Supplementary Information

· A system for fluid pumping by liquid metal multi-droplets

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1 Details of the PMMA channel processing

The microfabrication steps for the PMMA channel are illustrated below: Firstly, prepare the manufacturing drawing of the PMMA channel (Fig. S1a). Limited to the shape of the tool, the right angles are processed as rounded corners. Secondly, input the drawing information into the engraving machine, and select the appropriate tool (diameter of 1.5 mm), and rotational speed (8000 r/min). Then, start the engraving machine. Finally, two plastic plates are placed at the rounded corners to form right angles (Fig. S1b). The final shape of the PMMA channel is shown in Fig. S1c.

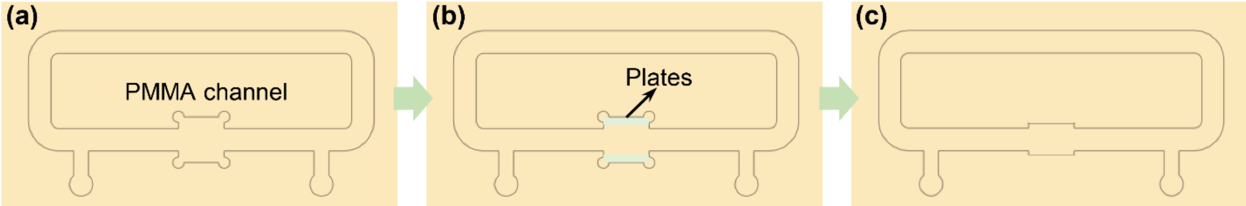

Fig. S1 (a) The manufacturing drawing of the PMMA channel; (b) The PMMA channel with two plates; (c) The final shape of the PMMA channel.

2 Dimensions of the single-droplet cuboid pump and the pillars

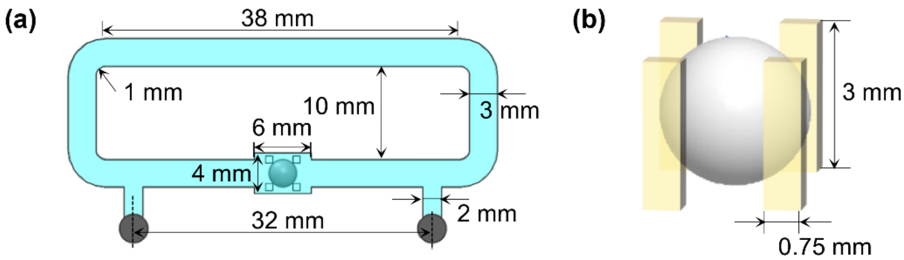

Fig. S2 (a) Structure and dimensions of the single-droplet cuboid pump, and the height of the microfluidic channel is 6 mm; (b) Dimensions of the pillars.

3 Droplet morphologies in the cylindrical and cuboid pumps

The morphologies of the droplet in the cylindrical and cuboid pumps without and with an electric field are shown in Fig. S3. Under an electric field, the droplet in the cylindrical pump moves towards the positive electrode side and undergoes significant deformation. The surface of the droplet of the positive electrode side is squeezed against the cylindrical walls (yellow oval position). The morphologies of droplet in the cuboid pump without and with an electric field are similar.

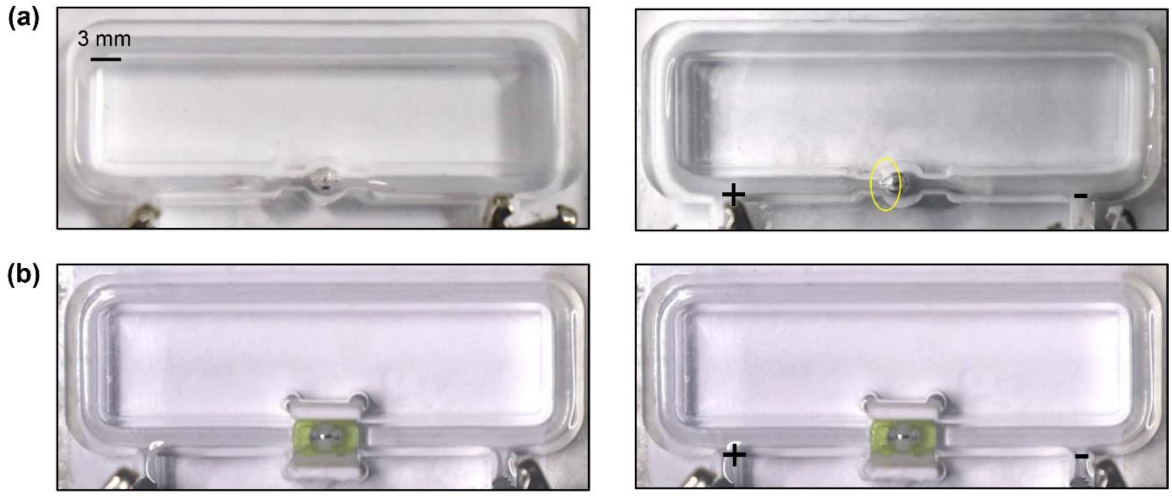

Fig. S3 (a) The morphologies of the droplet in the cylindrical pump without (left) and with (right) an electric field. (b) The morphologies of the droplet in the cuboid pump without (left) and with (right) an electric field. $V_{\text{p-p}}$ is 10 V, V_{DC} is 5 V, f is 50 Hz, and C is 0.4 M.

4 Derivation of the flow velocity of the solution on the droplet surface

The non-uniform distribution of charges within the electric double layer (EDL) at the interface of a liquid metal droplet generates a surface tension gradient on the surface. Consequently, this gradient induces surface flow on the droplet, thereby propelling the flow of the solution. To obtain the expression of the flow velocity of the solution on the surface of the droplet, it is necessary to determine the surface tension gradient of the droplet through the potential distribution in the channel of the pump system.

The following assumptions are made for the liquid metal pump system. First, the electric potential is distributed one-dimensionally along the flow direction of the solution. Second, the resistance of the liquid metal droplets is ignored. To facilitate analysis, the solution area is divided into 6 parts according to the characteristics of the pump system (Fig. S3a). L_1~L_6 are the lengths of each area where L_5=L_3, L_6=L_1, and the corresponding resistance is R_1~R_6 where R_1=R_6, R_3=R_5. a, b, and e are the widths of the channels at different positions. r is the radius of the droplet. The equivalent circuit of the liquid metal pump system is drawn (Fig. S3b). R_{e1} and R_{e2} are the resistances at the contact surfaces between the graphite electrodes on both sides and the solution, and the sum is R_e.

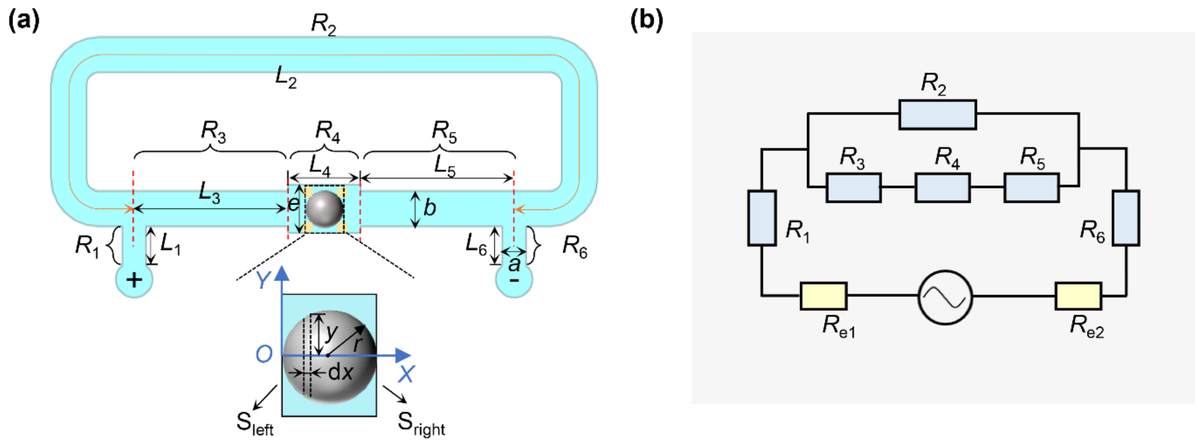

Fig. S4 (a) Structural size and area division of the liquid metal pump; The solution area is divided into 6 areas according to the characteristic of the pump system; R_1~R_6 are the resistances of each area; S_{left} and S_{right} refer to the tangent planes on the left and the right of the droplet. (b) Equivalent circuit diagram of the liquid metal pump; R_{e1} and R_{e2} are the resistance at the contact surfaces between the graphite electrodes on both sides and the solution, and R_2 is connected in parallel with R_3~R_5.

The resistance of each solution region is directly and inversely proportional to the length and cross-sectional area of the region, respectively. When the depth of the solution is h, the resistance values of each region are

$$R_1 = R_6 = \frac{L_1}{\sigma a h} \tag{S1}$$

$$R_2 = \frac{L_2}{\sigma b h} \tag{S2}$$

$$R_3 = R_5 = \frac{L_3}{\sigma b h} \tag{S3}$$

Given the spherical shape of the droplet, the shape of the solution interface between the droplet and the wall is irregular. To analyze this region, a coordinate system is established (Fig. 3a). The radius y and cross-sectional area A_d of the droplet at any x-section in this region can be expressed as

$$y = \sqrt{r^2 - (x-r)^2} \tag{S4}$$

$$A_d = \pi y^2 = \pi\left[r^2 - (x-r)^2\right] \tag{S5}$$

then the cross-sectional area A_s of the solution is expressed as:

$$A_s = eh - A_d = eh - \pi\left[r^2 - (x-r)^2\right] \tag{S6}$$

The surface area $dA_{d\text{-}s}$ of the droplet in the micro-element area dx can be expressed as:

$$dA_{d\text{-}s} = 2\pi y\,dx = 2\pi\sqrt{r^2 - (x-r)^2}\,dx \tag{S7}$$

The solution resistance R_4 can be determined by the integral method:

$$R_4 = \frac{L_4 - 2r}{\sigma eh} + \frac{1}{\sigma}\int_0^{2r}\frac{\mathrm{d}x}{A_s} = \frac{L_4 - 2r}{\sigma eh} + \frac{1}{\sigma}\int_0^{2r}\frac{\mathrm{d}x}{eh - \pi\left[r^2 - (x-r)^2\right]}$$
$$= \frac{L_4 - 2r}{\sigma eh} + \frac{2}{\pi\sigma}\sqrt{\frac{\pi}{eh - \pi r^2}}\arctan\left(r\sqrt{\frac{\pi}{eh - \pi r^2}}\right) \tag{S8}$$

For ease of the expression, denote $k = \sqrt{\pi/\left(eh - \pi r^2\right)}$, and eqn (S8) is simplified:

$$R_4 = \frac{L_4 - 2r}{\sigma eh} + \frac{2k}{\pi\sigma}\arctan(kr) \tag{S9}$$

The resistance of the solution between the tangent planes on the left of the droplet S_{left} and the x-section (i.e., 0~x) is:

$$R_x = \frac{1}{\sigma}\int_0^{x}\frac{\mathrm{d}x}{A_s} = \frac{1}{\sigma}\int_0^{x}\frac{\mathrm{d}x}{eh - \pi\left[r^2 - (x-r)^2\right]}$$
$$= \frac{k}{\pi\sigma}\left\{\arctan\left[k(x-r)\right] + \arctan(kr)\right\} \tag{S10}$$

R_3~R_5 are connected in parallel with R_2, so the total resistance of the circuit is:

$$R_{\mathrm{total}} = 2R_1 + R_e + \frac{R_2\left(2R_3 + R_4\right)}{R_2 + 2R_3 + R_4} \tag{S11}$$

The potential difference across R_3~R_5 or R_2 is:

$$V_2 = \frac{R_2\left(2R_3 + R_4\right)}{\left(R_2 + 2R_3 + R_4\right)R_{\mathrm{total}}}V_{\mathrm{E}} \tag{S12}$$

The surface tension γ of the droplet is related to the capacitance c per unit area of the EDL and the potential difference $V_{\mathrm{t}-x}$ across the EDL:

$$\gamma = \gamma_0 - \frac{1}{2}cV_{\mathrm{t}-x}{}^2 \tag{S13}$$

where γ_0 is the surface tension of the droplet when $V_{\mathrm{t}-x}=0$.

Before the electric field is applied, the charges on the droplet are evenly distributed, forming an original electric double layer. The initial charge density is represented as q_0, and the potential difference V_0 in the original electric double layer of the droplet can be expressed as follows:

$$V_0 = \frac{q_0}{c} \tag{S14}$$

Due to its high conductivity, the droplet in the NaOH solution can be regarded as an equipotential body when the electric field is applied, leading to changes in the potential difference at each location in the EDL, and resulting in an uneven induced double layer. The potential difference V_x of the induced double layer at any position x on the surface of the droplet is the difference between the potential φ_x in the solution and the potential φ_0 of the electrostatic body of the droplet:

$$V_x = \varphi_x - \varphi_0 \tag{S15}$$

φ_x and φ_0 can be expressed as follows:

$$\varphi_x=\left(1-\frac{R_1+R_{\mathrm{e1}}}{R_{\mathrm{total}}}\right)V_{\mathrm{E}}-\frac{R_3+R_x+\dfrac{L_4-2r}{2\sigma eh}}{2R_3+R_4}V_2 \tag{S16}$$

$$\varphi_0=\frac{\int_0^{2r}\varphi_x \mathrm{d}A_{\text{d-s}}}{\int_0^{2r}\mathrm{d}A_{\text{d-s}}}=\frac{\int_0^{2r}2\pi\left[\left(1-\frac{R_1+R_{\mathrm{e1}}}{R_{\mathrm{total}}}\right)V_{\mathrm{E}}-\frac{R_3+R_x+\dfrac{L_4-2r}{2\sigma eh}}{2R_3+R_4}V_2\right]\sqrt{r^2-(x-r)^2}\,\mathrm{d}x}{\int_0^{2r}2\pi\sqrt{r^2-(x-r)^2}\,\mathrm{d}x}$$
$$=\left(1-\frac{R_1+R_{\mathrm{e1}}}{R_{\mathrm{total}}}\right)V_{\mathrm{E}}-\frac{R_3+\dfrac{L_4-2r}{2\sigma eh}}{2R_3+R_4}V_2-\frac{k\arctan(kr)V_2}{\pi\sigma(2R_3+R_4)} \tag{S17}$$

Combining eqs (S16), and (S17), eqn (S15) can be written as:

$$V_x=\frac{kV_2\arctan(kr)}{\pi\sigma(2R_3+R_4)}-\frac{R_xV_2}{2R_3+R_4} \tag{S18}$$

The potential difference $V_{\text{t-}x}$ of the EDL can be expressed as:

$$V_{\mathrm{t}-x}=V_0+V_x \tag{S19}$$

The shear stress at x on the surface of the droplet:

$$\tau_x=\frac{\partial\gamma}{\partial x} \tag{S20}$$

Combining eqs (S13) - (S15) and (S19), eqn (S20) can be written as:

$$\tau_x=\left[\frac{kV_2\arctan(kr)}{\pi\sigma(2R_3+R_4)}-\frac{R_xV_2}{2R_3+R_4}+\frac{q_0}{c}\right]\frac{cV_2}{\sigma(2R_3+R_4)\left\{eh-\pi\left[r^2-(x-r)^2\right]\right\}} \tag{S21}$$

Therefore, the flow velocity of the solution with a dynamic viscosity of μ at any x on the droplet surface is:

$$u_x=\frac{\tau_x r}{\mu}=\left[\frac{kV_2\arctan(kr)}{\pi\sigma(2R_3+R_4)}-\frac{R_xV_2}{2R_3+R_4}+\frac{q_0}{c}\right]\frac{cV_2r}{\mu\sigma(2R_3+R_4)\left\{eh-\pi\left[r^2-(x-r)^2\right]\right\}} \tag{S22}$$

The derivation of the flow velocity of the solution on the droplet surface in a single-droplet cuboid pump is further extended to a multi-droplet cuboid pump (Fig. S4). The number n of droplets affects R_4:

$$R_4=\frac{L_4-2nr}{\sigma eh}+\frac{2nk}{\pi\sigma}\arctan(kr) \tag{S23}$$

The surface tension gradient on the surface of the droplet is the same because each droplet has the same potential difference between its tangent planes on the left (S_{left}) and tangent planes on the right (S_{right}), as well as the same solution shape between the droplet and the wall. The shear stress on the surface of the droplet is only determined by its surface tension gradient, so every droplet has the same shear stress on its surface. Shear stress on the i-th droplet surface:

$$\tau_{x_i}=\frac{\partial\gamma_i}{\partial x_i}=\left[\frac{kV_2\arctan(kr)}{\pi\sigma(2R_3+R_4)}-\frac{R_{x_i}V_2}{2R_3+R_4}+\frac{q_0}{c}\right]\frac{cV_2}{\sigma(2R_3+R_4)\left\{eh-\pi\left[r^2-(x-r)^2\right]\right\}} \tag{S24}$$

where R_{x_i} is:

$$\begin{aligned}R_{x_i}&=\int_0^{x_i}\frac{\mathrm{d}x}{\sigma A_s}=\int_0^{x_i}\frac{1}{\sigma\left\{eh-\pi\left[r^2-(x_i-r)^2\right]\right\}}\mathrm{d}x\\&=\frac{k}{\pi\sigma}\left\{\arctan\left[k(x_i-r)\right]+\arctan(kr)\right\}\end{aligned} \tag{S25}$$

Then, the velocity of the solution on the surface of i-th droplet is:

$$u_{x_i}=\frac{\tau_{x_i}r}{\mu}=\left[\frac{kV_2\arctan(kr)}{\pi\sigma(2R_3+R_4)}-\frac{R_{x_i}V_2}{2R_3+R_4}+\frac{q_0}{c}\right]\frac{cV_2r}{\mu\sigma(2R_3+R_4)\left\{eh-\pi\left[r^2-(x-r)^2\right]\right\}} \tag{S26}$$

When the resistance of the pump system, the initial charge density q_0 in the EDL of the droplet surface, the capacitance c per unit area, the dynamic viscosity μ of the solution, the droplet radius r, and the DC bias V_{DC} remain unchanged, the eqs (S11), (S12) and (S23) can be substituted into (S26) to obtain the relationship between the velocity of the droplet surface solution and the AC voltage amplitude of the and the number of droplets:

$$u_{x_i}\sim\frac{k_1V_{\text{p-p}}^2+k_2V_{\text{p-p}}+k_3}{k_4n^2+k_5n+k_6} \tag{S27}$$

where k_1~k_6 are the coefficients.

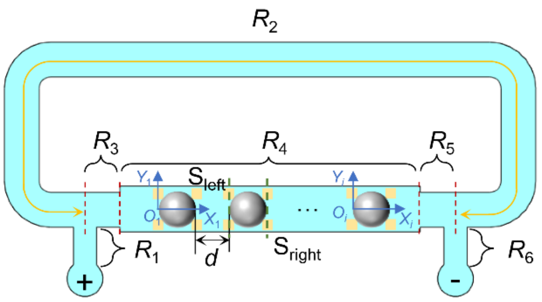

Fig. S5 Schematic diagram of a multi-droplet cuboid pump. d is the distance between two adjacent droplets. Changes in the number of droplets affect the shape of the solution in the corresponding area of R_4.

5 The relationship between the flow rate of the pump and the velocity of the solution on the surface of the droplet

The flow rate q of the pump is related to the flow velocity U of the solution in the channel:

$$q\sim U \tag{S28}$$

The relationship between U and the flow velocity of the solution on the droplet surface $\bar{u}_{x_i}$ is: [1]

$$U \sim \bar{u}_{x_i} \tag{S29}$$

and:

$$\bar{u}_{x_i} \sim u_{x_i} \tag{S30}$$

Thus:

$$q \sim u_{x_i} \tag{S31}$$

According to eqn (S27):

$$q \sim \frac{k_1^{'} V_{\text{p-p}}^2 + k_2^{'} V_{\text{p-p}} + k_3^{'}}{k_4^{'} n^2 + k_5^{'} n + k_6^{'}} \tag{S32}$$

where $k_1{}'$~$k_6{}'$ are the coefficients.

6 The transport of saline by the multi-droplet cuboid pump

Fig. S6 The flow of saline.

7 Dimensions of the "Y"-shaped PMMA channel

The "Y"-shaped PMMA channel is also processed by an engraving machine, and the fabrication details are similar to that of the ring PMMA channel (Fig. S7). The main dimensions of the "Y"-shaped flow channel are shown in Fig. S8.

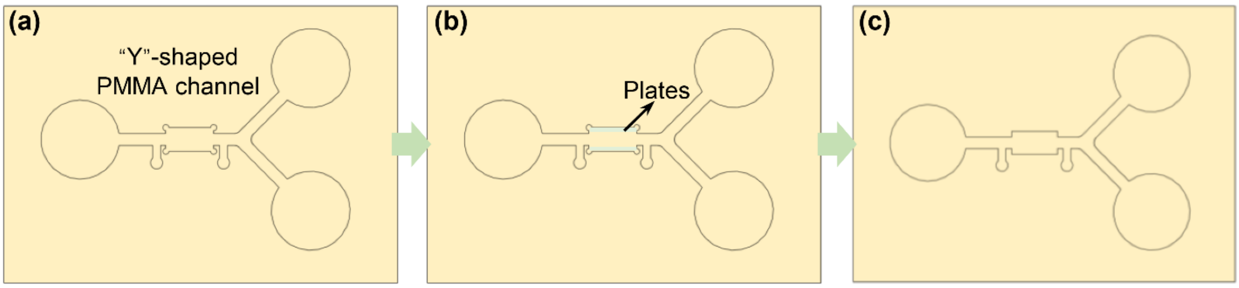

Fig. S7 (a) The manufacturing drawing of the "Y"-shaped PMMA channel; (b) The "Y"-shaped PMMA channel with two plates; (c) The final shape of the "Y"-shaped PMMA channel channel.

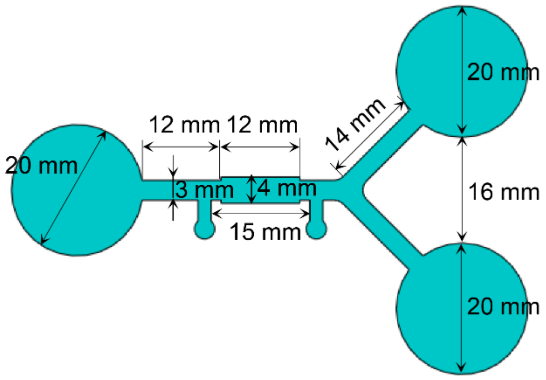

Fig. S8 Dimensions of the “Y”-shaped PMMA channel

References

1. T. Papanastasiou, G. Georgiou and A. N. Alexandrou, *Viscous fluid flow,* CRC Press, Boca Raton, USA, 2021.