

## Liquid Film Instability on a Single Fiber

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Due to surface tension of a liquid, liquid films coating fibers or the inner surface of capillary tubes are unstable. They spontaneously break up into periodic array of droplets. Only very thin films can be stabilized by long-range van der Waals forces. This phenomenon is of big importance for textile materials, because they are being subjected to different kinds of liquids during various manufacturing and finishing processes where the stability of a liquid film is required. In this paper we present the simulation of liquid film instability using an Ising model combined with Monte Carlo computer simulation technique. The results of simulation are compared with the experimental ones.

*Keywords:* wetting, liquid film, instability, simulation.

### INTRODUCTION

Analysis and evaluation of fiber's wetting behavior is substantial in prediction of fibrous structures' performance. Those structures are textiles, fiber reinforced composites, etc. Wetting characteristics of fibers are determined by the value of contact angle, the work of adhesion and the surface energy. Those characteristics affect processability and the performance of structures while contacting fluids during their final application.

The instability of cylindrical liquid films was first studied by J. Plateau and L. Rayleigh [1]. The wetting of surfaces depends upon two criterions: chemical parameters of both liquid and solid, and upon geometry of the solid body. Geometry can be local and global. Local geometry is in the form of roughness and local pattern. Global geometry describes the shape of the solid body and can be in the form of cones, spheres, cylinders, etc. Textile fibers are, or often are assumed to be cylindrical [2–4].

In this paper computer simulation of liquid film instability phenomenon on a fiber is presented. Ising model combined with Monte Carlo technique is used for computer simulation. The simulation results are compared with the experimental ones: behaviour of viscous liquid (glycerine) on a cylindrical body (PA monofilament) is analysed.

### WETTING OF SOLIDS

Thickness of the film deposited on a flat surface is fixed by the competition between long-range forces (generally only van der Waals forces are taken into account) and Laplace pressure, which tends to thicken the film. This thickness  $e$  can be expressed through the spreading parameter  $S$  [1, 5]:

$$S = P(e) + e\Pi_d(e), \quad (1)$$

where  $P(e)$  is a contribution of long-range van der Waals forces, and is equal to:

$$P(e) = \frac{A}{12\pi e^2}, \quad (2)$$

where  $A$  is an effective solid-liquid Hamaker constant on the order of a few  $k_B T$  ( $k_B = 1.38 \cdot 10^{-23}$  J/K is a Boltzmann constant,  $T$  is a temperature).

If  $A$  is positive, the interaction is attractive (it is always positive between two identical bodies). In the case of wetting  $A$  is negative, because solid and gas repel each other through liquid.  $P(e)$  has its minimum for an infinite liquid thickness ( $e = \infty$ ), thus long-range forces tend to

thicken the film.  $\Pi_d(e) = -\frac{dP}{de}$  is a description of long-range forces through disjoining pressure or pressure that must be applied on a film to maintain it at constant thickness. In wetting conditions  $A < 0$  and  $\Pi_d(e)$  is positive. This pressure can be supplied by Laplace pressure. It is induced when the solid body is curved – for example, when wetting film is deposited on a fiber [1, 5].

If a liquid drop is deposited on a thin cylinder of radius  $b$  (that is at least one order of magnitude larger than liquid film thickness  $e$ ) the situation is quite different from wetting of plain surfaces because of the curvature of the solid surface, i.e. of its global geometry. In this case increase of the film thickness from  $e$  to  $e + \Delta e$  requires an additional surface energy of  $2\pi\Delta e\gamma_{LA}$  (where  $\gamma_{LA}$  is an interfacial tension between liquid and air) per unit length [1]. During wetting process the thickness of liquid film on a fiber cannot remain small because of this competition between disjoining pressure (pressure that helps to maintain constant thickness of a film) and Laplace pressure.

Fibers have convex surfaces, thus they imply a positive Laplace pressure that acts on the liquid-gas interface.

Very thin cylindrical film on a fiber has internal pressure that is composed of two terms:

$$p = \frac{\gamma_{LA}}{b+e} - \Pi_d(e), \quad (3)$$

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where the first term is Laplace pressure and the second one – pressure due to long-range interactions (van der Waals forces).

For the liquid film on a fiber to be stable this internal pressure  $p$  must be an increasing function of  $e$ :

$$\frac{\partial p}{\partial e} > 0. \quad (4)$$

This implies small thickness of the film [1]:

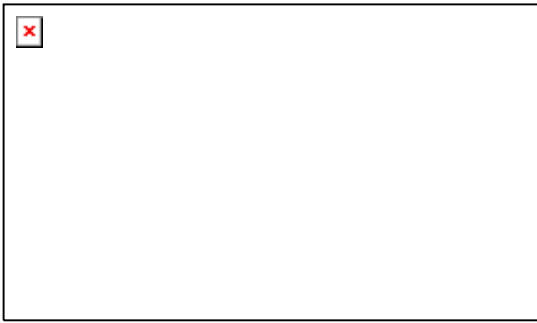
$$e < e_c = \sqrt{a \cdot b}, \quad (5)$$

where  $e_c$  is a critical thickness of a liquid film, and  $a$  is a convenient length on the order of 1 Å.

For liquid film on fibers of radius  $b$  ranging between 10 and 100 μm stability can be assured only if its thickness is smaller than few hundred angstroms. Thick film remains unstable – see Fig. 1. Competition between capillary and adhesion forces determines equilibrium configurations of liquid films and droplets. The distance between single drops can be calculated using Rayleigh equation:

$$\lambda_R = 2.88 \cdot \pi \cdot r, \quad (6)$$

where  $r$  is a radius of the column “fiber-liquid film” [1, 6 – 8].



**Fig. 1.** Thick liquid film spontaneously starts to undulate and later breaks down into single drops

## COMPUTER SIMULATION

In this step we tried to simulate previously theoretically described physical phenomena.

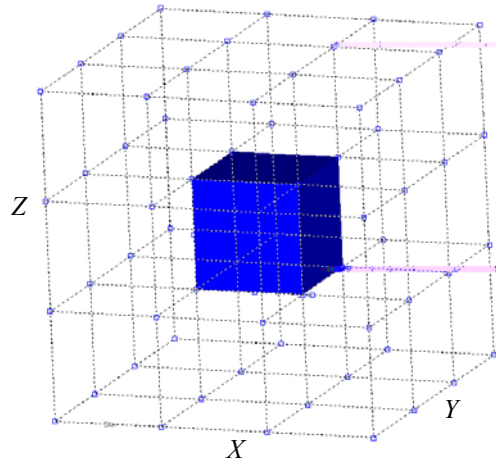
Simulation of liquid film behaviour on a cylindrical body was performed using modified 3D Ising model (or so called Auto-model) [9] and Monte Carlo method.

The simulation area was a cubic lattice consisting of  $80 \times 80 \times 80$  cells. Value  $c = 0, 1$  or  $2$  is attributed to every cell depending upon its content – each of cells contained either gas or liquid or fiber respectively. Each cell interacted with 26 neighboring cells forming so called supercube (see Fig. 2). At the initial moment of simulation every cell  $c_i$  had a certain value of energy that depended on the relationship of this cell with surrounding cells  $c_j$  in a supercube.

Cells containing fiber stayed immovable during simulation, only cells containing liquid or gas did or did not exchange their positions depending on their interaction energies as the system tended to have the smallest value of total energy. For the calculation of cell's  $c_i$  energy following equation was used:

$$E_i = G \cdot c_j + \sum_j C_{ij}, \quad (7)$$

where the first term was a gravitational part ( $G$  – gravity) of energy and the second term was a sum of exchange energies between cell  $c_i$  and its surrounding cells  $c_j$ .



**Fig. 2.** Single cell and its surrounding media forming so called supercube consisting of 27 cells

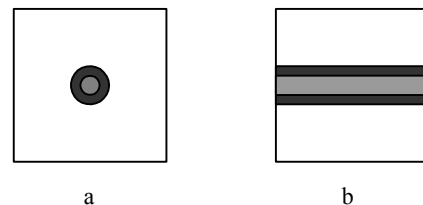
At each step of simulation two cells – one containing liquid and one containing air – were randomly chosen from the simulation area at the liquid-air interface and the sum of energies of those two cells using equation (7) were calculated. Then positions of those cells were exchanged and energies calculated again. The change of energy values was compared. If newly obtained energy was lower than previous one, cells stayed in their new positions. If it was higher, then Boltzman's law was used to calculate the probability of system's transformation from the first configuration into the second one:

$$W = e^{\left(\frac{-\Delta E}{\tau}\right)}, \quad (8)$$

where  $\tau$  was a value proportional to the thermodynamic temperature,  $\Delta E$  – the difference between energy of first and second configuration.

A number  $N$  was randomly generated and compared with calculated probability value  $W$ . If the latter was bigger than generated number, the cells stayed in their new positions; if it was smaller, cells were put back into their initial positions; and then finally there was no change in energy.

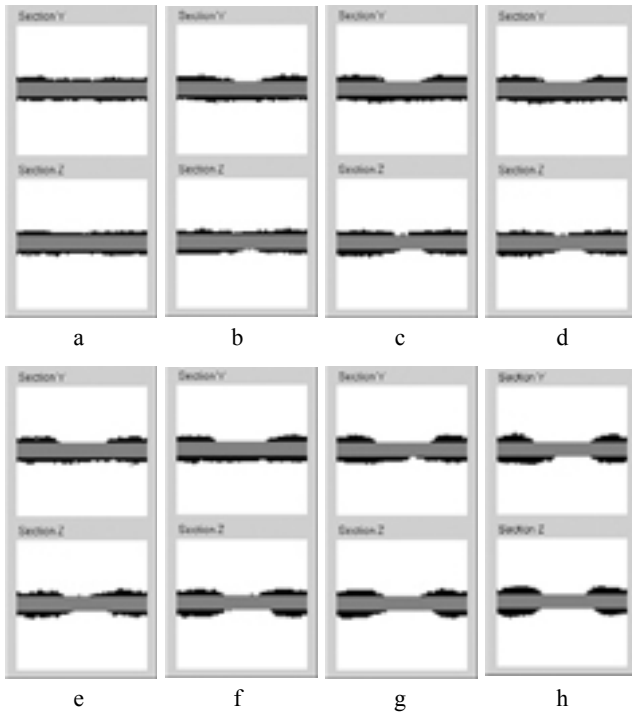
Initial setup of the system contains a horizontal fiber (parallel to  $X$  axis), uniformly coated by a thin liquid film and surrounded by air (see Fig. 3).



**Fig. 3.** Initial setup of the system. Cross-sections: (a) – perpendicular to  $X$  axis; (b) – perpendicular to  $Y$  and  $Z$  axes; colors: grey – fiber, black – liquid, white – air

Three cross-sections of simulation area were observed: perpendicular to  $X$ ,  $Y$  and  $Z$  axes. They passed through the center of the fiber, thus in the pictures (see Fig. 4) the diameter of the fiber is presented. Snapshots of the cross-sections were made at specific moments in time. There are presented snapshots of cross-sections perpendicular to  $Y$  (upper picture) and perpendicular to  $Z$  (lower picture) axes – the ones that are the most important.

It is well seen that liquid film starts to undulate first and then finally breaks down into separate drops of similar shape and size.



**Fig. 4.** Snapshots of simulation area: a – after 16 Monte Carlo steps; b – 26; c–g – after every next 2 steps; h – after 42 MCS. Upper picture – cross-section perpendicular to  $Y$ , and lower picture – perpendicular to  $Z$  axis

It should be noticed that the resulting conformation of liquid film is the same for various diameters of the fiber and various thicknesses of the film. Only in the case of very thick film influence of gravity becomes significant – then cells that contain water move downwards.

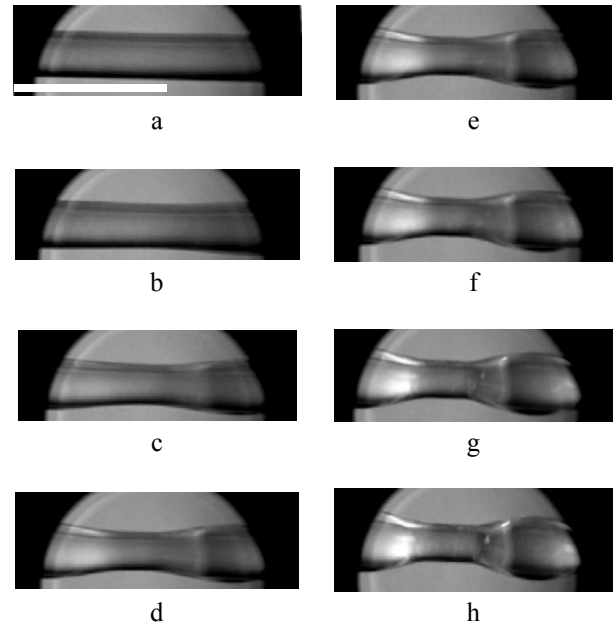
## EXPERIMENTAL

To have a better picture of this physical process and to be able to evaluate the results obtained from computer simulation we performed a set of experiments. In this section one experiment as a sample of liquid film instability is presented in details.

Experiments of this phenomenon were performed using a microscope and a camera. PA fishing line of diameter 0.18 mm was used as a cylindrical body and test liquid glycerine was assumed to be the right one, because high viscosity was desirable. Uniform glycerine film on PA monofilament was obtained by slowly drawing it horizontally out of a small diameter plastic cylinder filled with the test liquid. In order to ensure smaller liquid film

thickness the speed of the withdrawing was low – 60 mm/min (0.001 m/s). After pulling glycerine-coated monofilament in front of microscope equipped with camera, the motor was stopped and the development of liquid film instability was recorded. It was done from the side, to be able to evaluate the influence of gravity. Every 2 seconds pictures of the system (magnified for 20 times) were taken. Examples of obtained pictures are presented in Fig. 5.

In Fig. 6 an example of the change of system's "fiber-liquid film" diameter – the formation of single drops – is presented.



**Fig. 5.** Side view of the system: a – initial situation, b – after 10 s, c – 14 s, d – 18 s, e – 22 s, f – 26 s, g – 38 s, h – 50 s; length of bar – 1000  $\mu\text{m}$

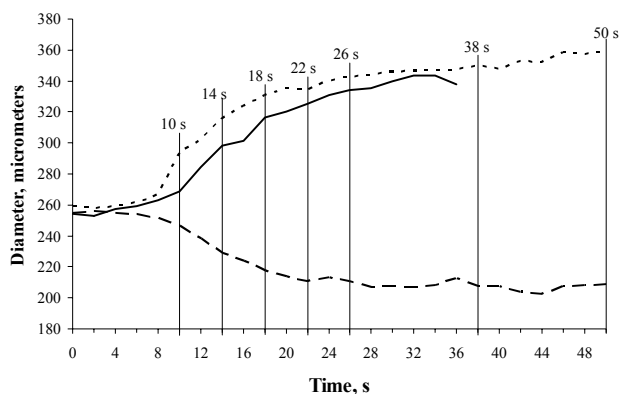
In Fig. 6 it is seen that the curve representing the development of the first drop breaks at certain point (38<sup>th</sup> second in this example). The problem we faced was that the distance between the drops was constantly increasing and at a certain moment first drop went out of our observation area.

The curves show general flow of the process: liquid film on the filament breaks down into two drops of similar size with regular spacing in between. At a certain point - we could call it critical point - the process becomes very intensive. This is the moment (at approximately 9<sup>th</sup> – 10<sup>th</sup> second as it is shown in Fig. 6) when destabilizing action of Laplace pressure takes place [6].

Experiments showed that the surface of the liquid film follows the sine curve as it starts to undulate. We both measured and calculated using (6) the wavelength (or the periodicity) of drops on a monofilament. Experimental wavelength was  $\lambda_{measured} = 1088.3 \mu\text{m}$ , and calculated value:

$$\lambda_{calculated} = 2.88 \cdot \pi \cdot r = 2.88 \cdot \pi \cdot 127 = 1148.5 \mu\text{m}$$

The difference between those values is only 5.24%. So it can be said that theoretical and experimental results are in a good agreement.



**Fig. 6.** Change of system's "fiber-liquid film" diameter as a function of time. Curves of one measurement: — – first drop, - - - - – trough, ..... – second drop

Experiments also showed that the influence of gravity is not very significant – in general, drops are deposited symmetrically to the filament's longitudinal axis. Despite this fact, in computer simulation influence of gravity was taken into account.

It should be noticed that there is one small difference between the results obtained from computer simulation and the results obtained from the experiments. From the snapshots of simulation a bare fiber between two liquid drops is seen, but in the pictures obtained from the experiments there is still thin liquid film between two drops present (after 50 seconds it was still in the range of 10 μm). This film is also called *precursor film*. It becomes thinner in time, but after all it is still present because due to the influence of adhesion forces (van der Waals forces). Equilibrium configuration of liquid droplets and precursor film on a single fiber is determined by this competition between capillary forces (Laplace pressure) and adhesion between liquid and solid.

## CONCLUSIONS

In general mathematical simulation of the processes showed good congruity with the experimental results – liquid film spontaneously breaks down into drops. The usage of Ising model (or so called Auto-model) and Monte Carlo method for those simulations complement the

behavior of a real system. Experiments also showed that liquid film on the fiber is not stable - it is well seen from presented figures. The initial thickness of the liquid film, presented in those figures is ~37 μm and does not imply on the stability of the system. Unfortunately viscous liquids, used in many processes of textile technology (and also with different textile products) can not ensure the required (few hundreds of angstroms) thickness of the film. Thus further research on those features should be performed. Above described simulation could be further used for the analysis of liquid film instability on a cylindrical body – single fiber.

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