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The effect of aluminium nanocoating and water pH value on the wettability behavior of an aluminium surface



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ABSTRACT

Experimental investigation was performed to highlight the influence of ionic bounding and surface roughness effects on the surface wettability. Nanocoating technique via e-beam physical vapor deposition process was used to fabricate aluminium (Al) film of 50, 100, and 150 nm on the surface of an Al substrate. Microstructures of the samples before and after deposition were observed using an atomic force microscopy. A goniometer device was later on used to examine the influence of surface topography on deionised water of pH 4, 7 and 9 droplets at a temperature ranging from 10 °C to 60 °C through their contact angles with the substrate surface, for both coated and uncoated samples. It was found that, although the coated layer has reduced the mean surface roughness of the sample from 10.7 nm to 4.23 nm, by filling part of the microstructure gaps with Al nanoparticles, the wettability is believed to be effected by the ionic bounds between the surface and the free anions in the fluid. As the deionised water of pH 4, and 9 gave an increase in the average contact angles with the increase of the coated layer thickness. On the other hand, the deionised water of pH 7 has showed a negative relation with the film thickness, where the contact angle reduced as the thickness of the coated layer was increased. The results from the aforementioned approach had showed that nanocoating can endorse the hydrophobicity (unwitting) nature of the surface when associated with free ions hosted by the liquid.

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1. Introduction

Nanoparticles have gained wide recognition in a variety of industrial and commercial applications over the years, such as sunscreen products [1], medicine [2,3], electronics [4], transportation [5], reduction of buildings pollution [6], magnetic sealing [7], microbial fuel cells [8], space and defence [9], and structural applications [10].

Alumina (Al₂O₃), in specific, possesses a variety of industrial and commercial uses and has become one of the important elements that is used in manufacturing commercial ceramic materials [11,12]. On the nano scale, nanoparticles of Al₂O₃ have been used to produce nanocomposites [13,14], polymer modification [15],

textiles functionalization [16], wastewater treatment [17], heat transfer fluids [18], surface coating [19], and as catalysis [20–22].

Surface friction and wettability are important in many of these applications, however, they require further advancement. Whereas in piping systems, the inner pipe surface friction plays a prominent role in the determination of the pressure drop through the head losses along the pipelines for flows of turbulent state. This is theoretically proven using the non-linear relationship between the Reynold number (*Re*) and the implicit Colebrook–White equation [23], which are expressed in form of a formulation as,

$$Re = \frac{V \cdot D}{D} \tag{1}$$

$$\frac{1}{\sqrt{f}} = 1.14 - 2\log_{10}\left(\frac{\varepsilon/D}{3.7} + \frac{2.523}{Re\sqrt{f}}\right)$$
 (2)

where v, D, V, f, and ε are kinematic viscosity, inner pipe diameter, inner pipe average flow velocity, Darcy-Welsbach friction factor, and roughness height, respectively.

On the other hand, surface wettability is determined by the angle of contact between a liquid droplet and the surface in contact

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to it [24]. Where the surface is called "super-hydrophilic" if the angle is less than 5°, "hydrophilic" if the angle is between 5° and 90°, "hydrophobic" if the angle is between 90° and 150°, and "su per-hydrophobic" if the angle is greater than 150° [24,25]. The term hydrophilic reflects the tendency of the fluid to form a strong bond with the surface, where the term hydrophobic indicates the propensity of the fluid to repel from the surface. Once a liquid of an ionic nature, such as water, comes in contact with an aluminium (Al) surface, an ionic reaction occurs in any of the following forms [26,27].

$$Al + 3H_2O \rightarrow Al(OH)^3 + 3H^+ + 3e^-$$
 (3)

$$Al + 3/2H_2O \rightarrow Al_2O_3 + 3H^+ + 3e^- \eqno(4)$$

$$Al + 2H_2O \rightarrow AlO(OH) + 3H^+ + 3e^-$$
 (5)

Such reaction leads to a reduction of oxygen (O₂) atoms and production of hydroxyl ions (H⁺) and hence changes the hydrophobicity/hydrophilicity nature of the surface [19].

Fluid-solid interaction has its own importance in a range of applications such as designing of water repelling surfaces [28] to fluid flow manipulation in piping systems [24] and inhabitation of machinery corrosion [29].

Kang et al. [24] studied in their work the effect of surface nanocoating on the reduction of liquid pumping power by modifying the contact angle (CA) of the riser surface. The CA's examined were between 23.7° and 153.8° with the highest pumping power efficiency obtained at a CA of 90.3° for the silicon dioxide (SiO₂) coating of concentration 6.67×10^{-3} wt%.

Zhang et al. [30] considered the improvement of heat exchanger, which consist of fins and tubes, thermal performance by depositing titanium dioxide (TiO_2) film on an Al substrate. Baking temperatures of 150 °C, 250 °C, 350 °C, 450 °C, and 600 °C were implemented using an electric muffle furnace for surface treatment. For the same Re and relative air humidity (RH) condition, the heat transfer coefficient was found to be the highest after baking the coated substrate at a temperature of 250 °C. But decreased when the baking temperature increased above 250 °C due to the reduction in coated film area.

Phan et al. [31] investigated experimentally, using nanocoating techniques, the surface wettability effect on nucleate boiling heat transfer. Water CA on a stainless steel grad 301 substrates was varied from 22° to 112° by depositing different coating materials. They found that greater surface wettability decreased the bubble emission frequency but raised the vapor bubble departure radius. Moreover, lower superheat temperature was required to generate the bubbles growth on a hydrophobic surfaces. They also noticed the tendency of bubbles to merge together forming a vapor blanket on the hydrophobic surface leading to critical heat flux (CHF).

Akbari et al. [32] studied the enhancement of saturated pool boiling of distilled water under atmosphere pressure where they formed a layer of silver nanoparticles on a copper substrate by boiling silver nanofluid of 0.025 and 0.05 vol%. They found that higher particles concentration increased the clustering deposition and surface hydrophobicity, however stability of the deposition was reduced. Their results also showed that, the heat transfer coefficient (HTC) and CHF improved by reaching a nanocoated polished surface state.

In this study, the CA and surface roughness of an Al substrate were modified through electron beam physical vapor deposition (e-beam PVD) coating technique to enhance the ionic interaction between the surface and the water droplet. An atomic force microscopy (AFM) device was used to measure the reduction in surface roughness after coating the substrate with 50, 100, and 150 nm thick layers. Furthermore, a goniometer devices was used in an

attempt to understanding the influence of free ions imbedded in the liquid on the attached surface CA.

2. Experimental

2.1. Materials

All chemicals were used as-received without further purification. Hydrochloric acid (HCl $\sim\!\!37\%$) grad ACS reagent and acetone (CH $_3$ COCH $_3 \geq 99.5\%$) grad ACS reagent were purchased from SIGMA-ALDRICH, and sodium hydroxide pellets (NaOH $\sim\!\!98\%$) grad AR was purchased from LOBA Chemie. Al pellets, 3.175 mm diameter and 6.35 mm height, of 99.99% purity was purchased from Kurt J. Lesker Co. Four cylindrical shaped, 25 mm diameter and 15 mm height, substrates of 92.5% Al were manufactured using a computer numerical control (CNC) machine.

2.2. Preparation of aluminium coatings

Aluminium pellets were placed in a 8 cm³ graphite crucible to be used as the coating material source and the Al substrate was cleaned by a Soniclean company digital benchtop ultrasonic cleaner filled with acetone for 20 min after which it was wiped carefully before tightly adjusted to the sample holder and positioned vertically inside the e-beam PVD device. The e-beam PVD device chamber was then vacuumed to a pressure of 346.64×10^{-6} pa to insure the removal of all particle contaminations within it and to control the level of evaporation. The Al pellets were later on partially evaporated from the crucible and the Al vapor deposited on the substrate surface with a deposition rate of 0.1 Å/s to form a 50, 100, and 150 nm thick layers. After the particle deposition process completion, the substrate was left in the chamber for 4 h to cool down before removal. The coating procedure used for the preparation of the Al layer on the substrate is shown in Fig. 1.

2.3. Characterization

Elemental analysis of the Al substrates was performed three times and averaged using a BRUKER TITAN S1 X-ray fluorescent (XRF) handheld analyser to insure that the bulk components in the manufactured substrate were of Al base. This was done by placing the substrate on the working station and adjusting the XRF device lens vertically on the substrate before starting the measurements which required 10 s to complete for each measurement. Additional elemental test was performed through a 9 kW Rigaku SmartLab, Japan, X-ray diffraction (XRD) analyser and its software, SmartLab Guidance, using a Cu Kα X-ray source with a diffraction angle of 2θ and an incidence beam angle of 0.02° to determine the Bragg's peaks of each element contained in the substrate. The diffraction scanning angle ranges were from 20° to 80° at a scanning rate of 2°/min. A Keysight Technologies 5600LS AFM in tapping mode was used to illustrate the changes in surface topography of the coated and uncoated substrates. The particles size of Al₂O₃ film and surface roughness were determined using Mountview software. Surface wettability was measured by preparing three beakers of 250 ml that were filled with 150 ml of deionised water (DIW) of pH 7 in each. Two of the three DIW, contained in the beakers, pH levels were adjusted to 4 by adding HCl and 9 by adding NaOH. The pH level in each beaker was measured using a HACH HQ11D portable pH meter with accuracy of 0.002 pH. The samples were then used separately to fill one of three 500 µl glass syringes, purchased from Hamilton company, each time. The three syringes containing DIW of pH 4, 7, and 9 were then placed on the Dataphysics OCA 100 contact angle goniometer device automatic multi liquid dispenser. The surface

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