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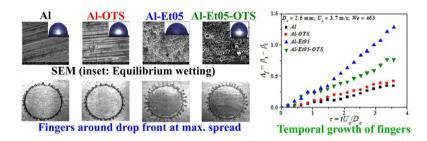
Impact dynamics of high Weber number drops on chemically modified metallic surfaces



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GRAPHICAL ABSTRACT



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ABSTRACT

Hydrophobic/superhydrophobic metallic surfaces prepared via chemical treatment are encountered in many industrial scenarios involving the impingement of spray droplets. The effectiveness of such surfaces is understood through the analysis of droplet impact experiments. In the present study, three target surfaces with aluminum (Al-6061) as base material-acid-etched, Octadecyl Trichloro Silane (OTS) coated, and acid-etched plus OTS-coated-were prepared. Experiments on the impact of inertia dominated water drops on these chemically modified aluminum surfaces were carried out with the objective to highlight the effect of chemical treatment on the target surfaces on key sub-processes occurring in drop impact phenomenon. High speed videos of the entire drop impact dynamics were captured at three Weber number (We) conditions representative of high We (We>200) regime. During the early stages of drop spreading, the drop impact resulted in ejection of secondary droplets from spreading drop front on the etched surfaces resembling prompt splash on rough surfaces whereas no such splashing was observable on untreated aluminum surface. Prominent development of undulations (fingers) were observed at the rim of drop spreading on the etched surfaces; between the etched surfaces the OTS-coated surface showed a subdued development of fingers than the uncoated surface. The impacted drops showed intense receding on OTS-coated surfaces whereas on the etched surface a highly irregular receding, with drop liquid sticking to the surface, was observed. Quantitative analyses were performed to reveal the effect of target surface characteristics on drop impact parameters such as temporal variation of spread factor of drop lamella, temporal variation of average finger length during spreading phase, maximum drop spreading, time taken to attain maximum spreading, sensitivity of maximum spreading to We, number of fingers at maximum spreading, and average receding velocity of drop lamella. Existing models for maximum drop spreading showed reasonably good agreement with the experimental measurements on the target surfaces except the acid-etched surface.

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1. Introduction

The phenomenon of liquid drop interaction with solid surfaces is seen in numerous practical cases: self-cleaning [1], engineering applications involving the impingement of spray droplets such as spray cooling, spray painting, fuel spray impact inside IC engines, and pesticide spraying [2–4], ink jet printing [5], drop liquid transport in microfluidic devices [6], etc. Outcomes of drop-surface interaction process assume great significance in these practical cases. An impinging liquid drop on a solid surface spreads rapidly in the radial direction, reaches a maximum spreading diameter, and then recedes. The comprehensive reviews by Rein [7], Yarin [8] and Marengo et al. [9] provide salient details associated with the drop impact phenomenon. The final outcome of drop impact process can be broadly classified into deposition, splashing, and bouncing and it is mainly governed by a combination of surface and drop liquid characteristics [8,9].

The physical and chemical features of solid surface play a pivotal role in determining the outcomes of an impacting drop. These features include the physical surface quality of solid surface (often quantified in terms of mean surface roughness, R_a) [10] and the chemical nature of solid surface (often quantified in terms of contact angle of drop liquid on the surface) [11]. Such surface properties alter wettability characteristics of solid surface, thereby modifying the outcomes of drop impact process [12]. An appropriate tuning of the physical and chemical features of solid surface helps to control and manipulate the behavior of drop liquid on the solid surface in order to obtain desired outcomes of drop-surface interaction [13-15]. This idea has been effectively used in the development of superhydrophobic surfaces which find use in several practical applications [16-20]. Superhydrophobic solid surfaces exhibit high equilibrium contact angle (>150°) and low contact angle hysteresis (measured as the difference between upper and lower limits of contact angle on the surface). By mimicking the surface morphological structure of lotus leaves [16], superhydrophobic solid surfaces have been prepared through a combination of hierarchical micro/nano textures and chemical hydrophobicity [21,22]. Metallic surfaces are hydrophilic in nature. Superhydrophobic surfaces with metallic base have often been prepared with micro/nano surface roughness along with low surface energy coating. The fabrication technique, the so-called two-step process, involves physical creation of desired surface roughness and coating of low surface energy material on the base metallic surface [23–28]. The desired surface roughness is imparted to the metal surface via techniques such as chemical bath deposition, acid-based chemical etching, galvanic exchange reactions, etc. As the second step process, the roughened metallic surfaces are exposed to low surface energy materials such as fluoroalkylsilane molecules to create chemical bond between metal atoms and low surface energy molecules. Recently, Saleema et al. [29,30] prepared superhydrophobic aluminum alloy surfaces in a single step process by immersing aluminum alloy substrates in a solution containing NaOH and fluoroalkyl-silane (FAS-17) molecules. This technique has further been optimized by Bernagozzi et al. [31].

Currently, there is an upsurge in the development and fabrication of metallic hydrophobic/superhydrophobic surfaces for various engineering applications. The above mentioned studies [23–30] on the fabrication of superhydrophobic surfaces characterized the prepared metallic surfaces only via static wetting experiments using sessile drops. Since the physical surface quality is significantly altered in these fabrication methods, the characterization of surface via static wetting experiments alone is not sufficient to estimate the outcomes of drop impact. Understanding the dynamic behavior of impacting drops on such metallic surfaces is very crucial to determine the effectiveness of these fabrication methods and the number of published works in this context is

very less. Antonini et al. [32] studied the impact of water drops on 9 different solid surfaces of different wettability with advancing contact angle of drops ranging from 48° to 166°. Several of these surfaces were prepared using the abovementioned fabrication methods. Measurements were presented on maximum spread factor, spreading time (time to reach the maximum spread factor), time at maximum spreading, and drop rebound time (time lapse from the start of impact to the instant of drop rebound) [32,33]. Two different regimes were identified based on the analysis of these measurements: a moderate Weber number, We regime (30 < We < 200) in which surface wettability affects both maximum spreading diameter and spreading time; a high We regime (We>200) in which the effect of surface wettability is relatively small. An in-depth analysis on the role of target surface characteristics for the impact of high We drops is still lacking in their study. In a similar study of drop impact (25 < We < 585) involving aluminum superhydrophobic surfaces [34], it was observed that the drop rebound is mainly governed by the receding contact angle of the drop on target surface.

The present study investigates the impact of water drops on chemically modified aluminum surfaces prepared using the above mentioned fabrication process involving the creation of micro/nano surface roughness and low surface energy coating. Three different chemically modified surfaces, identified here based on the surface quality as etched surface, coated surface, and etched plus coated surface, were prepared from commercially available (Al-6061 grade) bare aluminum surface. The dynamics of drop impact on the chemically modified aluminum surfaces was compared with that on the bare aluminum surface. By considering inertia dominated drop impacts encountered in practical spray impact scenarios, the present work focuses only on the impact of high *We* drops.

2. Experimental

2.1. Target surfaces

Four target surfaces with aluminum (Al-6061) as base material of size $38 \text{ mm} \times 20 \text{ mm} \times 1 \text{ mm}$ were prepared. The target surfaces are identified in the present study as Al (bare surface), Al-OTS (coated surface), Al-Et05 (etched surface), and Al-Et05-OTS (etched plus coated surface). Surface characteristics of the target surface Al is as obtained from the vendor. The target surface Al-OTS is aluminum surface coated with a low surface energy material. The coating material was Octadecyl-Trichloro-Silane CH₃(CH₂)₁₇SiCl₃ (often referred as OTS) obtained from Sigma Aldrich (CAS No. 112-04-9). Since OTS reacts violently with water [35], sufficient care was taken to remove moisture content from the surface just before the start of coating process. The surface was dipped in OTS present in a petri-dish for about 2 h. Thereafter, the surface was cleaned with excess hexane to remove uncoated OTS left over the surface and dried in a hot air oven at 60 °C for about 15 min to remove moisture content. The target surface Al-Et05 is a roughened aluminum surface. The surface roughness was imparted via acid-based chemical etching process. The acid etchant was Beck's dislocation etchant [23]. It comprised of 37 wt% hydrochloric acid (HCl), distilled water (H₂O), and 40 wt% hydrofluoric acid (HF) thoroughly mixed in the volume ratio 16:5:1. The bare aluminum surface was kept immersed in the pool of acid etchant for a time period of approximately 5 s. Then the surface was cleaned with excess of distilled water and dried in a hot air oven at 80 °C for about 2 h to remove moisture content. Al-Et05-OTS is a rough aluminum surface coated with OTS. For preparing this surface, the bare aluminum surface was subjected to the above mentioned chemical etching process for a time period of approximately 5 s followed by the

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