DIFFUSIOPHORESIS OF GOLD IN SILICA DRIVEN BY THE NANOMETRIC INTERFACIAL LAYER

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ABSTRACT

This paper describes a model to describe diffusiophoresis of a gold nanoparticle inside a solid silica substrate. The motion of the particle is powered by the flow of a liquid-silica interfacial layer due to a gradient of gold concentration inside this layer. This mechanism can help explain the experiments of de Vreede *et al.* [1] in which the authors observed that gold nanoparticles become engulfed into the solid silica substrate at high temperature. Indeed our model describes precisely their experimental measurement.

KEYWORDS: Diffusiophoresis, Gold Nanoparticles, Surface Diffusion, Amorphous Silica

INTRODUCTION

In the experiments performed by de Vreede *et al.* [1], a gold nanoparticle is placed on top of a fused-silica substrate heated below the melting temperature of gold and silica. After a few hours the gold particle has been engulfed into the substrate. We suggest here a diffusiophoretic model for silica transport along the particle surface toward the pore in contact with air.

THEORY

We model the observed fast surface diffusion at the interface of silica [1-4] by assuming a liquid nanolayer at the silica interface. According to experiments from literature [5], we assume in the present model that gold diffuses in the silica, in the interfacial nanometric layer only. Gold evaporation and surface diffusion decreases the concentration of gold at the pore opening and creates a gradient in the nanometric layer (Figure 1).
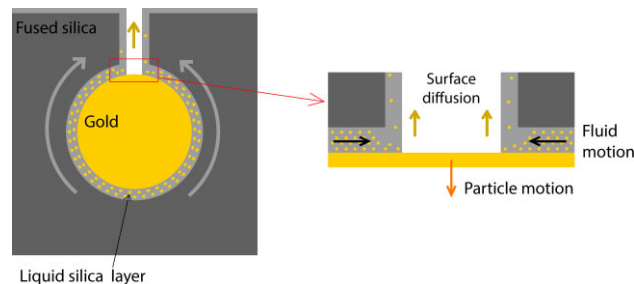

Figure 1: Sketch of the mechanism. Gold (yellow) diffuses in the nanometric interfacial liquid silica layer (light gray), and to the top. The gradient of concentration generates a motion of the liquid silica that pushes the particle away from the interface.

A concentration gradient induces a gradient of free energy and therefore a body force in the silica layer, as detailed by Vladimirova *et al.* [6]. This body force pulls the silica toward the free surface and the resultant of the pressure forces pushes the particle away from the surface.

RESULTS AND DISCUSSION

We calculated that diffusion of gold in silica occurs significantly faster than the motion of the particle. We therefore performed simulations of the diffusion profile for a static particle. These simulations show that the gradient of concentration extends on a length scale comparable to the depth of the interfacial layer (Figure 2a).
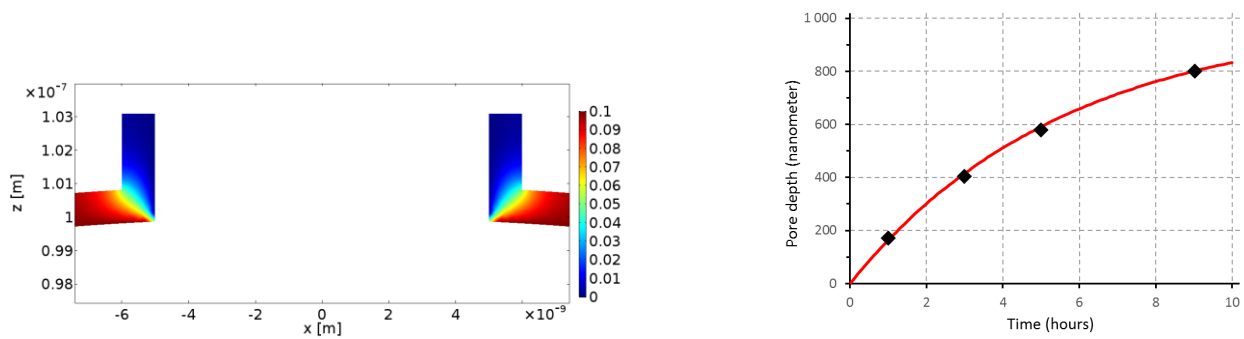
Figure 2: (a) Gold-fraction profile in the liquid layer according to our simulations. The picture show the gold molar fraction in the upper part of the layer. We impose a known gold fraction of 0.1 at the interface of the particle (calculated according to reference [7]), and a very low concentration in the pore. The gradient extends on a very short distance of the order of the layer thickness. (b) Pore depth along time. Comparison of experimental data (black square, from reference [1]: gold in silica) with the result of the model (red line, for a viscosity in the interfacial layer η= 0.7 MPa.s and a layer thickness h = 1 nm). The speed decreases due to a decrease of the pore diameter which is measured experimentally.

Therefore, according to the previously described model, we calculated the speed of a cylindrical gold particle sinking in silica, and we can describe the experiments with reasonable value of the parameters, as presented in Figure 2b.

CONCLUSION

We showed that the engulfing of gold nanoparticles in silica can be explained by a diffusiophoretic effect due to surface diffusion of gold at the surface of silica. We need now to confront our theory with further measurements to find evidence of this phenomenon. This mechanism should allow a probing of the properties of the silica interfacial nanolayer. Moreover, the understanding of this phenomenon paves the way of new routes for silica nano-patterning.

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