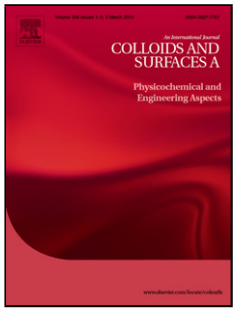

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Capillary rise dynamics of liquid hydrocarbons in mesoporous silica as explored by gravimetry, optical and neutron imaging: Nano-rheology and determination of pore size distributions from the shape of imbibition fronts

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Abstract

We present combined gravimetrical, optical, and neutron imaging measurements of the capillarity-driven infiltration of mesoporous silica glass (Vycor) by hydrocarbons. Square-root-of-time Lucas-Washburn invasion kinetics are found for linear alkanes from n-decane (C10) to n-hexacontane (C60) and for squalane, a branched alkane, in porous monoliths with 6.5 nm or 10 nm mean pore diameter, respectively. Humidity-dependent experiments allow us to study the influence on the imbibition kinetics of water layers adsorbed on the pore walls. Except for the longest molecule studied, C60, the invasion kinetics can be described by bulk fluidity and bulk capillarity, provided we assume a sticking, pore-wall adsorbed boundary layer, i.e. a monolayer of water covered by a monolayer of flat-laying hydrocarbons. For C60, however, an enhanced imbibition speed compared to the value expected in the bulk is found. This suggests the onset of velocity slippage at the silica walls or a reduced shear viscosity due to the transition towards a behaviour typical of polymer-like flow in confined geometries. Both, light scattering and neutron imaging, indicate a pronounced roughening of the imbibition fronts. Their overall shape and increase in width can be resolved by neutron imaging. The fronts can be described by a superposition of independent wetting fronts moving with pore size-dependent square-root-of-time laws and weighted according to the pore size distributions obtained from nitrogen gas sorption isotherms. This finding indicates that the shape of the imbibition front in a porous medium, such as Vycor glass, with interconnected, elongated pores, is solely determined by independent movements of liquid menisci. These are dictated by the Young-Laplace pressure and hydraulic permeability variations and thus the pore size variation at the invasion front. Our results suggest that pore size distributions can be derived from the broadening characteristics of imbibition fronts.

Keywords: imbibition, alkanes, radiography, nanopore, wetting, porosimetry, porous media