Wrinkles provide a simple method for determination of elasticity and thickness of an ultrathin film

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It is a common observation that under purely planar tension, a thin sheet deforms out of plane to form wrinkles. This happens because the elastic energy required to stretch a sheet is reduced by the out-of-plane bending that follows wrinkling. Wrinkles are observed on several occasions on different objects – we notice them on our skin as it is stretched by smiling, or ageing. A wound on healing contracts, thickens and wrinkles the neighbouring skin forming a scar[1]. Apples, mangoes, plums and several fruits wrinkle as they become old and dehydrate. The cream that floats on warm milk invariably forms wrinkles. Washed clothes wrinkle during drying.

Because thinner and lighter films are extensively used in technological applications involving coatings, optical reflectors and filters, dielectric stacks, lithographic resists, packaging, etc., there is a growing need for testing platforms to rapidly determine the thickness and mechanical properties of thin films, particularly thin film polymers. Only a few options are available to measure their mechanical properties and thickness[2]. For instance, the thickness of films may be ascertained by X-ray reflectivity measurements. In this context, it is of interest to note a recent report[3] which describes how with the help of a low-magnification microscope and a dish of fluid, measurements of both elastic modulus and thickness of ultrathin films can be accomplished by studying wrinkles formed by the capillary force exerted on the films. Huang et al.[3] prepared polystyrene (PS) films of thickness varying from 31 to 233 nm. To vary the elastic modulus of PS, different amounts of di-octylphthalate were added to PS as plasticizer. A circle of diameter 22.8 mm of each film was placed on water taken in a petri dish. Due to the hydrophobic nature of PS, the film floated and it was stretched flat by the surface tension of the air–water interface. An elegant method for the formation of wrinkles was adopted by placing a drop of water (~0.2 mg) using a micropipette in the centre of the film (Figure 1). The images of the wrinkling pattern shown in Figure 1 were obtained using a consumer digital camera mounted on a low-magnification microscope.

Wrinkles can also be induced by placing a solid disk at the centre of the film or by poking the film with a sharp point. An important difference lies between these two ways of producing the wrinkles and that using a drop of water. In the latter case, the wrinkling is not due to the weight of the drop, but due to the capillary force exerted on the film by the surface tension at the air–water–PS contact line. For a given film when the radius of the drop (a) was increased by adding more water, both the number (N) and length (L) of the wrinkles increased. On the other hand, N is smaller in thicker films (Figure 1). There are 111, 68, 49 and 31 wrinkles in films of thickness (h) 41, 72, 118 and 233 nm respectively (Figure 1). L is measured from the edge of the water droplet to the white circle, as shown at the top left in Figure 1.

The dependence of N on a and h leads to a scaling relation[3]: $N \sim a^{1/2}h^{-3/4}$, as a good linear relationship exists between N and $a^{1/2}h^{-3/4}$ for different film thickness (Figure 2).

Based on an earlier mathematical treatment on wrinkling observed in a stretched, slender elastic sheet cut-out of a polyethylene sheet by Cerda and Mahadevan[4], Huang et al.[3] obtained a relation:

$$N = C_N \left[\frac{12(1-\Lambda^2)\gamma}{E} \right]^{1/4} a^{1/2}h^{-3/4}, \quad (1)$$
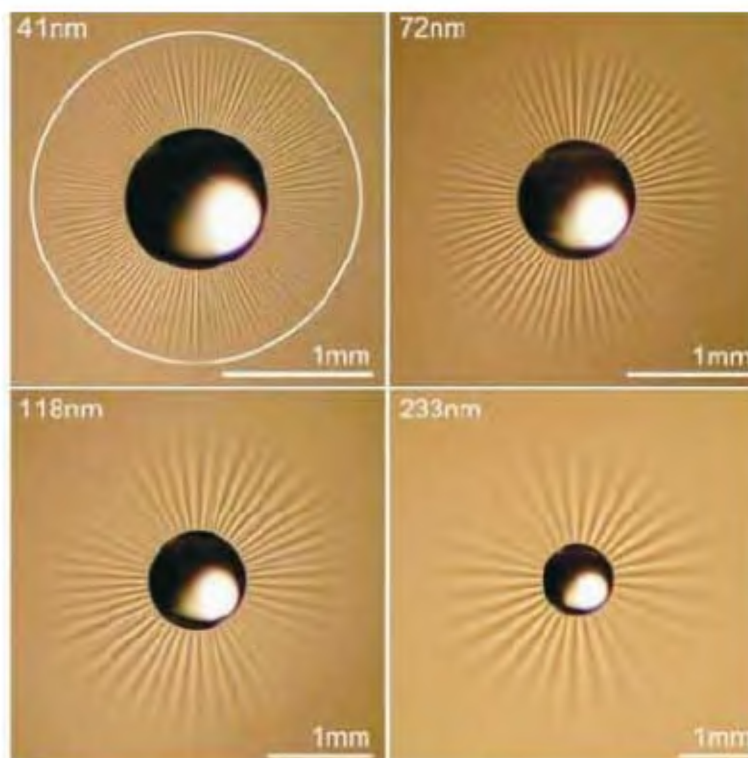

Figure 1. Wrinkles formed by placing ~0.2 mg (radius 0.5 mm) of water on four polystyrene films of diameter 22.8 mm. Thickness of films is indicated on the left hand corner for each case. The scale varies between images, whereas the water droplets are approximately the same shape. From Huang et al.[3]. Reprinted with permission from AAAS.
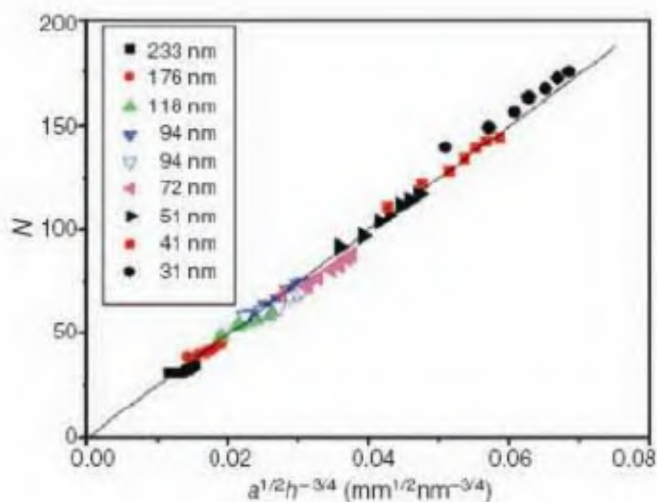
Figure 2. The number of wrinkles N as a function of a scaling variable, $a^{1/2}h^{-3/4}$. Data for different film thicknesses h (indicated by symbols in the legend) collapse onto a single line. The extent of reproducibility is indicated by the open and solid inverted triangles, which are taken for two films of the same nominal thickness. From Huang et al.[3]. Reprinted with permission from AAAS.
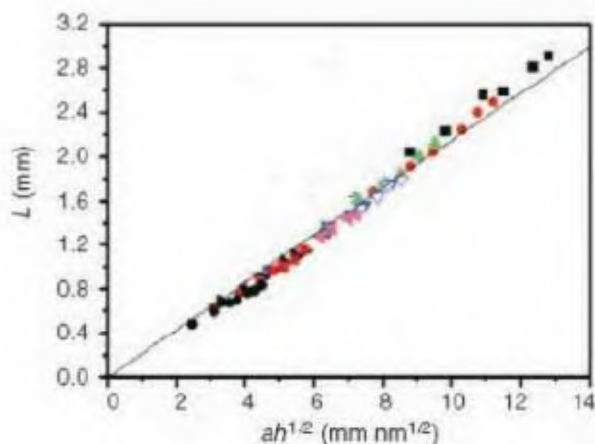

Figure 3. An approximate data collapse is achieved by plotting L against the variable $ah^{1/2}$. From Huang et al.[3]. Reprinted with permission from AAAS.

where C_N is a constant. From the slope of the linear fit and known values of Young's modulus ($E = 3.4$ GPa) and Poisson's ratio ($\Lambda = 0.33$) for PS and surface tension of water ($\gamma = 72$ mN/m), the value of C_N has been obtained as 3.62.

Huang et al.[3] also observed that L varies linearly with $ah^{1/2}$ (Figure 3) and therefore suggested the following equation (2) for the dependence of L on the radius of water drop and thickness of the film.

$$L = C_L(E/\gamma)^{1/2}ah^{1/2}. \qquad (2)$$

Thus counting N and measuring L of the wrinkles formed on a thin film due to the capillary force provides a simple and cost-effective method for the evaluation of the thickness and elasticity of the ultrathin film employing eqs (1) and (2).

This study is likely to shed light on how thin films behave and will probably have implications for understanding the behaviour of many materials, from membranes in water purification systems to the artificial skins used in the treatment of severe burns.

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