

VISCOELASTIC LEIDENFROST DROPS

Volfango Bertola*

*University of Edinburgh, School of Engineering and Electronics,
The King's Buildings, Mayfield Road, Edinburgh EH9 3JL, United Kingdom

ABSTRACT

The effect of polymer additives on the dynamic Leidenfrost phenomenon (rebound of liquid drops impacting on very hot walls, where a thin vapour cushion separates the liquid from the surface) is studied experimentally by high-speed imaging. Drops of a dilute solution (200 p.p.m) of Polyethylene Oxide (PEO) were compared with drops of pure water, with surface temperatures ranging from 150°C to 400°C and impact Weber numbers between 7 and 160. Two main effects of the additive were observed: (i) the complete suppression of secondary atomization, which affects the conventional definition of Dynamic Leidenfrost temperature, and (ii) a significant enhancement of the maximum height reached by the drop centre of mass after the first rebound, which implies that the energy dissipation in viscoelastic drops is smaller than that of Newtonian drops. Because of the vapour film between the drop and the surface, wetting effects can be neglected, and this result can be used to get a deeper insight of the effect of additives on the drop impact dynamics on hydrophobic surfaces.

INTRODUCTION

Leidenfrost drops are created by deposition on very hot surfaces, at temperatures significantly above the boiling point of the liquid. As it is well known, such drops float on a vapour cushion which separates the liquid and the hot surface; because the thermal conductivity of gases is much smaller than that of liquids and solids, the thin vapour film acts as a thermal insulator, which strongly reduces the rate of evaporation of the rest of the liquid [1]. The minimum temperature of the surface at which this phenomenon occurs is called the Leidenfrost temperature [2].

Something similar can be observed when a drop impacts on a hot surface with a certain velocity: in this case, the vapour film between the drop and the surface not only prevents a rapid evaporation of the liquid, but allows the drop to bounce on the surface [3], which is known as “dynamic Leidenfrost phenomenon” [4]. After the initial spreading immediately after impact, surface forces cause the drop to retract in order to retrieve its spherical shape; however, the vapour film prevents the drop from wetting the surface during retraction, which reduces the energy dissipation due to shear flow and leaves more of the initial kinetic energy available for bouncing. Moreover, if the temperature of the surface is very high, the elasticity of the vapour film and the momentum exchange between the drop and the film are also believed to contribute propelling the drop away from the surface [5].

Roughly speaking, the dynamic Leidenfrost phenomenon occurs for low values of the Weber number (i.e., when the ratio between the kinetic energy and the surface energy is small enough to prevent the drop from breaking-up into smaller droplets) and temperatures high enough to allow the formation of the vapour film. In general, the temperature of the wall must be significantly higher than the Leidenfrost

temperature for the same fluid/surface combination, because in the dynamic case the vapour film tends to collapse under the combined action of the drop weight and the dynamic pressure rise during impact: thus, a higher rate of evaporation hence a higher wall temperature is required to ensure the stability of the vapour film.

If the vapour film is not stable, for example because the temperature of the surface is not high enough, the liquid may locally touch the surface and start boiling: then, the small vapour bubbles formed inside the drop quickly rise and burst on the drop surface, scattering all around small satellite droplets, which is known as “secondary atomisation” or “drop miniaturisation” [6].

Thus, one can define a dynamic Leidenfrost temperature as the lowest temperature for which the vapor cushion causes the drop bouncing without secondary atomization or splashing. This temperature has been shown to be a growing function of the impact Weber number [7]. A qualitative overview of the possible outcomes of drop impact on heated surfaces is shown in Figure 1 with respect to the impact Weber number and the temperature of the surface.

The dynamic Leidenfrost phenomenon is a rich source of problems for science, but is also important in many practical applications, such as spray cooling, firefighting, and steel quenching. In most cases, drop bouncing has a negative effect on the process, because when the drop is far from the surface its cooling or quenching efficiency is greatly reduced.

Although the fluids used in most applications are Newtonian, the interest for non-Newtonian liquids is rapidly growing. Very often, these fluids are obtained by adding tiny amounts of flexible polymers to a Newtonian solvent: this is sufficient to change the behaviour of the fluid, which becomes viscoelastic, without affecting significantly its shear viscosity and surface tension.

Polymer additives are known to have significant effects on the dynamics of drop impacts. For example, it has been shown that a small quantity (of the order of 100 p.p.m.) of polyethylene oxide (PEO) can reduce the tendency of drops to rebound after impacting on hydrophobic surfaces, which can be exploited to control spray applications [8],[9]. In addition, polymer additives increase the critical Weber number for drop break-up (splashing) [8],[10] and completely inhibit secondary atomisation during the impact on hot surfaces [10],[11].

These effects are believed to be related to the elongational viscosity of the fluid [12], which is defined as the ratio of the first normal stress difference to the rate of elongation of the fluid, and for a polymer solution can be two or three orders of magnitude higher than that of water [13]. In particular, it has been suggested that the elongational viscosity causes large energy dissipations, so that nothing of the impact kinetic energy is available to propel the drop off the surface. This seems to be confirmed by the retraction velocity of the drop after maximum spreading, which for polymer solutions is one or two order of magnitudes smaller than that measured for the pure solvent [8],[9]. However, although the relationship between the retraction velocity and the elongational viscosity seems to be well established, the actual physical mechanism is still far from being understood.

This work aims to investigate the effect of polymer additives on bouncing Leidenfrost drops. In particular, it is shown that by suppressing secondary atomisation polymer additives alter the conventional definition of dynamic Leidenfrost temperature.

For surface temperatures above the dynamic Leidenfrost point (i.e. when secondary atomisation is not observed), wetting effects are negligible because of the vapour layer between the drop and the surface: thus, from this study one can also get a deeper insight of the effect of additives on the drop impact dynamics on hydrophobic surfaces.

EXPERIMENTS

Apparatus and procedure

The experimental setup is schematically shown in Figure 2. Drops were created at the tip of a hypodermic needle with flattened bevel by a screw-driven syringe dispenser, and detached under their own weight. Two needles with inner diameters of 0.838 mm and 0.495 mm (gauge 18 and gauge 21, respectively) were used to create drops of different diameters.

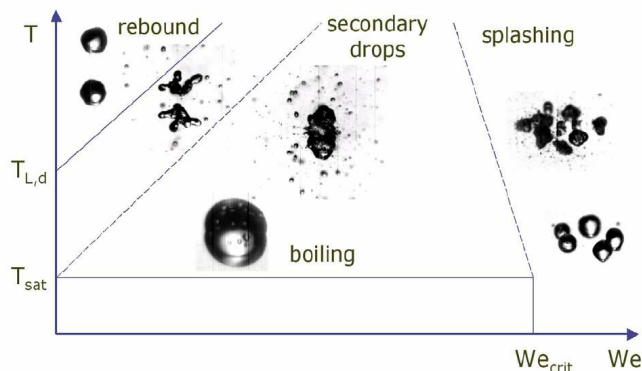


Figure 1. Qualitative map of drop impact regimes on a heated surface.

The dropping height (hence the impact velocity) was adjusted using a Vernier height gauge with a precision of ± 0.02 mm.

The needle was positioned above the surface of an aluminium square block (40 mm \times 40 mm) containing two electric cartridge heaters (100 W each) symmetric with respect to the point of impact to ensure a uniform temperature field. The surface was mirror polished with a chemical abrasive.

Temperature could be controlled within $\pm 1^\circ\text{C}$ by a PID controller driven by a K-thermocouple placed 1 mm below the point of impact. Drop impacts were recorded for constant wall temperatures ranging from 150°C to 400°C ; the higher limit was chosen to ensure stability of the vapour film and avoid the formation of secondary droplets [7],[11].

A high-frame rate CMOS camera (Mikrotron MC1310) equipped with a 18–108/2.5 macro zoom lens (Navitar Zoom 7000) and horizontally aligned with the surface recorded the impacts of single drops. Back-to-front illumination was provided by a LED backlight (Advanced Illumination) which ensured a uniform illumination intensity, and images with a resolution of 480×480 pixels were captured at 1,000 frames per second.

Magnification was kept constant throughout all experiments and lengths on the image could be calculated by comparison with a reference length (typical spatial resolution: 35 pixels/mm). To ensure a fine optical alignment, the camera, the heated surface and the backlight were fixed to an optical breadboard.

Quantitative data were extracted from images using proprietary software developed in LabView environment, which after background subtraction and image optimization measured the gap between the drop and the wall, as well as the drop dimensions in the vertical and in the horizontal directions.

Drop characterization

In this work, drops were created using two different fluids: de-ionized water and a 200 p.p.m. solution of poly-ethylene oxide in the same water. The PEO, supplied by Aldrich Chemicals under the form of granular powder, had an average molecular weight of 4 000 000 a.m.u. and a typical density of 1210 kg/m^3 .

To allow a homogeneous dispersion of the additive, solutions were prepared by gently dissolving the polymer into a batch of ultra-pure water, where a strong vortex was created by a magnetic stirrer.

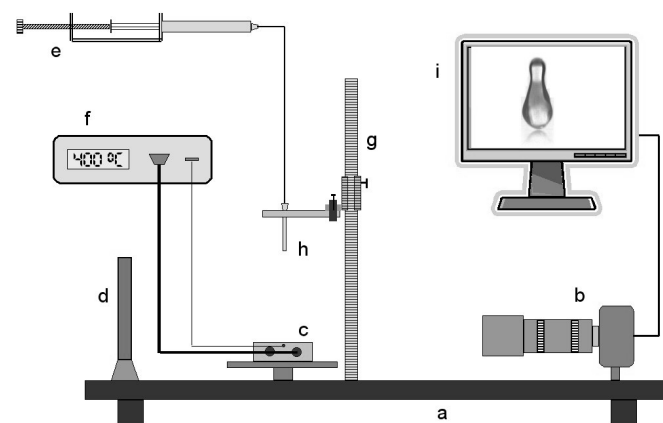


Figure 2. Experimental setup (see main text for description).

Drop weight measurements made with a precision balance (Mettler Toledo MT100) allowed calculation of the drop diameter at equilibrium:

$$D_0 = \sqrt[3]{6m/\pi\rho} \quad (1)$$

Statistical measurements over 50 samples gave values of water drops had diameters of 2.76 ± 0.14 mm and 3.49 ± 0.04 mm, whereas drops of polymer solution had diameters of 2.66 ± 0.1 mm and 3.32 ± 0.06 mm, respectively for the gauge 21 and the gauge 18 needles.

To determine the properties of the liquid during the impact (in particular, the surface tension), it is necessary to estimate the drop temperature with a reasonable precision. Because there is a co-existence of the liquid and the vapour phase, if one neglects the pressure wave caused by the impact [14] and other nonequilibrium effects, the drop should be at saturation (100°C for water at atmospheric pressure). However, the duration of the impact is so short (typically, about 5 ms for the initial spreading phase and about 50 ms for the subsequent retraction phase) that it may not be sufficient to bring whole drop in a condition of uniform temperature.

Since in the present experiments drops are released from the dispensing needle at ambient temperature, it is important to verify whether the duration of the contact with the hot surface is sufficient to heat the drop uniformly at the saturation temperature. A simple energy balance carried out assuming that the main heat transfer mode is thermal conduction in the vapour layer yields:

$$mcdT = \frac{k_{vap}}{\delta(t)} (T_{surf} - T) S(t) dt \quad (2)$$

where T is the average temperature of the drop, m its mass, c the heat capacity of water, k_{vap} the thermal conductivity of the vapour layer, $\delta(t)$ its thickness, and $S(t)$ the surface of the drop exposed to the heat flux.

To estimate an upper limit for the heating time (worst case), one can make the approximations $S(t) \approx \pi D_0^2/4$ and $\delta(t) \approx \delta_{max}$, where δ_{max} is proportional to $D_0^{4/3}$ [15], and integrate between the ambient temperature and the saturation temperature:

$$t_h < \frac{2\rho D_0 c \delta_{max}}{3k_{vap}} \ln \frac{T_{surf} - T_{amb}}{T_{surf} - T_{sat}} \quad (3)$$

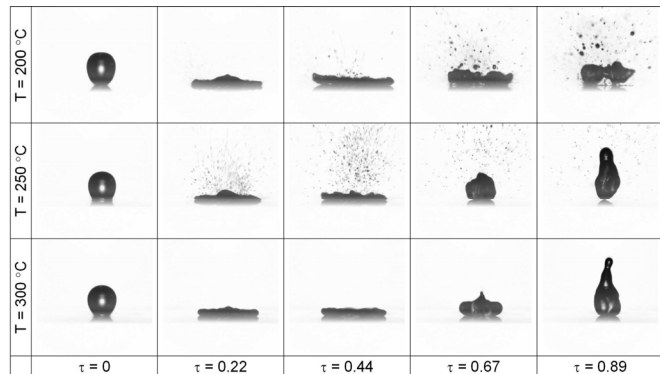


Figure 3. Impact of a drop of pure water ($We = 28$): secondary atomisation is observed for $T < T_{LD}$.

For a drop with a diameter of 4 mm impinging on a surface at 400°C , and taking $\delta_{max} = 50 \mu\text{m}$ [15], the time necessary to reach the saturation temperature is less than 5 ms, i.e. the same duration of the spreading stage after impact.

Because t_h estimates the heating time in excess, it is reasonable to assume that the drops considered in the present work attain the saturation temperature upon the first contact with the wall, before maximum spreading. Thus, in the following sections the relevant dimensionless numbers will be calculated using the fluid properties at 99°C .

The surface tension and the viscosity of the polymer solutions at saturation were measured by linear extrapolation of values measured in the range $20^\circ\text{C} - 80^\circ\text{C}$. Viscosities were measured with a rotational rheometer (Haake MARS II) equipped with a 60 mm plate/plate geometry and Peltier temperature controller, while equilibrium surface tensions were measured using a maximum bubble pressure instrument (Krüss PocketDyne).

In particular, the surface tension of water was 58 mN/m and its viscosity 0.28 Pa·s, while those of the polymer solution were 54 mN/m and 0.48 Pa·s, respectively.

For small distances of the dispensing needle above the impact surface ($H \leq 15$ cm), the impact velocity was found to be identical to the theoretical free-fall velocity, $u = \sqrt{2g(H - D_0)}$. Thus, the impact Weber number was calculated using the following expression:

$$We = \frac{\rho D_0 u^2}{\sigma} = \frac{2\rho g D_0 (H - D_0)}{\sigma} \quad (4)$$

The drops considered in the present work had impact Weber numbers in the range between 7 and 160.

RESULTS

Secondary atomization

Figure 3 shows examples of the impact of a pure water drop for different wall temperatures, as a function of the dimensionless time, $\tau = tu/D_0$. If the temperature is not sufficiently high, the first stages of impact are accompanied by secondary atomisation, which however becomes less important as temperature grows. Above the dynamic Leidenfrost temperature, no secondary atomisation can be observed.

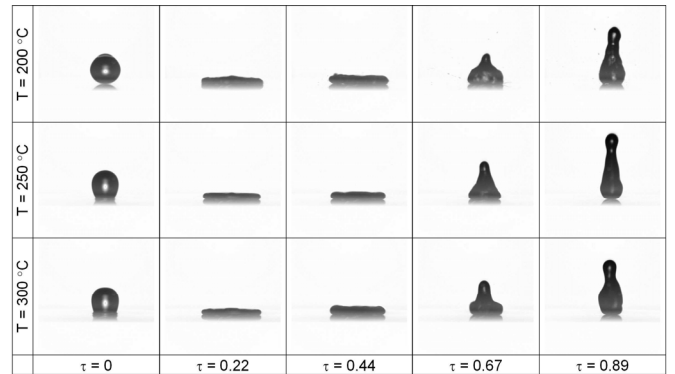


Figure 4. Impact of a drop containing 200 p.p.m. of PEO ($We = 28$): no secondary atomization is observed.

The behaviour of drop containing the polymer additive is completely different, as shown by the images of Figure 4. In this case, there is no secondary atomisation even for the lowest temperature.

Although the physical mechanism of such behaviour is still poorly understood at present, there is a general agreement about the fundamental role of the elongational viscosity, because this is the only parameter that changes significantly between the two fluids. In particular, the elongational viscosity is thought to improve the stability of the drop free surface during expansion on the hot wall immediately after collision, which prevents the nucleation of vapour bubbles as well as the ejection of satellite droplets.

The transition temperature where drop bouncing occurs without secondary atomisation, which conventionally defines the dynamic Leidenfrost temperature [5], is plotted with respect to the impact Weber number in Figure 5, for the two fluids considered. The polymer additive has a dramatic effect on T_{LD} , which is significantly lower than in the case of the pure solvent, and very close to the measured value of the Leidenfrost temperature of water on a polished aluminium surface, $T_L = 162 \pm 5$ °C [2].

Dry rebound

Above the dynamic Leidenfrost point, the vapour film between the drop and the hot surface is stable, and the liquid is never in contact with the wall. Thus, one can study drop bouncing in the absence of wetting effects, for both Newtonian and viscoelastic drops.

The maximum spreading diameter of drops after impact on a surface at 400°C is plotted in Figure 6 with respect to the impact Weber number. Theories based on the conservation of energy [16],[17] suggest that this quantity should scale as $We^{1/2}$.

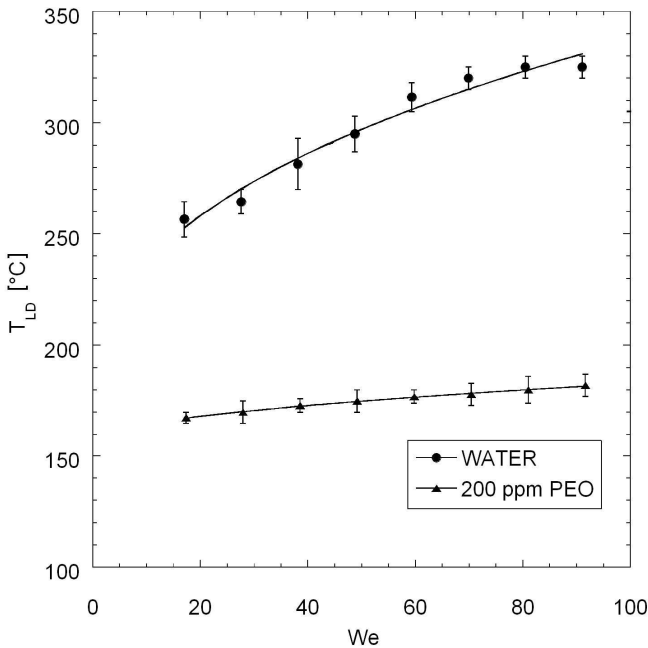


Figure 5. Dynamic Leidenfrost temperature for impacting drops of pure water and of polymer solution. Error bars indicate the temperature range where one may observe either secondary atomisation or dry rebound in a set of five experiments.

However, more recently it has been proposed that for $We \gg 1$ a model based on the conservation of momentum is more accurate, because it is difficult to quantify energy dissipation during impact [18],[19]. Whilst latter approach seems to be confirmed by the trends obtained for water drops, in particular for $We > 30$, the maximum spreading diameter of drops containing the polymer additive strictly follows the trend predicted by the conservation of energy approach.

In any case it must be observed that for $We > 20$ the maximum spreading diameter of viscoelastic drops is systematically smaller than that of water drops having the same impact Weber number. This means that the fraction of impact kinetic energy (which is proportional to the Weber number) converted into surface energy (which is proportional to the area of the drop surface at maximum spreading) is smaller.

In principle, such reduction of the surface energy at maximum spreading could be interpreted as the consequence of increased dissipation during the expansion stage due to the higher elongational viscosity of the polymer solution, providing an independent confirmation of the theory proposed to explain why polymer additives cause the suppression of drop rebound on hydrophobic surfaces [8],[9],[12]. However, this does not exclude that the missing surface energy may be stored elsewhere, for example as elastic energy.

In order to get a deeper insight of this phenomenon, one can observe what happens during the rebound stage, when surface energy is converted back to kinetic energy and propels the drop off the surface. In particular, the maximum height of the centre of mass of the drop allows one to calculate exactly the fraction of surface energy recovered as mechanical energy during this stage:

$$E_{rec} = mgH_{max} \quad (5)$$

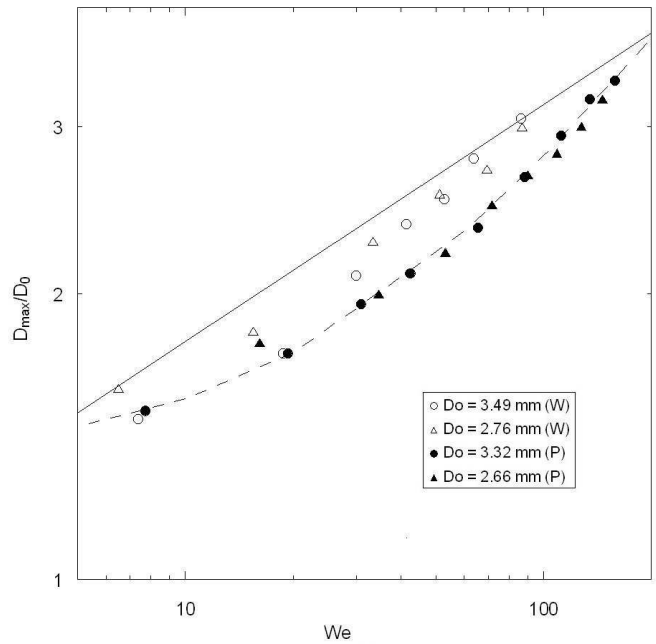


Figure 6. Maximum spreading diameter of Leidenfrost drops of pure water (open symbols) and polymer solution (filled symbols), impacting on an aluminium surface ($T = 400$ °C). The solid and dashed lines represent the trends $D_{max}/D_0 \sim We^{1/4}$ and $D_{max}/D_0 \sim We^{1/2}$, respectively.

Figure 7 shows the maximum height of the drop centre of mass during rebound. For $We \leq 50$, no significant differences can be observed between drops of pure water and viscoelastic drops. On the contrary, for $We > 50$ the maximum height reached by viscoelastic drops is significantly larger than that of Newtonian drops, irrespective of the drop diameter.

These results are somewhat surprising, because they clearly show that viscoelastic drops can recover a higher fraction of the initial impact kinetic energy even if they store less in the form of surface energy. This seems to be in contrast with the scenario of higher energy dissipation in the fast elongational flow of polymer solutions described above [8],[9],[12].

Neglecting the momentum transferred to the drop by evaporation and the elasticity of the vapour cushion [5] because their effects are similar for both the Newtonian and the viscoelastic drops at the same impact Weber number, one can write the following energy balance for two drops (one Newtonian and one viscoelastic) at maximum spreading after impacting on a hot surface with the same velocity:

$$E_{surf}^{(W)} + E_{diss}^{(W)} = E_{surf}^{(P)} + E_{diss}^{(P)} + E_{el}^{(P)} \quad (6)$$

Equation (6) implicitly postulates that a fraction of the impact kinetic energy is stored as elastic energy in the polymer solution. Since the results shown in Figure 6 suggest that the surface energy of viscoelastic drops is smaller than that of Newtonian drops, the relation between the energy dissipations in the two cases is:

$$E_{diss}^{(W)} < E_{diss}^{(P)} + E_{el}^{(P)} \quad (7)$$

However, Eq. (7) does not allow one to establish where the higher energy dissipation occurs. A more quantitative analysis can be obtained from the comparison between the mechanical energy recovered after the first rebound, given by Eq. (5), and the initial impact kinetic energy.

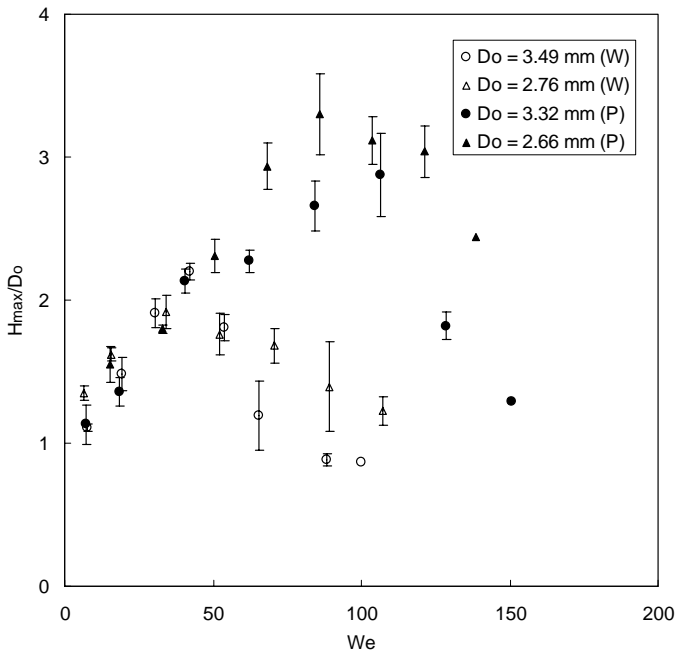


Figure 7. Maximum height of the centre of mass of Leidenfrost drops of pure water (open symbols) and polymer solution (filled symbols), after bouncing on an aluminium surface ($T = 400^\circ\text{C}$).

Figure 8 confirms that for $We > 50$ the mechanical energy recovered by viscoelastic drops is considerably higher than that of Newtonian drops, while no significant differences can be observed for $We < 50$. This obviously implies that the overall energy dissipation during the impact of viscoelastic drops is smaller.

Because these results have been obtained in a perfectly non-wetting case, one can argue that the suppression of rebound observed in drops of dilute polymer solutions impacting on hydrophobic surfaces is not due to a higher energy dissipation during impact, but rather to some still poorly understood effect of flexible polymers on the dynamic wetting behaviour of drops.

CONCLUSIONS

Small amounts of flexible polymers dissolved into a Newtonian solvent were shown to have important effects on the behaviour of bouncing Leidenfrost drops. In particular, drops containing polymer additives do not exhibit secondary atomization, which affects the conventional definition of dynamic Leidenfrost temperature (i.e., the minimum temperature for which no secondary atomization can be observed) for viscoelastic drops. Moreover, additives enhance drop rebounds observed for Weber numbers greater than 50.

Although a complete understanding of these effects remains elusive at present, it seems that polymers reinforce the stability of the drop free surface, which on one side prevents the liquid to make contact with the hot wall and initiate boiling, and on the other side prevents the surface instability that leads to the creation of satellite droplets.

Moreover, the enhancement of drop rebound observed for polymer solutions suggests that the overall mechanical energy dissipation during impact is smaller than that of Newtonian drops, which sheds some light on the understanding of the effect of polymer additives on the drop impact dynamics on hydrophobic surfaces.

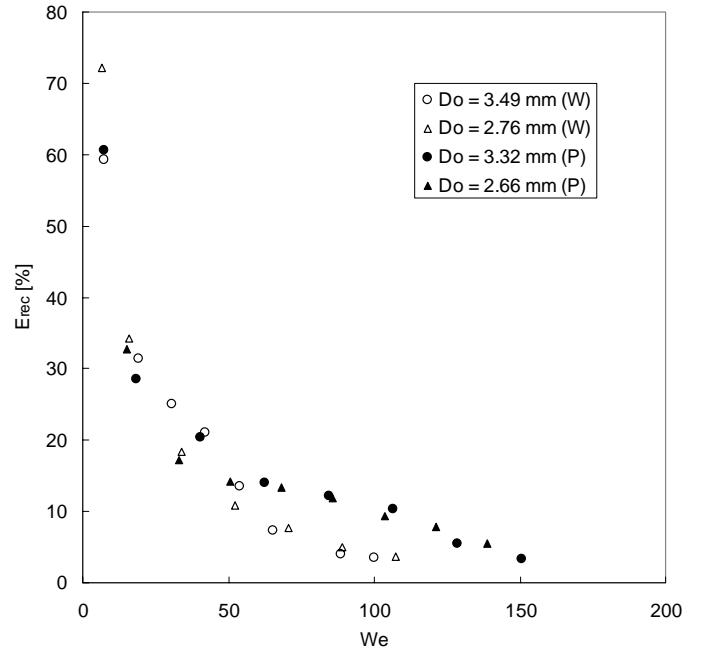


Figure 8. Mechanical energy recovered by Leidenfrost drops of pure water (open symbols) and polymer solution (filled symbols), after the first rebound on an aluminium surface ($T = 400^\circ\text{C}$).

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NOMENCLATURE

Symbol	Quantity	SI Unit
c	heat capacity	$\text{Jkg}^{-1}\text{K}^{-1}$
D_0	equivalent drop diameter	m
E	energy	J
g	gravity	m s^{-2}
H	falling height	m
H_{\max}	maximum height of center of mass	m
k_{vap}	thermal conductivity of vapour	$\text{Wm}^{-1}\text{K}^{-1}$
m	drop mass	kg
S	surface exposed to heat	m^2
t	time	s
T	temperature	K
u	impact velocity	m s^{-1}
We	Weber number	
Greek:		
δ	vapour layer thickness	m
η	viscosity	Pa s
ρ	density	kg m^{-3}
σ	equilibrium surface tension	N m^{-1}
τ	dimensionless time	

Subscripts and superscripts

amb	ambient
el	elastic
diss	dissipated
h	heating
L	Leidenfrost
LD	Leidenfrost, dynamic
(P)	polymer solution
rec	recovered
sat	saturation
surf	surface
(W)	water

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