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Mixing and Flow Control of Liquids in Nanochannel Liquid Phase Electron Microscopy

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We are developing a micro/nanofluidic device to study liquid phase processes *in situ* TEM. The new nanochannel liquid cell chip design alleviates the persistent issue of membrane bulging in clamped chip systems and offers the possibility of achieving high level flow control and mixing in the imaging region. The earlier version of the device were used to quantitively measure the mean inner potential (MIP) of liquid water [1] and the electron inelastic mean free path (IMFP) in liquid water [2].

The chips are made from two separately treated wafers that are bonded together and typically embody an array of nanochannels, each typically about 2 µm wide and 50-500 nm deep, see figure 1(a,b) [3]. Because the liquid containing channels of the chips have width down to 2 µm, the resulting bulging from the internal ambient pressure difference to the TEM vacuum is less than 10 nm, and the liquid layer thickness is well defined by the microfabrication process [2]. The nanochannel fluidic system has four inlets/outlets and thus offers the possibility of running experiments with numerous different gasses and liquids to observe reaction between them. The channel system can be lithographically designed to bring together flows of reagents in one place, or to affect how particles and molecules may be trapped, filtered and otherwise manipulated inside the channel system.

Flow mixing can be controlled in a nanochannel T-junction, aiming to create a steady state concentration gradient in the T-channel within the TEM field of view. A constant concentration gradient offers added experimental control in LPTEM not achievable in commercial LPTEM systems. The mixing of two laminar flowing liquids in a T-channel intersection is done solely via diffusion in the laminar flow with Reynolds' number $N = \frac{\rho v d}{\mu}$ <1. The Peclét number [4] indicates how far down a mixing channel a homogeneous solution is obtained: The diffusion coefficient of *e.g.* fluorescein is D=425µm^{2}s^{-1}. The time it takes for a liquid to diffuse across the channel with width w is approximately t ~ w^2/D [4], which with channel width 6 µm gives ~ 10 ms for the fluorescein to diffuse across the mixing channel. The corresponding diffusion velocity is 600 µms^{-1} and it indicates that if the flow velocity is v=600µms^{-1} then the fluid will have travelled just one channel width, L~6 µm, down the center channel when the fluid is completely mixed, corresponding to a Peclét number P=L/w=wv/D ≈1. For liquid with higher viscosities, or analytes with lower diffusion coefficients P will increase making a gradient clearly visible in the mixing channel at lower flow rates, while for aqueous solutions gradients are created by back diffusion upstream in the inlet channels.

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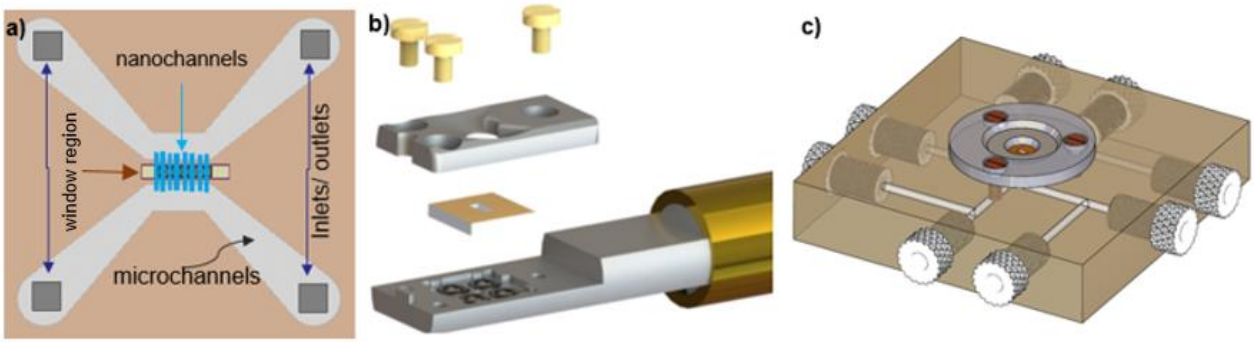

Figure 1. a) The nanochannel fluid cell chip contains four inlets/outlets, micro and nanochannels and one electron transparent window region in the middle b) TEM Nanochannel liquid cell holder, c) SEM liquid cell holder.

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