Supplementary Information

Investigating Physico-chemo-mechanical Changes of Eutectic Gallium Indium (eGaIn) Thin Films Induced by Fluorinated Aqueous Droplets

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Structure of the Cotton Swab used for Film Fabrication:

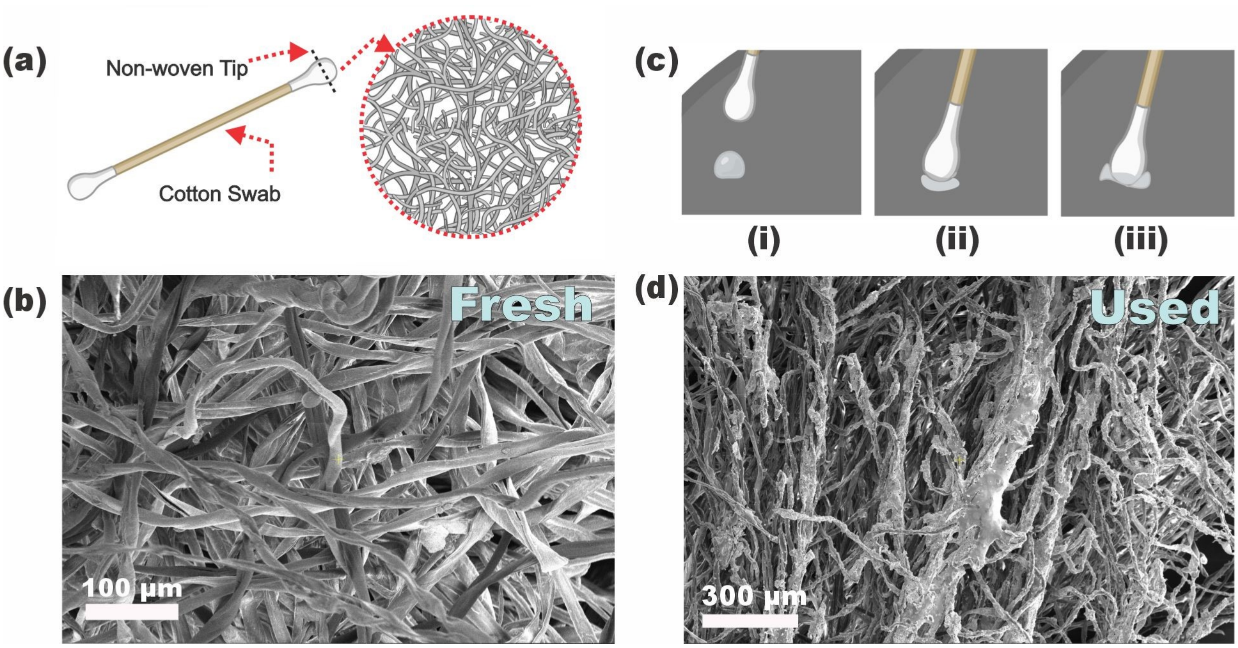


Fig S1: **Structure of the cotton swab used for film fabrication.** (a) Cartoon depiction of the woven cotton swab tip used for film preparation, (b) SEM topography of the woven structure of fresh cotton swab, (c) Three-step cartoon depiction of the scenario of pushing cotton swab right before patterning, (d) SEM topography of the woven structure of the cotton swab after patterning.

Figure S1(a) is a cartoon depiction of the cotton swab. Figure S1(b-d) are SEM images of the cotton swab surfaces (topography) before and after thin film fabrication. Figure S1(c) shows steps to dab on the droplet of liquid metal droplet before straining the thin film.

Film Thickness at different line scans using Atomic Force microscope (AFM):

Figure S2 depict thickness measurements of a thin film using atomic force microscopy (AFM).

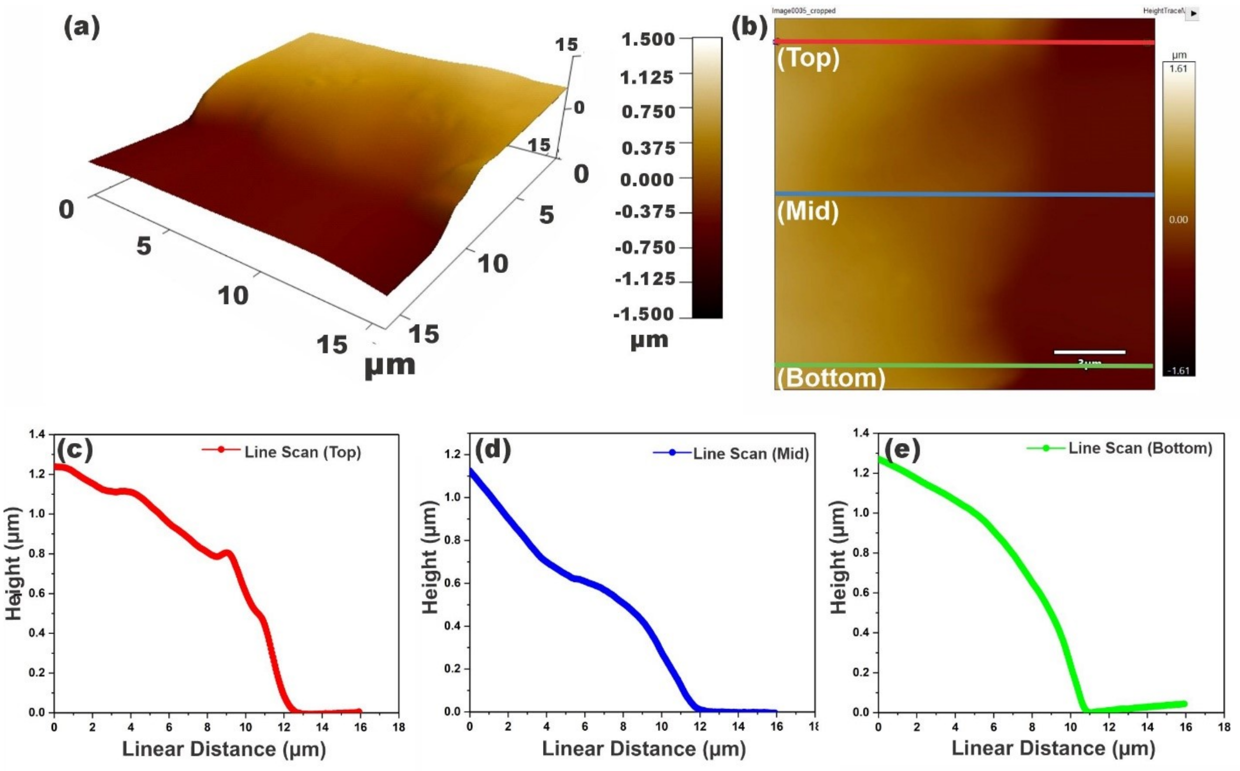


Fig S2: **Thickness measurement of eGaIn thin film using AFM.** (a) AFM topography of eGaIn thin film sample denoting a thickness of ~ 1.2 µm, (b) Line scan at different spots (top, mid. bottom) of the thin film shown in (a) denoting a thickness in between 1.1-1.3 µm, (c-e) Graphical representation of eGaIn thin film sample denoting a thickness in between 1.1-1.3 µm.

Reproducibility Test for Elemental Analysis on eGaIn Film

Figure S3 and S4 are two different EDS measurements done on the thin film. Elemental distributions are given on the insets.

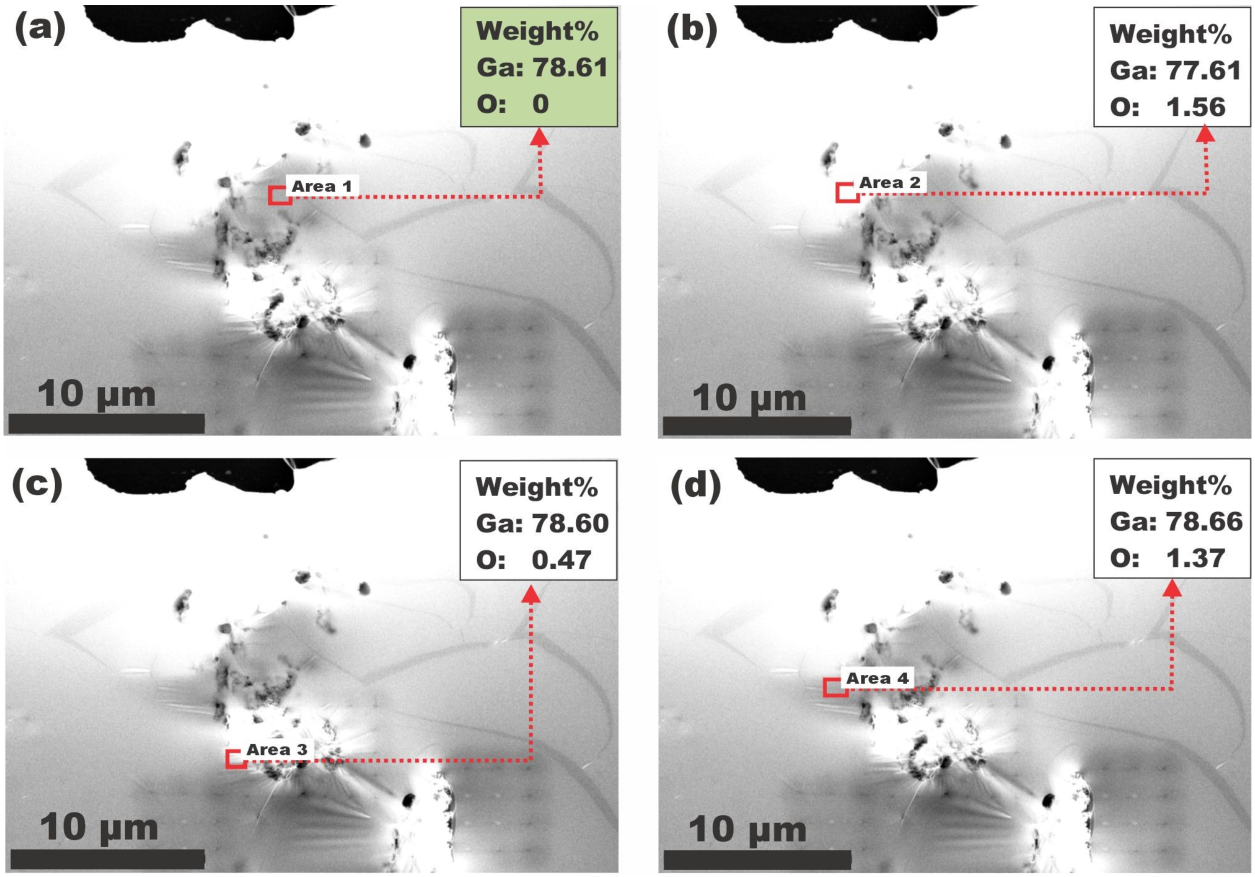


Fig S3: Elemental Analysis on eGaIn film. (a) Topography and EDS data of eGaIn thin film sample identifying the targeted area where the amount of oxygen is zero, (b) Topography and EDS data of eGaIn thin film sample identifying targeted area surrounding area shown in (a) where the amount of oxygen is non zero, (c) Topography and EDS data of eGaIn thin film sample identifying targeted area surrounding area shown in (a) where the amount of oxygen is non zero, (d) Topography and EDS data of eGaIn thin film sample identifying targeted area surrounding area shown in (a) where the amount of oxygen is non zero, (e) Topography and EDS data of eGaIn thin film sample identifying targeted area surrounding area shown in (a) where the amount of oxygen is non zero.

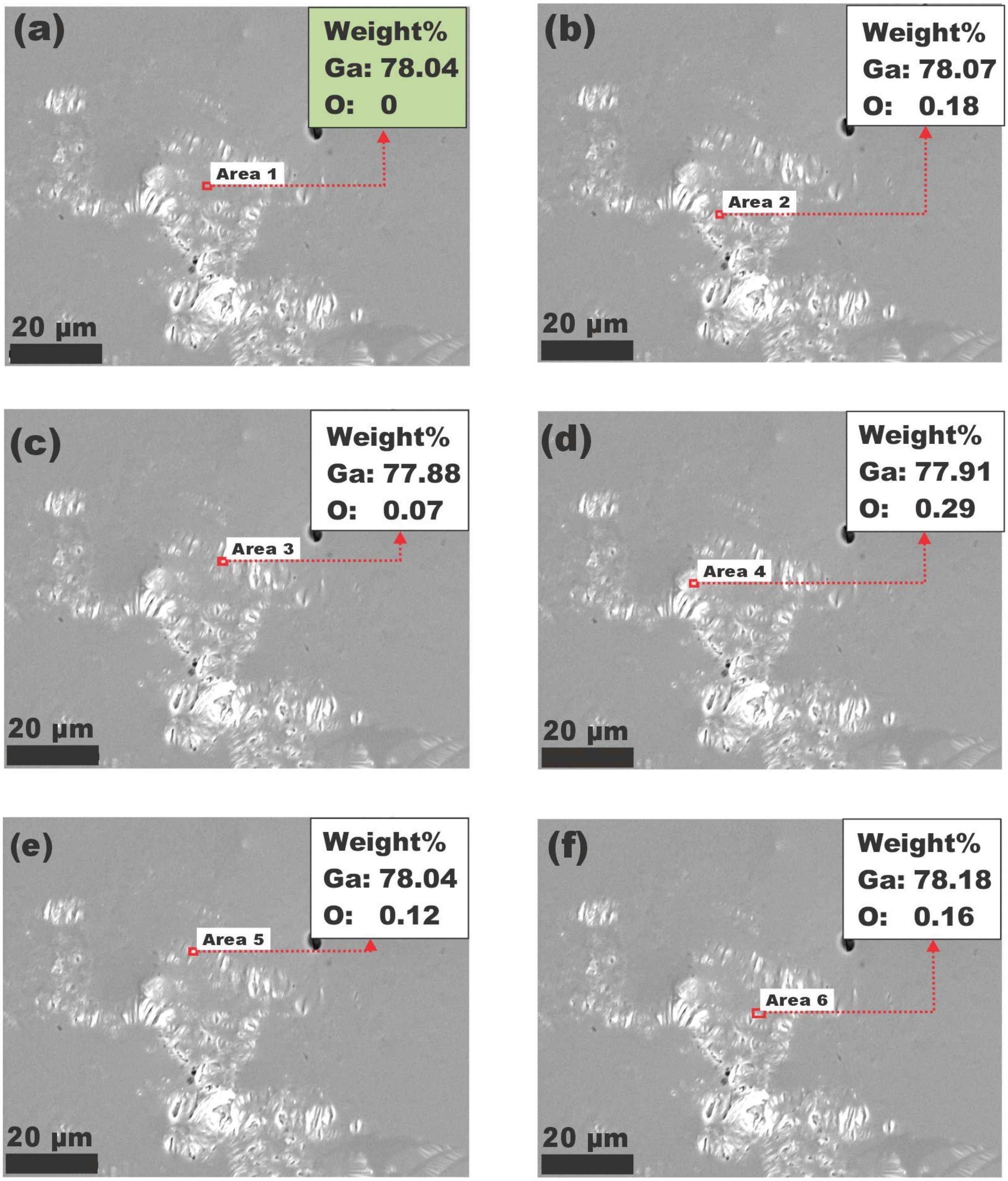


Fig S4: Elemental Analysis on eGaIn film. . (a) Topography and EDS data of eGaIn thin film sample identifying targeted area where the amount of oxygen is zero, (b) Topography and EDS data of eGaIn thin film sample identifying targeted area surrounding area shown in (a) where the amount of oxygen is non zero, (c) Topography and EDS data of eGaIn thin film sample identifying targeted area surrounding area shown in (a) where the amount of oxygen is non zero, (d) Topography and EDS data of eGaIn thin film sample identifying targeted area surrounding area shown in (a) where the amount of oxygen is non zero, (e) Topography and EDS data of eGaIn thin film sample identifying targeted area surrounding area shown in (a) where the amount of oxygen is non zero (f) Topography and EDS data of eGaIn thin film sample identifying targeted area surrounding area shown in (a) where the amount of oxygen is non zero.

Control RAMAN studies:

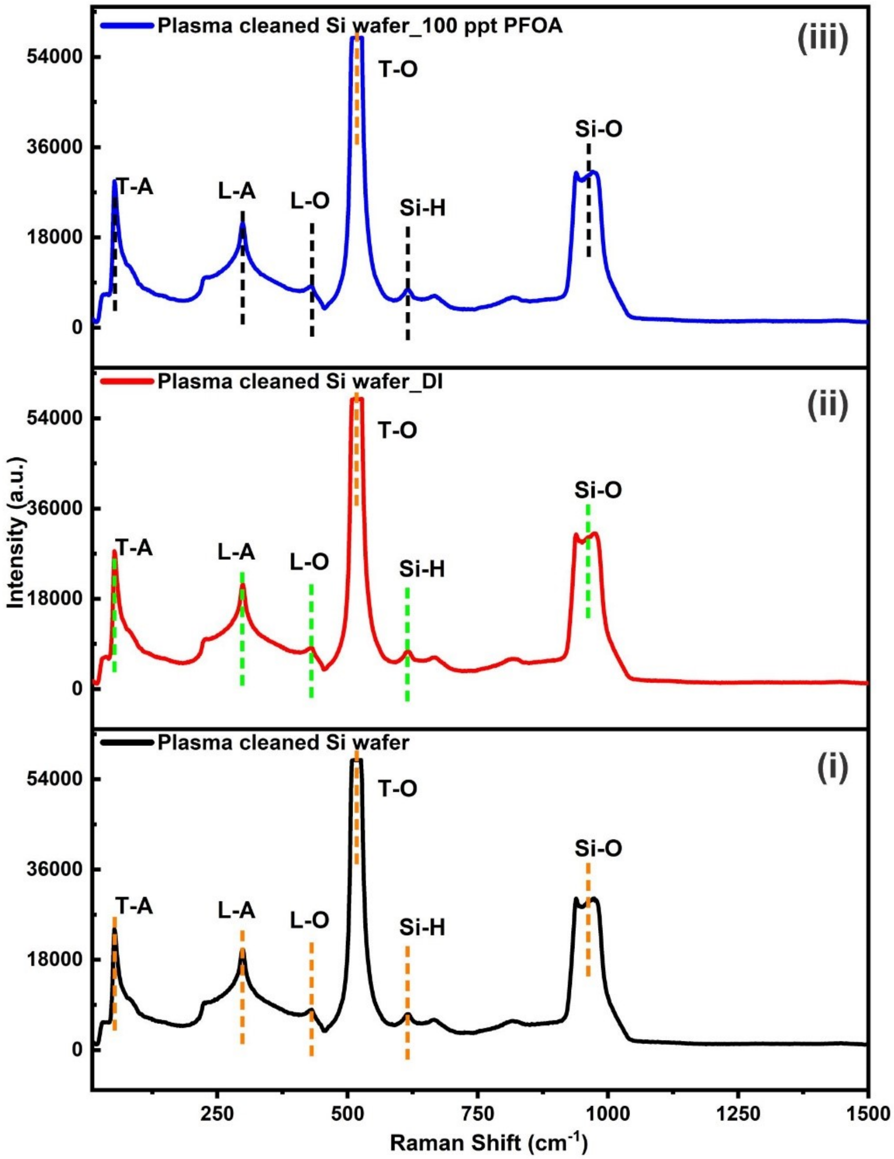


Fig S5: Control Raman Studies. Three different Raman spectra were collected on (i) a plasma-cleaned Si wafer, (ii) DI water coated with a plasma-cleaned Si wafer, and (iii) 100 ppt PFOA on a plasma-cleaned Si wafer.

The Raman spectrum of the bare Si wafer exhibits peaks of transverse acoustic (TA) mode at ~52 cm^{-1}, the longitudinal acoustic (LA) mode at ~297 cm^{-1}, the longitudinal optical (LO) mode at ~429 cm^{-1}, and the transverse optical (TO) mode at ~513 cm^{-1}. Additionally, Si-H wagging/rocking vibration modes in different bond topologies produced a little peak at ~612 cm^{-1}. The Si-O bond vibrations are typically attributed to another peak at ~925 cm^{-1}. This result is consistent with a previous study.[1] There are no more peaks observed after adding DI water and 100 ppt PFOA on the Si wafer surface. Therefore, a pristine Si wafer does not contribute to the shift in Raman spectra in the same way as a liquid metal thin film (i.e., a thin film of liquid metal is more sensitive).

More details on Raman Spectra for Figure 3

When we added a 3 μL water droplet containing 0.014 ppb PFAS, we identified a peak corresponding to the δ(CF_3) vibration at around 763cm^{-1} in **Figure 3b(iii)**.[42] We also found two nearby peaks at 706 and 804 cm^{-1}, which were present in the Raman peak of liquid metal. We found in literature that the peak of Ga-O-H is between 750 and 800 cm^{-1}. [41] Therefore, we hypothesize that the PFAS molecule was adsorbed on the surface of Ga_2O_3 so that we could detect the peaks for Ga_2O_3, gallium hydroxide, and the PFAS. Similar to the 0.001ppb PFAS sample, we observed a peak around 819 cm^{-1}, corresponding to the stretching vibration of the trifluoromethyl (CF_3) group ν(CF_3). Furthermore, peaks corresponding to the stretching vibrations of the carbon-fluorine bond ν(CF) at ~1370 cm^{-1} and the carbon-carbon bond (ν(CC)) in the range of ~1406 cm^{-1} were also identified.

When we added a 3 μL droplet of water contaminated with 0.001 ppb PFAS, we identified a peak corresponding to the δ(CF_3) vibration at around 763 cm^{-1} in **Figure 3b(iv)**. [42] We also found two nearby peaks at 706 and 804 cm^{-1}, which indicate the presence of Ga_2O_3 and Ga-O-H. [41] In this case, the PFAS molecule was also adsorbed on the surface of Ga_2O_3. We observed a peak around 819- cm^{-1}, corresponding to the stretching vibration of the -CF_3 group ν(CF_3). Furthermore, peaks corresponding to the stretching vibrations of the carbon-fluorine bond ν(CF) at 1370 cm^{-1} and the carbon-carbon bond (ν(CC)) in the range of 1406 cm^{-1} were also identified.

When we added a 3 μL water droplet with 200 ppm PFOA and analyzed the raman spectrum in **Figure 3b(v)**. The Raman spectrum shows a peak at 600, 706, 723, 763, 804, 819, 1210, 1300, 1370 and 1406 cm^{-1}. Literature suggests that peaks corresponding to the δ(CF_3) vibration fall within the 600-819 cm^{-1} range [42] due to the stretching and bending motions of the trifluoromethyl (-CF_3) group. A broad peak was observed around 1300 cm^{-1}, corresponding to the stretching vibration of the -CF_3 group, ν(CF_3). Furthermore, peaks corresponding to the stretching vibrations of the carbon-fluorine bond ν(CF) at 1370 cm^{-1} and the carbon-carbon bond (ν(CC)) in the range of 1406 cm^{-1} were also identified.

Reproducibility test of liquid metal (eGaIn) thin film after exposure to different concentrations of PFAS using SEM:

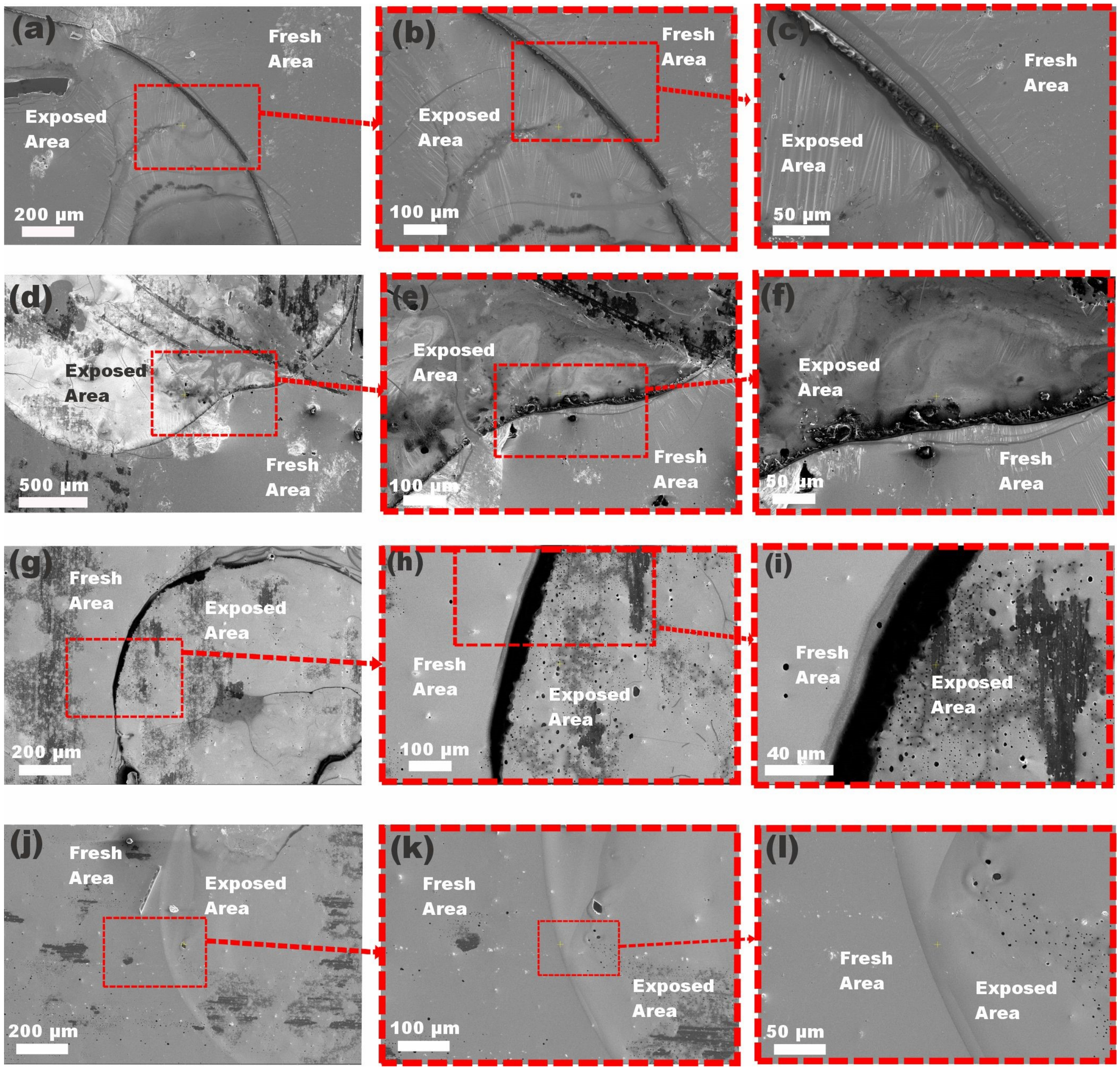


Fig S6: SEM Topography of liquid metal (eGaIn) thin film after exposure to different PFAS concentrations. (a) Topography of a eGain Thin film exposed to 0.001 ppb PFAS under scanning electron microscope (SEM) at 150X magnification, (b) Topography of a eGain Thin film exposed to 0.001 ppb PFAS under scanning electron microscope (SEM) at 250X magnification, (c) Topography of a eGain Thin film exposed to 0.001 ppb PFAS under scanning electron microscope (SEM) at 650X magnification, (d) Topography of a eGain Thin film exposed to 0.014 ppb PFAS under scanning electron microscope (SEM) at 80X magnification, (e) Topography of a eGain Thin film exposed to 0.014 ppb PFAS under scanning electron microscope (SEM) at 250X magnification, (f) Topography of a eGain Thin film exposed to 0.014 ppb PFAS under scanning electron microscope (SEM) at 500X magnification, (g) Topography of a eGain Thin film exposed to 200 ppm PFAS under scanning electron microscope (SEM) at 150X magnification, (h Topography of a eGain Thin film exposed to 200 ppm PFAS under scanning electron microscope

(SEM) at 500X magnification, (i) Topography of a eGain Thin film exposed to 200 ppm PFAS under scanning electron microscope (SEM) at 1000X magnification, (j) Topography of a eGain Thin film exposed to DI Water under scanning electron microscope (SEM) at 150X magnification, (k) Topography of a eGain Thin film exposed to DI Water under scanning electron microscope (SEM) at 350X magnification, (l) Topography of a eGain Thin film exposed to DI Water under scanning electron microscope (SEM) at 800X magnification.

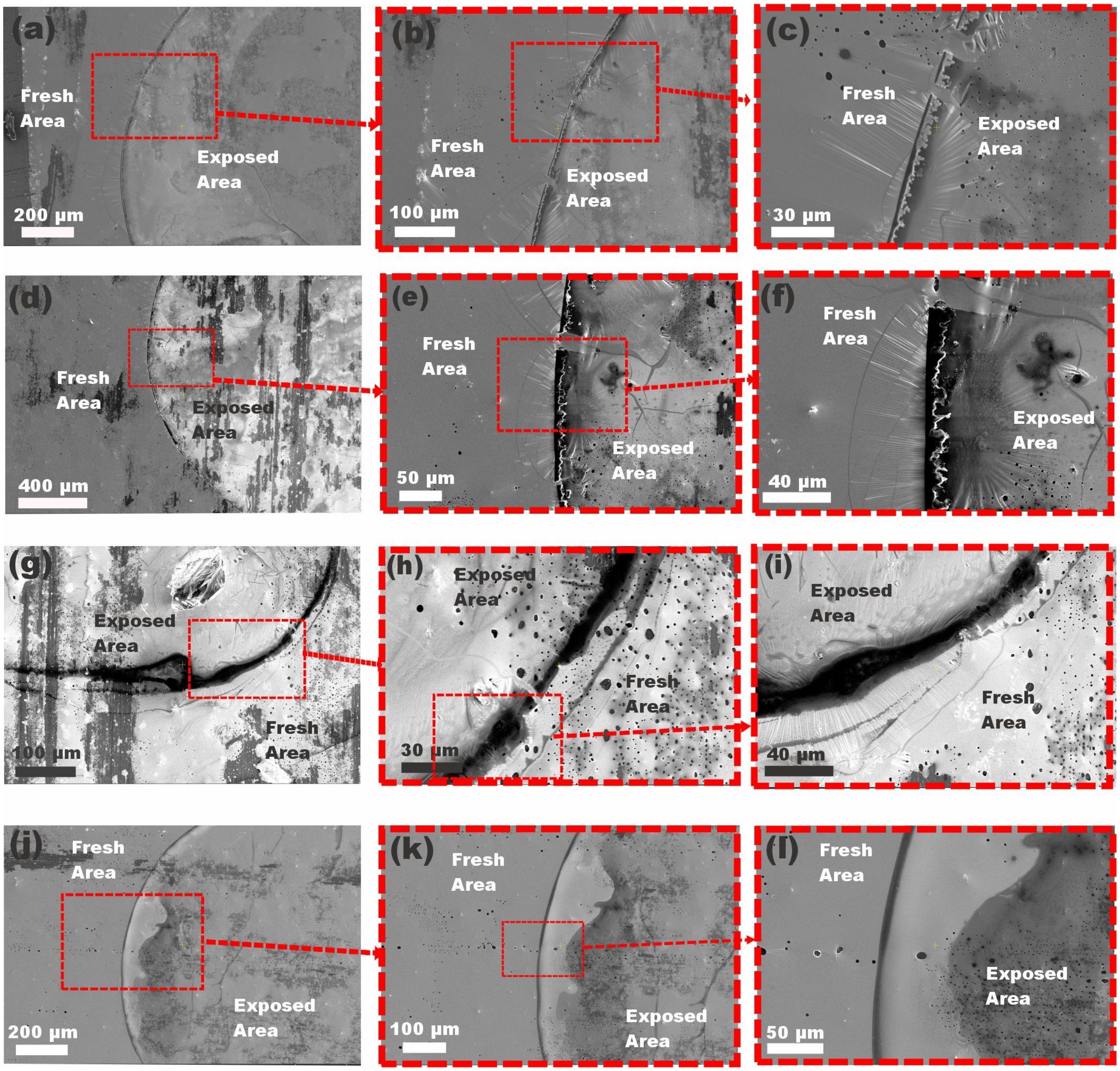


Fig S7: SEM Topography of liquid metal (eGaIn) thin film after exposure to different PFAS concentrations. (a) Topography of a eGain Thin film exposed to 0.001 ppb PFAS under scanning electron microscope (SEM) at 150X magnification, (b) Topography of a eGain Thin film exposed to 0.001 ppb PFAS under scanning electron microscope (SEM) at 250X magnification, (c) Topography of a eGain Thin film exposed to 0.001 ppb PFAS under scanning electron microscope (SEM) at 650X magnification, (d) Topography of a eGain Thin film exposed to 0.014 ppb PFAS under scanning electron microscope (SEM) at 80X magnification, (e) Topography of a eGain Thin film exposed to 0.014 ppb PFAS under scanning electron microscope (SEM) at 250X magnification, (f) Topography of a eGain Thin film exposed to 0.014 ppb PFAS under scanning electron microscope (SEM) at 500X magnification, (g) Topography of a eGain Thin film exposed to 200 ppm PFAS under scanning electron microscope (SEM) at 150X magnification, (h Topography of a eGain Thin film exposed to 200 ppm PFAS under scanning electron microscope (SEM) at 500X magnification, (i) Topography of a eGain Thin film exposed to 200 ppm PFAS under scanning electron microscope (SEM) at 1000X magnification, (j) Topography of a eGain Thin film exposed to DI Water under scanning electron microscope (SEM) at 150X magnification, (k) Topography of a eGain Thin film exposed to DI Water under scanning electron microscope (SEM) at 350X magnification, (l) Topography of a eGain Thin film exposed to DI Water under scanning electron microscope (SEM) at 800X magnification.

Reproducibility test of Elemental Analysis of liquid metal (eGaIn) thin film after exposure to different concentrations of PFAS and Water using EDS:

Two additional tests are given in Figures S8 and S9.

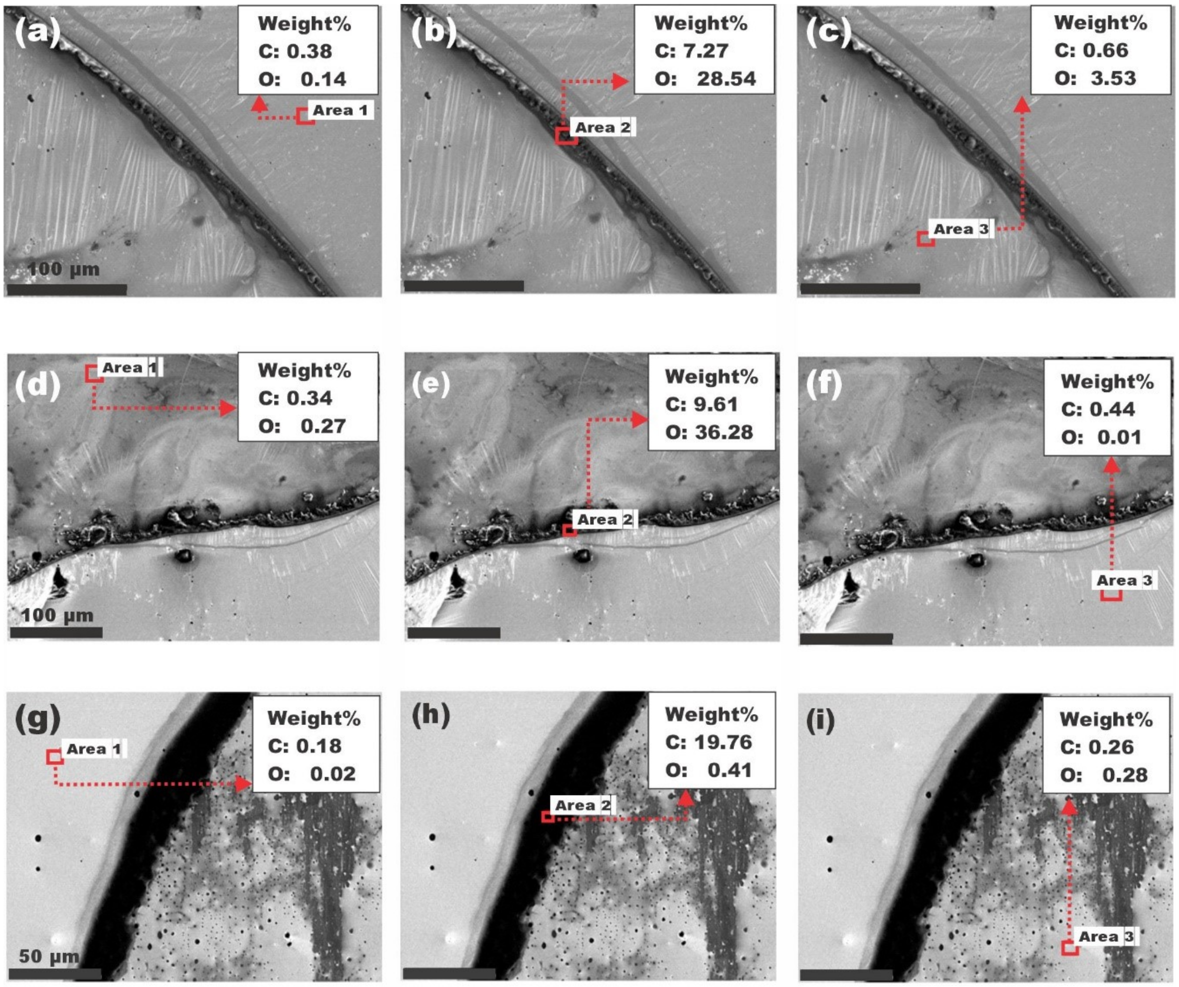
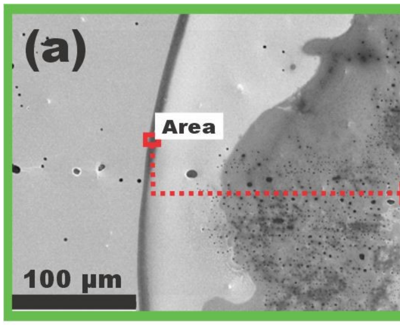
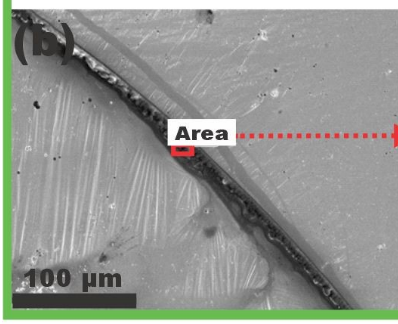


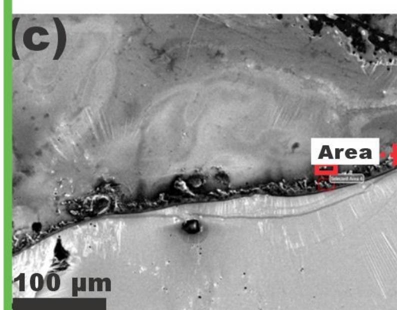
Fig S8: SEM Topography and EDS of liquid metal (eGaIn) thin film after exposure to different PFAS concentrations. (a) Quantitative data of carbon and oxygen on the fresh side of the film using EDS, (b) Quantitative data of carbon and oxygen at the interface of the droplet on the film using EDS, (c) Quantitative data of carbon and oxygen on the that was side exposed to 0.001 ppb PFAS using EDS shows the increased quantity of carbon and oxygen compared to fresh side, (d) Quantitative data of carbon and oxygen on the fresh side of the film using EDS, (e) Quantitative data of carbon and oxygen at the interface of the droplet on the film using EDS, (f) Quantitative data of carbon and oxygen on the that was side exposed to 0.014 ppb PFAS using EDS shows the increased quantity of carbon and oxygen compared to fresh side, (g) Quantitative data of carbon and oxygen on the fresh side of the film using EDS, (h) Quantitative data of carbon and oxygen at the interface of the droplet on the film using EDS, (i) Quantitative data of carbon and oxygen on the that was side exposed to 200 ppm PFAS using EDS shows the increased quantity of carbon and oxygen compared to fresh side,



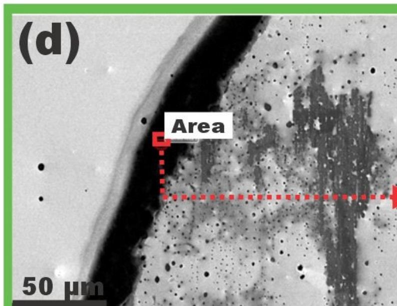
Element	Weight %	Atomic %	Net Int.
GaL	82.34	88.48	918.59
InL	17.66	11.52	82.22



Element	Weight %	Atomic %	Net Int.
C K	7.27	15.44	44.39
O K	28.54	45.52	424.57
F K	0	0	0
NaK	10.03	11.13	171.32
GaL	30.59	11.2	256.14
SiK	2.64	2.4	46.73
S K	3.45	2.74	44.36
ClK	4.28	3.08	45.71
K K	5.22	3.41	39.5
CaK	7.98	5.08	46.45



Element	Weight %	Atomic %	Net Int.
C K	6.84	12.88	42.22
O K	37.06	52.35	559.81
F K	0	0	0
NaK	10.79	10.61	173.22
GaL	17.75	5.75	141.97
MgK	2.87	2.67	49.84
SiK	3.77	3.04	66.47
S K	3.61	2.54	45.34
ClK	4.82	3.07	50.09
K K	2.93	1.69	21.45
CaK	9.56	5.39	53.69



Element	Weight %	Atomic %	Net Int.
C K	19.76	42.61	149.52
O K	0.41	0.66	6.69
F K	21.29	29.03	514.41
GaL	33.82	12.57	337.93
SiK	13.73	12.66	282.84
InL	11	2.48	45.49

Fig S9: EDS of the delaminated area on liquid metal (eGaIn) thin film after exposure to DI water and different concentrations of PFAS. (a) Topography and the elemental composition of an eGain thin film exposed to DI Water at the interface of the droplet and the film, (b) Topography and the elemental composition of an eGain thin film exposed to 0.001 ppb PFAS at the interface of the droplet and the film, (c) Topography and the elemental composition of a eGain thin film exposed to 0.0146 ppb PFAS at the interface of the droplet and the film, (d) Topography and the elemental composition of a eGain thin film exposed to 200 ppm PFAS at the interface of the droplet and the film.

Pattern delamination calculation for Exposing Thin Film to Different Concentration of PFAS:

PFAS	Total Area	Delaminated Area	Delamination %
0.001 ppb	74032	2464	3.33
0.014 ppb	128693	13549	10.53
200 ppm	246500	95788	38.86

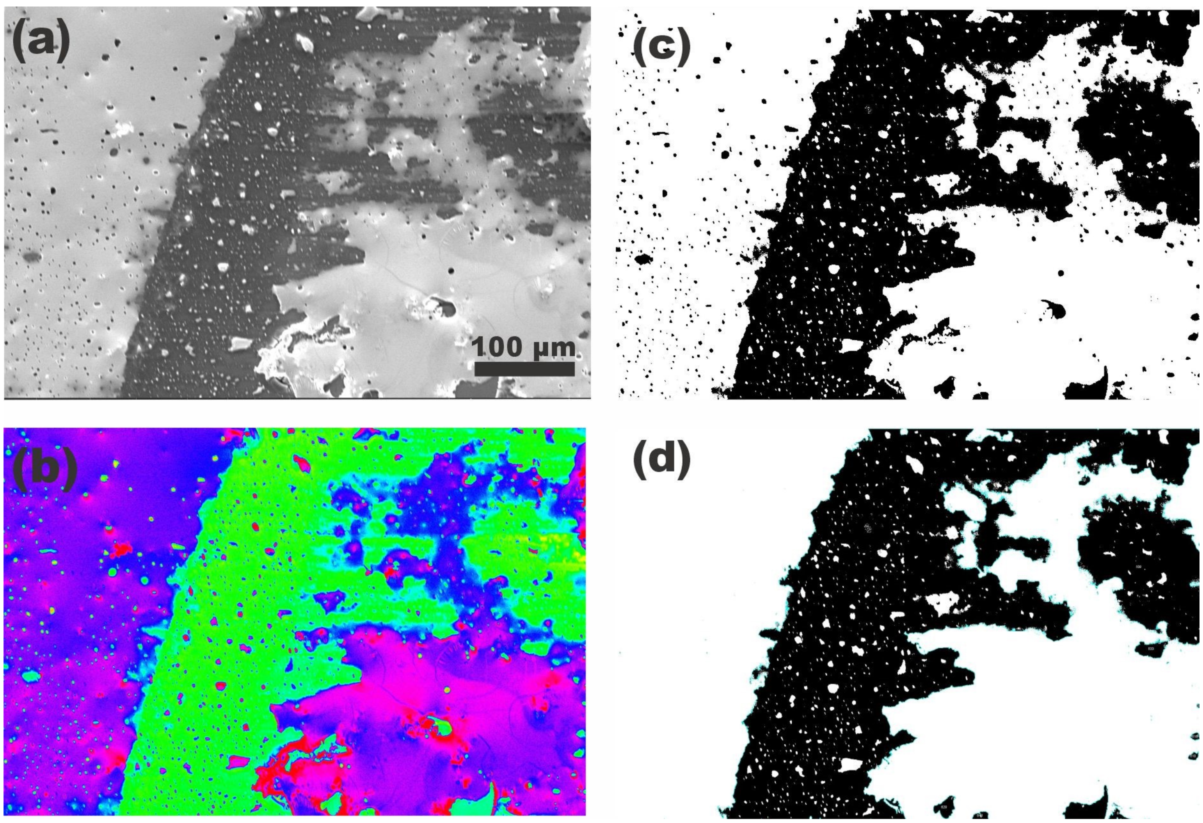


Fig S10: Area calculation of delaminated area. (a) Topography of eGain Thin film exposed to 200 ppm PFAS under scanning electron microscope (SEM), (b) Topography of an eGain Thin film exposed to 200 ppm PFAS under scanning electron microscope (SEM) with highlighted color for better visualization, (c) Binary Image of a eGain Thin film exposed to 200 ppm PFAS under scanning electron microscope (SEM), (d) Binary Image with cleanup of a eGain Thin film exposed to 200 ppm PFAS under scanning electron microscope (SEM) for area calculation in ImageJ.