

Electronic Supplementary Information for *Soft Matter* manuscript:

## X-ray scattering as an effective tool for characterizing liquid metal composite morphology

Erin R. Crater,<sup>‡ac</sup> Ravi Tutika,<sup>‡bc</sup> Robert B. Moore,<sup>ac</sup> Michael D. Bartlett<sup>\*bc</sup>

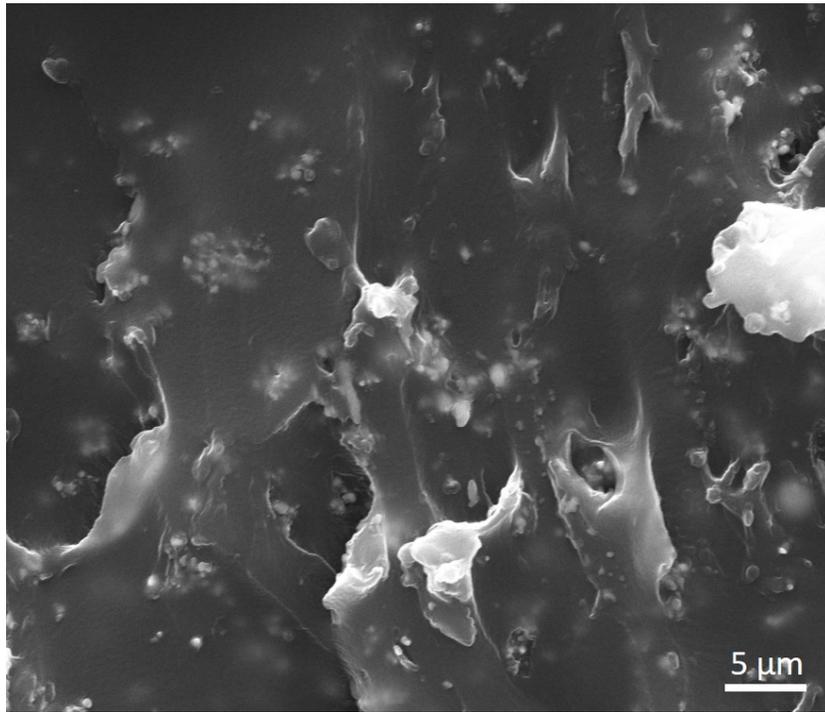
<sup>a</sup> Department of Chemistry, Virginia Tech, Blacksburg, VA 24061, USA.

<sup>b</sup> Department of Mechanical Engineering, Soft Materials and Structures Lab, Virginia Tech, Blacksburg, VA 24061, USA.

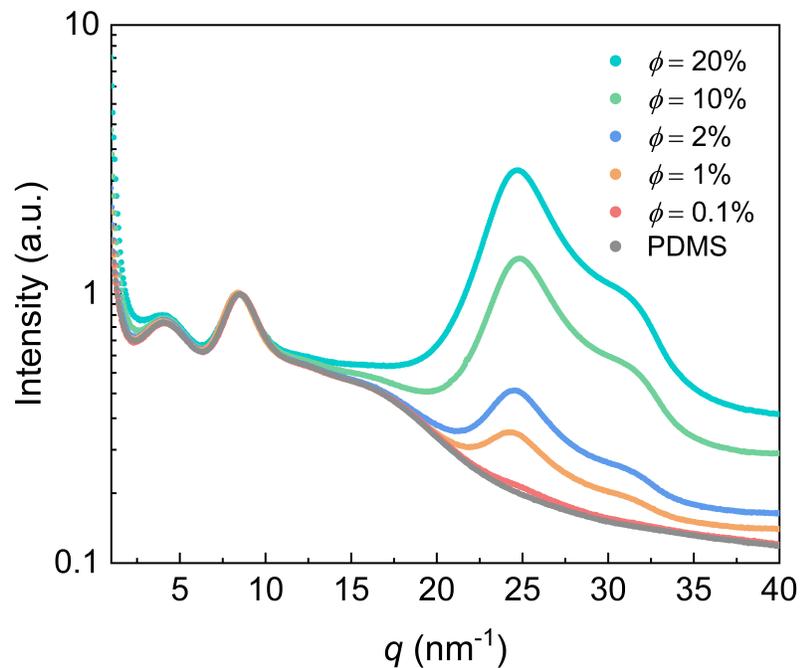
<sup>c</sup> *Macromolecules Innovation Institute (MII), Virginia Tech, Blacksburg, VA 24061, USA.*

<sup>‡</sup>Equal contribution, \*Corresponding author.

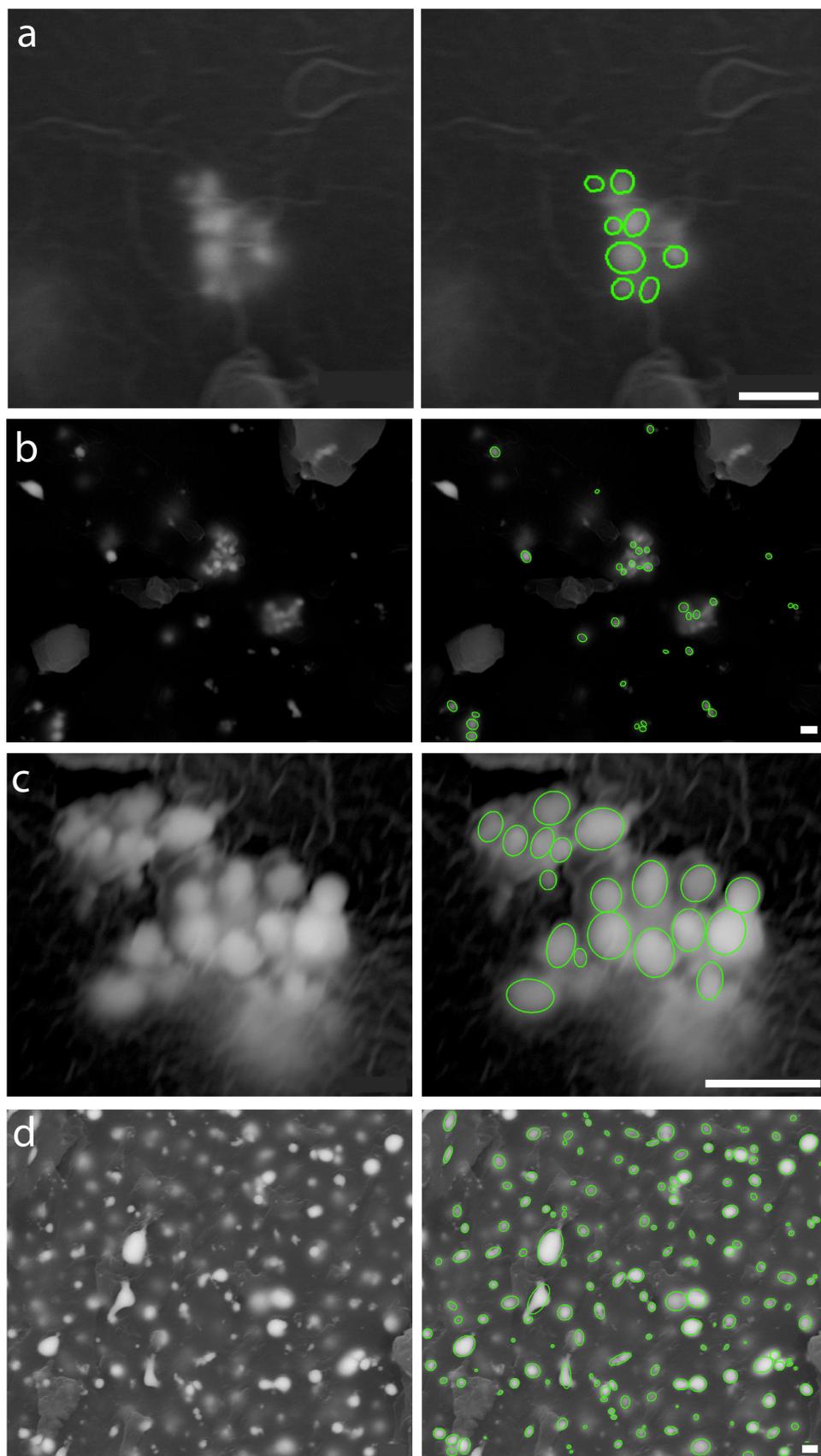
Email: mbartlett@vt.edu



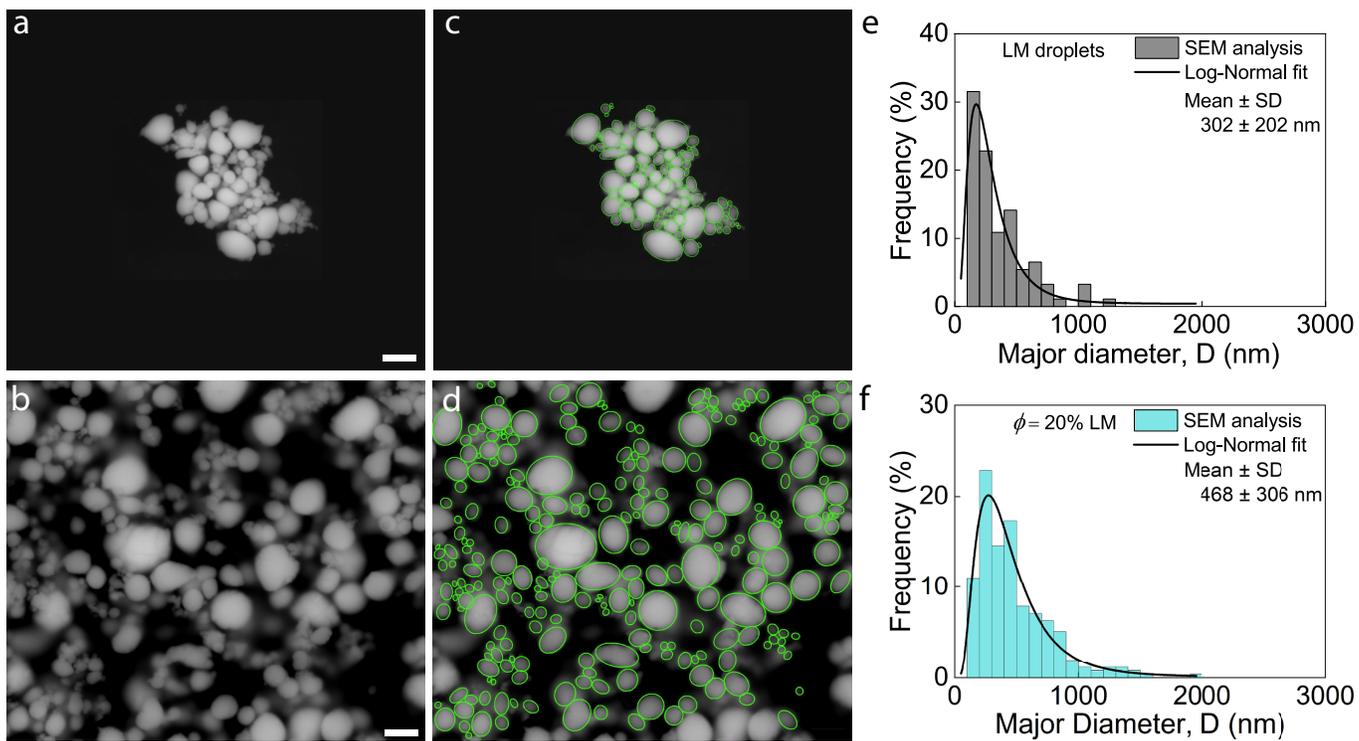
**Fig. S1** SEM image of the liquid metal composite ( $\phi = 2\%$ ) in the Secondary Electron (SE) mode.



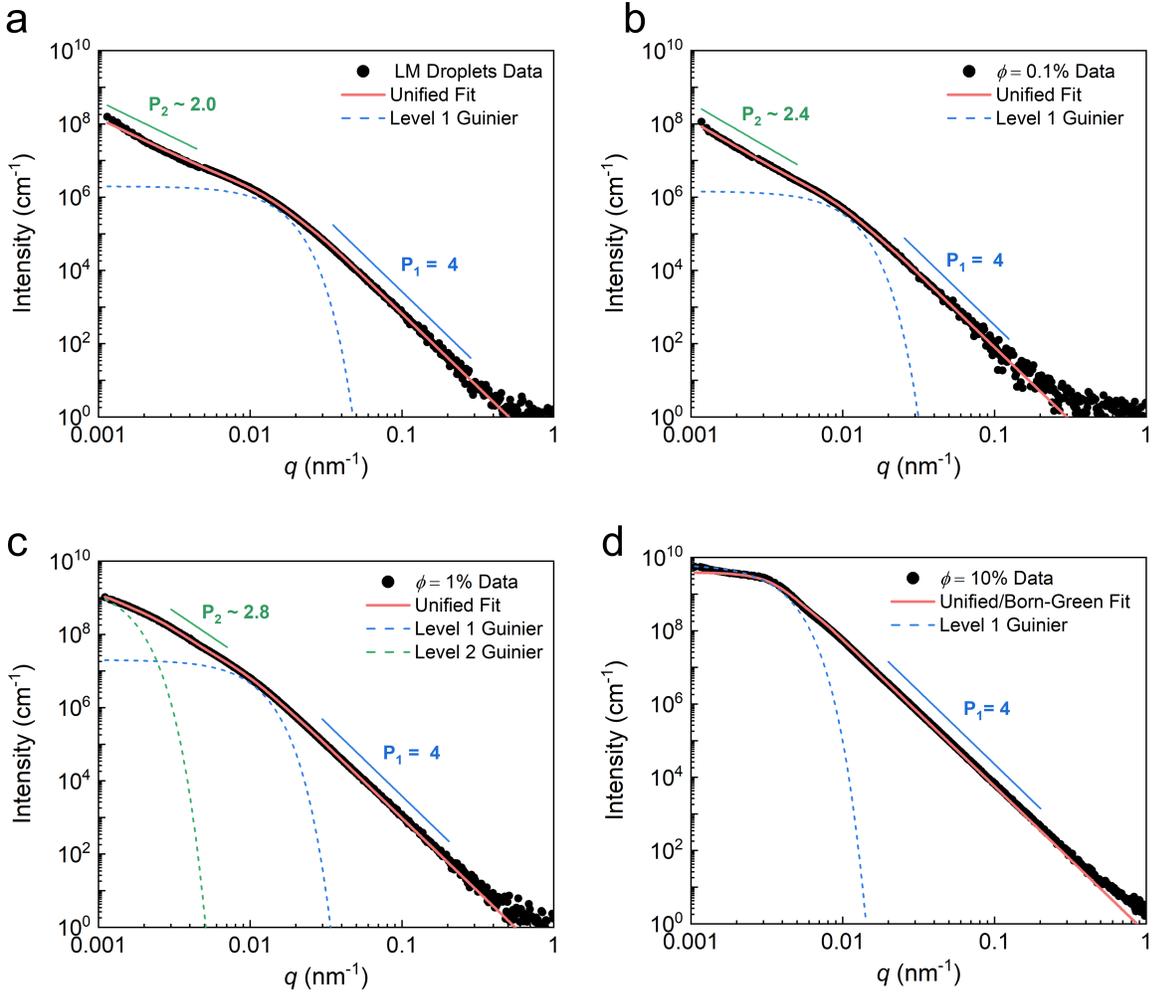
**Fig. S2** WAXS profiles of pristine PDMS elastomer and LM composites with  $\phi = 0.1, 1, 2, 10, 20\%$ . The data are normalized by the matrix feature at  $q = 8.5 \text{ nm}^{-1}$ . The WAXS profiles of the pristine PDMS and LM composites clearly indicate features that arise from the LM particles and the amorphous PDMS matrix. All samples exhibit features due to the matrix: a strong maximum at  $q = 8.5 \text{ nm}^{-1}$ , attributed to interchain spatial correlations, along with a shoulder around  $q = 15 \text{ nm}^{-1}$ , assigned to intrachain spatial correlations.<sup>68</sup> WAXS profiles of the composites display two broad features originating from the interatomic distances in EGaIn, with a maximum at  $q = 25 \text{ nm}^{-1}$  and a shoulder at  $q = 32 \text{ nm}^{-1}$ , consistent with prior analysis of LM-containing materials.<sup>17</sup> As expected, the intensity of these interferences increases with liquid metal content. The absence of sharp reflections confirms that EGaIn remains in the liquid state within the composite.<sup>69</sup>



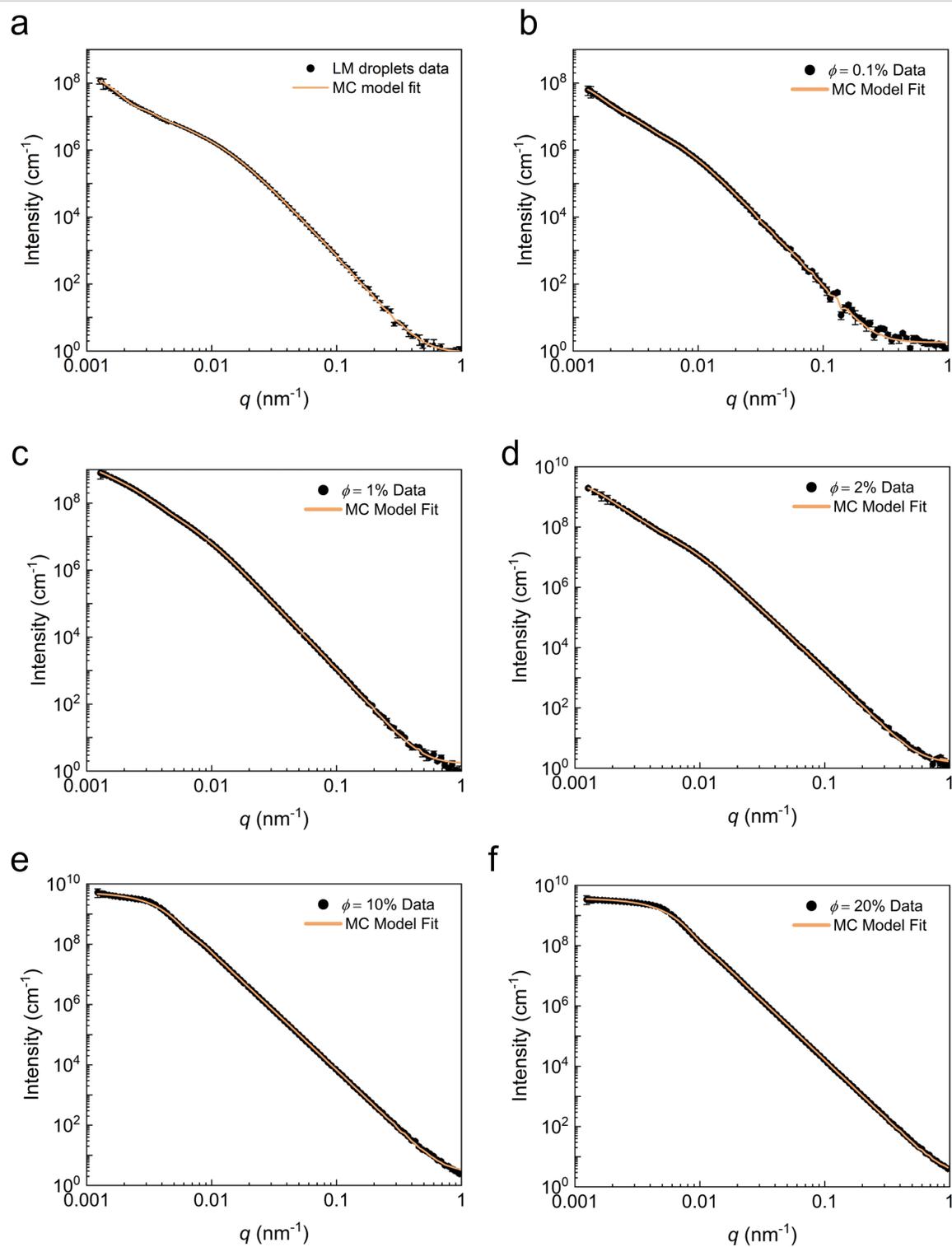
**Fig. S3** Ellipse fits as green outlines on the analyzed LM droplets for composites with a)  $\phi = 0.1\%$ , b)  $\phi = 1\%$ , c)  $\phi = 2\%$ , d)  $\phi = 10\%$ . Scale bars - 1000 nm.



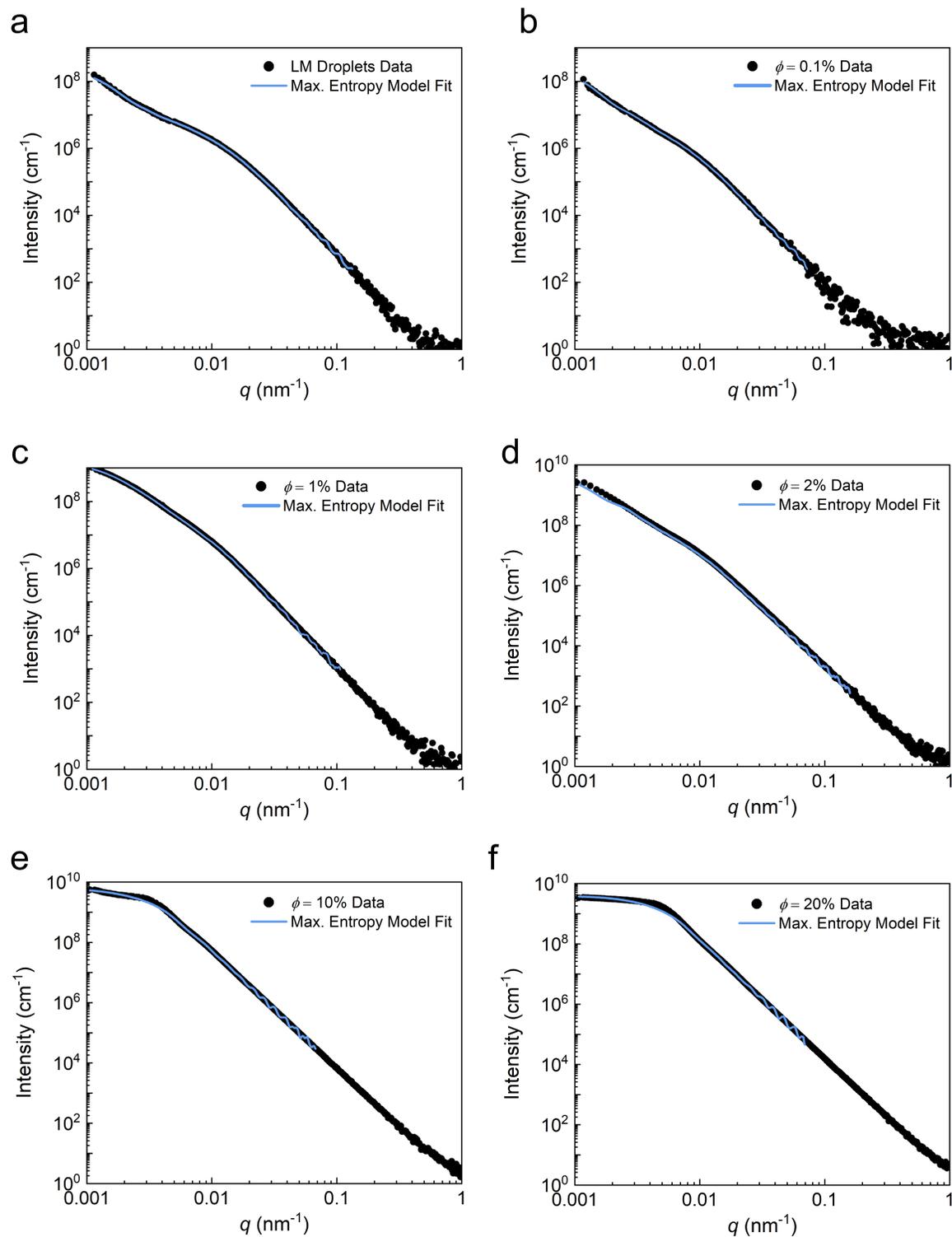
**Fig. S4** Analysis of the fabrication procedure. SEM image of a) LM droplets fabricated through the sonication technique. b)  $\phi = 20\%$  LM composite made by dispersing the droplets in PDMS by shear mixing at 800 rpm in a Flacktek speedmixer for 15 min. c,d) Ellipses fit to the LM droplets in (a) and (b) and overlaid on the actual image, e,f) Major diameter of the ellipses in (c) and (d) are plotted to obtain distribution histograms and mean size using a log-normal fit. Scale bars - 1000 nm.



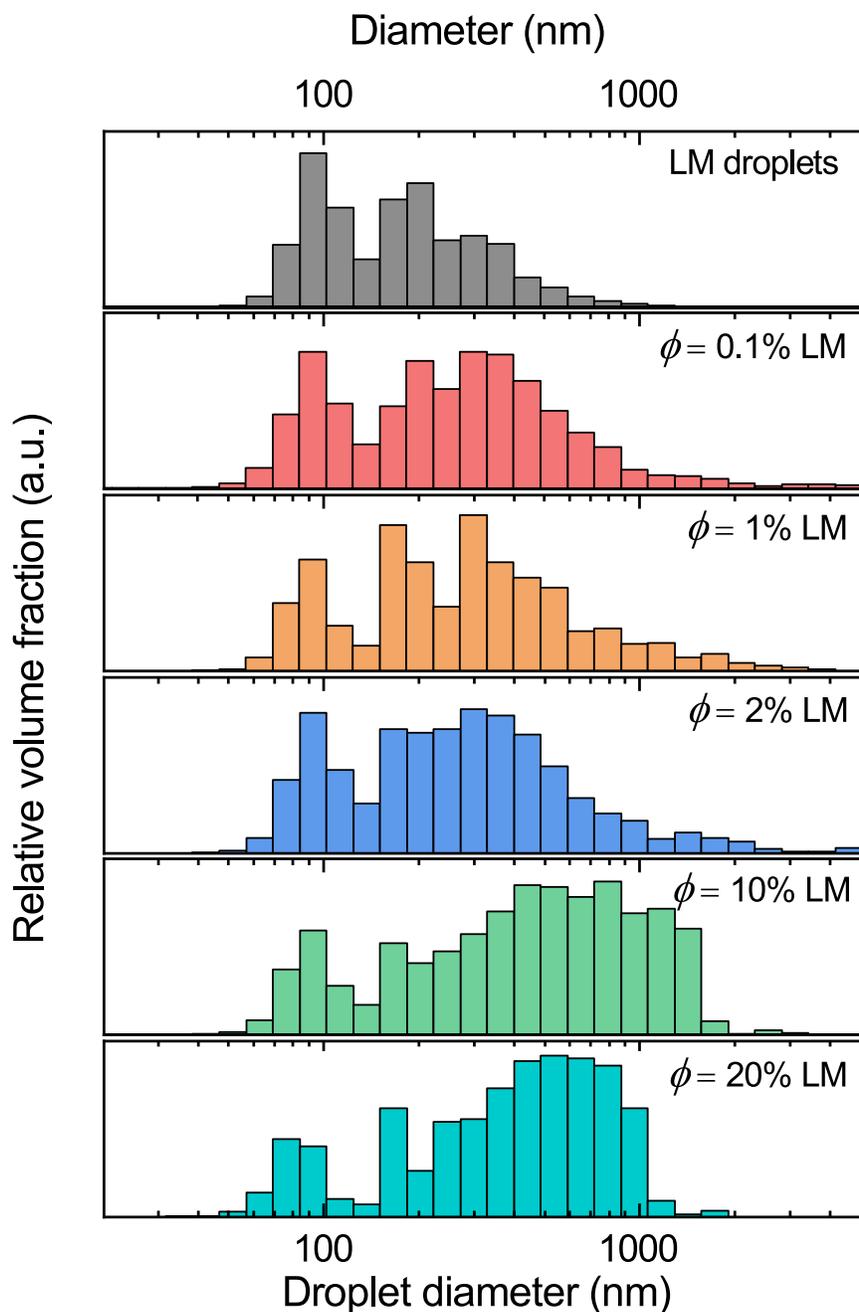
**Fig. S5** Experimental data and Unified model fit for the liquid metal droplets and composites at various liquid metal loadings ( $\phi = 0.1, 1, 10\%$ ).



**Fig. S6** Experimental data and Monte Carlo model fit for the liquid metal droplets and composites at various liquid metal loadings ( $\phi = 0.1, 1, 2, 10, 20\%$ ).



**Fig. S7** Experimental data and Maximum entropy model fit for the liquid metal droplets and composites at various liquid metal loadings ( $\phi = 0.1, 1, 2, 10, 20\%$ ).



**Fig. S8** Volume-weighted size distributions of the LM droplets and composites at various LM loadings based on the maximum entropy fits. In each of the size distributions, an additional population of small scatterers centered around  $D = 95$  nm appears. The origin of this population is likely a minor collection of relatively small LM droplets. The broad distribution of larger scatterers likely originates from a combination of primary particles and aggregates, however further experiments would be required to decouple particle and aggregate contributions.