

Supporting Information

Reactive Etching of Gallium Oxide on Eutectic Gallium Indium (eGaIn) With Chlorosilane Vapor to Induce Heterogeneous Wetting

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Thin film thickness measurement with AFM:

We used Atomic Force Microscopy model MFP3D bio infinity from Asylum Research- Oxford Instruments to measure the thickness of the film near the glass-LM interface. We created a 40 mm x 5 mm rectangular shape on the glass slide. We used a low-force cantilever beam to map the height difference along the interface.

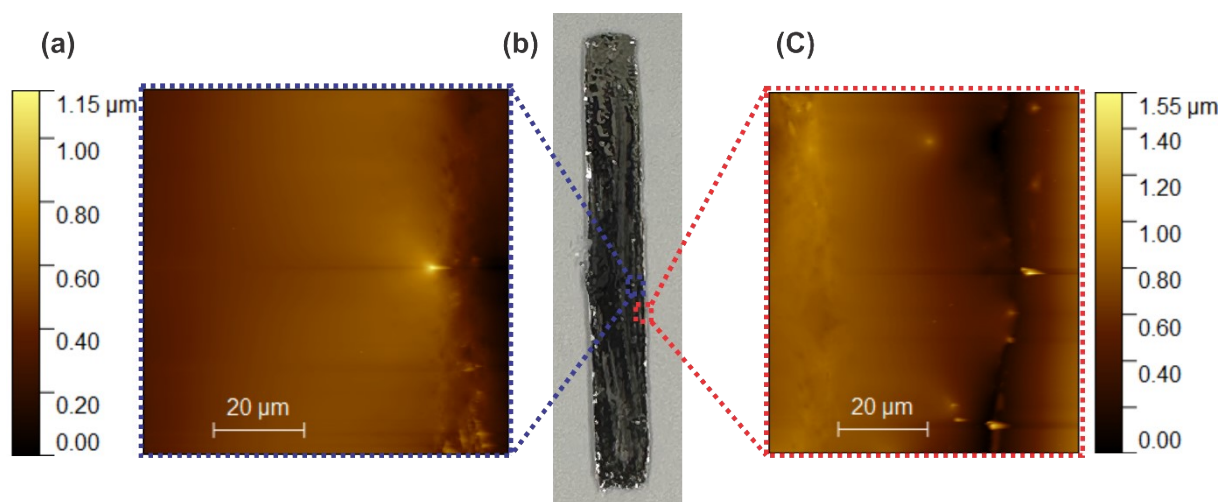


Figure S-1: AFM measurements to measure the thickness of LM film. (a) and (c) is the height variation along the XY plane on the representative area shown in (b). Height map (a) shows the height variation of the LM pattern starting from the very edge. Height map (c) captures the glass surface to the LM thin film edge.

We took measurements in two areas and marked them with blue and red dotted rectangles on the representative position on the LM pattern in (b). The red dotted rectangle captures and represents the glass-LM interface and clearly shows a variation of around $\sim 1.5 \mu\text{m}$ in the height map (c). The height variation is around $\sim 1 \mu\text{m}$ in the height map shown in (a), indicating an increase in height across the width of the film.

HCl vapor treatment:

We tested HCl vapor to delaminate the eGaln film; however, we did not notice a heterogeneous wettability similar to the chlorosilane vapor-treated surface. HCl vapor treatment also created a delaminated pattern with a distribution of metallic droplets. Both measured water contact angles on the pre-patterned and patterned area were hydrophilic.

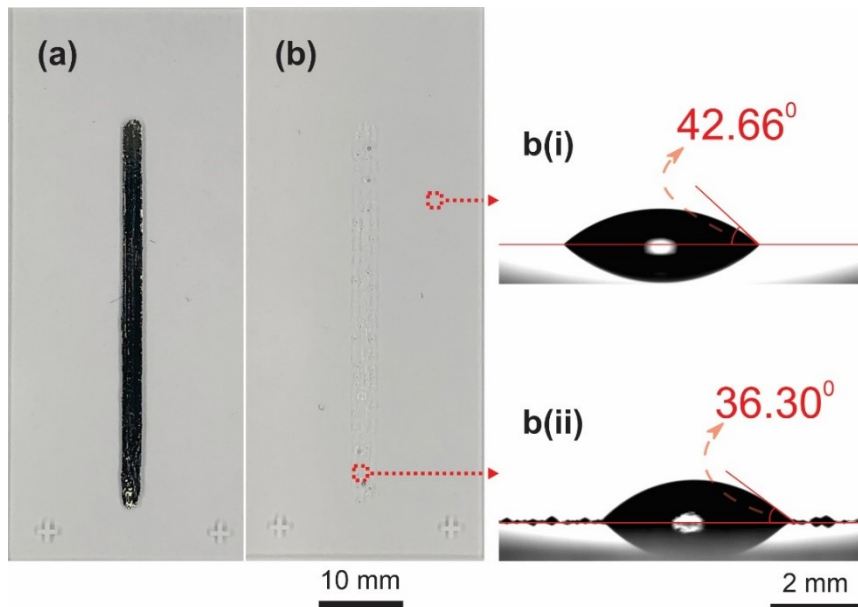


Figure S-2: Utility of HCl towards non-heterogeneous wetting demonstration. (a) eGaln pattern on a glass slide (top view), (b) HCl treated eGaln pattern on a glass slide (top view); the withdrawal seemed similar to chlorosilane treatment without heterogeneous wettability. Red dotted rectangles represent the approximate contact angle measurement area; b(i) water contact angle on the non-pattern side of the glass slide, b(ii) contact angle on the previously LM pattern side of the glass slide

EDS analysis to study the presence of chlorine:

We study the presence of elements on the sample using EDS ((model-JSM-7100FT FESEM). The samples after immediate FOTS treatment were compared with the FOTS-treated samples after rinsing. We observed a significant amount of chlorine on the LM droplet apart from their oxide layer change. The relevant EDS samples with their spectrum peaks are shown in Figure S-2.

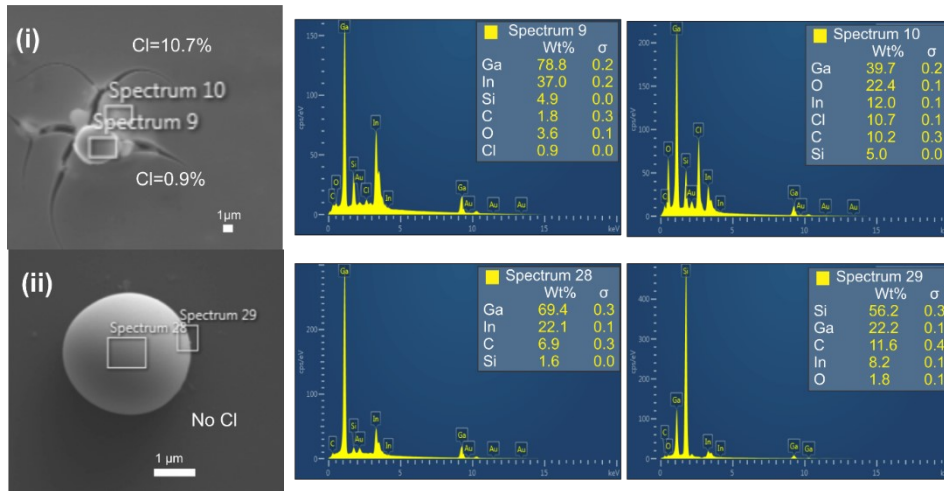


Figure S-3: Energy Dispersive Spectroscopic Analyses. (i) EDS analysis of eGaln droplet after FOTS treatment; the two spectra compare the available chlorine for etching. Spectrum 9 was targeted for the bulk amount of the metal, and spectrum 10 was targeted on the base of the metal droplet; After(ii) rinsing, DI water sample did not show any presence of chlorine. Spectrum 28 was targeted for the bulk amount of the metal, and spectrum 29 was targeted on the base of the LM droplet. Both droplets show completely different compositional variations. The scale bar is at the bottom of (i) and (ii).

We analyzed EDS in multiple areas to understand the surface and elemental composition of the sample. The presence of chlorine was significantly high near the oxide layer on the base of the metal droplet. We can infer that the chlorine amount was high because of the oxide presence. The presence of Oxide and Chlorine was significantly low in the center of the bulk metal droplet. On the other hand, there was no trace of chlorine on the rinsed sample. The DI water used during the rinsing process may have washed off the chlorine. This observation implies that chlorine may be responsible for the etching of gallium oxide.

Supporting video: SV-1 Demonstration of the FOTS etching process of LM pattern:

Here, we showed the LM withdrawal process by FOTS treatment. We used the video camera of an iPhone XS max to record the video. We set up the camera inside the desiccator and wrapped the camera with a transparent sheet to avoid silanization on the camera. Then, we started the vacuum pump. The video was fast-forwarded at 10X speed. We can observe slow etching randomly starting from various pinhole/damage of the LM pattern. An earlier study has also reported etching starting from similar pinholes.¹ Suddenly, the etching process gained speed, and the withdrawing process achieved completion with various LM drops randomly distributed on the glass slide.

Supporting video: SV-2 Microfluidic pattern under stereoscope:

The microfluidic demonstration of our fabricated device using the technique described in this work was studied under an Olympus SZ61 stereoscope and video recorded using a MU1000-HS camera from AmScope. AmScope software was used to record the video. The video shows that the blue dye is coming from the right inlet, and DI water is entering from the left channel. Then, both fluids flowed through T-shape microfluidic device and mixed throughout the length. The length and width of the whole T-shape were 40 mm and 20 mm, and the depth of the fluid flowing zone was 2 mm. If the water flow is too high or the length of the T-shape is too long, water can accumulate near the T-joint and show overflowing.

References

- 1 M. R. Khan, J. Bell and M. D. Dickey, *Advanced Materials Interfaces*, 2016, 3, 1600546.