

# Spreading, encapsulation and transition to arrested shapes during drop impact onto hydrophobic powders



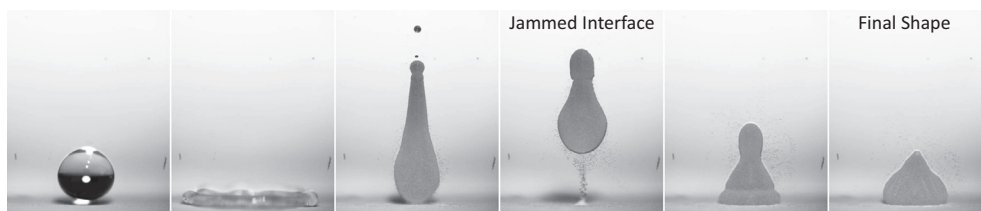
T. Supakar<sup>a</sup>, M. Moradiafrapoli<sup>a</sup>, G.F. Christopher<sup>b</sup>, J.O. Marston<sup>a,\*</sup>

<sup>a</sup>Department of Chemical Engineering, Texas Tech University, Lubbock, TX 79409, United States

<sup>b</sup>Department of Mechanical Engineering, Texas Tech University, Lubbock, TX 79409, United States

## GRAPHICAL ABSTRACT

Arrested shape formed during the impact of a 2 mm water droplet onto hydrophobic 25  $\mu\text{m}$  particles.



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## ABSTRACT

We present findings from an experimental study of the impact of liquid droplets onto powder surfaces, where the particulates are hydrophobic. We vary both the size of the drop and impact speed coupled with the size range of the powder in order to assess the critical conditions for the formation of liquid marbles, where the drop becomes completely encapsulated by the powder, and arrested shapes where the drop cannot regain its spherical shape.

By using different hydrophobization agents we find that a lower particle mobility may aid in promoting liquid marble formation at lower impact kinetic energies. From observations of the arrested shape formations, we propose that simple surface tensions may be inadequate to describe deformation dynamics in liquid marbles.

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## 1. Introduction

Superhydrophobicity has been an exciting topic of research for many years and many different techniques, typically incorporating both micro-texturing and vapor deposition, have been developed in order to render surfaces in such a state [1,2]. When water comes into contact with such a surface, it will exhibit a high contact angle and low degree of hysteresis. In particular, Water droplets resting on hydrophobic textured surfaces are typically in the Cassie–Baxter

state [3] meaning that the bottom surface of the droplet is supported only by the tops of the surface asperities (pillars). This ultra-low contact area yields a very low-friction state whereby droplets can roll easily across a surface.

A liquid marble, which is a liquid droplet encapsulated with solid particulate matter [4–7] essentially mimics this principle, whereby the marbles can roll and be transported in a very low-friction state [8–10]. In essence liquid marbles create a Cassie–Baxter state by having “pillars” (i.e. particles) embedded across the entire free-surface of the droplet.

Given their mobility and robustness, liquid marbles have been studied and tested for a variety of potential applications, such as

\* Corresponding author.

E-mail address: [jeremy.marston@ttu.edu](mailto:jeremy.marston@ttu.edu) (J.O. Marston).

for gas sensing [11], synthesis of compounds/composites [13], blood typing and cell culture [12], and miniature chemical and biological reactors [10].

The mechanisms and conditions for liquid marble formation have been investigated previously [14–18] with the aim of quantifying the dependence on various parameters such as surface tension, viscosity, and droplet kinetic energy. Post-production, various phenomena such as evaporation [17], condensation [19] and freezing [20] have been observed. In all of these studies, the key parameters studied are shape and size of the individual particles encapsulating the liquid droplets.

More generally, liquid marbles are a specific class of particle-laden interfaces, where solid particles lie at an interface [22] and induce interfacial mechanical properties beyond a simple surface tension [23]. In particular, particle-laden interfaces exhibit viscoelasticity and diminishing surface tension as they transition to a jammed state [25–28] and can form aggregates or “rafts” [24]. To date, only two studies [29,30] have reported jammed interfaces during liquid marble formations, which provides significant motivation for the present study; Given that liquid marbles are designed to be low friction transportable micro-reservoirs, their shape and mobility are key properties and understanding the transition towards a jammed interface is paramount.

In the context of liquid marble formation, cratering [31,32,21,33–35] could be an important consideration as the compliance of the powder bed will influence the spreading dynamics and ultimately, the final coverage of powder on the surface of the drop.

The spreading stage of a droplet during impact is typically quantified by the maximum spread,  $D_{max}$ , (e.g. [36–42]), often scaled in terms of initial drop diameter,  $D_0$ , and impact Weber number,  $We = \rho D_0 V_0^2 / \sigma$ . For the inviscid case, two primary scaling laws have been proposed: Firstly, where the kinetic energy at impact is completely transferred to surface energy at maximum deformation, whereby it can be derived that  $D_{max}/D_0 \sim We^{1/2}$  [36]. Secondly, when considering that the impact itself induces an acceleration term  $V_0^2/D_0$  and assuming a roughly cylindrical shape at maximum deformation, applying volume conservation leads to the scaling  $D_{max}/D_0 \sim We^{1/4}$  [37].

For powders, where the target surface can be deformed, the compliance of the powder bed has been incorporated through the bulk density,  $\rho_b$ , of the bed to render the scaling law  $D_c/D_0 \sim \frac{\rho_b}{\rho} We^{1/4}$  [33,34], where  $D_c$  is the crater diameter (equivalent to the maximum spread diameter). For low viscosity and low surface tension drops it was found [21,43] that the maximum spread  $D_{max}/D_0$  scaled as  $We^{1/5}$  onto dry, wettable powders for  $We \approx 10$ –1000, which decreased to  $We^{1/10-1/5}$  for pre-wetted powders, but for hydrophobic powders, the scaling  $We^{2/5}$  was observed [29] for  $We \approx 30$ –100. A summary of these observed scaling laws is presented in Table 1. Thus from previous theoretical arguments and empirical observations, the normalized maximum spread during impact onto hydrophobic powder beds should be described by

**Table 1**  
Summary of scaling for the normalized maximum spread diameter  $D_{max}/D_0$  for different target surfaces.

Target surface	Observed scaling	Reference
Dry solid (inviscid)	$We^{1/4}, We^{1/2}$	[36–38,42]
Dry solid (viscous)	$Re^{1/5} f(WeRe^{-2/5})$	[39,40]
Superheated solid	$We^{2/5}$	[41]
Dry wettable powder	$We^{1/5}$	[21,43]
Pre-wetted powder	$We^{1/10-1/5}$	[44]
Dry hydrophobic powder	$We^{2/5}$	[29]

the Weber number raised to some exponent  $\alpha = 0.2$ –0.5. However, there is no widely accepted scaling law for the impact of liquid drops onto loose, hydrophobic powder surfaces. This is an important aspect as it is pertinent to the formation of liquid marbles. As such, one focal area in this work was to conduct an experimental campaign to provide empirical evidence for a scaling law.

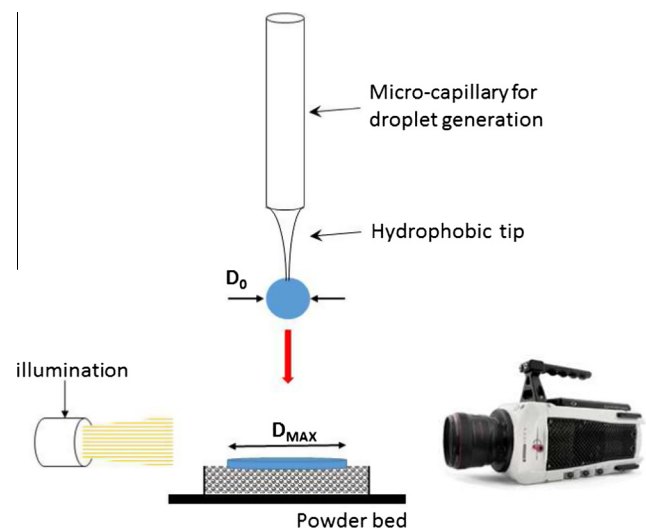
To achieve this, we have complemented, and extended, previous works by performing liquid marble formation experiments across a broad range of parameters, including the contact angle,  $\theta$ , between the droplet and the particles, particle diameter,  $d_p$ , initial droplet diameter,  $D_0$ , and impact speed,  $V_0$ . We have also examined grain mobility on the liquid droplet surface, which in turn affects the encapsulation of the liquid droplet with the particles. In doing so, we have expounded upon the threshold conditions for the formation of fully covered (spherical) and deformed (non-spherical) liquid marbles, originally reported in [29,30].

## 2. Materials and methods

The experimental setup, shown schematically in Fig. 1, consists of a small container filled with fine glass beads. We release a drop of pure water from heights  $h_r \approx 7.5$  cm up to 40 cm using hydrophobic glass capillaries to achieve highly repeatable droplet sizes,  $D_0 = 0.8$ –3 mm. The falling drops thus impact vertically with speed  $V_0 \approx \sqrt{2gh_r}$ , which varies from 0.61 to 2.6 m/s in our case. The impact, spreading and rebound of the impinging droplets are all captured in a single video sequence using a high-speed video camera (Phantom V711, Vision Research Inc.) equipped with a Nikon 60 mm micro-lens, which yielded a range of effective pixel sizes of 29–41  $\mu\text{m}/\text{px}$ . Frames rates of up to 10,000 fps were used.

### 2.1. Powder preparation and properties

The primary particles used in this study were glass beads (Pottery Industries Inc.), with a total particle diameter range of  $d_p = 25$ –500  $\mu\text{m}$ . A summary of the powder properties is given in Table 2. The untreated glass beads exhibit a contact angle of approximately 60° (e.g. [44,45]). As such, the glass beads were subject to hydrophobization using commercial agents – namely – Glaco mirror coat ‘zero’ (Soft 99 Co.) and Ultra every dry (UltraTech International Inc.). The former is an alcohol-based suspension of silica nanoparticles, which form micron and sub-micron sized roughness elements when heat-cured [29]. The Ultra ever dry is a two layer



**Fig. 1.** Schematic of the experimental setup used.

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