



# Studies of drag on the nanocomposite superhydrophobic surfaces



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## ABSTRACT

The nanocomposite thin films of stearic acid (SA)-functionalized ZnO nanoparticles incorporated in epoxy polymer matrix have been achieved. The X-ray diffraction (XRD) studies show the formation of zinc stearate on ZnO nanoparticles as the confirmation of SA-functionalization of ZnO nanoparticles in the thin films. Morphological analyses reveal the presence of micro-holes with the presence of irregular nanoparticles. The measured root mean square (*rms*) roughness of the thin film is found to be  $12 \pm 1 \mu\text{m}$  with the adhesion of 5B on both glass and aluminum substrates. The wetting property shows that the surface of the film is superhydrophobic with the contact angle of water of  $156 \pm 4^\circ$  having contact angle hysteresis (CAH) of  $4 \pm 2^\circ$ . The average terminal velocity in the water of the as-received glass spheres and superhydrophobic spheres were found to be  $0.66 \pm 0.01 \text{ m/s}$  and  $0.72 \pm 0.01 \text{ m/s}$  respectively. Consequently, the calculated average coefficients of the surface drag of the as-received glass sphere and superhydrophobic glass sphere were  $2.30 \pm 0.01$  and  $1.93 \pm 0.03$ , respectively. Hence, the drag reduction on the surface of superhydrophobic glass sphere is found to be approximately 16% lower than as-received glass sphere.

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## 1. Introduction

One the most studied phenomenon, at the present in the research in nanomaterials, is the wettability of the surfaces. The wetting phenomena can be categorized into four parts based on the contact angle of a water drop on a surface. The complete wetting signifies the contact angle on a surface is zero degree that refers as superhydrophilic. The contact angle below  $90^\circ$  is considered to be hydrophilic and the contact angle above  $90^\circ$  is considered to be hydrophobic. On the other hand, the surface said to be superhydrophobic when the water contact angle is more than equal to  $150^\circ$ . The superhydrophobic surfaces are surfaces that easily repel water. The ability of repelling water is a well-known phenomenon that occurs frequently in nature. Several insects and plants possess this ability. Among them the most studied and well understood surface is the lotus leaf, the symbol of purity, that has been analyzed through scanning electron microscope by Neinhuis and Barthlott in 1997 [1]. They observed that the surface of lotus leaf is not very smooth, as believed due to non-wetting properties, rather rough due to the co-existence of both micro and nano motifs. They also

observed that these motifs are also covered with a low surface energy waxy material [1]. The micro-nano pattern on the surface allows large amount of air to be entrapped into the pattern while the low surface energy waxy coating inhibits the interaction with the water. These two characteristics are the crucial parameters to obtain the superhydrophobic surfaces. Wenzel [2] as well as Cassie–Baxter [3] proposed two mathematical models to explain the wetting phenomena on rough surfaces that lead to superhydrophobic properties.

In Wenzel model [2], the water drop penetrates the surface irregularities. Mathematically, Wenzel equation is written as

$$\cos \theta' = R_w \cos \theta \quad (1)$$

where the roughness factor  $R_w$  is the ratio of the true and apparent (geometric) surface areas. According to the Wenzel model, when the true contact angle  $\theta$  of water on a smooth surface is less than  $90^\circ$ , the apparent contact angle  $\theta'$  is less than the true contact angle  $\theta$  on a rough surface, and when the true contact angle  $\theta$  is greater than  $90^\circ$ , the apparent contact angle  $\theta'$  is greater than the true contact angle  $\theta$  on a rough surface.

The Cassie–Baxter model [3] describes the effect of roughness on chemically heterogeneous structures where the apparent

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contact angle is mathematically derived from the Cassie equation as follows:

$$\cos \theta' = f_1 \cos \theta_1 + f_2 \cos \theta_2 \quad (2)$$

where  $\theta'$  is the apparent contact angle of the composite coating consisting of two components with contact angles  $\theta_1$  and  $\theta_2$  and corresponding area fractions  $f_1$  and  $f_2$ . In such a composite system  $f_1$  is assumed to be the solid surface and  $f_2$  is assumed to be air where  $\theta_2$  is  $180^\circ$  and as  $f_1 + f_2 = 1$ , Eq. (2) can be written as

$$\cos \theta' = f_1(\cos \theta_1 + 1) - 1 \quad (3)$$

This equation explains that on a rough surface with large amount of air entrapment, in the surface irregularities, one can achieve a highly superhydrophobic surface with a very small area fraction  $f_1$  of the surface in contact with the water drop. The configuration leads to a very high contact angle and a very low contact angle hysteresis leading to the rolling-off of water drops on the contacting solid.

Accordingly to the Cassie–Baxter model, a water drop on a pattern surface, e.g. surface of a lotus leaf, will be in contact with a very little solid fraction and rest on the entrapped air into the pattern that effectively increase the water contact angle more than  $150^\circ$ .

Each year, plenty of articles are published in the field of superhydrophobic surfaces. In general, various high surface energy materials like metals, oxides ceramics are used to obtain a surface with an optimum topography and further modified those surfaces with low surface energy polymers or organic molecules, to obtain superhydrophobic surfaces. Our group is actively working in this field and fabricated several superhydrophobic surfaces using fluorinated silica nanoparticles, methylated electromodified copper and aluminum surfaces and rf-sputtered Teflon coated etched aluminum surfaces [4–11].

The applications of the superhydrophobic surfaces are diverse. The main applications are the water repellency which is mainly used in the textile industries, the self-cleaning surfaces, now used on several building windows [4,5]. We have also shown that the superhydrophobic surfaces having the corrosion resistance properties as well as ice adhesion reduction properties [8,9]. Another important property of superhydrophobic surfaces is the reduction of surface drag. The drag reduction has the potential application in the energy consumption of moving vehicles in our daily life.

Drag is a frictional force that acts to the opposite to the motion of the moving fluids. The drag reduces the speed, and hence the efficiency of the moving cars, the aircrafts and even the boats [12]. The drag reduction is an emerging technology in automotive engineering. For reducing this effect, several geometries have been proposed to improve aerodynamic and hydrodynamic of the vehicles. The most recent development in this topic is related to the fabrication of nanostructured superhydrophobic surfaces [13]. Several authors proposed superhydrophobic coating methods that reduce drag and as much as evaluation methods for the drag reduction measurements over those surfaces [14–18]. The materials used to obtain the surfaces are varied, such as polymers [16,17,19], sol–gel method [20,21] and nanostructured metals [22,23].

Apart from the water-drag on the superhydrophobic surfaces, studies of air-drag are also of interest to the automotive and aerospace industries. It has been shown that the air drag can be reduced by 4–5% by engineering the surface with riblet structure of  $62 \mu\text{m}$  [24]. Similarly, Stenzel et al. proposed a paint based on aliphatic polyurethane resins that shows air-drag reduction of 6% [25]. A European patent shows an air-drag reduction of 10% with a composite coating, on a golf ball, consisting of silica particles in polyurethane [26]. Also, a United States patent shows that a hydrophobic coating can reduce the air-drag with a composite coating of acrylic and of fumed silica, hydrophobic titania, and zinc oxide [27].

In this article, we describe the studies of surface drag of water on a superhydrophobic surface. The surface is a nanocomposite thin film of epoxy polymer containing stearic acid functionalized ZnO nanoparticles prepared by spray deposition.

## 2. Experimental

The ZnO nanoparticles having average diameter of 70 nm (purchased from MK Nano, Ontario, Canada) were functionalized with stearic acid (SA) organic molecules using 0.1 M ethanolic solution in an ultrasonic bath. The SA-functionalized ZnO nanoparticles were separated from ethanol by centrifuging process and further dried in an oven at  $70^\circ\text{C}$  for more than 10 h. These functionalized ZnO nanoparticles were then dispersed into toluene utilizing an ultrasonic bath for 10 min. Furthermore, a commercial paint, based on an epoxy polymer (Bepoxy) has been added into the toluene solution that contains SA-functionalized ZnO nanoparticles. This mixture of epoxy polymer and SA-functionalized ZnO nanoparticles has been sprayed on both flat glass surfaces as well as on surface of spheres (glass) to obtain superhydrophobic nanocomposite thin film. The prepared flat substrates were used for the surface characterization on the other hand the spherical objects were used for drag studies. Sprayed substrate thin films coatings have been characterized using X-ray diffraction (Bruker D8 Discover System), and Scanning electron microscope (SEM: JEOL JSM-6480 LV). The chemical composition of surfaces was analyzed by infrared reflection absorption spectroscopy (IRRAS, Nicolet 6700FT-IR). The surface roughness of the films was measured using an optical profilometer (MicroXAM-100 HR 3D surface profilometer). The adhesion of the nanocomposite thin films coating has been evaluated using ASTM D3359-02: Standard Test Methods for Measuring Adhesion by Tape Test. ASTM D3359-02 is a test method for assessing the adhesion of the coatings to metallic substrates by applying and removing pressure sensitive tape over cuts made in the film. The adhesion of the films is evaluated by a rating from 0B where more than 65% of the coating is removed to 5B where 0% of the coating is removed, a perfect adhesion.

The wetting properties of the coating were performed by measuring both static and dynamic contact angles (Krüss contact angle goniometer) at five positions on each substrate using a  $5 \mu\text{L}$  deionized water drop. The static contact angle has been abbreviated as CA and dynamic contact angle or contact angle hysteresis has been abbreviated as CAH throughout the text. The CAH is the difference between the advancing and the receding angle. The CAH measurements were made at room temperature following a very standard and commonly used experimental procedure as reported in the literature [10,28,29]. In this method, a water drop of volume  $\sim 5 \mu\text{L}$  was suspended with the needle and brought in contact with the superhydrophobic surfaces using a computer controlled device as provided by Krüss GmbH. The contact CAH was measured by holding the water drop with a stationary needle in contact with the surface and moving the goniometer stage in one direction.

A water column of height 100 cm and diameter 22 cm was used for the studies of surface drag of a falling object. The falling objects were as-received and superhydrophobic glass spheres of diameter  $\sim 1.2 \text{ cm}$  with an average weight of 5.5 g. The time of falling of each sphere in the water column at room temperature is recorded using a high speed camera (Motionblitz EoSens Cube7 Mikrotron GmbH) having the recording speed of 300 frames per second. Ten as-received as well as ten superhydrophobic spheres were used to perform the drag test on hydrophilic (as water drop spread on it) and superhydrophobic surfaces.

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