

SPREADING DYNAMICS OF OIL FILMS ON AQUEOUS SOLUTIONS

ABSTRACT

KEYWORDS: Spreading, Hole nucleation, Hole Growth, Hole Coalescence, film coarsening, Rayleigh-Plateau instability in films.

Natural oil films and their dynamics in contact with aqueous substrates is interesting to several applications from digestion to emulsification. Natural oils are inherently complex fluids owing to their complex chemical as well as physical characteristics. The dynamics of such natural oil films on water or aqueous solution free surface is the subject of this experimental study. We examine the process of spreading and breakup of an oil film on water as well as on dilute aqueous solutions of Octanoic acid. Natural Oils from *Cocos Nucifera* (Coconut), *Sesamum indicum* (Sesame/Gingelly seeds), *Olea europaea* (Olive fruit), and *Ricinus communis* (Castor seeds) are used as the test fluids. Prior to all experiments, a range of physical as well as chemical properties of the test fluids has been measured. In each experiment a drop of test fluid (one of the natural oils) is introduced on to the free surface of the substrate fluid. The spatio-temporal evolution of the oil film is imaged and the characteristic features of the image were analyzed. In all cases, it was observed that the oil drop evolves into a film and spreads spontaneously because of the positive spreading coefficient. As the film continues to spread, its thickness decreases. At a critical film thickness, holes, which are voids in the oil film, begin to nucleate. When the hole area fraction approaches the random packing limit, these holes begin to coalesce resulting in a coarser structure of the oil film. Coalescence occurs when the liquid bridge separating a pair of holes is ruptured due to Rayleigh-Plateau instability. This process is completely spontaneous since it is driven by gradients in the interfacial energy landscape.

The oil film was observed to evolve through three phases viz., (i) spreading phase, (ii) hole nucleation and growth phase and (iii) hole coalescence phase. Quantitatively, the oil film diameters, number of holes in the oil film, diameter of each hole as well as the total triple line length have been measured. Natural oils are also very complex in their chemical nature. Typically, they are a mixture of several triglycerides of different fatty acids – saturated and unsaturated. The interfacial characteristics of natural oils with water are even more interesting owing to the fact that trace components of the oils are partially miscible in water. Additional solutal effects could arise as a result of the dissolution process. To study this effect, water was partially saturated with coconut oil and oil is saturated with water. These fluids partially mitigate solutal effects. Similar experiments were repeated with pure oil on partially saturated substrates as well as with partially saturated oil film on pure water. It was found that partially saturating the

substrate decreased the spreading rate of the oil film while saturating the oil increased the spreading rate of the oil film. Saturating the water substrate completely suppressed spreading. It was also observed that as the dissolution process progressed, solutal concentration gradients caused the interfacial tension to change, in turn influencing the dynamics. The coconut oil film diameter scales as $D_o \sim t^n$. The specific values are $n=0.5$, $n=0.43$ and $n=0.6$ for the experiments of pure coconut oil drop on pure water (PC PW), pure coconut oil drop on partially saturated water (PC SW) and saturated coconut oil drop on pure water (SC PW) respectively.

Experiments were also repeated by saturating the water substrate with Octanoic acid where the solubility level was known. These experiments allowed us to systematically study the effect of the dissolution process. From these experiments, a systematic decrease in spreading rate was observed on the oil film when the saturation level of OA in water substrate was increased. The coconut oil film scales as $D_o \sim t^n$. A generalization of the spreading law would involve two cases: oil on pure water ($\lambda = 0$) and oil film spreading on contaminated water ($\lambda > 0$). For $\lambda = 0$, D_o scales as \sqrt{t} . For $\lambda > 0$, D_o scales approximately as $t^{1/3}$.

It was observed that the spreading coefficient is reduced with increase in saturation value of substrate is increased. Because of this decrease in spreading coefficient, a proportionate decrease in spreading rate was observed. The spreading rate was measured as a change in the diameter of oil film with time. This oil film diameter is rescaled with the corresponding spreading coefficient and substrate fluid viscosity. This change in modified diameter of oil film of the different spreading experiments, on pure substrates and saturated substrates collapses into a single line. This collapse shows that spreading coefficient can be used to quantify the effects of saturations values on the spreading rate.

The hole nucleation process and the corresponding rate was observed to be affected significantly when the oil or water substrate was saturated. As far as the hole growth is concerned the diameter of the hole scales as $D_i \sim t^n$. The exponent (n) value is 0.5 for PC PW experiment and PC SW, SC PW and SC SW show exponents (n) of 0.3 respectively. Coalescence of the holes is observed by calculating the circularity of the coalescing hole at every time instant. It was observed that the hole coalescence rate is different for different sizes.

The motion and breakup of an Octanoic Acid drop (OA-drop) on the free surface of aqueous solutions (of Octanoic Acid and water) were also investigated. The OA drop spontaneously deforms and moves due to difference in surface tension (between the front and back of the drop). This difference in surface tension results in a solutal Marangoni force. This force also causes the drop to deform. This deformation could also lead to drop breakup into several daughter droplets for low concentrations of OA in the substrate fluid.