### Motion and Arrest Shape of Liquid on Cold Solid Targets

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# 1. Introduction

The Physics of non-isothermal spreading followed by phase change, unlike universal equations established for isothermal spreading and the extensive research done on the topic, is still a mystery [1,2]. The present work explores dynamic and thermal characteristics of liquid spreading and subsequent arrest due to solidification. Spreading of liquid was recorded, and the evolution of droplet diameter as well as the advancing contact angle were measured from the recordings of drop shape analyzer apparatus. The injection needle was used as a continuous liquid source to study the dynamics of motion and phase change on a smooth cold solid substrate at extremely low Reynolds numbers.

## 2. Experimental Setup and Materials

Experiments were carried out using water, hexadecane and pentadecane. Drop deposition experiments were conducted with Drop Shape Analyzer (DSA 100) manufactured by Krüss as shown in Fig. 1. Glass slides with dimensions of 50×24×0.15 mm<sup>3</sup> (VWR microcover glass) were rinsed successively in acetone, methanol and DI water. The glass slide was dried on a hot

<sup>&</sup>lt;sup>1</sup> Unpublished. ISCST shall not be responsible for statements or opinions contained in papers or printed in its publications.

plate and then placed into a Krüss drop shape analyzer chamber, in which substrate temperature and the relative humidity were accurately controlled. Glass windows covered three sides of the chamber and provided the observation porthole for a CCD camera and illumination source. Temperature of the glass substrate was adjusted by peltier element mounted at the bottom of the chamber with a range from -30 to 160 °C. Temperature measurements indicated in temperature controller unit were accurate to  $\pm 0.5$  °C established by both the FLIR<sup>®</sup> SC300 thermographic camera and a K-type thermocouple. The relative humidity was monitored and could range between 10 to 80% RH. The main stage, on which the peltier element and glass substrate were situated, was able to be precisely positioned in x, y and z direction.

The syringe driver unit dispensed liquid in two steps. In the first step, a syringe plunger extracted liquid from main storage bottles and stored it in a secondary cylindrical containment in a downward motion. In the next step, liquid was pushed through the tubes by the same plunger and through 0.5mm diameter injection needles. An injection syringe situated on a syringe revolver was brought down manually/automatically and entered thorough an opening at the chamber near the cover glass. The needle was brought close to the solid substrate to perform continuous dispensing flow rates range from 0.2 to 600 µl/min. A high-speed CCD camera capable of recording 90 fps was used as a visualization technique to study droplet evolution during spreading. The base diameter and contact angle of a liquid drop were measured at each frame by the Krüss analyzer using a tangent method. In this method, a profile of sessile drop in the region of the baseline is fitted to polynomial function. From the fitted parameters the slope of the three-phase contact point at the baseline is first determined and used to determine the contact angle. Consequently, base diameter is defined as a distance between to contact points.



Figure 1: Schematics of continuous liquid source spreading.

### 3. Experimental Observation

Generally, The non-isothermal spreading of the experiments generally can be subdivided into three regimes. First, there is a region in which instantaneous solidification occurs at the moment of contact between the liquid and the impermeable flat subcooled solid substrate. In this regime, solidification time is larger than spreading duration. In the second regime, which usually occurs at higher flow rates and higher substrate temperature, tangible spreading precedes solidification of a liquid droplet. When the drop arrests at contact line due to solidification, the contact base remains frozen on the target without any sign of further motion (not allowed to recede or advance) despite the continuing discharge of liquid from the injection needle. A thin frozen layer was formed at the base ascending from solid crust, while the bulk of the drop was still in a liquid state. Ultimately, the drop as a whole freezes on a much higher timescale. In the last and third regime, no solidification transpires during spreading and the droplet remained as a liquid during the entire spreading process, however, spreading rate differs from the isothermal case as generation of thermocapillary flows accelerates spreading. In this study, arrested contact angle ( $\theta^*$ ) and arrested base diameter (D\*), depicted in Fig. 2, are evaluated against two main contributing variables: fluid flow rate and Stefan number. Stefan number is defined as a ratio of sensible heat to latent heat. Our results suggest that the arrested contact angle and base diameter are not single-valued for given substrate temperature, but are strongly dependent on other operational conditions, such as flow rate and the wettability.



Figure 2: Arrested contact angle and arrested base diameter.

In addition, higher flow rates and lower Stefan numbers facilitate transition from the instantaneous solidification regime to the successive spreading-solidification regime. During spreading followed by solidification, water exhibited a larger variance in arrested base diameter values than other liquids due to its ability to undergo supercooling. Supercooling of water is a process in which the water temperature can be lowered beyond melting point without water forming into ice. In this region of successive spreading-solidification, icing is hindered with increasing flow rates leading to a bigger solidification foot print and lower  $\theta^*$ , whereas, D\* is less sensitive to flow rate changes in instantaneous solidification regime.

### 4. References

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