Diamond & Related Materials 52 (2015) 43–48

Contents lists available at ScienceDirect

Diamond & Related Materials

journal homepage: www.elsevier.com/locate/diamond

Graphene oxide in water lubrication on diamond-like carbon vs. stainless steel high-load contacts

Oskari Elomaa [a,*], Vivek K. Singh [a], Ajai Iyer [a], Timo J. Hakala [b], Jari Koskinen [a]

[a] Aalto University, School of Chemical Technology, Department of Materials Science and Engineering, Vuorimiehentie 2 A, 02150 Espoo, Finland
[b] VTT Technical Research Centre of Finland, P.O. Box 1000, FI-02044 VTT, Finland

A R T I C L E   I N F O

Article history:
Received 2 October 2014
Received in revised form 26 November 2014
Accepted 12 December 2014
Available online 20 December 2014

Keywords:
Graphene oxide
Diamond-like carbon
Water lubrication
Friction
Wear

A B S T R A C T

The friction and wear performance of 0–2 wt.% graphene oxide (GO) dispersions in water were studied on diamond-like carbon vs. stainless steel contact by a pin-on-disk tribometer. The wear surfaces were characterized using contact profilometry, X-ray spectroscopy, optical microscopy, scanning electron microscopy, energy dispersive spectroscopy, and Raman spectroscopy. The friction coefficient decreased 57% (from 0.14 to 0.06) compared to pure water, when 1 wt.% of GO and 10 N normal load were applied. A minimum wear for the counter ball was detected when pure water was used. GO additives reduced the corrosive effect on the counter ball. The main tribological mechanism of GO additives in water was observed to be the embedding of GO sheets on the counter ball surface forming a lubricating layer and binding water molecules into the contact.

1. Introduction

The limited non-renewable energy resources such as crude oil supplies and concern for the effects of CO_2 emissions on global temperature have raised the need for increased energy efficiency and development of renewable energy sources. The energy required to overcome friction is a major part of total energy consumption; in passenger cars, buses and trucks one third of the fuel energy is spent on overcoming friction [1,2]. The use of liquid lubricants to decrease friction is an efficient way to control friction and enable energy savings [1]. However, liquid lubricants are commonly based on mineral oils, which are by-products of the distillation of non-renewable petroleum. Water, on the other hand, is environmentally safe and globally available as a low cost lubricant. However, the low viscosity, corrosive properties and poor boundary lubrication properties [3] limit the use of water lubrication in mechanical applications that operate under high loads and are built of ferrous metals. It is possible to improve boundary lubrication properties of water and to provide better corrosion resistance for steel contacts by using soluble additives such as glycols, amines, ionic liquids, polymers and biomolecules [4] or insoluble additives such as graphite, nanodiamonds (NDs) and copper nanoparticles [4–8]. Diamond-like carbon coatings such as tetrahedral amorphous carbon (ta-C) have also been shown to provide low friction in water lubricated systems [9,10].

Graphene and graphene related materials have become an object of great scientific interest within the last decade due to their attractive multifunctional properties including; high surface area, low density, high electron transport properties, quantum hall effect, high thermal conductivity and ease of functionalization [11,12]. More recently graphene and its derivatives, such as graphene oxide (GO) and reduced graphene oxide have been studied for their lubricating properties as self-lubricating solids [13–16], as composites, [17–19] and as additives both in grease [20,21] and in liquid lubricants such as; oils [22,23], ethylene glycol [24] and water [25,26]. A thorough review on the lubricating properties of graphene and its derivatives has been written by Berman et al. [27]. Kinoshita et al. reported low friction coefficients of the level of 0.05 using monolayer graphene oxide sheets as water-based lubricant additives in steel–tungsten contact using a ball-on-plate setup [25]. Few other reports were found on the use of graphene or its derivatives as additives in water lubrication in contacts made of construction materials such as steel. For industrial purposes, understanding and controlling especially the macroscale tribological properties of graphene and its derivatives in potential lubricants for oil replacement such as water would be important in the future.

In this work the friction and wear performance of GO dispersions in water lubrication on ta-C vs. stainless steel contacts were studied. A commercial ND dispersion was also tested for comparison. Due to the polarity of water a good dispersivity of the polar GO particles was achieved. The ta-C coating was chosen due to its good tribological properties in water. Test parameters were selected to provide boundary lubrication conditions. The tribological tests were conducted using a pin-on-disk setup (POD) with a 0.02 m/s speed and 10 N load to ensure

* Corresponding author. Tel.: +358 50 460 2743.
E-mail address: Oskari.Elomaa@aalto.fi (O. Elomaa).

http://dx.doi.org/10.1016/j.diamond.2014.12.003

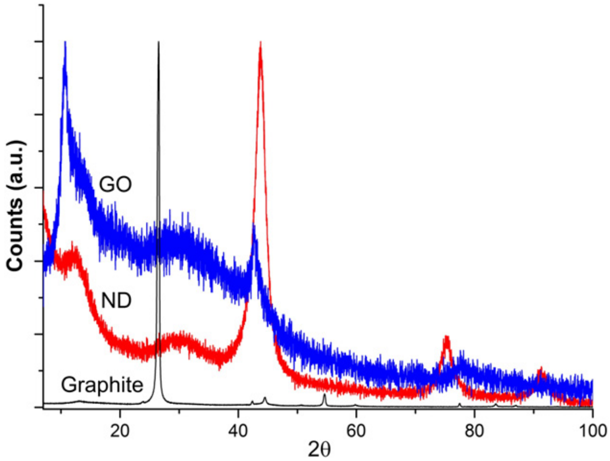

Fig. 1. XRD spectra of graphene oxide, nanodiamonds and synthetic graphite.

boundary lubrication. GO concentrations from 0 to 2 wt.% in water were employed as lubricant. The results will provide an understanding of the GO properties in water lubrication on the ta-C–stainless steel contact.

2. Materials and methods

In this part the experimental methods and materials used in the tests are discussed.

2.1. GO synthesis, ND details

GO was synthesized from synthetic graphite powder. The process and characterization of the GO is described in detail in our previous work [24]. The materials used in the processing were graphite powder (<20 μm synthetic) and potassium permanganate ($KMnO_4$, ACS reagent ≥ 99%, low in mercury), sulphuric acid (H_2SO_4, ACS reagent, 98%), phosphorus pentoxide (P_2O_5, ≥99.99% with trace metals) and potassium persulfate ($K_2S_2O_8$; ACS reagent, ≥99.0%), H_2O_2 (30 wt.% in H_2O, ACS reagent), and hydrochloric acid (ACS reagent, 37%) Dispersion with NDs in water (Andante 2 wt.%), was purchased from Carbodeon Ltd. The lower concentrations of NDs in water were prepared by diluting the original material with DI water. Ultrasonication was used to ensure even distribution of NDs after changes in concentration. The average particle size of the NDs according to the manufacturer was approximately 4 nm. GO and ND powders on silicon wafers were characterized using X-ray spectroscopy (XRD), Raman spectroscopy (Raman) and scanning electron microscopy (SEM).

2.2. Characterization methods

Raman spectra were taken on a Horiba Jobin-Yvon Labram HR Raman using 488 nm Argon laser excitation. Wide-angle X-ray diffraction (XRD) analyses were carried out on a Philips X'Pert PRO system by using Cu Kα (λ = 0.154 nm) radiation at 45 kV and 40 mA in the range of 2θ = 50–700. Scanning electron microscopy (SEM) images were obtained using TESCAN MIRA3 FEG-SEM and Hitachi S-4700 FEG-SEM. Profilometry was done by a Veeco Dektak 6M stylus profilometer. Optical microscopy was done by a Leica DRMX microscope.

2.3. Sample preparation

Steel disks (AISI 52100) with a diameter of 40 mm were ground with a P1200 grit paper and ultrasonically cleaned with acetone and ethanol. The ta-C films were deposited using a pulsed cathodic arc deposition system. The ambient pressure in the deposition chamber was approximately 3.0×10^{-4} Pa. The samples were sputter-cleaned prior to deposition using an Argon beam from a broad-beam ion source. A thin Ti intermediate layer was deposited on the substrate to improve adhesion using a metal plasma source from a filtered DC cathodic arc device. The ta-C film was deposited by a direct carbon arc. The growth rate was about 0.7 μm/h and the final coating thickness was approximately 0.3 μm as measured by a profilometer. The samples were rotating in a planetary motion in order to provide a uniform coating thickness to all specimens. The substrate temperature during the depositions was less than 60 °C leading to good quality ta-C as characterized by Raman spectroscopy (Supplementary data). The average surface roughness of the disks after the deposition was 30 nm, as measured by profilometry.

2.4. Tribological testing and characterization

The tribological properties of the GO and ND dispersions in water surfaces were studied using a POD tribometer designed and developed at Aalto University. All tests were carried out in a temperature controlled laboratory environment (temperature 22 ± 1 °C) using ta-C coated steel disks described in Section 2.3 as samples. POD test parameters were 10 N normal load, sliding velocity of 0.02 m/s and sliding distance of approximately 36 m. The average radius of the wear track was 15 mm and stationary stainless steel (AISI420) bearing balls of 10 mm diameter were used as pins. The concentration of GO dispersions in water was varied from 0 wt.% to 2 wt.%. For comparison purposes a ND dispersion of 1 wt.% was also tested. The total volume of the lubricant used in POD tests was 0.3 ml and the total test time was 30 min. The lubricant was applied onto the rings with the periodicity of 5 min (approximately 50 rounds) as droplets with a pipette. All tests were repeated at least three times and a few comparison tests were also conducted with chosen concentrations of GO using an increased amount of

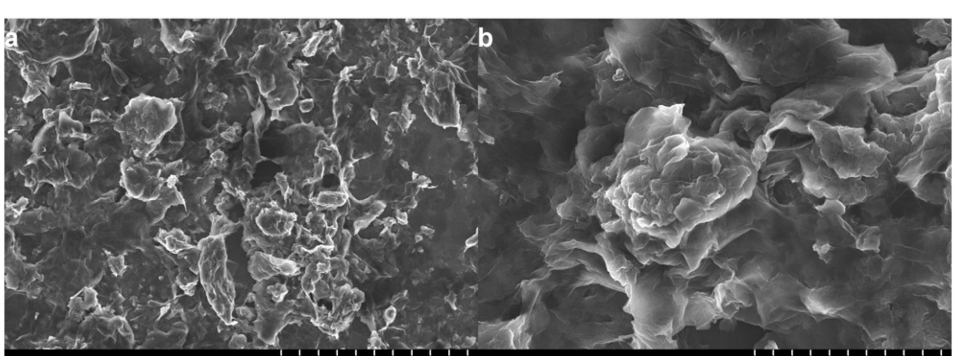

Fig. 2. SEM images of GO water dispersion dried on silicon wafer (a) showing in detail the layered morphology of GO sheets (b).