



# Liquid spreading on solid surfaces and penetration into porous matrices: Coated and uncoated papers



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## ARTICLE INFO

Available online 3 March 2015

### Keywords:

Liquid  
Wetting  
Spreading  
Penetration  
Power-law  
Models

## ABSTRACT

Liquid spreading on solid surfaces and penetration into porous matrices (powders and coated papers) are investigated. The influence of chemical and structural heterogeneity on equilibrium and dynamic surface wetting is evaluated both experimentally and theoretically. Single capillary systems are used to identify the predominating mechanisms for acceleration, momentum, inertial and viscous liquid penetration. Different stages of vertical and horizontal penetration of liquids from non-limited, restricted (sessile drop) and cut-off sources into powders and papers are evaluated with reference to a range of frequently used models. For all types of liquid transport power-law exponents are used to relate all observations. The applicability of models from which the exponents are derived is discussed. Results are compared to theoretical predictions for liquid penetration. Models are of general validity, but the focus is placed on probe liquid spreading on and penetrating into coated and uncoated papers. This sets a particular challenge, since papers are heterogeneous layered composites of powder compacts on fibrous network. For the evaluation of models published results are supported by extended original results.

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**1. Introduction**

In previous investigations wetting was reviewed and the processes involved evaluated [1] and compared to similar processes in solution [2]. The work of cohesion, adhesion, spreading and immersion were inter-related to the chemical nature of solids [3,4]. Later specific attention was focused on equilibrium and dynamic wetting of heterogeneous paper surfaces [5–7]. The chemical nature of surface sites was characterized with probe liquids and solids resulting in identification of dispersive (Lifshitz-van der Waals), acid and base contributions to the surface energy of surfaces [8–12]. In order to model the surface on a molecular level it is necessary to characterize the density and nature of the surface sites and to introduce models accounting for them [1,5,13]. Moreover, the influence of surface structure heterogeneities was characterized [14–16]. Equilibrium work of spreading relates to hydrodynamic movement of a multilayer film over the surface [17–23]. Dynamic wetting processes may also depend on monomolecular hopping and diffusion between sites [23–26]. In this report liquid spreading on solid surfaces and penetration into porous matrices is reviewed. A special focus is placed on the interconnection between initial surface spreading and the following penetration of liquid into the substrate. Although general for all porous materials, coated paper consisting of a dense powder layer on a porous fiber matrix is of key interest.

**2. Wetting of solid surfaces**

The spreading of a liquid on a surface is strongly dependent on the experimental conditions such as chemical properties and vapor pressure of the liquid [1,2], chemical nature of the solid surface [3,4] and the structure or degree of surface roughness [14–16]. The most important results on equilibrium and dynamic wetting from recent reviews [1,5] are reported in this section.

*2.1. Equilibrium wetting of solid surfaces*

The aim of this chapter is to define the symbols and concepts used throughout this review. Usually nearly ideal liquids and polymers are used when discussing wetting. In this chapter probe liquids with a large range of properties are used to characterize rather non-ideal solids (silica and titania) which are of interest in paper coating. The deviation from ideality is illustrated by plots, tables and suitable references. The limits of the corrections are briefly discussed.

*2.1.1. Basic definitions and characterizations*

Since non-relaxed solid surfaces are included in the considerations it is advisable to specifically distinguish the unique relaxed surface tension of liquid components denoted as  $\gamma$ /(mN/m) (previously dyne/cm, force) from the (strained) surface energies of solids denoted as  $\sigma$ /(mJ/m<sup>2</sup>) (previously erg/cm<sup>2</sup>, energy). Since their numerical values agree the symbols and units are frequently interchanged in literature. However, since environment influences the surface components it is necessary to state whether the measurements are made in vacuum, vapor (air) or saturated liquid vapor environment.

Assume that a pure liquid (L) is placed in contact with a smooth and homogeneous solid surface (S) maintaining temperature and pressure (and composition) constant. Then, in the absence of other work

functions, we may derive Young equation [1] in the following way (Fig. 1.II):

$$dG_{SL} = \sigma_{SV}dA - \gamma_{LV}(dA \cos\theta_{SL}) - \sigma_{SL}dA \tag{II.1}$$

where the subindex represent: V = vapor (air), L = liquid, S = solid. Note that Gibbs free energy is applied instead of Helmholtz free energy, since sharp interfaces without volume are assumed. At equilibrium:

$$dG_{SL}/dA = 0 = \sigma_{SV} - \gamma_{LV} \cos\theta_{SL} - \sigma_{SL} \tag{II.2}$$

This Young equilibrium is usually presented as vectors along an ideal smooth solid surface (Fig. 1.II).

The contact angle can be determined graphically or geometrically by assuming that the drop is represented by a hemisphere and by deriving it from the expression for the Laplace pressure. Note that pinning of the three-phase contact-line (tpcl) may be represented by the frictional surface tension vector directed perpendicular towards the surface. The change in free surface energy may also be expressed by the Dupr  equation for work of adhesion [1]:

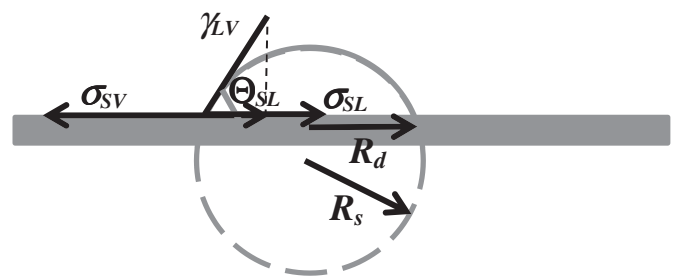
$$dG_{SL}/dA \approx \Delta G_{SL}/A = \Delta G_{SL}^S = -W_A = \sigma_{SL} - \gamma_{LV} - \sigma_{SV} \tag{II.3}$$

As shown work functions (W) are defined for separation of the interconnected phases (work done by the system), while the Gibbs free energy for adhesion is usually defined as uniting the surfaces. Combining the Young and Dupr  equations the work done at the interface may be defined as four key wetting (work) functions (omitting the integral difference symbol,  $\Delta$ ) [1]:

$$\text{Cohesion : } W_C = C_{LL} = 2\gamma_{LV} \text{ or } W_C = C_{SS} = 2\sigma_{SV} \tag{II.4a}$$

$$\text{Adhesion : } W_A = W_{SL} = \sigma_{SV} + \gamma_{LV} - \sigma_{SL} = \gamma_{LV}(\cos\theta_{SL} + 1) \tag{II.4b}$$

$$\text{Spreading : } W_S = S_{SL} = \sigma_{SV} - \gamma_{LV} - \sigma_{SL} = W_{SL} - C_{LL} = \gamma_{LV}(\cos\theta_{SL} - 1) \tag{II.4c}$$



**Fig. 1.II.** Contact angle of a hemispherical sessile drop on an ideally smooth and homogeneous surface is defined by the vectorial stress laid upon the three-phase (Solid–Liquid–Vapor) contact-line (tpcl) as defined by Young Eq. (II.2).  $R_s$ , radius of curvature of the sphere and  $R_d$ , radius of the drop base are not equal. The transversal component ( $-\gamma_{LV}\sin\theta_{SL}$ ) may be considered to represent the frictional pinning of the tpcl.

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