



Experiments on the contact angle of n-propanol on differently prepared silver substrates at various temperatures and implications for the properties of silver nanoparticles

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ABSTRACT

In this paper we present the results of contact angle measurements between n-propanol and silver substrates in the temperature range from $-10\text{ }^{\circ}\text{C}$ to $30\text{ }^{\circ}\text{C}$. The interest in a potential temperature dependence of contact angles originates from recent experiments by S. Schobesberger et al. (Schobesberger S., Strange temperature dependence observed for heterogeneous nucleation of n-propanol vapor on NaCl particles. Master's thesis, University of Vienna, 2008; Schobesberger S. et al., Experiments on the temperature dependence of heterogeneous nucleation on NaCl and Ag particles. In preparation.) investigating the temperature dependence for heterogeneous nucleation of n-propanol vapour on NaCl and on silver particles.

We determined dynamic advancing θ_a and receding θ_r angles on variously prepared silver probes. The Dynamic Wilhelmy method (Wilhelmy L., Über die Abhängigkeit der Capillaritäts-Constanten des Alkohols von Substanz und Gestalt des benetzten festen Körpers. *Ann. Phys. Chem.*, 199:177–217, 1863) was applied using a Krüss K12 Tensiometer, with a refrigerated double-walled glass top. With respect to its potential influence on heterogeneous nucleation mainly the advancing angle is of interest.

The uniform probe geometry required was achieved by accurate cutting and by multiple polishing stages up to the accomplishment of a $0.04\text{ }\mu\text{m}$ grain size. The original probes consist of 925 sterling silver including a 7.5% copper content. Additional coating with silver pro Analyti (p.A.) was applied making use of pure silver powder evaporation process via Physical Vapour Deposition (PVD).

Results show that a surface contamination by copper cannot be neglected for the specification of contact angles. It turned out that additional PVD coatings not only change the values of θ_a but also their temperature dependence. With increasing the number of coatings of a plate the contact angle decreases and its temperature dependence inverts. Since the contact angle hysteresis θ_{hyst} obtained for the variously often coated probes remained practically constant possible changes in surface roughness with increasing number of PVD layers could be excluded.

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1. Introduction

1.1. Relevance of the contact angle to heterogeneous nucleation

Atmospheric aerosols play an important role in connection with cloud formation, global radiative balance and the development of the global climate. The formation of aerosol particles is influenced by heterogeneous nucleation processes. According to Fletcher theory (Fletcher, 1958) heterogeneous

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nucleation on pre-existing seed particles is strongly dependent on the contact angle between seed surface and condensing liquid. Description of heterogeneous nucleation thus requires quantitative information on contact angles and their temperature dependence.

Recently Schobesberger et al. (Schobesberger, 2008; Schobesberger et al., in preparation) measured the temperature dependence for heterogeneous nucleation of n-propanol vapour on sodium chloride- and on silver- particles. A strange temperature dependence for the nucleation process of n-propanol vapour on NaCl was found.

This unexpected behaviour may be explained by a temperature dependent contact angle. Consequently we studied the contact angle as a function of temperature for both the NaCl/n-propanol and the Ag/n-propanol system. In the present paper we present experimental investigations of the contact angle for n-propanol on silver. Results obtained for the sodium chloride/n-propanol system are described in detailed elsewhere (Pinterich, 2009; Schobesberger et al., in preparation).

1.2. Temperature dependence of contact angle

So far only a few experimental studies have been reported investigating the temperature dependence of the contact angle for various liquids on metallic surfaces (Bernardin et al., 1997; Diesselhorst et al., 1997; Ponter and Boyes, 1973) while a number of investigations have been performed with non-metallic especially polymeric surfaces e.g. (Budziak et al., 1991; Petke and Ray, 1969). Typically the contact angle is reported to decrease with increasing temperature (Carey, 1992; Schonhorn, 1966) although an opposite temperature dependence e.g. for water on silicized glass (Neumann, 1974) or a water oil suspension on quartz (Dandina, 2002) was found as well. Numerical values for the temperature derivative of θ in a temperature range of 5 °C–100 °C were reported as $d\theta/dT \approx -0.01^\circ/\text{K}$ by Neumann et al. (Neumann et al., 1971) and Adamson (Adamson, 1973). Petke and Ray (1969) measured a decrease varying from $d\theta/dT \approx -0.01$ to $-0.18^\circ/\text{K}$. In addition to experimental determinations Adamson (Adamson, 1973) presented a theoretical model relating the molecular surface adsorption of a solid to the liquid–solid contact angle. In the course of his study he arrived at the following equation:

$$\cos\theta = 1 + C(T_\infty - T)^{a/(b-a)} \quad (1)$$

where T_∞ is a “pseudo-critical” temperature, or the temperature at which the contact angle approaches zero, C is a constant of integration and a and b are temperature-independent constants resulting from a balance of intermolecular forces. In contrast to the model predictions a few systems have been observed where the contact angle increased with temperature. As Adamson suggested, this may result from molecular interactions not accounted for in Eq. (1). But also nanostructural changes of the involved solid surface at high temperatures ranging from 100 °C to 400 °C cause an increase of contact angle with increasing temperature as reported in (Schwartz and Garoff, 1985).

With respect to the nucleation experiments performed by Schobesberger et al. (Schobesberger, 2008; Schobesberger et al., in preparation) the temperature dependence of the

contact angle for n-propanol on silver and on sodium chloride substrates is of interest. Only a few investigations (Hill, 2007; Ortner, 2000) have been performed so far considering those components, however, these studies did not focus on the temperature dependence. For the contact angle of n-propanol on silver Ortner (Ortner; 2000) reported a constant value of 28.3° at $T = 20$ °C and 0 °C whereas Hill (Hill, 2007) stated an advancing angle of $32.6 \pm 0.6^\circ$ at $T = 20$ °C.

In this paper we will focus on the silver/n-propanol system. The sodium chloride/n-propanol measurements and results are presented in detail elsewhere (Pinterich, 2009; Schobesberger et al., in preparation).

1.3. Contact angle hysteresis θ_{hyst}

According to the measurement method applied in the presented study not the equilibrium contact angle θ_e defined by Young’s equation (Young, 1805)

$$\sigma_{LV} \cos\theta_e = \sigma_{SV} - \gamma_{SL} \quad (2)$$

is measured but rather advancing θ_a and receding θ_r angles. The above stated equilibrium contact angle (Eq. (2)) results from a force balance along the three phase contact line (either liquid–vapour–solid or liquid–liquid–solid). Usually this equilibrium contact angle cannot be measured in real systems (Chibowski, 2007). If e.g. a solid plate is immersed into some liquid or a liquid droplet is settled on a solid surface the measured advancing contact angle θ_a is larger than the angle θ_r measured for the same system when withdrawing the probe or reducing the drop volume. This difference between θ_a and θ_r , the so called contact angle hysteresis $\theta_a - \theta_r$, results from imperfection of real surfaces. A relation between the idealized equilibrium contact angle, advancing and receding angle was developed e. g. by Wenzel (Wenzel, 1936) and Cassie (Cassie, 1948), but experimental determination of the equilibrium contact angle is still an open problem even when static measurement method such as the sessile drop method are applied. Contact angle hysteresis has been attributed to a large number of various effects. One may divide them into “solid-induced” factors such as surface roughness (Bartell and Shepard, 1953; Eick et al., 1975; Johnson and Dettre, 1964b; Oliver et al., 1980; Oliver et al., 1977), heterogeneity (Decker and Garoff, 1996, 1997; Dettre and Johnson, 1965; Good, 1952; Johnson and Dettre, 1964a), Marmur, 1994; Neumann and Good, 1972; Schwartz and Garoff, 1985), metastable states (Brandon and Marmur, 1996; Chen and McCarthy, 1997; Derjaguin, 1946; Fadeev and McCarthy, 1999; Johnson and Dettre, 1969; Sedev et al., 1993, 1996; Youngblood and McCarthy, 1999) including patch structures and “liquid-induced” factors such as chain length of the liquid under study (Lam and Ko, 2001). However for statements about liquid/solid interactions it is the advancing angle which is commonly used since in contrary to the dewetting process usually no liquid film is present in the movement direction of the liquid/solid interface (Lam et al., 2001) and thus the contact angle is a result of the apparent liquid solid interaction only.

Depending on the measurement method applied and its accuracy several contact angles can be distinguished (Bernardin et al., 1997). E.g. performing dynamic contact angle measurements either the static or the dynamic advancing/receding

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