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Supporting Information

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On-Chip Production of Size-Controllable Liquid Metal Microdroplets Using Acoustic Waves

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Figure S1: Experimental setup of the acoustic-based liquid metal microdroplets production platform.

Figure S2: Production of EGaIn microdroplets with different transducer activating frequencies. The figure shows the SEM images for microdroplets produced when the transducer is activated by a 175 V_{p-p} sinusoidal signals with the frequencies ranging from 6.8 to 8.2 kHz. The oxidative voltage applied is 0 V.

Figure S3: Production of EGaIn microdroplets with different transducer activating voltages. This figure shows the SEM images for microdroplets produced when the transducer is activated by a 7 kHz sinusoidal signals with V_{p-p} ranging from 70 to 175 V. The oxidative voltage applied is 0 V.

Figure S4: Size distribution of the EGaIn microdroplets produced when oxidative voltages of 20 and 25 V are applied to the EGaIn droplet.

Figure S6: Experimental setup for the electrochemical sensing of Pb2+ ions experiment.

To understand the acoustic field inside the PMMA well, we performed numerical simulations of our system using the perturbation expansion approach. The fluid response is governed by the standard Navier-Stokes equations for a linear viscous compressible fluid:

$$
\frac{\partial \rho}{\partial t} + \nabla \cdot (\rho v) = 0 \tag{S1}
$$

$$
\rho \frac{\partial v}{\partial t} + \rho (v \cdot \nabla) v = -\nabla p + \mu \nabla^2 v + (\mu_b + \frac{1}{3} \mu) \nabla (\nabla \cdot v)
$$
 (S2)

where ρ is the mass density, ν is the fluid velocity, ρ is the fluid pressure, and μ and μ_b are the shear and the bulk dynamic viscosities, respectively. We note that the term associated with the bulk viscosity has been retained in the formulation to fully account for viscous attenuation both inside the boundary layer as well as in the bulk fluid.

Using the perturbation approach, we divide each flow variable into corresponding firstand second-order components, such that the first-order terms represent the time-harmonic response of the system while the second-order terms represent the steady response of the system.

$$
v = v_0 + \varepsilon \widetilde{v}_1 + \varepsilon^2 \widetilde{v}_2 + O(\varepsilon^3) + \dots
$$

\n
$$
p = p_0 + \varepsilon \widetilde{p}_1 + \varepsilon^2 \widetilde{p}_2 + O(\varepsilon^3) + \dots
$$

\n
$$
\rho = \rho_0 + \varepsilon \widetilde{\rho}_1 + \varepsilon^2 \widetilde{\rho}_2 + O(\varepsilon^3) + \dots
$$
 (S3)

where ε is a non-dimensional smallness parameter.

This amounts to a linearization of the otherwise nonlinear Navier-Stokes equations (Eq. (S1) and (S2)). This linearization results into two sets of equations, which can be successively solved to obtain the first-order and the second-order fields. The first-order fields can be thought of as the acoustic response of the system, while the second-order terms represent the steady acoustic streaming response. More details of this approach can be found elsewhere.^[1]

We performed a 3D simulation of the first-order system to observe the acoustic fields inside the channel using COMSOL Multiphysics. However, the full resolution of the acoustic streaming response (the second-order equations) demands a mesh resolution of the order of viscous boundary layer width. For our device, owing to the large mismatch between the device dimensions (with substrate length of the order of 1 cm) and the characteristic length scales of the second-order solution (the viscous boundary layer width ~ 1 -10 μ m), even a fully resolved 2D simulation of the full system is computationally very expensive. Noting this, while we performed 3D simulations for the acoustic response of the system (the first-order system), we were limited to a 2D simulation (of a down-scaled system) for the second-order simulation. Nonetheless, a 2D simulation of the second-order fields provide valuable qualitative insights into the acoustic streaming flow formed inside the PMMA well, which was also observed experimentally as shown in Fig. S6A-C. Fig. S6D-E shows the results of acoustic streaming flow obtained from the numerical simulation. Considering the limitations mentioned above, these results demonstrate good qualitative agreement with the experiments. We note again that these simulations are instructive in nature with regards to the acoustic streaming flow and were performed only to gain qualitative insights into the formation of acoustic streaming flow inside the PMMA well.

Figure S6: Vortices induced by the transducer. (A-B) Color plot for the distribution of pressure field in the platform (blue minimum to red maximum) obtained from numerical simulations. (C) Experimental result shows the induced vortices by acoustic waves within the PMMA well. (D) The computational domain used for the 2D simulation. (E) Zoomed in image of the PMMA well showing the color plot of the magnitude of the second-order acoustic streaming velocity (blue minimum to red maximum) while the red lines indicate the streamlines of the flow.

Figure S7: Currents recorded at the preconcentration step with different transducer activating frequencies. The concentration of Pb2+ ions is 10 ppm.

Figure S8: (A) DPVs of Pb²⁺ ions obtained with EGaIn microdroplets produced with different oxidative voltages. (B) Overall comparison of DPVs obtained with a single EGaIn droplet, EGaIn microdroplets and mLMMs with or without the presence of acoustic waves.

Supporting Information Reference

[1] Nama N.; Barnkob R.; Mao Z.; Kähler C. J.; Costanzo F.; and Huang T. J., Numerical study of acoustophoretic motion of particles in a PDMS microchannel driven by surface acoustic waves, *Lab Chip*, **2015**,*15*, 2700-2709.