lubricant additive. Their investigation into the influence of hydrotalcite on friction characteristics revealed that LDHs could effectively enhance anti-wear and friction-reducing properties [13]. Since then, research into hydrotalcite as a lubricant additive has progressively increased. Previous studies have demonstrated that long-chain fatty acids (such as oleic acid OA and stearic acid SA) can be used for surface hydrophobic modification of binary layered double hydroxides (LDHs) like ZnAl-LDH and MgAl-LDH [14,15]. This enhances their dispersibility in non-polar media, thereby improving the wear and corrosion resistance of target products. However, research on surface modification of ternary LDHs remains relatively limited. The ternary system allows for the regulation of layer charge density and interlayer interactions, resulting in distinct interfacial properties after OA/SA modification compared to binary LDH systems.

In this study, we prepared ZnMgAl LDH via the coprecipitation method and successfully synthesised OA-ZnMgAl LDH and SA-ZnMgAl LDH by grafting oleic acid and stearic acid onto their surfaces through dehydration condensation. Surface modification aimed to enhance the dispersibility of LDHs in lubricating oils and we investigated the improvement in the anti-friction and anti-wear properties of wind turbine gearbox lubricants by adding LDHs. Subsequently, the effects of different LDH loading levels on lubricant anti-friction and anti-wear properties were compared to determine the optimal LDH dosage. Finally, based on these results, the anti-friction and anti-wear mechanism of LDHs was analysed.

## 2. Materials and Methods

### 2.1. Chemicals and Materials

$\text{Zn}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ ,  $\text{Mg}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ ,  $\text{Al}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$ , NaOH and  $\text{Na}_2\text{CO}_3$  were purchased from Aladdin (Shanghai, China), Oleic acid, stearic acid and n-Hexane were supplied by Macklin Biochemical Co., Ltd. (Shanghai, China). All reagents listed above are of analytical grade. The lubricant employed in the experiment was Mobil XMP-320 gear oil (Aniol St San Antonio, TX, USA), a commonly used grade for wind turbine gearboxes. Its physical and chemical properties, including viscosity and oxidation resistance, are well suited to the high-load, long-cycle operating conditions of wind turbines. This oil is suitable for evaluating the performance enhancement effects of nano-additives in actual wind power lubrication scenarios.

### 2.2. Synthesis of LDH Nanoparticles

The coprecipitation method was employed to synthesise ZnMgAl LDH in this study. The salt solution was prepared by stirring and dissolving  $\text{Zn}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ ,  $\text{Mg}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ , and  $\text{Al}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$  in 200 mL of deionised water, maintaining a  $(\text{Zn}^{2+} + \text{Mg}^{2+}):\text{Al}^{3+}$  ratio of 2:1 ( $\text{Zn}^{2+} = 0.005 \text{ mol}$ ). The alkaline solution was prepared by stirring and dissolving NaOH and  $\text{Na}_2\text{CO}_3$  in 200 mL of deionised water, yielding  $\text{OH}^- = 0.96 \text{ mol/L}$  and  $\text{OH}^-:\text{CO}_3^{2-} = 2.4$ . The salt solution and alkaline solution were placed into an ultrasonic cleaner and agitated to ensure complete dissolution of the chemical reagents. The salt solution was transferred to a three-necked flask, and the alkaline solution was transferred to a burette. The magnetic stirrer and pH metre were activated, and the burette flow rate was adjusted to one drop per second until the pH stabilised between 10 and 11. Stirring was continued for half an hour after the completion of the titration. Subsequently, the stirred suspension was transferred into a cleaned reactor vessel and aged for 15 h in a high-pressure reactor at  $100^\circ\text{C}$ . The resulting product was centrifuged, washed three times with deionised water and ethanol and dried at  $66^\circ\text{C}$  for 24 h. The product was ground and sieved through a 200-mesh screen to obtain ZnMgAl LDH.

### 2.3. Surface Modification of ZnMgAl LDH

Surface graft modification of ZnMgAl LDH was performed using oleic acid (OA) and stearic acid (SA). The synthesis schematic is depicted in Figure 1. ZnMgAl LDH was dispersed in n-hexane, OA was added, and the mixture reacted at  $60^\circ\text{C}$  for 12 h. The resulting product was centrifuged, washed three times with ethanol, dried for 28 h, then ground and sieved through a 200-mesh screen to complete the preparation of OA-ZnMgAl LDH. The preparation method for SA-ZnMgAl LDH was analogous to that of OA-ZnMgAl LDH.

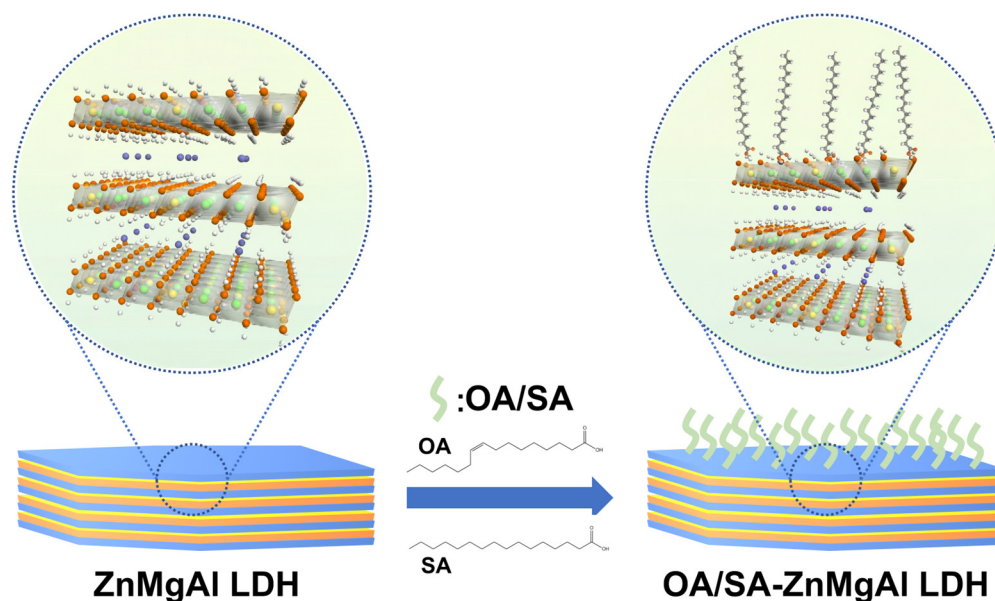


**Figure 1.** Schematic diagram of the synthesis of OA-ZnMgAl and SA-ZnMgAl.

#### 2.4. Characterisation of LDH Nanoparticles

X-ray diffraction (Bruker D8 Advance, Bremen, Germany) patterns were obtained using a diffractometer at  $2^\circ/\text{min}$  within the range of  $3^\circ$  to  $70^\circ$  with Cu K $\alpha$  radiation ( $\lambda = 0.15406 \text{ nm}$ ) to confirm the crystal structure; Fourier transform infrared spectroscopy (Thermo Fisher Scientific Nicolet iS20, Waltham, MA, USA) was employed to analyse the structure and surface functional groups of LDH powders, with a wavenumber range of  $4000\text{--}400 \text{ cm}^{-1}$ . The structure and dimensions of the nanosheets were determined using SEM (ZEISS Sigma 300, Oberkochen, Germany). The thermal stability of the nanosheets was assessed using an STA 449F3 (Nerz, Troisdorf, Germany) thermogravimetric analyser, heated at  $10^\circ\text{C}/\text{min}$  from  $25^\circ\text{C}$  to  $800^\circ\text{C}$  in air. Elemental composition and chemical valence states on the sample surface were analysed using X-ray photoelectron spectroscopy (Thermo Kalpha, Waltham, MA, USA), equipped with a monochromatic Al K $\alpha$  source, with the C 1s binding energy of  $284.8 \text{ eV}$  as reference. The contact angle of the nanosheets was measured using a Dataphysics OCA20 (Cangzhou, China) contact angle analyser. Surface charge data of the nanosheets were determined using a Malvern Zetasizer Nano ZS90 (Malvern, UK). To evaluate the dispersion stability of the nanosheets in oil, alongside visual inspection, the  $\zeta$ -potential of the oil samples was also measured using a  $\zeta$ -potential analyser (Malvern Zetasizer Nano ZS90).

#### 2.5. Anti-Wear and Friction Reduction Performance Testing

First, the LDH nanoparticles were dispersed into the lubricant oil following the steps described below. The required amount of LDH powder was accurately weighed and introduced into the base lubricant oil. The mixture was initially magnetically stirred for 1 h to promote preliminary dispersion, and subsequently ultrasonicated for 30 min to achieve a uniform and stable nanoparticle dispersion.

The diameter of the scuff marks was tested using an MS-10J four-ball friction tester (Xiamen, China) under the following conditions: friction duration of 60 min, rotational speed of  $1800 \text{ r/min}$ , load of  $196 \text{ N}$ , and temperature of  $54^\circ\text{C}$ . The coefficient of friction was measured using a Bruker (CETR) UMT-2 (Bremen, Germany) friction and wear tester under the following parameters: friction duration of 60 min, load of  $200 \text{ N}$ , friction rate of  $50 \text{ Hz}$ , and friction stroke of  $2 \text{ mm}$ . The COF test was conducted once under strictly controlled conditions. The depth of the wear scar was measured using a three-dimensional

optical profilometer (Bruker Contour GT-K 3D, Bremen, Germany). Elemental composition and distribution were analysed via X-ray photoelectron spectroscopy. Cross-sections of the friction film were prepared using FIB-SEM (Thermo Fisher Scios2, Waltham, MA, USA) to investigate the anti-wear and friction-reducing mechanism of LDHs.

### 3. Results and Discussion

#### 3.1. Characterisation Analysis of LDHs

The three LDHs were characterised using an XRD diffractometer to investigate their crystalline structures, with the results shown in Figure 2a. According to PDF#89-0460 in the powder diffraction library, the laboratory-prepared ZnMgAl LDH, OA-ZnMgAl LDH, and SA-ZnMgAl LDH all exhibit diffraction peaks characteristic of a hexagonal layered structure: (003) and (006). The (110) peak indicates the integrity of the internal structure in all three samples [16]. Table 1 presents the interlayer spacings  $d_{(003)}$  and  $d_{(110)}$  for the three nanosheets, calculated from the Bragg equation ( $n\lambda = 2d\sin\theta$ ). The unit cell parameters  $a$  and  $c$  were then determined using the relationships  $a = 2d_{(110)}$  and  $c = 3d_{(003)}$  [17].

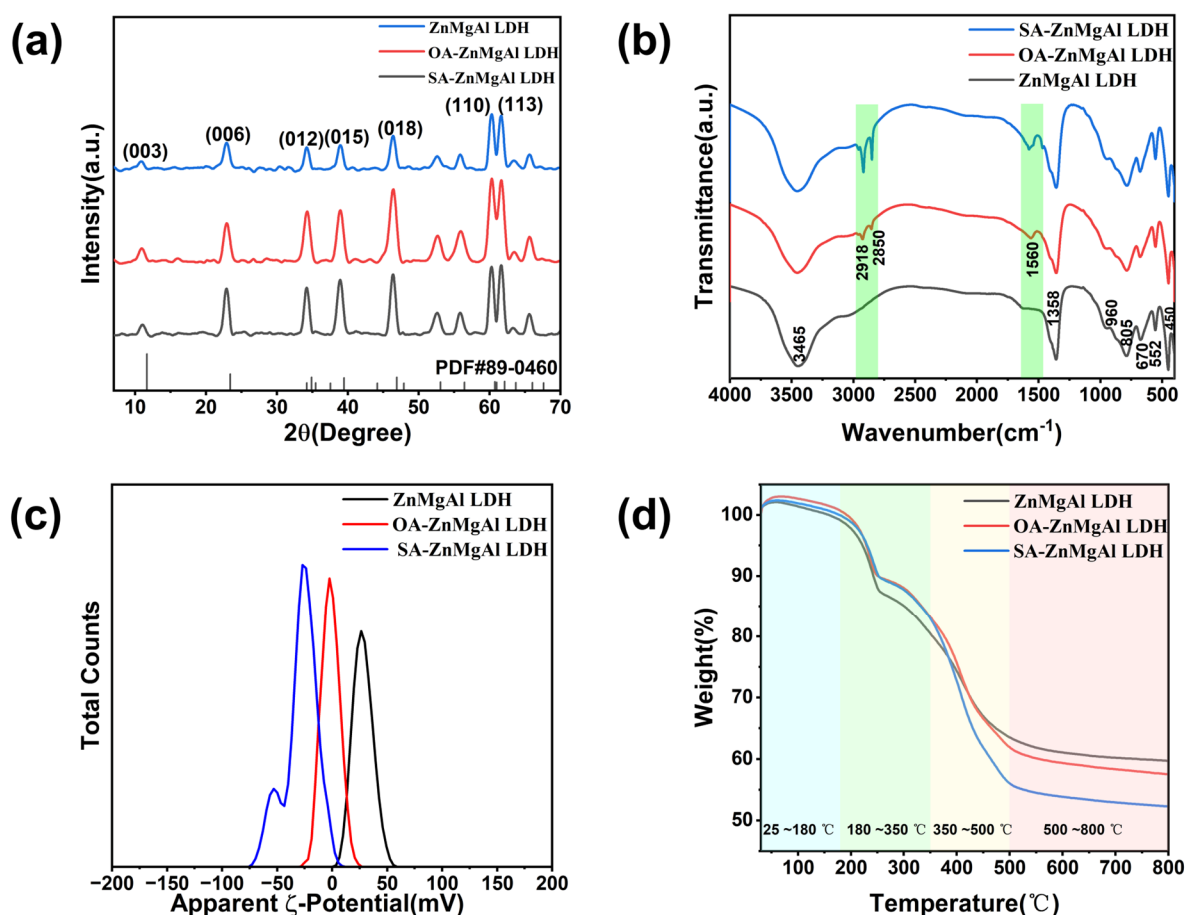


**Figure 2.** ZnMgAl LDH, OA-ZnMgAl LDH and SA-ZnMgAl LDH: (a) powder XRD images; (b) Fourier Transform Infrared (FTIR) spectra; (c) powder  $\zeta$ -potentials; and (d) thermogravimetric analysis (TGA) plots.

As shown in Figure 2a, under the same diffraction plane, the (003) and (110) diffraction peaks of OA-ZnMgAl LDH and SA-ZnMgAl LDH exhibit a negative angle shift compared to ZnMgAl LDH. Table 1 indicates that the interlayer spacing of ZnMgAl LDH increases following OA and SA surface modifications. This is attributed to the grafting of long organic carbon chains onto the ZnMgAl LDH surface, thereby increasing the interlayer spacing and enhancing its dispersibility in lubricating oils [18]. Concurrently, the grafting of these

chains onto the surface alters the lattice parameter  $a$  and the hydrotalcite layer thickness  $c$ . The increase in layer thickness also contributes to the expansion of the interlayer spacing.

**Table 1.** Comparison of layer spacing and lattice parameters of ZnMgAl LDH, OA-ZnMgAl LDH and SA-ZnMgAl LDH.

| LDHs Products | $d_{(003)}$ (Å) | $d_{(110)}$ (Å) | $a$ (Å) | $c$ (Å) |
|---------------|-----------------|-----------------|---------|---------|
| ZnMgAl        | 8.0561          | 1.5347          | 3.0694  | 24.1683 |
| OA-ZnMgAl     | 8.0991          | 1.5334          | 3.0668  | 24.2973 |
| SA-ZnMgAl     | 8.1717          | 1.5343          | 3.0686  | 24.5151 |

The Fourier transform infrared spectra of the three nanoplates are shown in Figure 2b. The broad, intense absorption peak at  $3465\text{ cm}^{-1}$  arises from the overlapping stretching vibrations of surface hydroxyl groups and interlayer water molecules in ZnMgAl LDH, OA-ZnMgAl LDH, and SA-ZnMgAl LDH [19]; The absorption peaks at  $2918\text{ cm}^{-1}$  and  $2850\text{ cm}^{-1}$  for OA-ZnMgAl LDH and SA-ZnMgAl LDH, respectively, correspond to the antisymmetric and symmetric stretching vibrations of the long carbon chains' C-H bonds in oleic and stearic acid molecules [20,21]. However, the vibration peak at  $1710\text{ cm}^{-1}$  corresponding to the carboxyl group (-COOH) in oleic and stearic acids disappears, replaced by a carboxylate vibration peak at  $1560\text{ cm}^{-1}$ . This indicates that both acids react with the surface hydroxyl (-OH) groups of the hydrotalcite-like phase to form carboxylates, achieving chemical adsorption and surface modification. The antisymmetric stretching vibration peak  $\nu_3$  of free  $\text{CO}_3^{2-}$  typically appears at a wavenumber of  $1415\text{ cm}^{-1}$ . However,  $\text{CO}_3^{2-}$  intercalated between hydrotalcite layers undergoes a shift in the  $\nu_3$  vibration peak towards the lower frequency of  $1358\text{ cm}^{-1}$  due to the influence of interlayer water molecule hydrogen bonds [22]. Absorption peaks at low frequencies of  $960\text{ cm}^{-1}$  and  $670\text{ cm}^{-1}$  correspond to the interlayer  $-\text{CO}_3^{2-}$  out-of-plane bending vibration  $\nu_2$  and antisymmetric bending vibration  $\nu_4$ , respectively. The absorption peaks at  $805\text{ cm}^{-1}$  and  $552\text{ cm}^{-1}$  are attributed to interactions between the interlayer  $\text{CO}_3^{2-}$  and the layered structure; the peak at  $450\text{ cm}^{-1}$  arises from vibrations of the  $[\text{AlO}_6]^{3-}$  group or Al-O single bonds within the layered structure [23,24]. The infrared spectra confirm the successful modification of ZnMgAl LDH by OA and SA.

The  $\zeta$ -potential distribution of the three nanoplate samples is shown in Figure 2c.  $\zeta$ -potential is an important indicator of the colloidal stability and surface charge characteristics of nanoparticles. The unmodified ZnMgAl LDH exhibited a relatively high positive  $\zeta$ -potential of  $+25.5\text{ mV}$ , which can be attributed to the presence of surface hydroxyl groups that readily generate positively charged layers. After surface modification, the  $\zeta$ -potentials of OA-ZnMgAl LDH and SA-ZnMgAl LDH decreased significantly to  $+1.03\text{ mV}$  and  $-29.0\text{ mV}$ , respectively. The near-neutral value of OA-ZnMgAl LDH suggests reduced electrostatic repulsion and the introduction of hydrophobic organic chains, whereas the negative  $\zeta$ -potential of SA-ZnMgAl LDH reflects the adsorption of stearate anions on the LDH surface. These changes confirm successful surface modification and indicate improved compatibility and dispersion behaviour of the LDHs in non-polar lubricant media. Thermogravimetric analysis was employed to characterise the thermal stability of the nanosheets, with results presented in Figure 2d. The mass loss of the three LDHs primarily comprises three stages. The first stage spans from room temperature to  $180^\circ\text{C}$  [25], during which mass loss is mainly attributed to the evaporation of surface-adsorbed water molecules and interlayer water molecules, exhibiting slight weight loss at a low rate. However, the surface-modified LDHs exhibited reduced water loss, indicating that the grafting of organic long chains partially suppressed the adsorption and retention of water

molecules. The second stage spans 180–350 °C, where mass loss results from partial hydroxyl group removal and interlayer  $\text{CO}_3^{2-}$  release, causing a marked increase in weight loss rate. The third stage, 350–500 °C, exhibits the most pronounced weight loss. Ultimately, as temperature rises, the layered structure of LDHs collapses, converting them into oxides (LDO). Subsequently, the process enters a terminal plateau phase where mass loss flattens. As shown in the figure, ZnMgAl LDH exhibits the highest residual mass, indicating that organic modification increases combustible components. The results demonstrate that all three LDHs exhibit favourable thermal stability under lubricant operating conditions (approximately 80 °C), meeting the requirements for lubricant nanoadditives.

The microstructures and particle size distributions of the three types of nanosheets were characterised using SEM, with experimental results shown in Figure 3. Figure 3a,c,e reveal that all three nanoplates exhibit a typical hexagonal layered structure. The unmodified LDH nanoplates possess higher surface energy and exhibit more pronounced agglomeration. Following OA and SA modification, the surface energy of ZnMgAl LDH decreases due to partial hydroxyl group reduction via dehydration condensation, thereby mitigating agglomeration. Figure 3b,d,f reveal that the lateral dimensions of the three LDH nanosheets largely follow a normal distribution. The median widths for ZnMgAl, OA-ZnMgAl, and SA-ZnMgAl were 95 nm, 81 nm, and 84 nm, respectively, indicating relatively uniform size distributions.

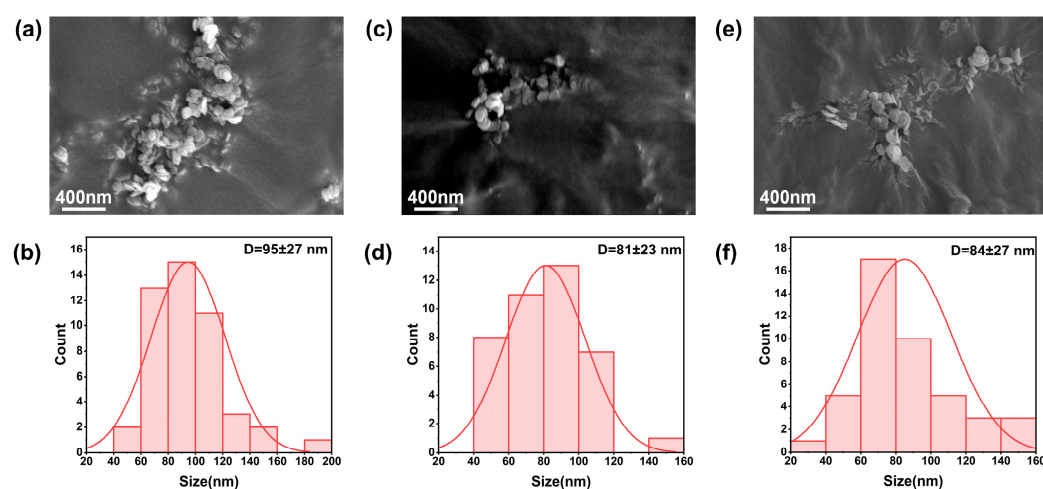


**Figure 3.** SEM images of (a) ZnMgAl LDH, (c) OA-ZnMgAl LDH, and (e) SA-ZnMgAl LDH powders. (b) Particle size distribution plots of ZnMgAl LDH, (d) OA-ZnMgAl LDH, and (f) SA-ZnMgAl LDH powders.

The contact angle is a primary parameter characterising a material's wettability, reflecting the compatibility between additives and the original base oil. ZnMgAl LDH powder exhibits hydrophilicity and oleophobicity, with a contact angle below 5°. Following surface modification, the contact angles of OA-ZnMgAl LDH and SA-ZnMgAl LDH are shown in Figure 4, measuring 92.2° and 137.9°, respectively, indicating superior hydrophobicity in both cases. This is attributable to the grafting of hydrophobic long-chain alkyl groups (OA and SA) onto the ZnMgAl LDH surface via dehydration condensation of carboxyl and hydroxyl groups, thereby enhancing material compatibility.

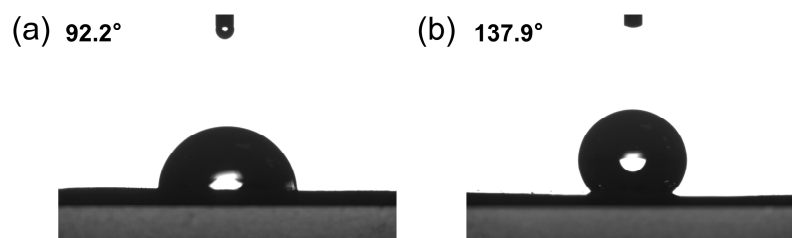


**Figure 4.** Contact angles of (a) OA-ZnMgAl LDH and (b) SA-ZnMgAl LDH powders.

The X-ray photoelectron spectra of the three nanoplates are shown in Figure 5. The positions of the Zn 2p, Mg 1s, and Al 2p peaks are consistent across all three (Figure 5a), corresponding to 1021.92 eV, 1303.96 eV, and 74.22 eV, respectively [26]. However, the peak intensities are reduced, attributed to the grafting of organic compounds onto the nanoplate surfaces, forming a coating that attenuates the signal. The C 1s peak of ZnMgAl LDH, OA-ZnMgAl LDH, and SA-ZnMgAl LDH was fitted using peak resolution, with the results shown in Figure 5b. The results indicate that the intensities of the C 1s peaks undergo significant changes following modification. The peaks at 284.7 eV, 285.5 eV, and 288.6 eV correspond to C-C, C-OH, and  $\text{CO}_3^{2-}$  bonds in the carbon group, respectively. The strong C 1s peak at 284.7 eV, characteristic of alkyl chains, confirms the successful grafting of oleic acid and stearic acid onto the ZnMgAl LDH surface.

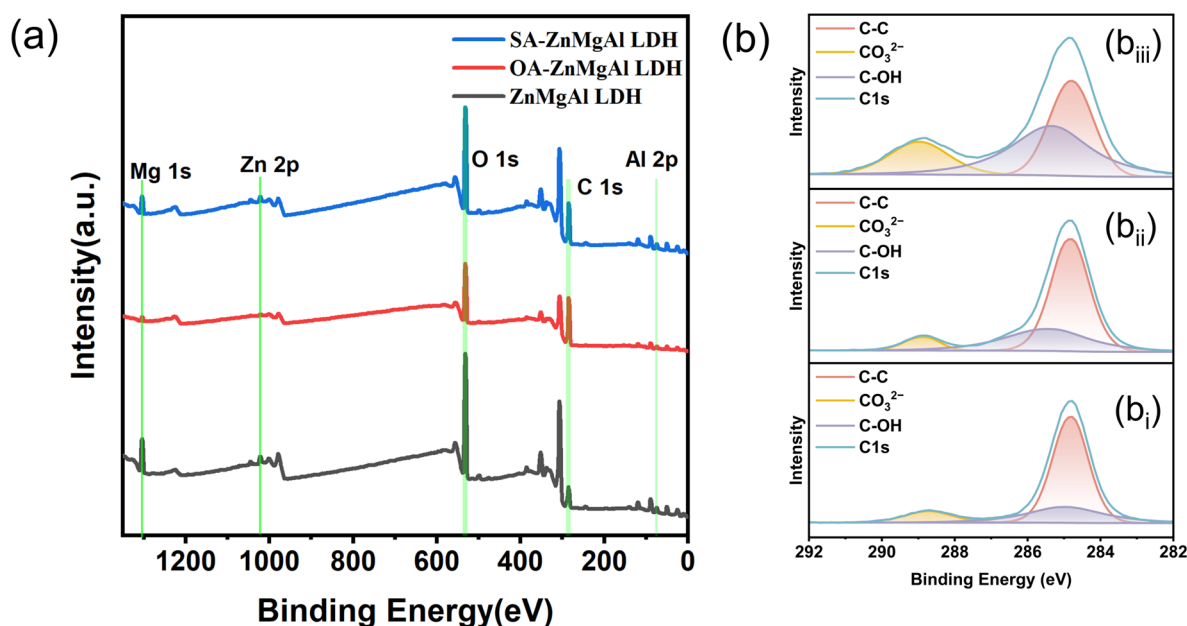


**Figure 5.** (a) XPS spectra of LDH nanosheets and (b<sub>i</sub>–b<sub>iii</sub>) C 1s split-peak fitting results of ZnMgAl LDH, OA-ZnMgAl LDH and SA-ZnMgAl LDH.

### 3.2. Study on the Dispersibility of Gear Lubricants Containing LDH Nanoadditives

To investigate the dispersion stability of three types of LDHs in Mobil XMP-320 gear oil, ZnMgAl LDH, OA-ZnMgAl LDH, and SA-ZnMgAl LDH were added at a mass fraction of 0.15 wt.% to the lubricant. The samples were then left to settle in a cool, dry location for observation of sedimentation (as shown in Figure 6). At initial preparation, all lubricants appeared relatively clear. By day 4, the unmodified ZnMgAl LDH exhibited pronounced agglomeration and sedimentation at the container base. By day 8, the SA-ZnMgAl LDH also displayed relatively dispersed deposits. In contrast, the OA-ZnMgAl LDH showed no significant sedimentation even after 12 days, demonstrating excellent dispersion stability.

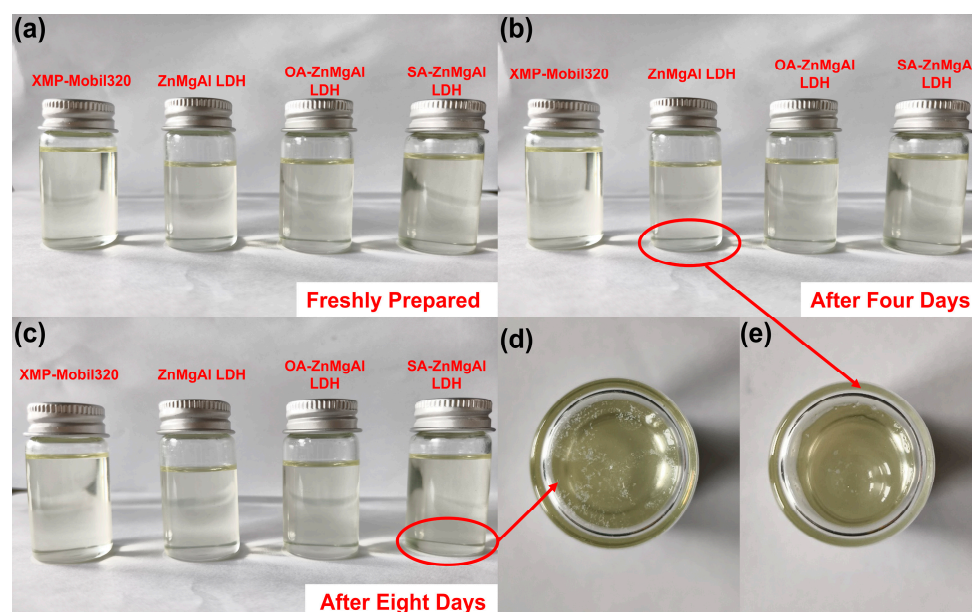


**Figure 6.** (a) Optical photographs of freshly prepared XMP-Mobil 320 oil samples without LDHs and with the addition of three LDHs, (b) optical photographs after four days of preparation, (c) Optical photographs after eight days of preparation, (d) precipitation optical photographs of oil samples with the addition of SA-ZnMgAl LDH, and (e) oil samples with the addition of ZnMgAl LD.

To further investigate the dispersion stability of LDHs in lubricating oils,  $\zeta$ -potential measurements were conducted on lubricants containing three types of LDH nanoadditives, with results presented in Figure 7. The  $\zeta$ -potential of the unmodified ZnMgAl LDH was 7.04 mV, whose absolute value fell significantly below 15 mV, indicating poor stability. In contrast, the modified OA-ZnMgAl LDH and SA-ZnMgAl LDH exhibited  $\zeta$ -potentials of  $-21.9$  mV and  $-26.7$  mV, respectively, with absolute values substantially exceeding 15 mV. This indicates enhanced electrostatic repulsion between particles and improved dispersion stability. Consequently, surface modification with OA and SA significantly enhances the dispersion and anti-agglomeration properties of LDH nanoadditives within oil.

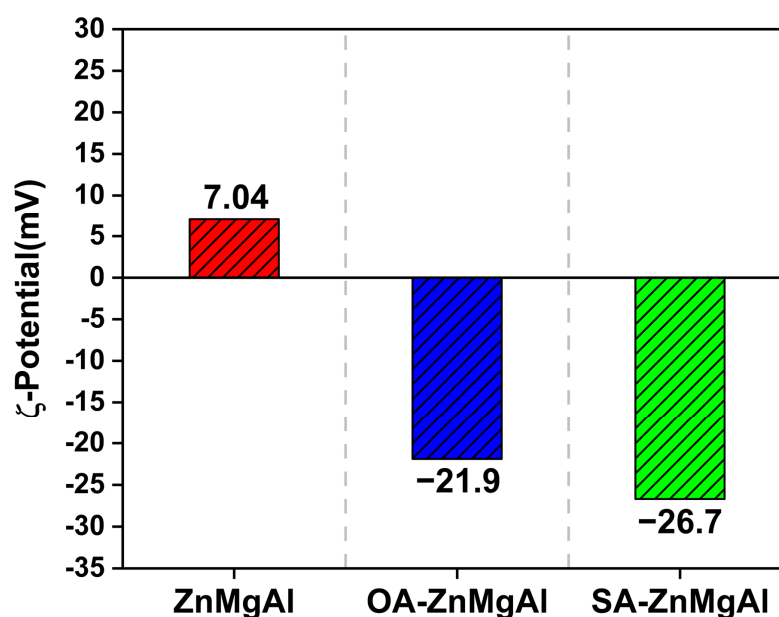


**Figure 7.**  $\zeta$ -potentials of ZnMgAl LDH, OA-ZnMgAl LDH and SA-ZnMgAl LDH lubricants with a mass fraction of 0.15 wt.%.

### 3.3. Anti-Friction and Wear Resistance Performance Testing

#### 3.3.1. Anti-Friction Performance Testing

First, the influence of three types of LDHs on the coefficient of friction (COF) at the same concentration (0.15 wt.%) was investigated and compared. The experimental results are shown in Figure 8a. During the initial phase of the experiment, the COF of all lubricant samples exhibited a trend of first increasing and then decreasing. This was attributed to the lubricant not yet fully penetrating the friction pair, preventing the COF from reaching a stable state. The COF of the lubricant without LDHs increased from 0.11 to 0.135 before decreasing and stabilising near 0.12, with an average of 0.117. Upon adding ZnMgAl LDH, the initial COF was lower with minimal fluctuation, and the average decreased to 0.113, indicating enhanced anti-friction properties. The OA-ZnMgAl LDH sample exhibited enhanced friction-reducing performance from the outset, with COF rapidly decreasing from 0.105 to 0.09 and stabilising throughout the test period. The average COF was 0.094, representing a reduction of approximately 20% compared to the base oil. The SA-ZnMgAl LDH sample initially rose slightly to 0.12 before rapidly declining, subsequently stabilising long-term around 0.095. Its average COF was 0.098, representing a reduction of approximately 16%. Overall, all three LDH types enhanced the lubricant's anti-friction capability and produced smoother friction curves. Among these, OA-ZnMgAl LDH demonstrated the most favourable performance, while SA-ZnMgAl LDH exhibited superior stability.

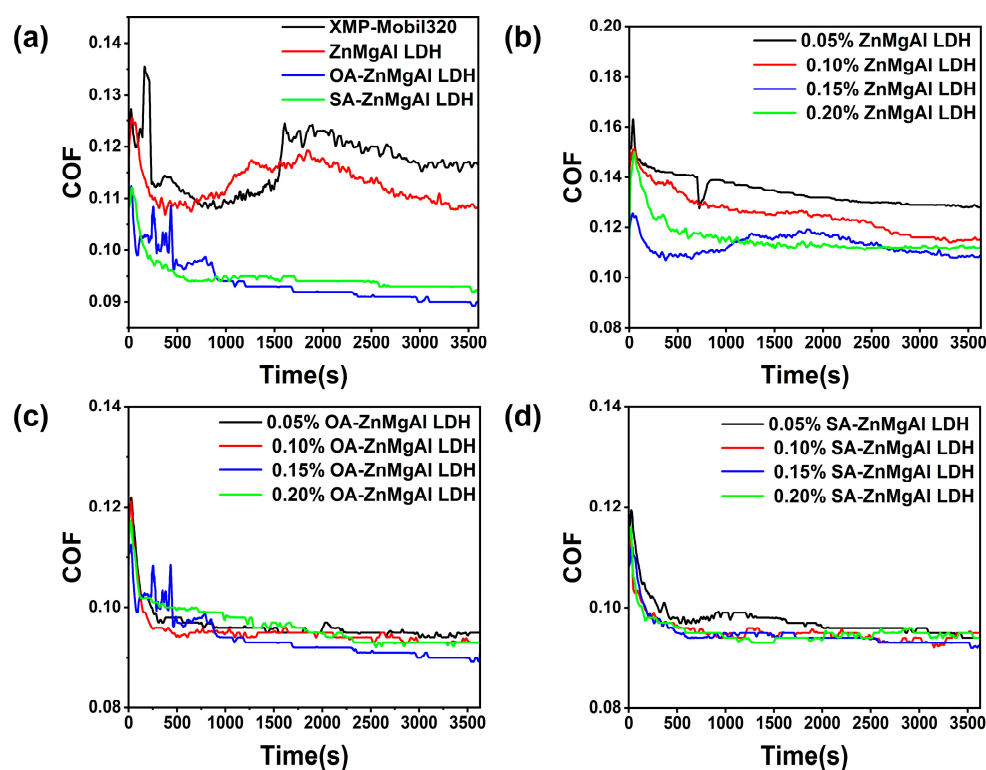


**Figure 8.** (a) COFs of XMP-Mobil 320, ZnMgAl LDH, OA-ZnMgAl LDH, and SA-ZnMgAl LDH. (b) COFs of ZnMgAl LDH as an additive to Mobil 320 (additions from 0.05 wt.% to 0.2 wt.%). (c) COF of OA-ZnMgAl LDH as an additive to Mobil 320 (additions from 0.05 wt.% to 0.2 wt.%). (d) COF of SA-ZnMgAl LDH as an additive to Mobil 320 (additions from 0.05 wt.% to 0.2 wt.%).

Subsequently, the influence of three nano-additive concentrations on lubricant anti-friction properties was investigated, as illustrated in Figure 8b–d. Figure 8b shows that unmodified ZnMgAl LDH achieved the lowest COF and optimal performance at 0.15 wt.%. At 0.05 wt.%, insufficient concentration prevented full anti-friction effect, resulting in the

highest COF. At 0.2 wt.%, despite increased loading, agglomeration between particles intensified, leading to diminished anti-friction performance.

In Figure 8c, OA-ZnMgAl LDH exhibited enhanced friction-reducing performance even at just 0.05 wt.%. As the concentration increased, the COF gradually decreased, reaching its lowest value at 0.15 wt.%. Subsequently, it rose slightly at 0.2 wt.%, again due to agglomeration effects diminishing the anti-friction performance. Figure 8d shows similar experimental results for lubricants containing SA-ZnMgAl LDH.

In summary, all three LDHs improve the friction properties of lubricating oils at different addition levels, though the effect is significantly influenced by concentration. At lower concentrations, the anti-friction effect is insufficient, while excessively high concentrations impair particle distribution at the friction interface due to agglomeration, leading to performance degradation.

### 3.3.2. Anti-Wear Performance Testing

A comparative study of the anti-wear properties of four lubricating oils was conducted using the four-ball test. As shown in Figure 9a, the base oil exhibited a wear scar diameter of 0.287 mm. Upon incorporation of 0.15 wt.% unmodified ZnMgAl LDH, the wear scar diameter decreased to 0.273 mm, representing a reduction of 5% and indicating enhanced anti-wear performance. Upon adding 0.15 wt.% OA-ZnMgAl LDH, the wear scar diameter further decreased to 0.247 mm, representing a 14% reduction, with anti-wear performance significantly superior to the unmodified sample. SA-ZnMgAl LDH also demonstrated better anti-wear effects than the unmodified sample, though slightly inferior to OA-ZnMgAl LDH.

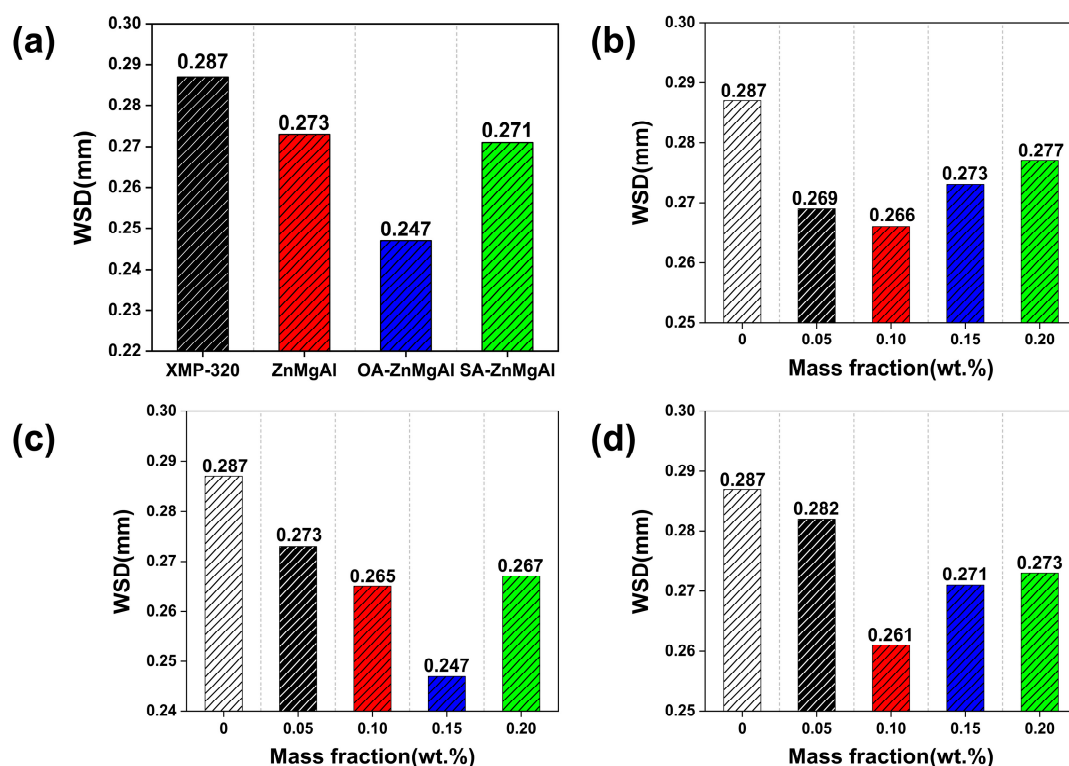


**Figure 9.** Effect of (a) additive type, (b) mass fraction of ZnMgAl LDH, (c) mass fraction of OA-ZnMgAl-LDH, and (d) mass fraction of SA-ZnMgAl-LDH on diameter of abrasive spots.

Figure 9b–d reveal the influence of concentration on anti-wear performance for the three distinct nano-additives. As shown in Figure 9b, unmodified ZnMgAl LDH already exhibits favourable anti-wear effects at a low addition level (0.05 wt.%), with a scuff

diameter of 0.269 mm. Optimal anti-wear performance was achieved at 0.1 wt.%, reducing the wear scar diameter to 0.266 mm—a 7% decrease. However, at 0.2 wt.%, the wear scar diameter markedly increased and anti-wear performance deteriorated, indicating that excessively high concentrations cause nanoparticle agglomeration, thereby impairing anti-wear properties.

The experimental results for OA-ZnMgAl LDH are shown in Figure 9c. Its anti-wear performance surpassed that of the unmodified sample at all concentrations. The trend resembled that of unmodified LDHs, but the scuff diameter reached its minimum value of 0.247 mm at an addition of 0.15 wt.%, representing a reduction of 14% and significantly outperforming other samples. At a concentration of 0.2 wt.%, anti-wear performance showed a slight decline, demonstrating its excellent anti-friction potential. As depicted in Figure 9d, the anti-wear performance trend of SA-ZnMgAl LDH mirrors that of OA-ZnMgAl. Optimal performance was achieved at 0.1 wt.%, with a wear scar diameter of 0.261 mm and a reduction of 9%. However, performance declined at excessively high concentrations.

In summary, all three LDH powders effectively reduce scuffing diameter and enhance lubricant anti-wear performance. The anti-wear efficacy of nano-additives is markedly concentration-dependent: insufficient concentrations yield inadequate nanoparticle distribution for optimal performance, while excessive concentrations promote agglomeration and uneven distribution, thereby diminishing anti-wear effects.

In the SRV tests, increasing the concentration of ZnMgAl LDH and SA-ZnMgAl LDH to 0.15 wt.% promotes the formation of a more continuous and lubricious boundary film. The greater availability of LDH nanosheets and organic species facilitates the generation of the tribofilm, thereby reducing the COF. In contrast, the four-ball configuration is subjected to higher Hertzian pressure and distinct lubrication conditions. At 0.1 wt.%, the LDH additives can already form a sufficiently protective tribofilm, leading to the minimum WSD. Further increasing the concentration to 0.15 wt.% may induce particle aggregation or produce a thicker but softer film, which can be disrupted under high load and may even cause slight three-body abrasion, preventing additional wear reduction.

Compared with pristine ZnMgAl LDH and SA-ZnMgAl LDH, the OA-ZnMgAl LDH benefits from the unsaturated structure of oleic acid. The cis-double bond imparts higher molecular flexibility and interfacial activity, which not only improves dispersion stability but also enhances the adaptability of OA-ZnMgAl LDH at elevated concentrations. As a result, at 0.15 wt.% the additive is able to form both a lubricious carbonaceous layer and a mechanically robust tribofilm, yielding simultaneous optimisation of friction reduction and wear resistance.

Furthermore, the superior COF reduction of OA-ZnMgAl LDH relative to SA-ZnMgAl LDH (20% vs. 16%) can also be attributed to these structural characteristics. While OA's unsaturation facilitates more effective adsorption and rapid interfacial film formation, the fully saturated and rigid alkyl chain of stearic acid limits its interfacial interaction capability, leading to comparatively weaker tribological performance

### 3.4. Analysis of Tribological Mechanisms

SEM/EDS characterisation of the four lubricated wear scars following the four-ball test was conducted to analyse their microstructure and chemical composition. The experimental results are presented in Figure 10. The base oil without nano-additives produced larger scuff marks with pronounced central scratches, indicating severe abrasive wear. Adding 0.15 wt.% ZnMgAl LDH slightly reduced the scuff diameter and mitigated central scratches, though minor marks remained observable (as shown in Figure 10b). In contrast, Figure 10c,d demonstrate that lubricating oils containing 0.15 wt.% OA-ZnMgAl LDH

and SA-ZnMgAl LDH exhibited not only significantly reduced wear scar dimensions but also shallower central scratches, alongside the emergence of minor spalling pits on the surface. These spalling pits may represent reaction products generated by the hydrotalcite nanoadditives during friction, or they could result from the detachment of powder particles during the friction process. EDS analysis indicates that without the addition of unmodified ZnMgAl LDH, elemental distribution was non-uniform with pronounced agglomeration. Following modification, LDH dispersion improved, mitigating agglomeration. Furthermore, on the worn surfaces lubricated with LDH-containing oil, Zn, Mg, and Al elements exhibited enrichment slightly deeper within the scratches. This suggests the presence of LDH particles or a protective film generated by LDH during friction at these locations.

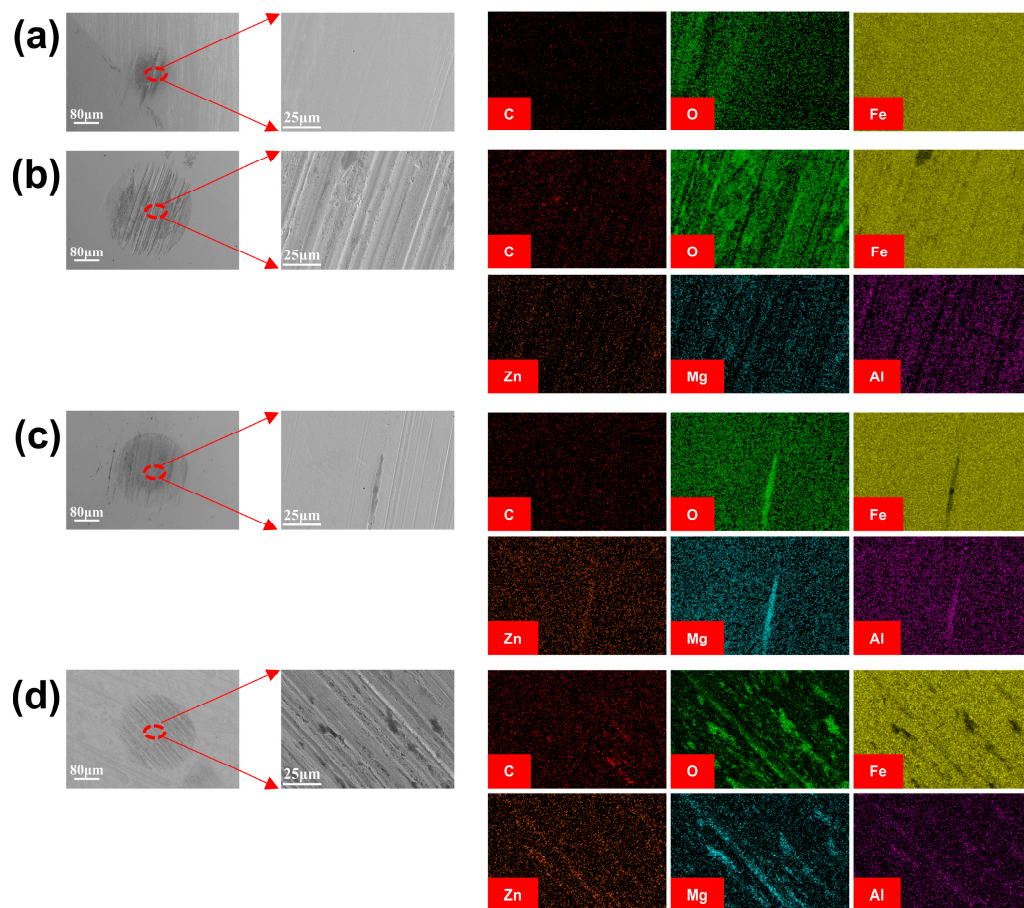


**Figure 10.** SEM/EDS Characterisation of Four-Ball Test Scratch Surfaces: (a) Mobil XMP-320 Base Oil, (b) ZnMgAl LDH, (c) OA-ZnMgAl LDH, and (d) SA-ZnMgAl LDH.

To further compare the effects of four different additives on scuff marks, their three-dimensional profiles were characterised using an optical profilometer. The experimental results are shown in Figure 11. Following lubrication with the base oil, pronounced deep grooves (blue regions) appeared at the centre and periphery of the scuff marks, accompanied by elevated ridge-like structures. This indicates inadequate oil film lubrication, resulting in direct contact between the friction surfaces. After adding ZnMgAl LDH (Figure 11b), wear conditions improved, though deep grooves remained observable at the centre. whereas after adding OA-ZnMgAl LDH and SA-ZnMgAl LDH (Figure 11c,d), particularly OA-ZnMgAl LDH, the wear scar surface became nearly flat with only minor abrasion marks. This demonstrates that modified additives significantly enhance the lubricating performance of the oil, exhibiting excellent synergistic lubrication effects with the base LDH.

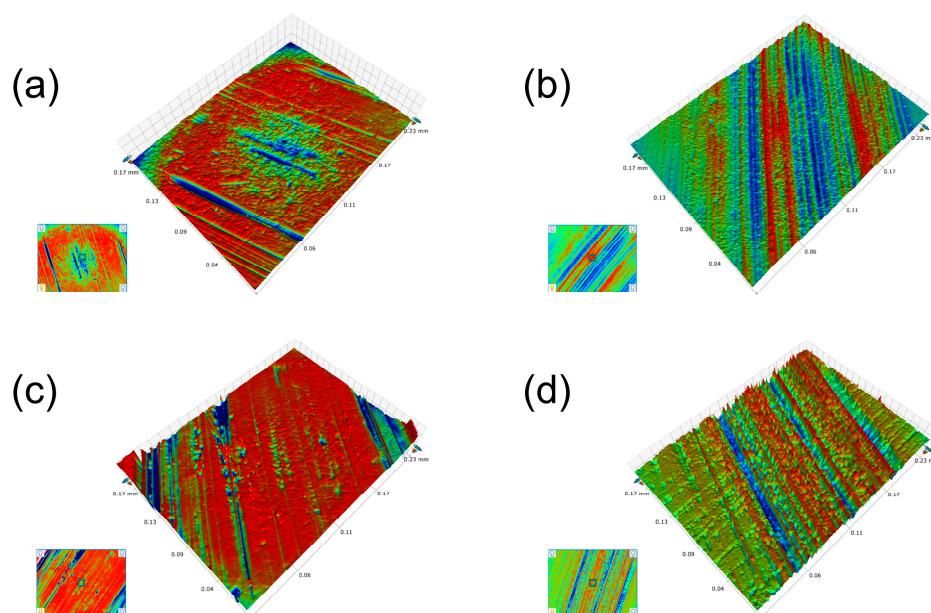


**Figure 11.** Three-dimensional optical images of four-ball test wear scars under constant parameters: (a) Mobil XMP-320 base oil, (b) ZnMgAl LDH, (c) OA-ZnMgAl LDH, and (d) SA-ZnMgAl LDH.

To further investigate the chemical composition of the friction film on the scuffed surface and elucidate the chemical constituents of residual elements on the scuffed surface, XPS scanning was performed on the modified additive scuff. The experimental results are shown in Figure 12. Figure 12a–f display the C 1s, O 1s, Fe 2p, Zn 2p, Mg 1s, and Al 2p spectra of the wear surface after the four-ball test. Peaks at 709.28, 711.48, and 715.00 eV in the Fe 2p spectrum confirm the presence of iron oxides ( $\text{Fe}^{2+}$ ,  $\text{Fe}^{3+}$ ), consistent with the iron oxide peak (529.48 eV) in the O 1s spectrum. This indicates that friction oxidation reactions occurred on the steel surfaces of all three samples. Two peaks observed in the C 1s spectrum of the worn surface confirm the presence of C–C fatty chains (284.8 eV) and C–O bonds (286.22 eV). This indicates that LDH was adsorbed onto the worn surface following lubrication with OA-ZnMgAl LDH and SA-ZnMgAl LDH. The 1303.80 eV peak in the Mg 1s spectrum and the 75.84 eV peak in the Al 2p spectrum are attributed to the formation of  $\text{MgAl}_2\text{O}_4$ . This results from the evolution of ZnMgAl LDH into LDO under the thermal effects generated during friction.

To further investigate the friction and wear mechanisms of lubricants, this study employed FIB-SEM to conduct cross-sectional observation and elemental analysis of the friction film formed during the four-ball test. Figure 13a illustrates the preparation process of the friction film. Figure 13b demonstrates that under lubrication conditions incorporating 0.15 wt.% OA-ZnMgAl LDH, a friction film approximately 2.5  $\mu\text{m}$  thick formed on the steel substrate surface. Compared to conventional thin films, combined with XPS analysis of the wear scar, this thicker friction film indicates that the friction interface undergoes not only transient physical adsorption but also accompanying friction chemical reactions. These reactions cause active components in the lubricant to react and deposit with the metal substrate, promoting gradual film growth and achieving continuous coverage. The results demonstrate that OA-ZnMgAl LDH exhibits high reactivity at the friction interface, promoting gradual film growth and continuous coverage. This film also prevents direct contact between friction pairs to a certain extent, thereby enhancing friction performance.

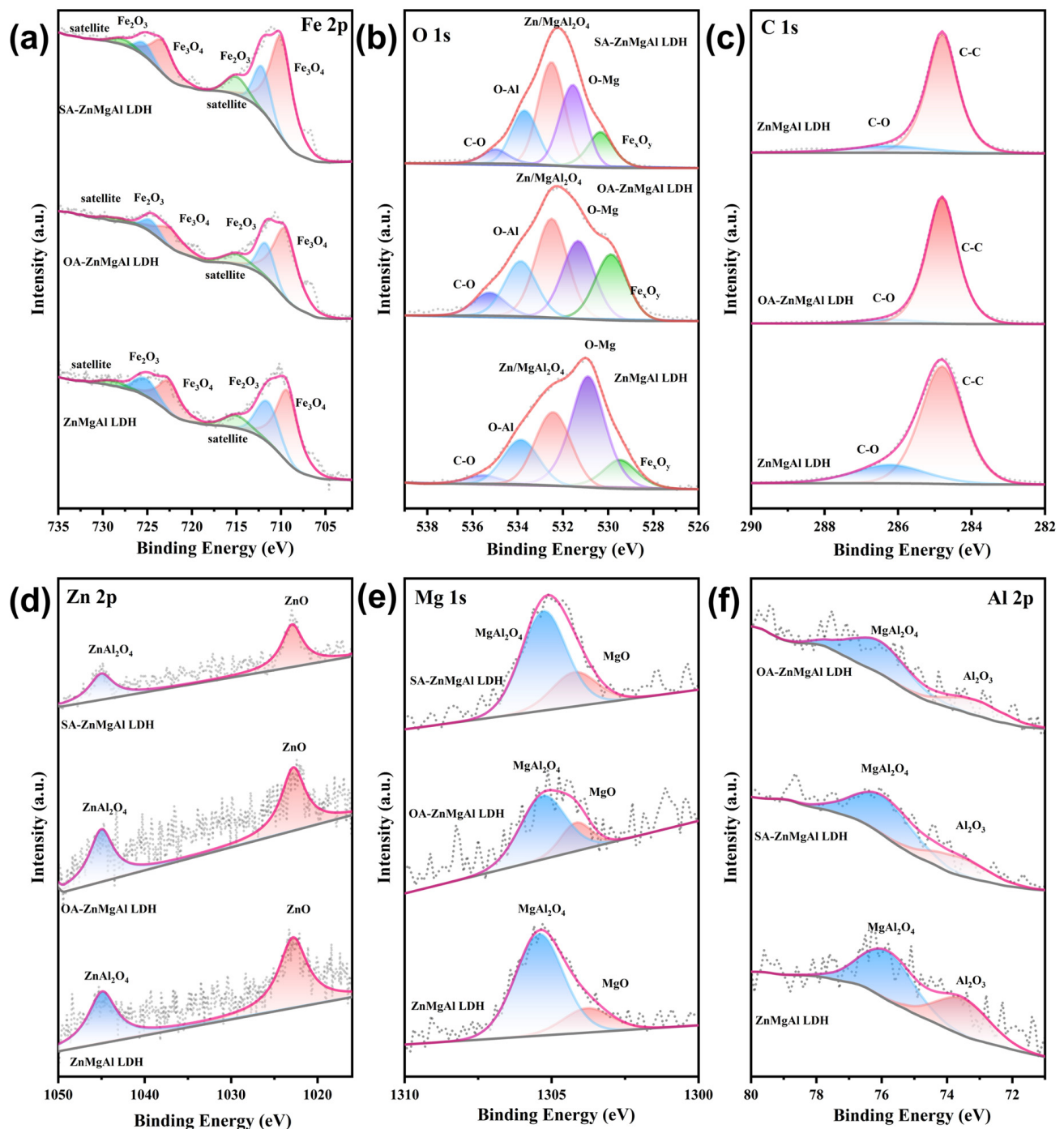


**Figure 12.** XPS images of the four-ball test wear scar under constant parameters: (a) Fe 2p, (b) O 1s, (c) C 1s, (d) Zn 2p, (e) Mg 1s, and (f) Al 2p.

EDS linear analysis along the scratch depth direction revealed significant enrichment of Fe and O elements within the friction film. This indicates that oxides such as  $\text{Fe}_2\text{O}_3$  and  $\text{MgO}$  formed and deposited during friction, enhancing the film's stability. In contrast, Zn, Mg, and Al elements exhibited sparser distribution. This is attributed to the limited incorporation of OA-ZnMgAl LDH and insufficient deposition at the interface. Despite their lower content, these elements still effectively induce oxide film formation.

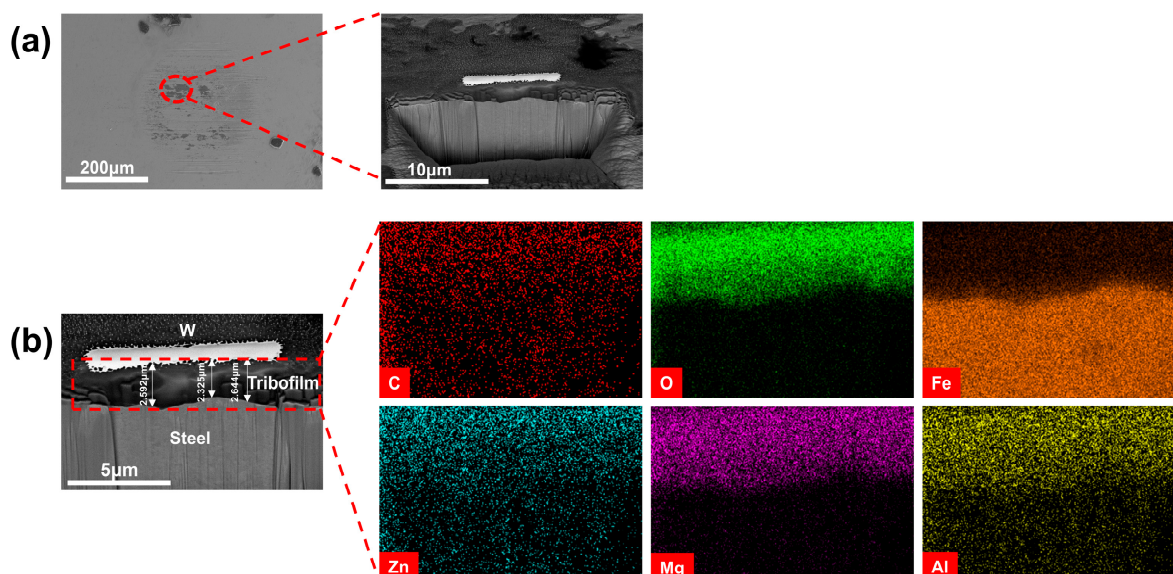


**Figure 13.** (a) Preparation Process of Friction Membrane Sections and (b) Morphology of the tribofilm and EDS elemental distribution along its depth.

Furthermore, EDS analysis indicates the presence of a graphite layer on the surface of the friction film, likely formed through friction-induced chemical reactions triggered by localised high temperatures. The graphite layer detected on the worn surface originates from both the organic modifiers (oleate/stearate) and the base oil [27,28]. Under shear and elevated temperature, the long-chain fatty acids grafted on OA-LDH and SA-LDH provide additional carbon-rich precursors, which facilitate tribochemical carbonization [29]. Compared with unmodified LDH, the modified LDHs may facilitate the formation of a more continuous carbonaceous tribofilm. This behaviour can be attributed to the improved dispersion stability of the modified LDH in oil, which enables the nanosheets to more readily and continuously enter the contact interface and facilitate the formation of a more uniform carbonaceous tribofilm. This graphite layer provides an interface with low shear strength, further reducing the coefficient of friction and mitigating surface wear. Multiscale characterisation combining SEM and EDS reveals that the friction interface, aided by OA-ZnMgAl LDH, forms a multilayer composite protective film structure comprising an inner oxide layer and an outer graphite layer. This composite film not only enhances lubrication conditions but may also induce redistribution of interface atoms and structural rearrangement, thereby playing a pivotal role in improving friction performance. Furthermore, LDHs not engaged in chemical reactions fill the worn surface, acting as rolling bearings and providing filler repair. The rolling bearing effect occurs when LDH nanoparticles distribute across the friction interface, transforming the sliding friction between the friction pair into rolling friction. This significantly reduces friction resistance at the contact interface and enhances the lubricating performance of the lubricant. The filling and repair effect arises from the extremely small size and large specific surface area of nanoparticles, enabling them to readily undergo physical adsorption on friction surfaces and fill damage areas such as microcracks and pits. This enhances the smoothness of the friction contact interface [30,31], hereby comprehensively improving the anti-friction and anti-wear properties of lubricants. Figure 14 comprehensively illustrates the anti-friction and anti-wear mechanism of LDHs.

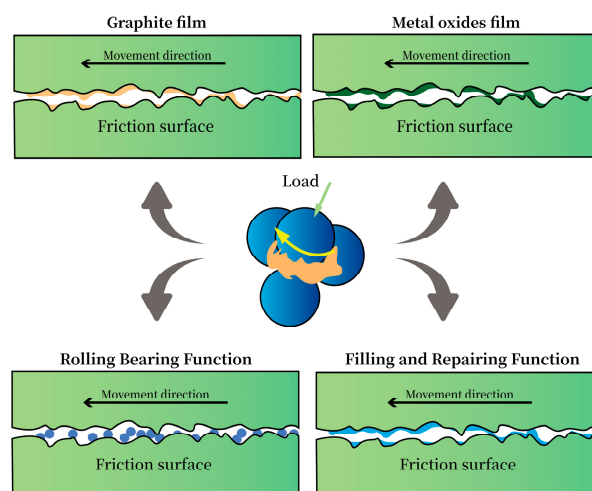


**Figure 14.** Anti-friction and Anti-wear Mechanism of LDHs.

#### 4. Conclusions

(1) ZnMgAl LDH was successfully synthesised via the coprecipitation method and further surface-modified with oleic acid and stearic acid through dehydration condensation reactions. Compared with unmodified ZnMgAl LDH, both OA-ZnMgAl LDH and SA-ZnMgAl LDH exhibited significantly improved dispersion stability. This improvement is attributed to the grafting of long-chain hydrophobic alkyl groups, which enhanced surface hydrophobicity and modified interparticle electrostatic interactions, enabling stable and uniform dispersion of LDHs in lubricating oils and effectively preventing agglomeration.

(2) Tribological results demonstrate that all three LDHs effectively improve the anti-friction and anti-wear performance of the base lubricating oil. The tribological performance strongly depends on the additive concentration. At low concentrations, insufficient protective film formation limits performance improvement, whereas excessive loading leads to particle agglomeration and deteriorates lubrication behaviour. The optimal anti-friction performance was achieved at an additive concentration of 0.15 wt.% for all three LDHs, with maximum COF reductions of 3.4%, 19.66%, and 16.24%, respectively. ZnMgAl LDH and SA-ZnMgAl LDH exhibited optimal anti-wear performance at 0.1 wt.% with wear scar reductions of 7.3% and 9.1%, while OA-ZnMgAl LDH achieved the best anti-wear performance at 0.15 wt.% with a wear scar reduction of 13.9%.

(3) Based on surface morphology and chemical composition analyses of the worn surfaces, a synergistic anti-friction and anti-wear mechanism is proposed for OA-ZnMgAl LDH and SA-ZnMgAl LDH. Under contact stress and shear during sliding, LDHs participate in tribochemical reactions to form a dense and continuous multilayer composite protective film consisting of an inner oxide layer and an outer graphite layer, effectively preventing direct asperity contact. Meanwhile, unreacted LDHs provide rolling bearing function and filling/repair functions. The combined action of these mechanisms significantly reduces friction resistance and wear at the contact interface. (4) From an industrial perspective, the excellent dispersion stability, low optimal additive dosage, and significant tribological performance enhancement of OA-ZnMgAl LDH and SA-ZnMgAl LDH demonstrate their strong potential as high-efficiency, low-content lubricant additives. These modified LDHs provide a promising strategy for improving the service life, energy efficiency, and reliability of mechanical systems under boundary lubrication conditions, indicating broad application prospects in advanced lubrication and industrial tribology.

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**Conflicts of Interest:** Authors Hao Xiao, Yihao Zhang, Xueqiang Ding, Mingmin Zheng, Qiuya Tua and Xin Zhang were employed by the company State Power Investment Corporation Research Institute Co., Ltd and author Yuan Xu was employed by the company Inner Mongolia Electric Power (Group) Co., Ltd. The remaining authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

## Abbreviations

The following abbreviations are used in this manuscript:

LDHs Layered double hydroxides

OA Oleic acid

SA Stearic acid

COF Coefficient of Friction

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