## Liquid Pockets Encapsulated in MoS<sub>2</sub> Liquid Cells

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Liquid cell transmission electron microscopy has attracted a lot of attention recently, since it has enabled direct observation of many dynamic processes in real-time with high spatial resolution, such as crystallization in liquids [1], self-assembly of nanoparticles [2], electrochemical processes in liquid electrolytes [3] and so on. Significant advances in the development of liquid cells have been achieved and various thin membranes have been used for the fabrication of liquid cells. For example, besides silicon nitride [4], graphene [5] and  $MoS_2$  [6] have been incorporated as the liquid cell windows, which offer the advantages of atomic layer thickness, high mechanical strength and flexibility, or being the functional substrates as well as the supporting membrane. The behavior of liquids including how liquids being encapsulated inside a liquid cell can impact the outcomes of experiments significantly. Although there have been many reports on liquids inside silicon nitride liquid cells [7] and graphene liquid cells [8], there are limited studies on the formation of liquid pockets in a  $MoS_2$  liquid cell.

In this work, we report the encapsulation of liquid pockets in a MoS<sub>2</sub> liquid cell. A MoS<sub>2</sub> liquid cell was fabricated using MoS<sub>2</sub> of three atomic layer thickness as one side of membrane and about three layers of graphene as the other side of membrane. The two membranes were sandwiched together with liquids in the middle. We used a liquid mixture of 1,2-dichlorobenzene (99%, Sigma-Aldrich, US) and oleylamine (technical grade, Sigma-Aldrich, US) with the volume ratio of 10:1 (1,2-dichlorobenzene: olevlamine) for the study. For systematic studies, the liquid encapsulation in a MoS<sub>2</sub> liquid cell was compared with the same liquid solution encapsulated with a graphene liquid cell and a SiNx liquid cell. As shown in Figure 1, the uniform liquid thin film was achieved in the SiNx liquid cell. Bubbles were observed from electrolysis of the liquid under electron beam irradiation. We found the liquid film thickness may vary significantly even using the same thickness of spacer, as indicated by the variations in electron transparency of the liquid cell. It suggests that effort is needed to control the gap thickness between two SiNx membranes during liquid cell fabrication. For liquids insides a graphene cell, many round liquid pockets or patches of liquid thin films were found. Some liquid pockets were too large and thick to be electron transparent. It implies that the flexibility of the graphene offers many local regions ideal for imaging and at the same time the exact thickness of the liquids can be hard to identify. For liquids encapsulated inside a MoS<sub>2</sub> liquid cell, it is interesting that many elongated liquid packets were found.

In order to understand the formation of unique liquid pockets, we further did high resolution imaging of a liquid pocket encapsulated inside a  $MoS_2$  liquid cell. As shown in Figure 2, the  $MoS_2$  membranes around the liquid pocket show different crystal orientations. It suggests that the liquid droplet prefers to settle at the grain boundaries. This is likely driven by the minimization of system energies. Other factors, such as bending at the grain boundaries of a  $MoS_2$  thin film, may also contribute to the liquid

pocket formation around the grain boundaries. Further study of liquid dynamics under various applied forces through thermal heating or electric biasing may yield valuable fundamental information on the nanoscale liquid behavior and the liquid interactions with MoS<sub>2</sub> substrate. An understanding and further controlling of the liquid encapsulation inside a liquid cell open many opportunities for further studies [9].

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Figure 1. The shape of liquid pockets encapsulated in various TEM liquid cells. (a-d) TEM image showing the liquid pockets in  $SiN_x$  (a), graphene (b,c), and  $MoS_2$  (d) liquid cells.



**Figure 2.**  $MoS_2$  lattice orientation around a liquid pocket. (a) False-colored TEM image of a liquid pocket in a  $MoS_2$  liquid cell. The regions with different  $MoS_2$  grains are highlighted by blue (region 1), green (region 2), and red (region 3). (b-d) High-resolution masked inverse FFT images for each region. The corresponding areas are marked by white boxes in each region.