



# The impact and deformation of a viscoelastic drop at the air–liquid interface

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## ABSTRACT

The parameters that influence the deformation of viscoelastic drops impinging on a viscoelastic bath, and that lead to the detachment of these drops from the surface were investigated. A range of PEO solutions with different viscosities and molecular weights was used and the deformation of each drop during impact was observed using a high speed camera. The use of image analysis to measure the evolution of interfacial areas between the drop and the cavity formed during impact allowed the estimation of the potential and interfacial energies. This gave valuable information for the understanding of drop detachment. The drops need to retain a sufficiently high kinetic energy after impact in order to pass through the surface. It is therefore necessary to limit the deformation of the drop as well as the deformation of the bath (i.e. cavity depth) by increasing the drop viscosity. Reducing the kinetic energy of the drop at the moment of impact also limits deformation and promotes detachment of the drop.

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## 1. Introduction

Many applications involve the impact of liquids on solids, such as ink jet printing, spray cooling, fire suppression via sprinkler systems, the bouncing of drops on hydrophobic surfaces, spraying of pesticides, or the splashing of raindrops on dispersions of seeds or microorganisms. Other applications involve the impact of solids on liquids such as in metallurgical or plastics processes where solids are added to melted materials, or in the case of hydro-ballistics. The impact of liquids on liquids is also of importance for the formation of capsules or gelled beads, such as the formation of alginate beads gelled in a solution of calcium chloride. We are also all familiar with the noise of rain drops on the surface of a liquid/air interface [1,2], such as a lake, or the air entrainment of a steady jet into a pool of liquid [3]. These applications involve deformation of liquid drops or liquid surfaces and in some cases the formation of cavities.

Many studies have thus investigated the deformation of drops upon impact with solids [4–6]. During the impact of liquid droplets or solid spheres [7] on liquid surfaces, various phenomena are observed such as the Worthington jet [8,9], the formation of a crown with dissipative waves [10–12] at the liquid surface, vortices [13], air entrapment [3,14,15] and the spreading of droplets on surface [16]. Some studies have also been performed on drop deformation when going through liquid–liquid interfaces [17,18] or when submitted to flow [19–22].

In many applications, liquid drops or baths are made of non-Newtonian materials, where the solution contains a viscous and an elastic component. During impact, the viscous component dictates the amount of energy dissipated and the elastic component dictates the amount of energy stored. In the current study, we focus on the deformation and conditions of detachment of viscoelastic droplets after impact with a liquid surface. The cavity formed at the surface of the bath, the deformation of the drop, and the drop/air interface at the surface of the bath, have been studied for a range of drop viscosities and elasticities. Control of many parameters such as the viscoelasticity of the drop and bath solutions, the kinetic energy of the drop at the moment of impact with the surface of the bath, and the surface tension is essential in order to obtain the detachment of drops from the surface of the bath.

Dimensionless ratios can be used to describe the behaviour during impact such as the Weber, Reynolds and Froude number which compare the relative importance of inertial forces due to the velocity of the drop, with the interfacial forces, due to the surface tension of the drop, the viscous forces or the gravitational forces. These parameters are defined as follows: The impact velocity ( $u_0$ ) required to perform the parameterisation can be found using a simple Newtonian calculation based on air friction and the gravitational force

$$u_0 = \sqrt{\left( \frac{g(1 - e^{-2AH})}{A} \right)},$$

where  $H$  is the fall height,

$$A = \frac{3C_f \rho_{\text{air}}}{\rho_{\text{drop}} R_0},$$

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$C_f$  friction coefficient = 0.7796,  $\rho_{\text{air}}$  density of the air,  $\rho_{\text{drop}}$  density of the drop,  $R_0$  radius of the drop.

Weber number (We):

$$We = \frac{\text{inertial forces}}{\text{interfacial forces}} = \frac{\rho_{\text{drop}} u_0^2 2R_0}{\sigma},$$

where  $\sigma$  is the surface tension of the drop.

Reynolds number (Re):

$$Re = \frac{\text{inertial forces}}{\text{viscous forces}} = \frac{\rho_{\text{drop}} u_0 2R_0}{\eta}.$$

Froude number (Fr):

$$Fr = \frac{\text{inertial forces}}{\text{gravitational forces}} = \frac{u_0^2}{grR_0}.$$

In the current study, the drops had a radius of 1.1 mm and fell from a height of 15 cm leading to an impact velocity of 1.44 m/s. The surface tension used was the surface tension of the PEO solution, 62.1 mN/m. The values of Re ranged between 20 and 1050, dependent on the viscosity of the drops. For drops with low viscosity, the value of Re ( $\sim 1050$ ) suggests that the effect of inertial forces will have a much greater effect on the drop deformation than the viscosity of the drop. However, at higher viscosity,  $Re \sim 20$  and we can expect viscous forces to have a greater effect. When the height of the fall is not changed, Fr and We, calculated at the moment of impact, stay constant as the gravitational and interfacial forces are the same for all drops. When falling from 15 cm,  $Fr \sim 96$  and  $We \sim 74$  which suggests that the effect of gravitational and interfacial forces is low compared to the effect of inertia. In summary, the main criteria suspected to have a great effect are the kinetic energy of the drop as it impacts the surface, and the viscosity of the drop.

## 2. Materials and methods

### 2.1. Materials

Poly(ethylene oxide) (PEO) samples with different molecular weights (Fluka 81310 polyethylene glycol,  $M_w \sim 35,000$  g/mol; Sigma-Aldrich 372773 poly(ethylene oxide),  $M_w \sim 400,000$  g/mol; Sigma-Aldrich 372781 poly(ethylene oxide),  $M_w \sim 1,000,000$  g/mol) were used in order to explore solutions with different elasticities. Polyethylene oxide solutions with different concentrations were used to study the effect of viscosity on the drop formation.

0.1 w/w% toluidine blue was added to the drop solutions in order to have good optical contrast between the drop and the bath, and enable observation of the drops. This did not alter any of the material properties of the drops.

### 2.2. Material characterisation

#### 2.2.1. Rheology

The viscosity, storage ( $G'$ ) and loss ( $G''$ ) shear moduli were measured with a TA AR2000 rheometer. Parallel plate geometry was used with a narrow gap (150  $\mu\text{m}$ ) to allow accurate and reliable measurements. In order to have reliable measurements of  $G'$  and  $G''$ , and make sure we are in the linear regime,  $G'$  and  $G''$  have been measured for shear stress from 0.23 to 50 Pa and frequencies from 0.001 to 30 Hz. In order to have an idea of the range of viscosities obtained with the different molecular weights of PEO, we also measured the zero shear viscosity of solutions at different concentrations.

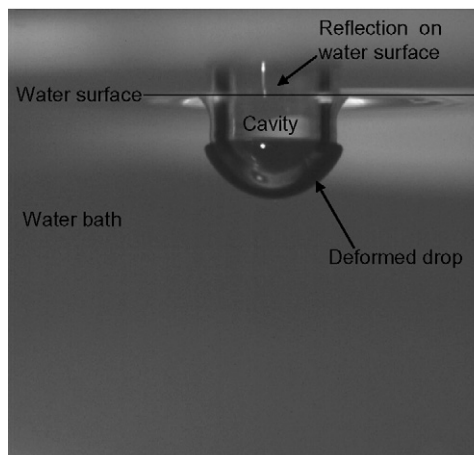


Fig. 1. Image taken under the surface after impact of a PEO drop in water.

#### 2.2.2. Surface tension

Dynamic surface tensions were measured using a SITA (SITA Messtechnik GmbH Gostritzer Str. 61–63, 01217 Dresden, Germany) online t60 bubble pressure tensiometer. This can measure surface tension in a range from 10 to 100 mN/m with a resolution of 0.1 mN/m and a range of bubble lifetimes from 30 ms to 60 s with a resolution of 1 ms.

#### 2.2.3. Density

The densities of the PEO solutions were measured using an Anton Paar DMA 35N handheld Density Meter at room temperature. This can measure density from 0 to 1.999 g/cm<sup>3</sup> with an accuracy of  $\pm 0.001$  g/cm<sup>3</sup>, in a range of temperatures from 0 to 40 °C.

#### 2.2.4. Drop deformation observation

Movies of drops falling into solutions have been made using a Photo-Sonics (Photo-Sonics International Ltd, 5 Thame Park Business Centre, Wenman Road, Thame, OX9 3FR, Oxfordshire) Phantom V5 high speed camera with a speed of 2000 frames per second (fps). The frames were recorded with a resolution of 512  $\times$  512 pixels. The drops were made using a 20 mm diameter syringe on a “genie” syringe pump (Kent Scientific Corporation, 1116 Litchfield St, Torrington CT 06790), that extruded the solution through a needle with an internal diameter of 0.5 mm, at a rate of 0.1 mL/min. The drop size obtained was 2.2 mm diameter in all cases. The distance between the end of the needle and the surface of the bath was typically 15 cm, although other heights were explored in order to vary the energy of the impact. Observation of the drops that detached from the needle showed that the drops’ solutions did not form a thread which would have slowed down their fall. The impact velocity was thus calculated using the height and the size of the drops measured when they detached from the needle. It was not verified experimentally using images. The bath solution was placed in a square beaker in order to have good visualisation with the camera, and the depth of the bath was 5 cm.

#### 2.2.5. Image analysis

The frames of the video have been separated to allow analysis of each frame. The time difference between two consecutive frames was 0.5 ms. Several parameters were analysed for each image in the sequence. The depth of the cavity formed during the impact of the drop has been measured on the pictures. Fig. 1 shows what is typically observed after impact of a drop on the surface of water. The contact area air/drop was estimated using an image analysis package (Inkscape) to draw on the image the visible 2D interface. A cylindrical rotation of the two halves of the draw-

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