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Effect of Different Fluids on Rectified Motion of Leidenfrost Droplets on Micro/Sub-Micron Ratchets

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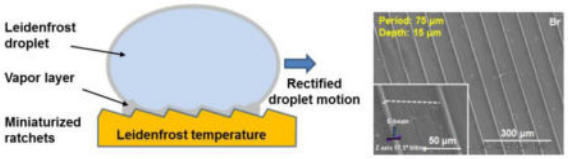
Abstract

Leidenfrost droplets, liquid droplets placed on a hot flat surface above the Leidenfrost temperature of the liquid, are an interesting model system to understand and achieve frictionless motion of droplets on a surface. Controlled unidirectional motion of otherwise random Leidenfrost droplets can be achieved by replacing the flat surface by a surface with topological ratchets. In this study, we show how an increase in the vapor layer thickness below the Leidenfrost droplet influences the droplet motion for underlying ratchets with various periods ranging from 1.5mm down to 800nm. This was exploited by systematically studying the Leidenfrost droplet motion of various liquids with low boiling points including acetone, isopropanol, and R134a on the aforementioned various ratchets. For all liquids with boiling points lower than water, no unidirectional motion was observed for 800 nm. This indicates that the asymmetric vapor flow beneath the Leidenfrost droplet becomes negligible due to the large vapor layer thickness relative to the ratchet depth. However, unidirectional droplet motion was still observed for the micron and millimeter scale ratchets even when the ratchet surface temperature was increased up to 360°C and 230°C for acetone and isopropanol, respectively. This can be attributed to the insulating property of the thick vapor layer which prevent the droplet from producing more vapor with increasing temperature. We also report the effect of the ratchet period on the droplet motion at room temperature using R134a droplets.

Graphical abstract



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Leidenfrost droplets on ratchet surfaces show rectified droplet motion without friction at the droplet/ratchet interface. Here we investigated the Leidenfrost droplet motion on miniaturized ratchets with the ratchet periods ranging from 800 nm to 1.5 mm using different fluids. We learn that the vapor layer thickness beneath the droplet with respect to the ratchet depth presents the limitation in obtaining the rectified motion.

Keywords

Leidenfrost droplets; rectified motion; micro and nanoscale ratchets; micro-milling

1. INTRODUCTION

Liquid droplets placed on a hot flat surface above the Leidenfrost temperature of the liquid moves vehemently on the surface.[1–3] A vapor layer existing between the droplet and hot surface levitates the droplet from the surface, leading to a state where no friction at the droplet/surface interface exists. The evaporation of the droplet is extremely slow due to poor heat transfer in the vapor layer. Such droplets are called Leidenfrost droplets and the temperature where the total evaporation time of a droplet is longest is defined as Leidenfrost temperature. Leidenfrost droplets are of great importance in many practical applications such as spray cooling in the heat treatment of metal alloys, impingement of oil drops on turbine engines and re-wetting of fuel rod in nuclear reactor.[4] Recently, the Leidenfrost droplets have attracted significant research interest as a model system of non-wetting surfaces to understand and achieve extreme mobility of droplets without friction, similar to droplet dynamics on surfaces with superhydrophobicity.[5–8]

Leidenfrost droplets on a flat surface usually show random motion, as can be observed when water is spilled on a hot frying pan. Control over the directionality of Leidenfrost droplets can be achieved by replacing the flat surface by a surface with topological ratchets. This phenomenon was initially demonstrated by Linke et al. with millimeter scale ratchets.[9] In their work, liquid droplets dispensed onto 1.5 mm period ratchets resulted in unidirectional motion with the droplet velocity in the range of several cm/s. The unidirectional motion was attributed to a viscous drag exerted by asymmetric vapor flow in the ratchet trenches. Following Linke's work, several research groups attempted to theoretically and experimentally reveal the driving mechanism relevant to this unidirectional droplet motion.[10–14] The Quéré group supported the viscous drag mechanism proposed by Linke et al. by showing that Leidenfrost solids directly sublimating on a hot substrate also self-propel by surface ratchets.[10, 11, 13] On the other hand, Würger et al. proposed using a Stokes hydrodynamics model in which thermal creep can be a mechanism to contribute to the unidirectional motion of Leidenfrost droplets on ratchet surfaces.[14]

While all of the aforementioned work has been performed with millimeter scale ratchet surfaces, our group extended the work to miniaturized ratchets with the period ranging from 1.5 mm down to sub-micrometers.[15] Interestingly, as the ratchet period decreases, the maximum velocity of water droplets dramatically increases, even reaching a droplet velocity larger than 40 cm/s with 800 nm period ratchets which has never been achieved with any

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chemical and topological gradient surfaces including millimeter scale ratchets. In addition, unidirectional motion is observed for all ratchet samples extending to the maximum temperature (360°C) investigated.

Despite the achievement of such a fast droplet motion, our previous results [15] raised two additional questions. First, as the size of ratchets decreases, the velocity and effective Reynolds number of the vapor flow in the ratchet valleys become smaller than those in the vapor layer between the ratchet peaks and the bottom of droplet, leading to reduction of net asymmetric flow of the vapor.[14] Therefore, the rectified droplet motion is expected to decrease and ultimately disappear. In contradiction, the Leidenfrost droplets still show an increase droplet velocity as the ratchet size decrease down to 800 nm in period. Second, due to the same reason, rectified droplet motion is expected to disappear as the vapor layer thickness becomes significantly larger than the ratchet depth, which is the case at a ratchet surface temperature significantly larger than the Leidenfrost temperature. However, the rectified motion is still observed at a ratchet surface temperature of 360°C which corresponds to a vapor layer thickness of 50–100 μm.[3] Considering that the ratchet depth for 800 nm period ratchets is 200 nm, the results indicate that asymmetric vapor flow in such shallow ratchet valleys with nanometer scale depths can still produce drag force enough to induce a motion of droplet over the tens of micrometer thick vapor layer. Hence, it is interesting to examine the limitation on the unidirectional motion of Leidenfrost droplets driven by miniaturized ratchets, ultimately allowing for a better understanding of the driving mechanism.

In this study, in order to answer the aforementioned two questions, we designed experiments in such a way that the vapor layer thickness underneath Leidenfrost droplets relative to the ratchet depth are varied in a broader range than that used in our previous work.[15] Due to the difficulty in fabricating large area nanoscale ratchets, we use different liquids with lower boiling points than that of water in order to achieve a significantly larger vapor layer thickness underneath the droplet for an equal temperature to that of the water case. Our results show that the rectified motion of Leidenfrost droplets disappeared with sub-micron ratchets. However, the rectified motion was still observed for large ratchets even with the use of low boiling point liquids.

2. EXPERIMANTAL

2.1. Fabrication of nickel and brass miniaturized ratchets

Ratchets with periods ranging from 1.5 mm down to 800 nm (800 nm, 15 μm, 75 μm, 150 μm, and 1.5 mm) were fabricated in metals (either brass or nickel) and polymers (such as poly(methyl methacrylate) (PMMA)) using various micromachining techniques. Brass ratchets with micrometer scale period were produced via milling with a micromilling machine (KERN MMP2522, KERN Micro- and Feinwerktechnik GmbH & Co, KG, Germany) while nickel ratchets with 800 nm period were produced by replicating optical gratings and subsequent nickel electroplating. Details on the ratchet fabrication can be found in our previous paper.[15] The areas of the fabricated ratchet surface were 5×10 cm^2 for brass ratchets and 5.2×5.2 cm^2 for the sub-micron nickel ratchets. In order to investigate the effect of ratchet period, the ratchet aspect ratio, defined as the ratio of ratchet depth to

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period, was kept at a similar range (0.2 for micrometer scale brass ratchets and 0.25 for sub-micron nickel ratchets). To produce ratchets in polymers such as poly(methyl methacrylate), the fabricated brass or Ni ratchets were replicated via nanomprint lithography. In order to study the influence of a hydrophobic coating on ratchet surfaces on the Leidenfrost droplet motion, some of the brass and nickel ratchets were coated by a fluorinated silane molecule, 1H,1H,2H,2H–perfluorodecyltrichlorosilane ($C_{10}H_4Cl_3F_{17}Si$) in a custom made chemical vapor deposition (CVD) chamber.

Prior to the study of impact and motion of droplet, morphologies of the fabricated ratchet surface were inspected using various metrology tools such as optical microscope, surface profilometer, scanning electron microscope (SEM), and atomic force microscope (AFM). Figure 1 shows example SEM, AFM and optical micrographs for 75 μm and 800 nm period ratchets. The images clearly show the existence of topological asymmetric profiles. The root mean square roughness for the surfaces produced by the micromilling technique was typically 100–300 nm.[16]

2.2. Investigation of Leidenfrost droplet motion

The experimental setup used to investigate Leidenfrost droplet motion consists of three parts: a hot plate for heating ratchet samples, a micropipette for injection of droplets, and a video camera for recording the droplet motion. For investigating the Leidenfrost droplet motion, a ratchet sample was placed on a digital ceramic hot plate (Isotemp, Fisher Scientific) at a set temperature. Once a constant temperature was achieved, we waited for an hour to ensure that thermal equilibrium was reached and proceeded to measure ratchet surface temperatures at four corners as well as at the center with a K-type thermocouple (TP 873/TP 882, EXTECH) and thermometer (ML720, EXTECH). The average temperature was used as the ratchet surface temperature. The accuracy of temperature measurements with the thermocouple and thermometer was ±0.3% according to manufacturer specifications. Usually, the difference between temperatures measured at the four corners is in the range of 0 – 15 °C. Droplets of a constant volume were then dispensed using a commercial micropipette (Eppendorf) with the volume in the range of 3 – 6 μL. The height of the pipette tip was manually controlled within the range of 2 – 5 mm from the ratchet surface. Various liquids including acetone, isopropanol and R134a were used.

Droplet trajectory was captured using a video camera (Sony DSC-V1, 16 frames per second) with the Windows Movie Maker (Microsoft) software for tracking and processing the captured videos. Since the acceleration of the droplet could not be properly monitored due to the resolution limit of the video setup and an unachievable equilibrium velocity within the length of the fabricated ratchets, we simply took the mean velocity for data analysis. This value was obtained by dividing the distance travelled by the droplet before it escaped from the ratchet surface by time. At least the motion of 10 droplets was captured for each ratchet surface temperature and the velocity values were averaged to obtain a single data point.

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3. RESULTS AND DISCUSSION

3.1. Effect of various liquids on motion of Leidenfrost droplets

The physical properties of the liquids used in this study and water are shown in Table 1. Two important parameters to note in the table include the boiling temperature (T_b) and the capillary length (*a*) which is calculated by $a=\sqrt{\gamma/\rho g}$, where γ and ρ are the surface tension and density of the liquid, and *g* is the gravitation constant. *a* provides a criterion to determine the size of the droplet for the experiments. If the droplet radius is smaller than the capillary length, the droplet is nearly spherical from the lateral view except for the droplet bottom where it is flattened by the contact with the substrate surface. The volume of the liquid dispensed for each experiment (3 – 6 μL) is comparable to or smaller than the critical volume of the liquids obtained, thus expecting nearly spherical droplets in our experiments. On the other hand, T_b determines the ease of evaporation and thus the thickness of the vapor layer formed beneath the droplet. Acetone, isopropanol and R134a have T_b significantly lower than that of water and thus the vapor layer thickness underneath a droplet is expected to be larger at the same temperature compared to that of the water droplet. In the Leidenfrost temperature regime, an insulating vapor layer produced under a droplet flows down, split by the ratchet peaks. Due to the asymmetric topological profile of ratchets, the vapor on the less inclined side exerts more shear stress on the droplet bottom than the other side, resulting in rectified motion.[9–11, 13, 15] The vapor above the ratchet peaks predominantly flows out radially from the droplet bottom; thus, negligibly contributes to the asymmetric vapor flow except for the vapor portion dragged by the asymmetric vapor flow induced by ratchets. Consequently, as the thickness of the vapor layer increases, the magnitude of the asymmetric vapor flow leading to a net motion is expected to be smaller.

Various liquids with different boiling temperatures and vapor pressure including acetone, isopropanol and R134a were used to examine the limitation in rectified Leidenfrost droplet motion on miniaturized ratchets. Figure 2 shows an example droplet trajectory for a motion of acetone droplet on brass ratchets of 150 μm period at 113.0±1.0°C. The droplet exhibits rectified motion in the direction of the less inclined side of the ratchets, which is in agreement with the water case in our previous work.[15] We set t = 0 when the droplet touches the ratchet surface. After initial contact the droplet becomes accelerated and reaches a saturated velocity. However, due to the poor resolution of our camcorder, we did not analyze the acceleration, but just obtained the mean velocity calculated from Δl (the total travel distance) / Δt (the time to travel the distance). Figure 3(a) shows the mean droplet velocity of acetone droplets with a volume of 5 μL as a function of ratchet surface temperature for various ratchet periods. Overall, the mean droplet velocity versus surface temperature curves show a similar behavior to those for DI water[15] where the velocity initially increases after droplets starts to show rectified motion, reaches a maximum velocity, and then decreases to a constant velocity. However, the differences from the DI water case are three fold. First, the maximum mean velocity was observed at slightly below 100°C. Second, the mean velocity fell down sharply after reaching the maximum velocity. Third, the saturation velocity reached at the high temperature regime lasts even up to the maximum temperature (360°C) used for the experiments with small variations. The first two differences can be understood in relation to a low Leidenfrost temperature of acetone. A

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small increase in the surface temperature results in a rapid increase in the thickness of the vapor layer, leading to the steep decrease of the mean velocity after the maximum velocity. The third difference of still showing unidirectional motion even at 360°C indicates that the vapor layer thickness between the droplet and ratchet surface does not further increase with temperature after a certain value. This can be explained by the fact that the insulating property of the vapor layer prevents the liquid droplet from producing more vapor with increasing temperature.

For IPA, a similar trend compared to that of acetone was observed, specifically with 1.5 mm ratchets (Figure 3(b)). The reproducibility of the results was also examined by repeating the temperature ramp cycle four times with the same droplet volume. The variations in the mean velocity were negligible, confirming the reproducibility of our results. Table 2 provides the comparison of maximum mean velocity for different liquid droplets (for 5 μL) on various dimensions of ratchets. For both solvents, however, the directional motion was not observed for two smallest ratchets of 15 μm and 800 nm periods. Instead, droplets show random and slow motion, still levitating from the ratchet surface. This clearly indicates that as the vapor layer thickness with respect to the size (predominantly the depth) of the ratchets becomes larger than a critical value, the radial vapor flow above the top of the ratchets is dominant over the asymmetric vapor flow at the ratchet surface and thus does not lead to a rectified droplet motion. From this result, we can also deduce that in our previous work with water droplets [15], the rectified droplet motion would disappear if ratchets smaller than 800 nm period could be fabricated and used.

Figure 4 shows the mean velocity for R134a droplet motion at room temperature as a function of ratchet period with and without a fluorinated silane coating. The boiling temperature of R134a is −26.1°C. The maximum velocity appears with 75 μm period ratchets. Upon a hydrophobic silane coating on the ratchet surface, the droplet velocity increases by 2 – 5 cm/s. This indicates that direct contacts exist between the ratchet peaks and the bottom of droplets which is in agreement with our previous observation.[15] Conversely, no directional motion was observed with ratchets with 800 nm period, as was the case with acetone and isopropanol. The results also support our conclusion that there is a critical vapor layer thickness relative to ratchet depth above which the radial vapor flow from the bottom of droplet dominates over the asymmetric vapor flow within the ratchet grooves and thus the droplet sees the surface as flat. Conclusively, the ratio of the vapor layer thickness to the ratchet depth seems to be an important parameter to determine rectified motion of droplets.

3.2. Effect of ratchet materials on motion of Leidenfrost droplets

We also performed the same experiments with ratchets made of other materials. Directional motion of Leidenfrost droplets was observed for the metal ratchets in the period range from 1.5 mm down to 800 nm. However, when R134a droplets were put on ratchets made in polymer sheets fabricated by imprinting into PMMA, PC or a 100 μm thick SU-8 layer coated on Si substrate, droplets immediately spread out horizontally and evaporate instead of forming Leidenfrost droplets and moving in one direction. Even after coating a thin metal (Au 30 nm/Cu 10 nm) on the polymer ratchets, Leidenfrost droplets of R134a could not be

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generated. This observation disagrees with the results by Linke et al. showing that self-propelled Leidenfrost droplet motion is independent of ratchet materials. In the work by L. Melling,[17] motion of nitrogen droplets was observed on plastic (Plexiglas or PMMA) macroscale ratchets (p: 1.5 mm, d: 0.2 mm) at room temperature, but only for a very short time duration of around 0.3 second. Even though nitrogen droplets quickly evaporated on Plexiglas ratchets, the terminal velocity was in the range of 6 – 10 cm/s.[17] There are several possible reasons why it is difficult to form Leidenfrost droplets on polymer substrate those of which include their porous chain structures, poor thermal conductivities, and the chain interaction which depends on liquid polarity.

4. CONCLUSIONS

Motion of Leidenfrost droplets with various liquids on miniaturized ratchets with period ranging from 1.5 mm down to 800 nm was studied. When liquids with low boiling temperatures such as acetone, isopropanol and R134a were used, no rectified droplet motion was observed for 800 nm ratchets. The results indicate that there is a critical vapor layer thickness above which the influence of the asymmetric vapor flow in ratchets is negligible in the motion of the Leidenfrost droplets. Therefore, the ratio of the vapor layer thickness to the ratchet depth is an important parameter to determine rectified motion of droplets. To understand the critical vapor layer thickness is important in designing the optimal dimensions of the ratchets which provides the maximum droplet velocities. A theoretical or experimental study of the critical vapor layer thickness will also help understanding the fundamental physics for the Leidenfrost droplet motion.

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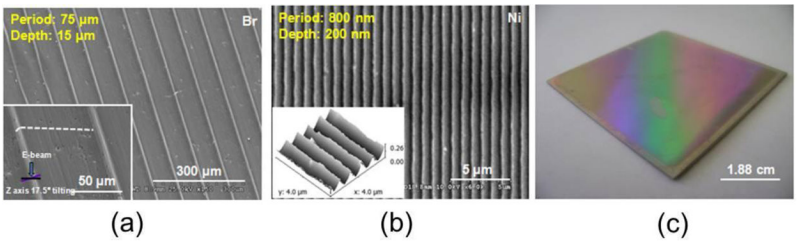

Fig. 1.

(a) Scanning electron micrographs of micromilled brass ratchets with periods of 75 µm and (b) scanning electron and atomic force micrographs and (c) photograph of replicated nanometer scale ratchets with 800 nm period.

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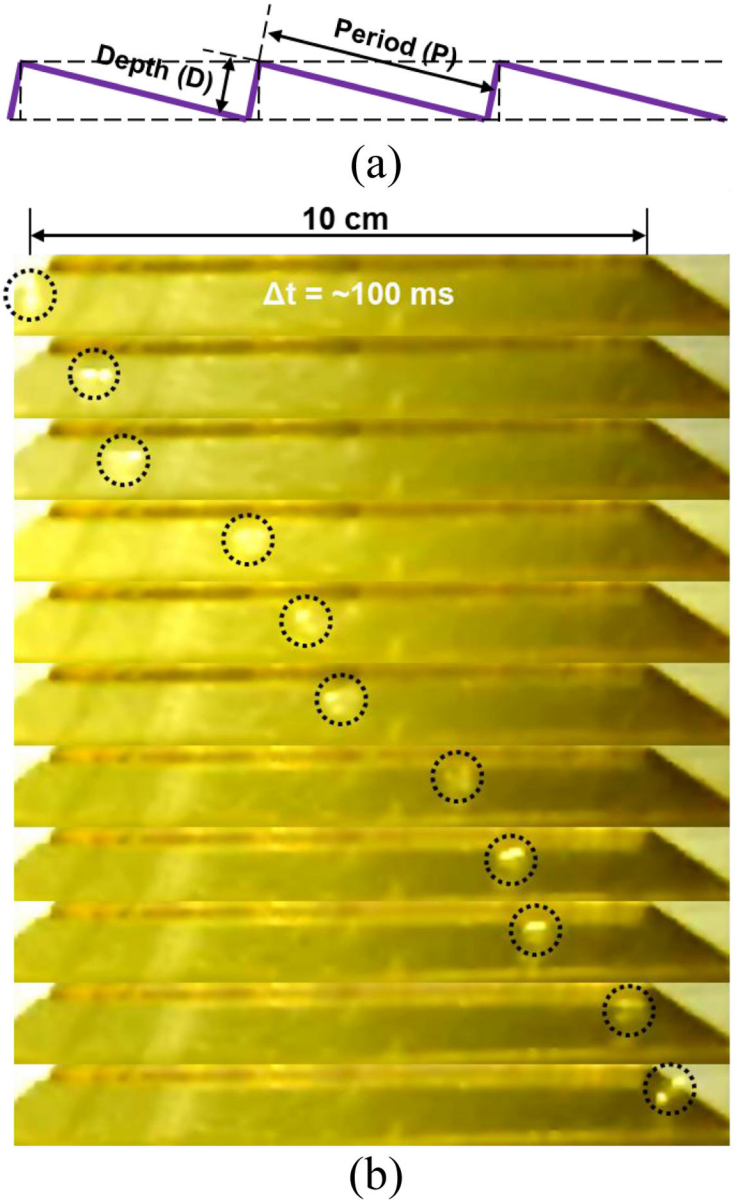

Fig. 2.

(a) A schematic showing definitions of ratchet depth and period. The schematic is not scaled. (b) A sequence of images captured during translocation of an acetone droplet on micromilled brass ratchets with 150 μm period at 113.2±1.1°C. Due to the poor contrast of the video images, dotted circle are added to represent the position of the droplet.

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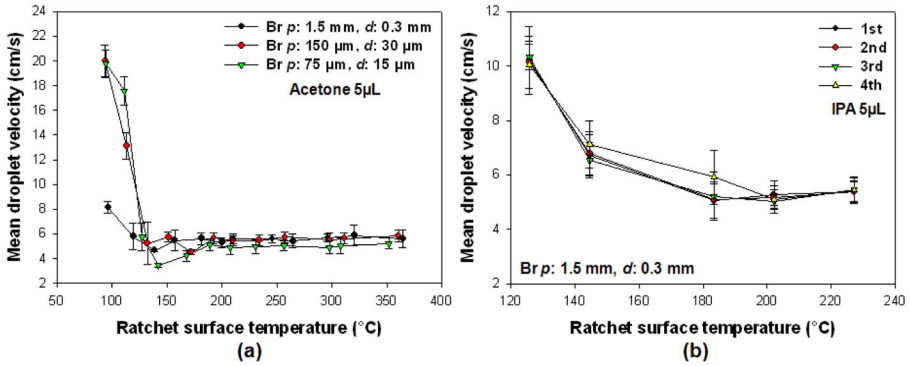

Fig. 3.

The motion of solvents droplets (V = 5 µL): (a) the mean velocity of acetone (T_b: 56.3°C) droplets as a function of ratchet surface temperature for different brass ratchet periods of p = 75 µm, 150 µm and 1.5 mm and (b) the mean velocity of isopropanol (T_b: 82°C) droplets as a function of ratchet surface temperature for 1.5 mm ratchets.

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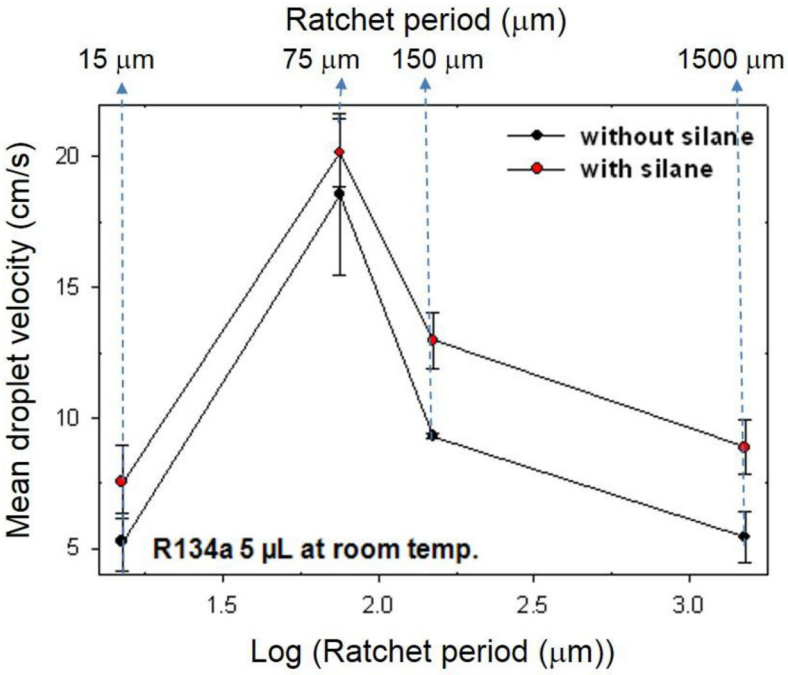

Fig. 4.

Mean velocity of 5 μL R134a droplet versus a logarithm of ratchet period at room temperature.

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Table 1

Physical properties of liquids at their boiling temperature (T_b) [18, 19]. *a* is the capillary length which decides droplet shape as spherical or puddle and V_a is the critical volume of the liquid based at its *a*.

Liquids\Properties	T_b (°C)	ρ (kg/m^3)	γ (mN/m)	*a* (mm)	V_a (μL)
Water	100.0	958.4	58.9	2.50	65.5
Acetone	56.0	747.1	19.7 (at 50°C)	1.64	18.5
IPA (iso-propanol)	82.3	711.3	17.0 (at 75°C)	1.56	15.9
R134a	−26.1	1,392.2	14.9	1.05	4.9

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Table 2

Mean maximum velocities (v_{max}) for different liquid droplets (5 μL) on various dimensions of ratchets.

Liquid\$v_{max,\ p}$: x	$v_{max,\ p}$: 800 nm	$v_{max,\ p}$: 15 μm	$v_{max,\ p}$: 75 μm	$v_{max,\ p}$: 150 μm	$v_{max,\ p}$: 1.5 mm
Acetone	–	–	20.8±1.1	20.0±1.3	8.15±0.5
IPA	–	–	–	–	10.1±1.0
DI Water	40.6±1.7	30.9±3.6	29.7±4.0	21.8±1.9	19.7±1.2