Electronic Supplementary Material (ESI) for Soft Matter.
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Electronic Supporting Information for Oxide-Mediated Mechanisms of Gallium Foam Generation and Stabilization during Shear Mixing in Air

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S1. Preparation of Gallium Foam

Gallium metal (99.99% purchased from Rotometals) was heated on a hot plate between 35 and 40°C to liquify the material. 100 g of liquid gallium was transferred into a plastic 50 mL beaker using a syringe and stirred at 600 rpm using an industrial mixer with a 3D-printed cross-shaped impeller as shown in Fig. S1a. Mixing times for each 100 g batch of liquid metal varies as follows (in minutes): 0, 2, 3.5, 5, 7.5, 10, 15, 30, 60, 90, and 120. While mixing, the liquid metal was kept warm to maintain its liquid state by blowing air from a heat gun. The impeller for mixing the liquid metal was custom designed to fit with the beaker. The fabrication of the impeller was accomplished by a MakerBot Replicator 3D printer using 1.75 mm PLA filament. An isometric view and the dimensions of impeller are shown in Fig. S1b. A high-speed camera (Photron FastCam mini UX-100, Type: 800K-M-16GB with Sigma 18-250 mm F3.5-6.3 DC MACRO OS HSM lens) was used to image the surface of the LM during the mixing at a 45-degree angle (see example consecutive images of surface waves in Fig. S2). The imaging was performed at 4000 frames per second. To provide enough light for high speed imaging, several lamps were mounted around the apparatus.

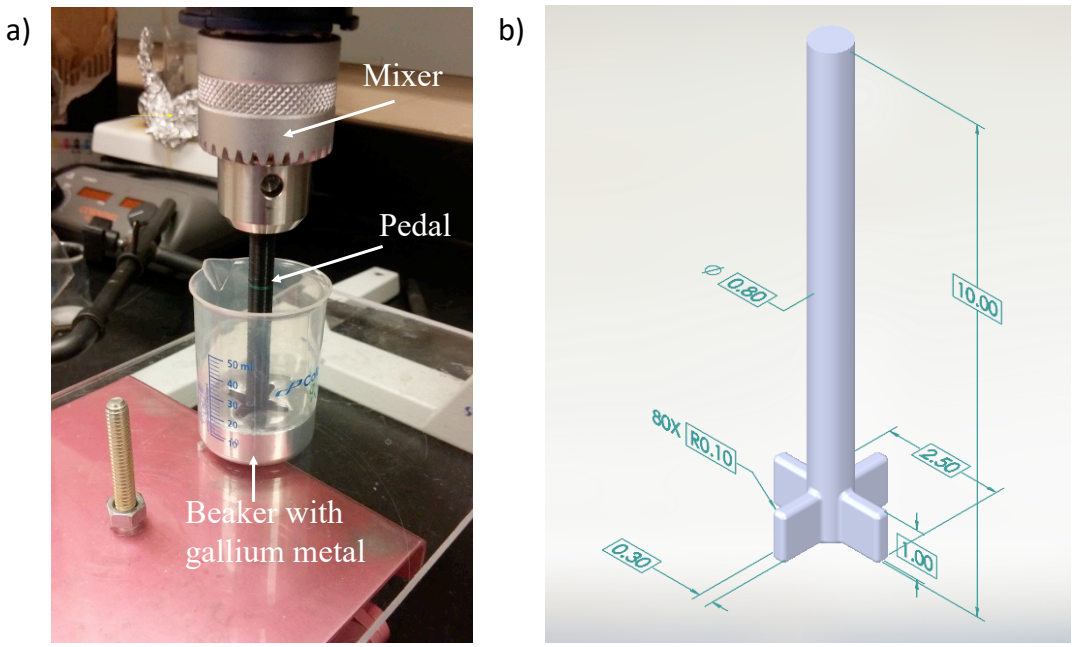

Fig. S1. Setup for preparation of gallium metal foam: (a) mixing arrangement of liquid metal, (b) isometric view of the impeller used for mixing gallium (all the dimensions are in cm). Note that all edges on the cross-shaped impeller have fillets of 0.10 cm radius.

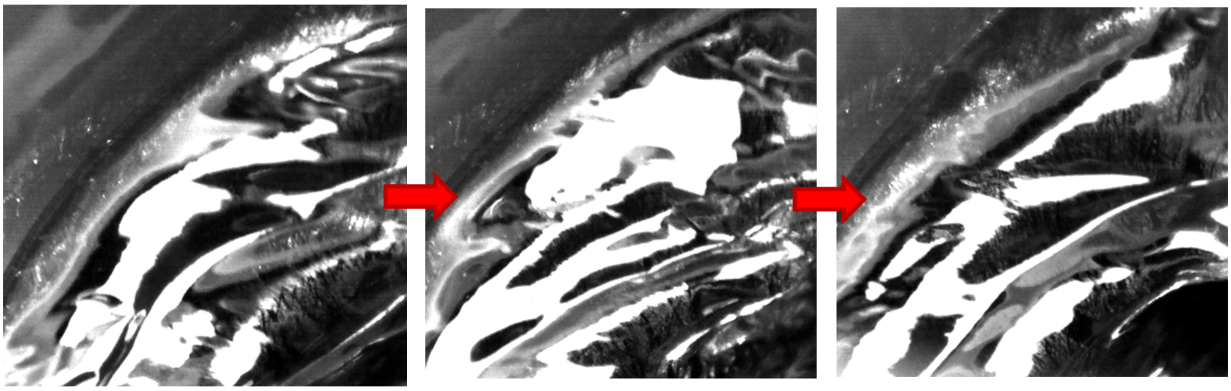

Fig. S2. Consecutive high-speed images of LM surface as the waves and ripples fracture the surface oxides floating on top.

S2. Materials Characterization

Thermal Characterization

The thermal conductivity of gallium foams was characterized using a thermal reference bar testing method following a modified ASTM D5470 standard.[1] The schematic of the testing apparatus is shown in Fig. S4a. The LM-foam was applied onto the copper reference bar and compressed to a 1.5 mm thickness and ~0.1 MPa for each measurement. A Teflon gasket was used to keep the LM-foam in place while testing. Please note that the low-density foam in the prepared samples mixed at shorter times tend to rise from the bulk LM, causing stratification. Effort was made to minimize the effect of stratification by rapidly stirring the sample to homogenize the sample before applying onto the reference bar. During the measurement, the samples were at about 70 to 75 °C.

Density Characterization

The density of each gallium foam sample was determined following the Archimedes principle.[2] A circular disk of gallium samples (2 cm diameter and 6 mm thick) is cast using a polymer mold and the buoyant force is measured by suspending the disk in a 50 mL beaker of water on a microbalance as shown in Fig. S4b.

Rheological Characterization

All rheological experiments conducted for this report were performed with a TA AR-G2 rotational rheometer. A parallel plate of 40 mm diameter and a gap height of 800 μm were used as the geometry for the rheological experiments. The upper plate was attached with sandpaper and the liquid metal was placed on a petri dish attached on the bottom plate to avoid metal-metal contact with the liquid metal samples. All tests were performed at 40 °C, which is above the

melting temperature of gallium, to avoid the liquid metal from solidification. Two tests were performed to measure the storage modulus and viscosity of the samples including oscillatory frequency sweep test and flow sweep test. Oscillatory frequency sweep test was performed at 2% strain with a frequency sweep from 0.1 to 100 rad/s. Flow sweep test was performed by varying the shear rate from 0.1 to 100 s^{-1}. The error bars shown represent the standard deviation between multiple samples and across a range of angular frequencies (0.1 to 100 $rad.s^{-1}$).

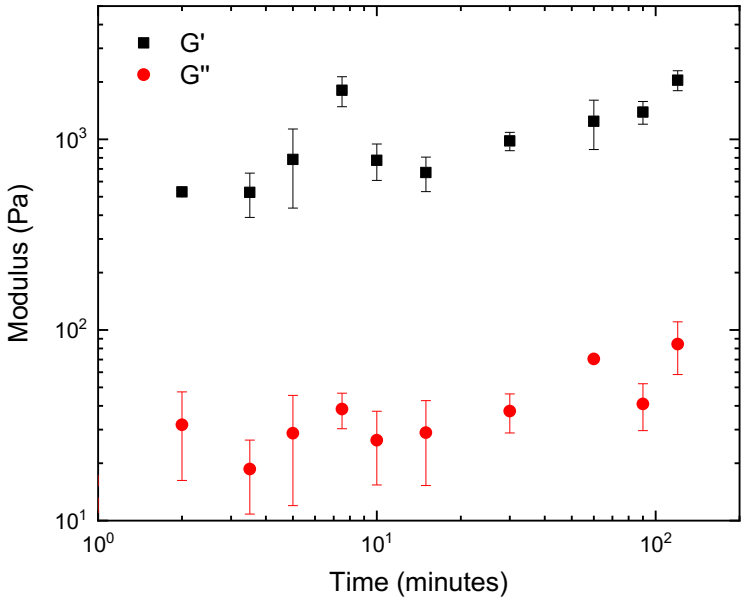

Fig. S3. Comparison of the storage and loss modulus of the liquid metal at increasing mixing times.

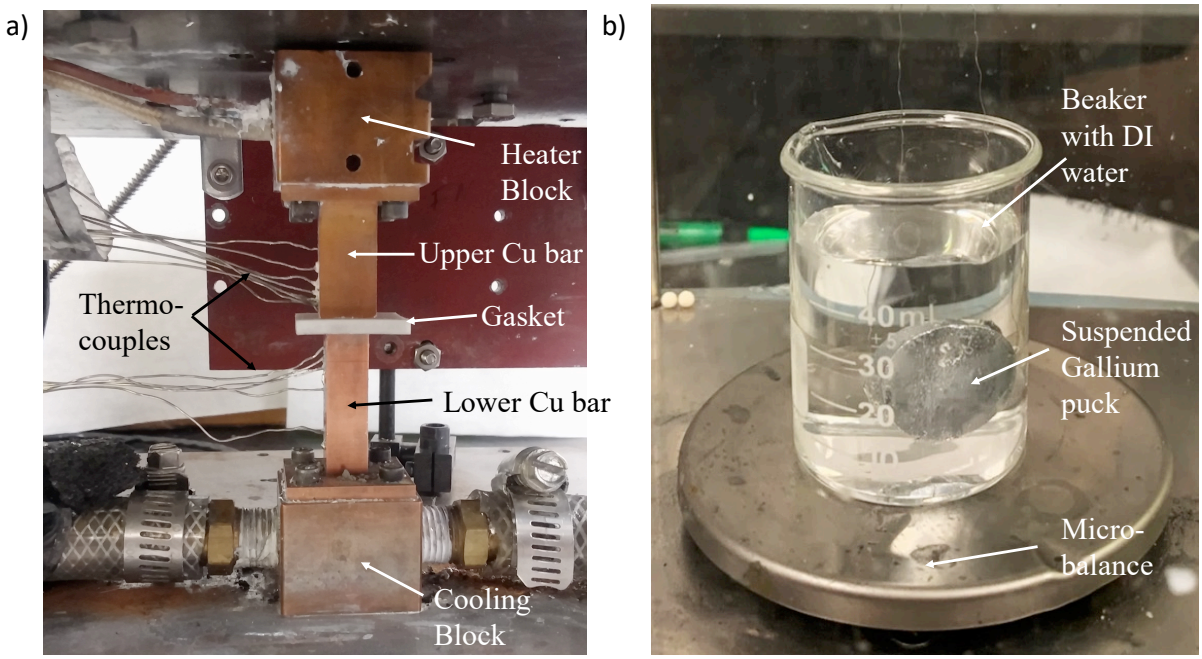

Fig. S4. Characterization of gallium foams (a) schematic of thermal conductivity testing apparatus; (b) schematic of density measurement using Archimedes' method.

S3. Cross-Sectioning of Oxide Flakes

Microscopic images were collected with an Amray 1910 FESEM with 20 kV accelerating voltage. Cross sections of imaged frozen sample blocks (at room temperature) were prepared by cleaving the surface with a razor blade edge. A focused ion beam (FEI NOVA 200 FIB-SEM) with a gallium ion column is used to cross section small sections of gallium stirred in air and show the crumpled oxide flakes within (Fig. S5). Cross sections of larger samples were cleaved manually with a razor blade. Higher magnification SEM images of solidified gallium cross sections mixed at various times is shown in Fig. S6.

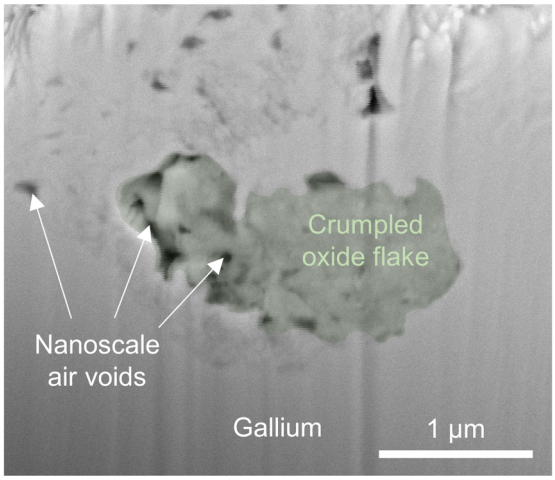

Fig. S5. Cryogenic Focused Ion Beam-Scanning Electron Microscope cross-sectional image of a crumpled oxide flake (colored in light green) in a gallium foam that was stirred for ~5 minutes. The milling and imaging processes were performed using our typical procedure, that is described in detail elsewhere.[3,4]

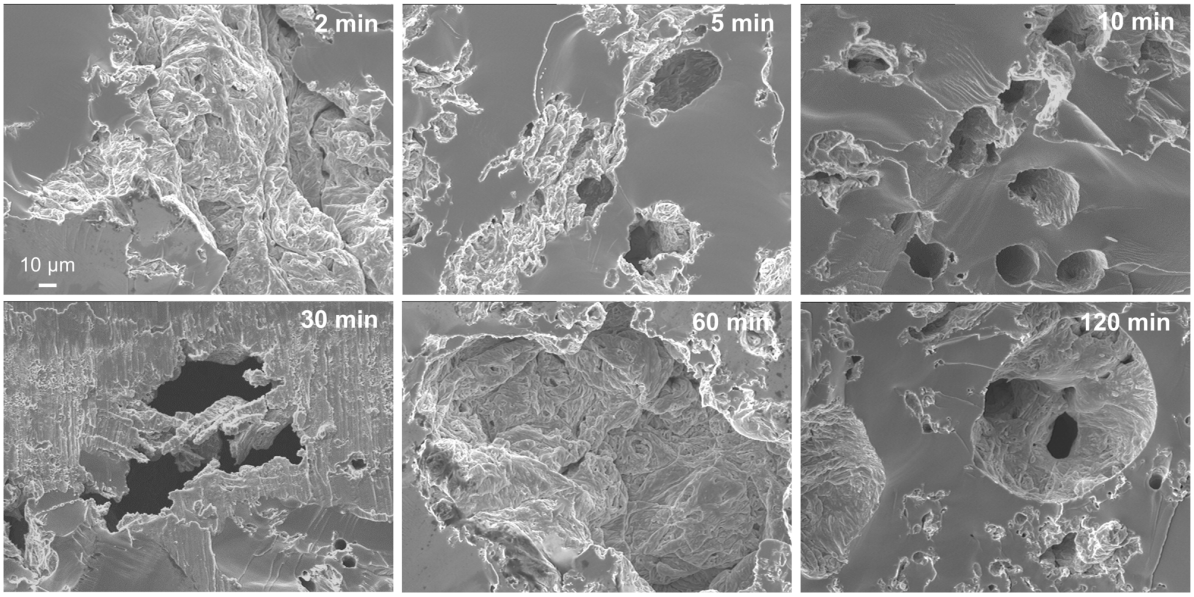

Fig. S6. SEM images of solidified gallium foam cross sections at high magnification.

S4. Expanded Results

Fig. S7 shows the density data which is used to calculate the mean density as in Fig. 2b of the main text. It can be noted that for density measurement, three LM-foam pucks were made

corresponding to each stirring time. Fig. S8 shows the total data points collected for thermal conductivity. For each LM-foam sample, at least five thermal tests were performed. The error bars in Fig. 2c represent the standard deviation from the averaged standard deviations of each individual data point shown here. The storage modulus of the different LM-foam samples is plotted against the porosity (Fig. S9) which is calculated from the ratio of the average density of each sample to the density of the pure gallium sample.

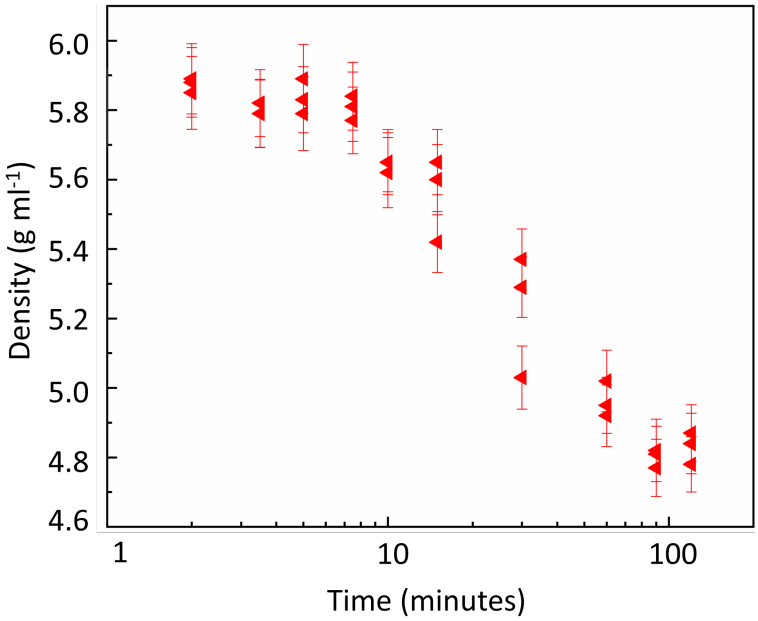

Fig. S7. All collected LM foam density data.

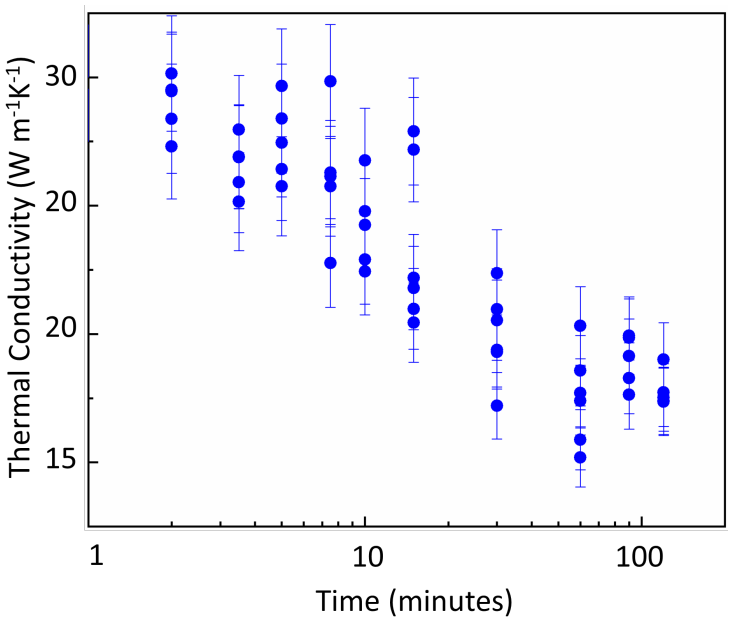

Fig. S8. All collected LM foam thermal conductivity data.

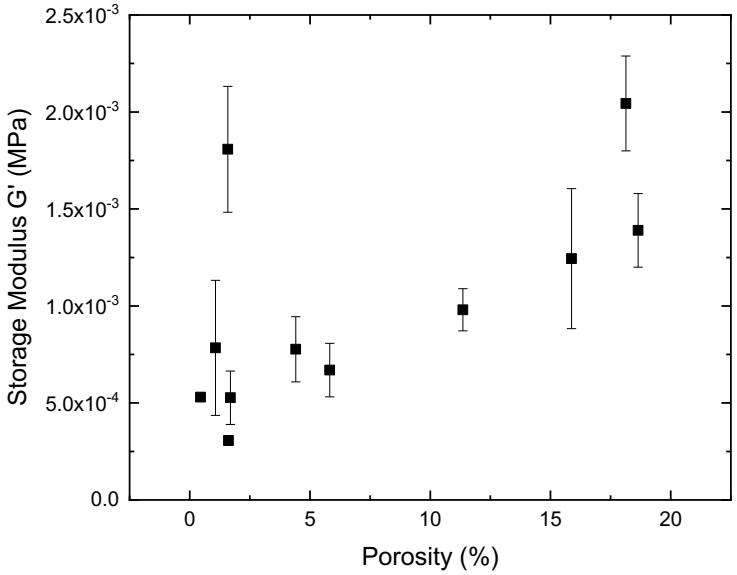

Fig. S9. Storage modulus vs calculated gallium foam porosity.

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