Contact line dynamics of gravity driven spreading of liquids

Alireza Mohammad Karim[1,2,*] , Keita Fujii[3] and H Pirouz Kavehpour[1]

[1] Department of Mechanical and Aerospace Engineering, Complex Fluids and Interfacial Physics Laboratory, UCLA, Los Angeles, CA 90095, United States of America
[2] The Nanoscience Center, University of Cambridge, Cambridge CB3 0FF, United Kingdom
[3] Department of Aerospace Engineering, Graduate School of Engineering, Nagoya University, Chikusa-ku, Japan

E-mail: alireza.m.k.2010@gmail.com

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Abstract

The spreading dynamics of the gravity-driven liquid motion on an inclined solid surface was studied by considering two fundamental physical models: the molecular kinetic theory and the hydrodynamic theory (HDT). The molecular kinetic theory is the most appropriate model to describe the gravity driven spreading mechanism investigated in this study. The gravity driven spreading which is one form of the forced spreading mechanism was compared with the spontaneous spreading for the same liquid/solid system from previous study by Mohammad Karim *et al* (2016 *Langmuir* **32** 10153). Unlike the gravity driven spreading, the HDT was appropriate model to define the spontaneous spreading. This finding reveals the importance of the mechanism of spreading which are the forced and the spontaneous on the suitability of the physical model

* Author to whom any correspondence should be addressed.

such as the molecular kinetic theory and the HDT to describe the spreading dynamics.

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1. Introduction

Understanding dynamic wetting behaviour is important for a vast range of applications such as industrial processes, coating and painting technology (Joanny and de Gennes 1984, Pomeau and Vannimenus 1985, Lafuma and Quere 2003, He *et al* 2004, Ou *et al* 2004, Bhushan and Jung 2007, Daniello *et al* 2009, Dorrer and Ruhe 2009, Heck and Papavassiliou 2013, Smyth *et al* 2013, Mohammad Karim *et al* 2016b, 2017a, 2017b, 2018b, 2018c, 2018d, 2019, 2020, Abe *et al* 2017). If the thickness of the liquid film can be controlled, the coating and painting processes would be optimized in terms of the speed and the quality. When highly viscous liquid is deposited on an inclined smooth solid surface, it begins sliding down and spreading on the surface under influence of the droplet density. During the droplet's downward slide on an inclined plate, the droplet leading edge, liquid contact line, develops a span-wise instability as it becomes long and thin, and an unwetted region appears on the surface, as illustrated in figure 1. This instability phenomenon occurs when the liquid film is too thin. This is an undesirable condition for many manufacturing processes and operating conditions of coating technology.

Experiments on the spreading of a viscous liquid sheet with a fixed volume on an inclined plate surface have been reported by Huppert (1982). It was observed that the contact line of the deposited liquid film was distorting in the span-wise direction after the contact line of the highly viscous liquid film had spread over a certain distance down the inclined plate surface. Huppert reported development of two distinct forms of instability: the parallel-sided fingers and the triangular structures (i.e. zig-zag shapes) in the span wise direction of the contact line of the liquid film flowing down on the inclined surface (Huppert 1982). The theoretical/numerical studies on these two types of the instabilities of the highly viscous liquid during downward slide on the inclined plane surface were conducted by Hocking (1990). He concluded that the observed instability of the shape of the liquid contact line is attributed to the dynamics of the liquid flow within the formed upward bulge near the leading edge. Numerical study by Hocking also revealed that the nonlinear development of a small disturbance to the position of a straight contact line show the incipient development of parallel-sided fingers of highly viscous liquid with a width that does not depend on the channel size (Hocking *et al* 1999). Silvi and Dussan (1985) studied on the influence of the static contact angle of the deposited liquid film on the onset of instability of the liquid film during the advancing motion of the thin liquid film on an inclined solid surface which played an important role on the rewetting process on the inclined surface. They reported that the triangular structure instability along the leading edge of the liquid film is attributed to the small dynamic contact angles and parallel-sided fingers along the leading edge of the liquid film are related to the large dynamic contact angles. When the static contact angle is very small, the dynamic contact angle is dominated due to the capillary effect as increase in the capillary number results in the dynamic contact angle increase. They reported that when the capillary number is large, it might be possible to obtain parallel-sided fingers even for complete wetting liquids (Silvi and Dussan 1985, Goodwin and Homsy 1991). Numerical and analytical studies by Goodwin and Homsy (1991)
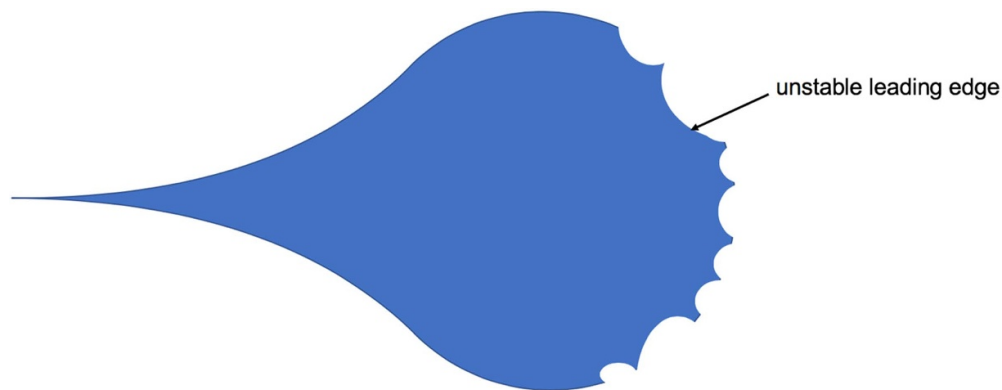
Figure 1. An illustrative image to depict the instability along the long and thin droplet leading edge on the solid surface.

presented a solution for the two-dimensional flow of a liquid film flowing down on an inclined solid surface over a wide range of three parameters: the dynamic contact angle, the inclination angle, and the capillary number. They demonstrated that a hump arises due to the kinematic considerations and does not depend on the liquid contact line singularity. Similar finding was also obtained through numerical study by Moriarty *et al* (1991). Perazzo and Gratton (2003) derived the governing equations for a power law non-Newtonian liquid flow on an inclined plane assuming the lubrication approximation. These equations were used as a starting point to study the influence of rheology on the fingering instability. López *et al* (1997) presented a steady-state downward motion of a thin liquid film on an inclined plane surface assuming the lubrication approximation by considering the effect of inertia force in the analysis via applying the Karman–Pohlhausen approximation. Moreover, they theoretically predicted the characteristic wavelength of the disturbance, causing the flow instability, as a function of the inclination angle of the solid surface. Pascal and D'Alessio (2007) numerically represented the effect of the predefined surface shear stress on the initiation and the structure of the flow instability with the power-law relation for the liquid film flowing downward on an inclined solid surface using the linear stability analysis and reported that the shear stress could either stabilize or destabilize the flow of the liquid film depending on the direction of the action of the shear stress. They deduced that the shear stress stabilizes the flow of the liquid film when applied uphill while the applied surface shear stress destabilizes the flow when acting downhill direction (Pascal and D'Alessio 2007). There have been tremendous studies on the instability of the liquid contact line and the form of humps. However, there has not been enough attention on how the dynamic contact angle depends on the velocity of the contact line of the liquid film during its downward motion on an inclined solid surface. Recent works by Le Grand *et al* (2005) reported experimental results on the shape and motion of millimetre-size droplets during their downward slide on an inclined plane for the condition of partial wetting liquids in which they also investigated the dynamics of the spreading of the droplets via considering different physical models: molecular-kinetic model, hydrodynamic model, de Gennes' model, and finally a linear model relating the dynamic contact angles to the capillary number.

The spreading of the liquid films and droplet(s) on inclined solid substrates are frequently encountered in reality. Unlike the axisymmetric droplet spreading on horizontal substrates, the gravity driven spreading of droplets undergo a non-axisymmetric and much complicated morphological structure. Hence it is very important to thoroughly understand the mechanism

of spreading of droplets under influence of gravity which happens usually on complex morphological substrates. The mechanism of spreading can be described via investigating on the dependency of dynamic contact angle along the leading edge of the droplet on the speed of the downward motion of the droplet under the influence of the gravity. The goal in this study is to analyse the correlation between dynamic contact angle of the liquid film flowing down an inclined solid surface with the velocity of the leading edge of the liquid film, known as the liquid contact line, and compare the findings with previous works.

2. Experimental method

To study the dependency of the dynamic contact angle of highly viscous liquids on an inclined solid surface on the liquid contact line dynamics of the advancing liquid, a silicone oil was deposited on an inclined solid surface as illustrated in figure 2. Three inclination angles, 30°, 45° and 60° were tested in this experimental study. Two different silicone oils with different kinematic viscosity were used in the experiments. Table 1 lists the physical properties of the two silicone oils used in the experiments. The densities were measured using the tensiometer. The surface tension were also measured using the tensiometer, applying the Wilhelmy plate method. The experiments were conducted at room temperature. Videos of the downward motion of the silicone oils on the inclined solid surface were recorded from the side view to determine the variation of the dynamic contact angle of the leading edge of the liquids at different speeds of the motion of the leading edge (liquid contact line). The dynamic contact angle and the corresponding instantaneous downward distance travelled by the liquid during the spreading were measured from the picture frames of the recorded video. The dynamic contact angles were measured using the ImageJ software for each frame of the video. Velocity of the leading edge of the liquid (liquid contact line velocity) was determined from instantaneous downward distance travelled during the spreading and the corresponding time of each picture frame. In order to do the calibration from picture frames, a ruler was placed in perpendicular direction to the inclined solid surface, as illustrated in figure 2. The experiment was conducted for each case of the spreading of each silicone oil on the inclined acrylic solid surface. The experiments were repeated five times for each inclination angle.

3. Theory

There are two fundamental approaches to determine the dependency of the dynamic contact angle on the liquid contact line velocity: the hydrodynamic theory (HDT) and molecular kinetic theory (Hoffman 1975, Voinov 1976, Tanner 1979, de Gennes 1985, Cox 1986, Cazabat 1987, Berg 1993, de Ruijter *et al* 1999, Mohammad Karim and Kavehpour 2014a, Mohammad Karim 2015, Mohammad Karim *et al* 2016a, Kavehpour *et al* 2017).

The HDT describes the relation between the dynamic contact angle and the speed of the moving liquid contact line through focusing on the dynamics of the bulk liquid moving on the solid surface. The HDT assumes the viscous dissipation caused by the motion of the bulk liquid on the solid surface via application of the lubrication assumption on the Navier–Stokes equations (Cox 1986, Eggers and Stone 1999, Mohammad Karim and Kavehpour 2014a, 2014b, 2015, 2018, Mohammad Karim 2015, Kavehpour *et al* 2017, Mohammad Karim *et al* 2018a). Therefore, the dependency of the dynamic contact angle of the moving liquid on the speed of the moving liquid contact line is defined by HDT by equation (1) (Cox 1986):

$$\theta_A{}^3 - \theta_0{}^3 = \frac{9\mu U}{\sigma} \ln\left(\frac{L}{L_s}\right) \tag{1}$$
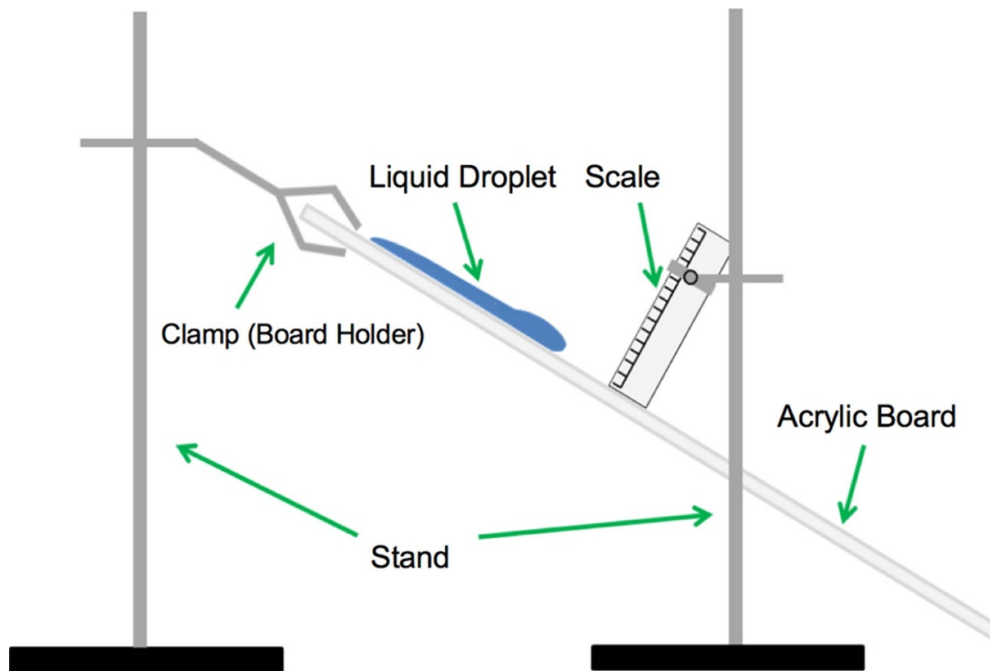
Figure 2. Experimental set up for procedure of forced spreading of a liquid due to gravity effect based on the liquid density.

Table 1. Physical properties of the liquids used in the experiments.

Physical property	Silicone oil 100 cSt	Silicone oil 1000 cSt
Dynamic viscosity (Pa s)	9.64×10^{-2}	9.69×10^{-1}
Surface tension (N m^{-1})	20.9×10^{-3}	21.2×10^{-3}
Density (kg m^{-3})	9.66×10^{2}	9.71×10^{2}

in which, θ_A is the dynamic contact angle, θ_0 is the equilibrium contact angle (i.e. static contact angle), μ is the dynamic viscosity, σ is the surface tension, L is the characteristic length of the flow, L_s is the slip length over which the no-slip condition of the flow at the solid/liquid interface can be relaxed, and U is the contact line velocity of the advancing liquid. Previous studies have revealed that $\frac{L}{L_s}$ is a contact line velocity dependent parameter based on capillary number (Eggers and Stone 1999, Mohammad Karim *et al* 2016a). It has been shown that $\frac{L}{L_s}$ is dependent on the capillary number, Ca, (i.e. $\frac{L}{L_s} = \alpha \, \mathrm{Ca}^{2/3}$), in which α is a power law factor to be determined by the fitting analysis, for the case of completely wetting liquids by considering the slip boundary condition over a small slip length L_s in the region near the liquid contact line.

In HDT, the following assumptions have been applied in order to describe the dynamics of the liquid contact line motion:

(a) The liquid/air interface near the liquid contact line is nearly flat: The HDT is applied to the vicinity of the liquid contact line over which the curvature of the liquid/air interface is approximately negligible.

(b) The lubrication approximation: The liquid thickness in the vicinity of the liquid contact line is assumed to be very small. Therefore, the motion of the bulk of the liquid near the liquid contact line is approximated to be one-dimensional in the polar coordinate system.

(c) The pressure variation is caused by capillary and van der Waals forces: The pressure gradient which causes the motion of the liquid bulk in the vicinity of the liquid contact line is driven by the capillary force and the van der Waals force.

The molecular kinetic theory describes the dependency of the dynamic contact angle on the velocity of the advancing motion of the liquid contact line based on the molecular displacements of the liquid molecules on the solid surface near the liquid contact line through considering both solid and liquid physical properties (Eyring 1938, Blake and Haynes 1969, Blake and Berg 1993). The absolute activated reaction rate theory proposed by Glasstone, Laidler, and Eyring (Eyring 1938) was used by Blake (Blake and Haynes 1969, Blake and Berg 1993) to describe the relation between the dynamic contact angle and the liquid contact line velocity as illustrated in equation (2) (Blake and Haynes 1969, Blake and Berg 1993, Eyring 1938):

$$\theta = \cos^{-1}\left[\cos\theta_0 - \frac{2k_{\mathrm{B}}T}{\sigma\lambda^2}\sinh^{-1}\left(\frac{U}{2K_{\mathrm{w}}\lambda}\right)\right] \tag{2}$$

in which, θ is the dynamic contact angle, θ_0 is the equilibrium contact angle (i.e. static contact angle), k_{B} is the Boltzmann constant, T is the absolute temperature of the environment, σ is the surface tension, and U is the contact line velocity of the advancing liquid. λ is known as the average molecular distance between two adjacent absorption sites, known as the locations on the solid surface, where the liquid molecules attach or detach from. Alternatively, λ is also defined as the average molecular displacement near the moving liquid contact line region. K_{w} is the called the frequency of the molecular attachment and molecular detachment on and from the solid surface at the state of equilibrium.

The combination of the molecular kinetic theory and HDT known as the molecular-hydrodynamic spreading dynamics model has been proposed by Petrov and Petrov (1992). The combined molecular-hydrodynamic model takes into account the energy dissipation due to viscous force happened in the bulk of the liquid as well as the energy dissipation due to friction force occurred in the immediate vicinity of the three-phase liquid contact line (leading edge of the spreading droplet) (Petrov and Petrov 1992). The combined molecular-hydrodynamic model considers the non-hydrodynamic (i.e. molecular kinetic theory) dependency of equilibrium advancing contact angle to the liquid contact line velocity (Petrov and Petrov 1992), $\theta_0(U)$, in the HDT of the spreading dynamics model (equation (3)):

$$\theta_0(U) = \cos^{-1}\left[\cos\theta_{\mathrm{Y}} - \frac{2k_{\mathrm{B}}T}{\sigma\lambda^2}\sinh^{-1}\left(\frac{U}{2K_{\mathrm{w}}\lambda}\right)\right] \tag{3}$$

in which θ_{Y} is known as the Young's static contact angle when the liquid droplet is at stationary condition on the solid surface. The combined molecular-HDT is stated as following (equation (4)):

$$\theta_{\mathrm{A}}{}^3 - (\theta_0(U))^3 = \frac{9\mu U}{\sigma}\ln\left(\frac{L}{L_{\mathrm{s}}}\right) \tag{4}$$

4. Results and discussion

The contact line dynamics of two silicon oils with kinematic viscosities of 100 cSt and 1000 cSt on an acrylic solid surface inclined at three different inclination angles, 30°, 45°, and 60° were
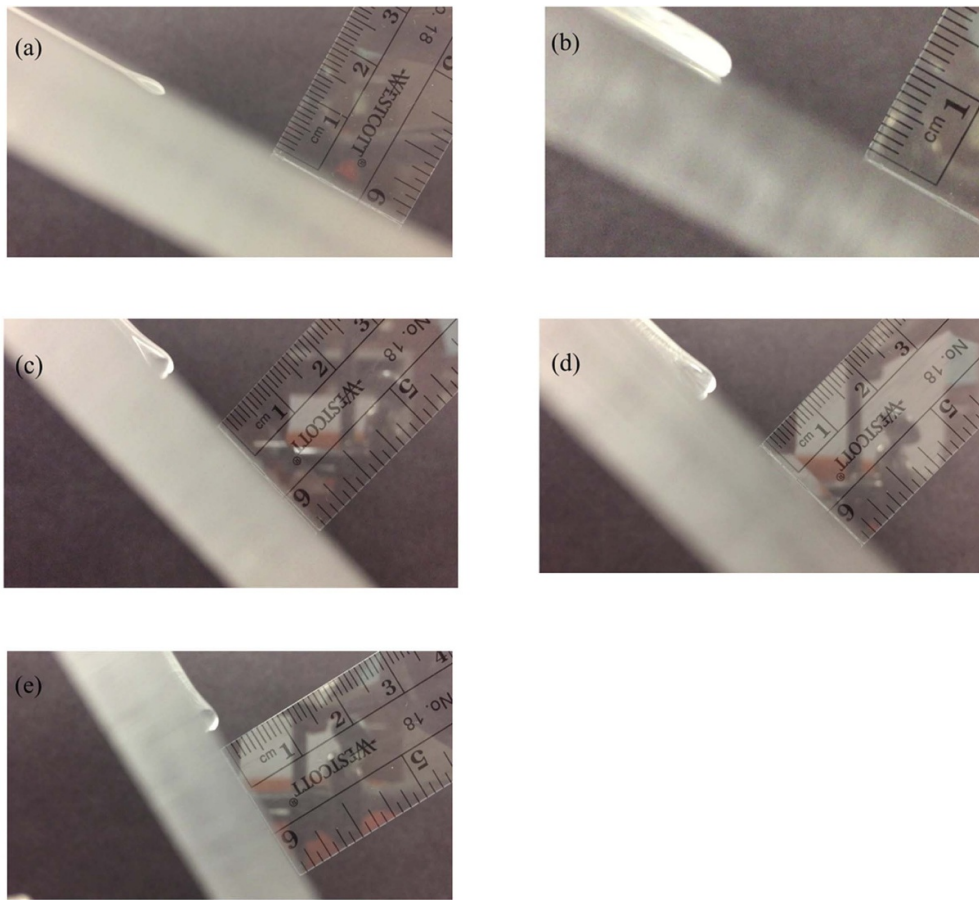
Figure 3. Side view images of the silicon oil droplets moving down on the acrylic surface at different inclination angles. (a) Silicone oil 100 cSt flowing on the 30° inclined solid surface. (b) Silicone oil 1000 cSt flowing on the 30° inclined solid surface. (c) Silicone oil 100 cSt flowing on the 45° inclined solid surface. (d) Silicone oil 1000 cSt flowing on the 45° inclined solid surface. (e) Silicone oil 1000 cSt flowing on the 60° inclined solid surface.

experimentally studied. The instantaneous advancing dynamic contact angle and the corresponding liquid contact line velocity were measured for each case. Figure 3 illustrates some of the picture frames of the experiments for silicon oils flowing downward on the acrylic surface. The results for 100 cSt silicon oil on 60° inclination could not be obtained due to the fact that the liquid did not spread and it just rolled over on the inclined surface. This could be caused by the presence of air entrainment as capillary number was too high ($5.40 < Ca < 7.65$) with a smooth contact line as well as it could be due to the large contact line velocity as would be indicated by a broken contact line. This may be due to the fact that at 60° inclination angle the contact line velocity becomes very large which accompanied by the large viscous stress contribution in the balance of the viscous force with the capillary force through capillary number. Furthermore, this causes the magnitude of the viscous bending of the interface become very large to exceed the critical limit at which it leads to the entrainment of the air bubbles along the leading edge of the moving droplet.
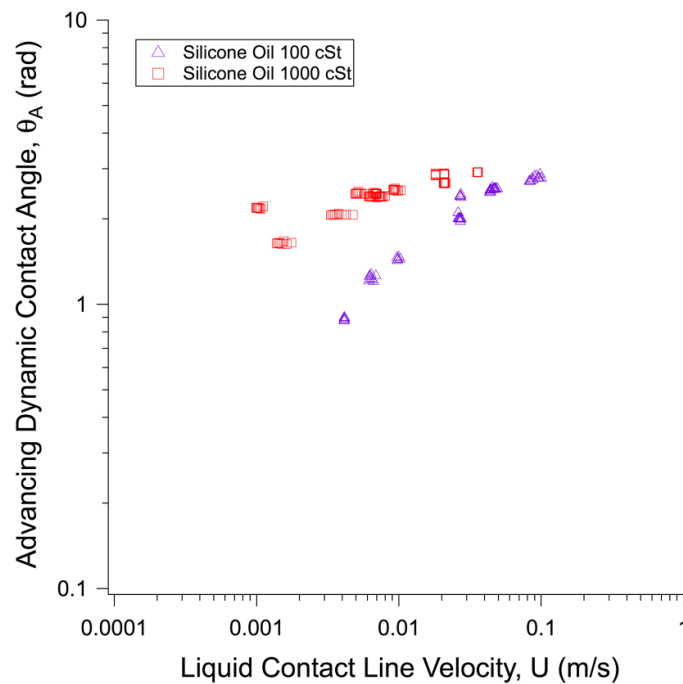
Figure 4. Advancing dynamic contact angle variation due to the change in liquid contact line velocity due to the gravity effect.

Figure 4 shows the supporting information text S1 (available online at stacks.iop.org/FDR/53/035503/mmedia) the change of the advancing dynamic contact angle caused by change in the silicone oil (i.e. 100 and 1000 cSt) contact line velocity during the spreading on the inclined solid surface due to effect of the gravity. The results indicate that 1000 cSt silicone oil has larger advancing dynamic contact angle compared to the advancing dynamic contact angle obtained for the case of spreading of silicon oil 100 cSt over the same liquid contact line velocity range. As the liquid contact line velocity increases the difference in the advancing dynamic contact angle obtained from the two silicon oils become smaller. The possible physical reason for this finding may be related to the condition that as the contact line velocity becomes too large, the influence of the viscous force is almost comparably the same for both liquids as they can merge to the case where the rolling of the leading edge of the droplet may happen (i.e. the situation at which the speed of the moving droplet is similar for both liquids). The range of capillary number for the case of silicone oil 100 cSt was between 0.02 and 0.46 and for the case of silicone oil 1000 cSt was between 0.05 and 1.66.

The molecular kinetic theory and the HDT were applied on the results (i.e. correlation between the advancing dynamic contact angle and the corresponding liquid contact line velocity) to determine the most suitable spreading dynamics model for the gravity driven spreading mechanism. The analytical results obtained from the molecular kinetic theory are shown in figures 5 and 6. From the fitting analyses, the equilibrium frequency of molecular displacement, K_w, and the molecular displacement, λ, have been obtained by fitting the experimental data with the equation of the molecular-kinetic theory (equation (2)). In the fitting analyses on the experimental data using the molecular-kinetic theory and the HDT, the physical properties of the silicone oils were held fixed and the only parameters which were set to be free to change
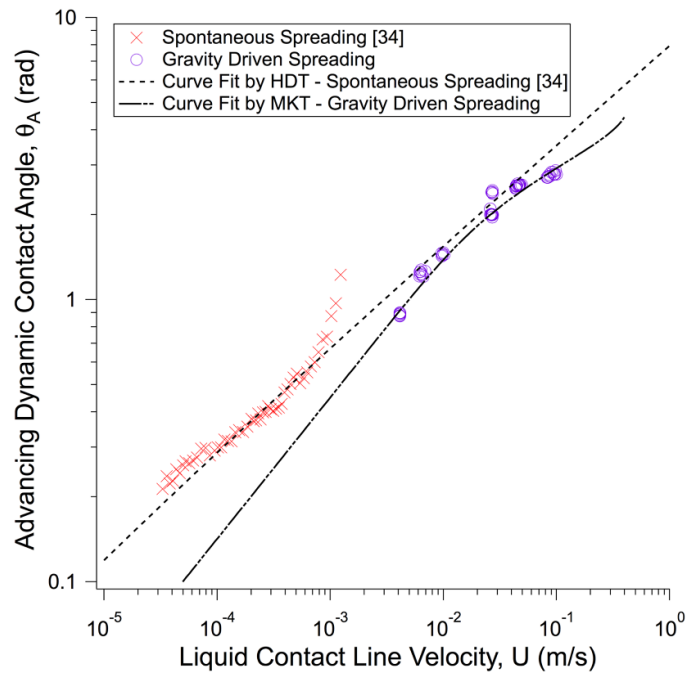
Figure 5. Instantaneous advancing dynamic contact angle of the silicone oil, with 100 cSt kinematic viscosity, during spreading on a smooth inclined acrylic surface and on a smooth glass surface (Mohammad Karim *et al* 2016a) as a function of velocity of the liquid contact line. HDT: hydrodynamic theory; MKT: molecular kinetic theory. The experimental data for the spontaneous spreading of silicone oil, with 100 cSt kinematic viscosity, on a smooth glass surface in figure 5. Reprinted with permission from Mohammad Karim *et al* (2016a). Copyright (2016) American Chemical Society.

were the molecular displacement, the equilibrium frequency of molecular displacement, and the $\frac{2}{3}$ power law factor, α, introduced in the equation of the HDT for the $\frac{L}{L_s}$ term, $\frac{L}{L_s} = \alpha \, \mathrm{Ca}^{2/3}$ (see table 2).

The HDT was not an appropriate model to describe the dynamics of motion of the liquid contact line for the gravity driven spreading mechanism. That was due to the fact that the characteristic lengths, which were defined as the size of the liquids that have been deposited on the inclined solid surface to spread, were much larger than the capillary length ($L_{cap} = \sqrt{\sigma/\rho g}$) whose values for silicone oils (i.e. 100 and 1000 cSt) was estimated to be ∼1.5 mm. The macroscopic characteristic length used in the equation of the hydrodynamic model must be equal to the size of the liquid being spread on the solid surface and its size was much larger than the capillary length of the silicone oils. This large macroscopic length scale caused the violation of the application of the lubrication assumption on the gravity driven spreading mechanism. Therefore, the HDT was not an appropriate physical model to describe the dynamics of spreading (i.e. correlation between the advancing dynamic contact angle and liquid contact line velocity) for the gravity driven spreading mechanism.

Given that the gravity driven spreading mechanism is one form of the forced spreading mechanism, the contact line dynamics of the gravity driven spreading mechanism were compared with the contact line dynamics of the spontaneous spreading mechanism obtained from the experimental data provided from the spreading of the silicone oils (i.e. 100 and 1000 cSt)
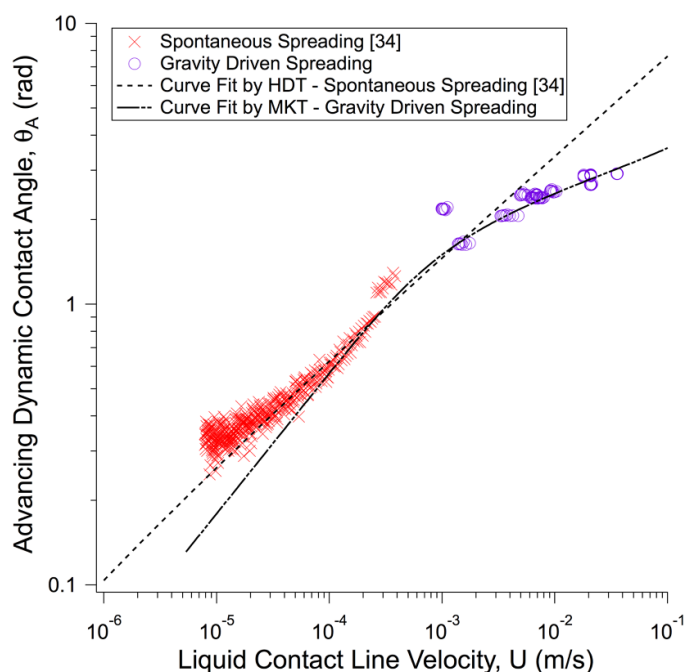
Figure 6. Instantaneous advancing dynamic contact angle of the silicone oil liquid, with 1000 cSt kinematic viscosity, during spreading on a smooth inclined acrylic surface, as a function of velocity of the liquid contact line. The experimental data for the spontaneous spreading of silicone oil, with 1000 cSt kinematic viscosity, on a smooth glass surface in figure 6. Reprinted with permission from Mohammad Karim *et al* (2016a). Copyright (2016) American Chemical Society.

Table 2. The values of the fitting parameters obtained from fitting the equation of the molecular kinetic theory on the gravity driven spreading of the liquids on an inclined solid surface.

Liquid	θ_0 (rad)	σ (mN m^{-1})	T (K)	λ (nm)	K_w (kHz)
Silicone oil 100 cSt	0	21	298	0.9269 ± 0.0477	1321.8 ± 654.0
Silicone oil 1000 cSt	0	21	298	1.1729 ± 0.0064	29.8 ± 2.2

on smooth horizontal glass solid surface reported by Mohammad Karim *et al* (2016a). As shown in figures 5 and 6, the HDT is the most appropriate model to describe the dynamics of spontaneous spreading mechanism unlike the gravity driven spreading mechanism. Table 3 shows the results of the fitting analyses for the spontaneous spreading of the silicone oils on horizontal solid surface based on the HDT. This study reveals the fact that one cannot use the same physical model (i.e. the molecular kinetics theory or the HDT) to describe the spreading dynamics, via the dynamics of the liquid contact line, of two different spreading mechanisms (i.e. spontaneous and forced) even for the same liquid/solid system as confirmed by previous study conducted by Mohammad Karim *et al* (2016a).

Table 3. The values of the fitting parameters obtained from study conducted by Mohammad Karim *et al* (2016a) via fitting the equation of the hydrodynamic theory on the spontaneous spreading of the liquids on a horizontal solid surface.

Liquid	θ_0 (rad)	μ (mPa s)	σ (mN m^{-1})	α
Silicone oil 100 cSt	0	96.4	20	37183
Silicone oil 1000 cSt	0	969.0	20	9767.5

The combined model which is the combination of the molecular kinetics theory and the HDT was also tested on the experimental data from both spontaneous and gravity driven spreading mechanism. The combined model did not fit with any of the data. That shows the fact that either spreading mechanism only follows one physical model (i.e. either the molecular kinetics theory or the HDT).

It is important to note that the previous work by Le Grand *et al* (2005) on the shape and dynamics of the silicone oil droplets sliding downward on an inclined solid surface reported the experimental results and fitted them with the molecular-kinetic model and the hydrodynamic model. They claimed the best consistency of the hydrodynamic model with their experimental data (Le Grand *et al* 2005). Moreover, they have also reported the suitability of the molecular-kinetic model within the scatter of their data (Le Grand *et al* 2005). The capillary number in their experimental data was very small (Ca < 0.02) much less than the range of capillary number investigated in our study which was between 0.02 and 1.66. The main difference between their findings and ours is the range of the capillary number resulting to draw different conclusion on choosing the best spreading dynamics model for the contact line dynamics of the gravity driven spreading mechanism. Moreover, the hydrodynamic model generally is not an appropriate model for large capillary number which explains the main reason on unsuitability of the hydrodynamic model to explain the contact line dynamics of our data.

5. Conclusions

Two silicone oils with different kinematic viscosities (i.e. 100 and 1000 cSt) spreading on a smooth acrylic solid surface inclined at three inclination angles (i.e. 30°, 45°, and 60°) were tested. The dynamics of motion of the liquid contact line on the inclined surface was described through exploring the dependency of the advancing dynamic contact angle on the liquid contact line velocity. The dynamics of spreading of the silicone oils on the inclined solid surface was studied by considering two fundamental physical models of spreading dynamics (i.e. the molecular kinetic theory and the HDT). The advancing dynamic contact angle for silicone oil with lower kinematic viscosity was smaller than the advancing dynamic contact angle for silicone oil with higher kinematic viscosity over the same range of liquid contact line velocity measured in this study. The HDT could not explain the dynamics of gravity driven spreading. Molecular-kinetic theory was found to be the appropriate physical model to describe the dynamics of gravity driven spreading mechanism.

The fitting analysis revealed that the HDT was not suitable to the dynamics of the gravity driven spreading. That may be attributed to the size of liquids being spread on the inclined solid surface which would violate the lubrication assumption which requires the liquid film to be thin, to be smaller than the capillary length related to the physical properties of the silicone oils.

Comparing the analytical findings for the gravity driven spreading mechanism which is one form of the forced spreading mechanism with the previous study on the spontaneous spreading

mechanism reveals the fact that one cannot use the same physical model (i.e. the molecular kinetics theory or the HDT) to explain the spreading dynamics of both mechanisms even the liquid/solid system is the same which was also confirmed by Mohammad Karim *et al* (2016a). Spontaneous and gravity-driven spreading dynamics of liquids with different rheological characteristics such as shear thinning and viscoelastic models need to be investigated to examine the extent of the applicability of the conclusion stated in this study and the hypothesis demonstrated in the previous work conducted by Mohammad Karim *et al* (2016a).

Data availability statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

ORCID iD

Alireza Mohammad Karim   https://orcid.org/0000-0002-2031-9057

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