Electronic Supplementary Material (ESI) for Lab on a Chip.

Lab on a Chip

Electronic Supplementary Information

In-situ generation of plasma-activated aerosols via surface acoustic wave nebulization for portable spray-based surface bacterial inactivation

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To model the generation and propagation of the acoustic waves in the piezoelectric substrate, we employ the time-domain constitutive equation governing the motion of a piezoelectric solid:[1-5]

$$\frac{\partial D_i}{\partial t} = e_{ijkl}\frac{\partial S_{kl}}{\partial t} + \varepsilon_{ik}^{S}\frac{\partial E_k}{\partial t}, \qquad (S1)$$

$$\frac{\partial T_{ij}}{\partial t} = c_{ijkl}^{E}\frac{\partial S_{kl}}{\partial t} + e_{ijkl}\frac{\partial E_k}{\partial t}, \qquad (S2)$$

where $D_i = \varepsilon_{ik}E_k$ is the electric displacement, E_k is the electric field, ε_{ik} are the dielectric coefficients, t is the time, e_{ijkl} are the piezoelectric stress coefficients, T_{ij} are the stress components, and c_{ijkl} are the elastic stiffness coefficients; the superscripts S and E denote that these quantities are measured at constant strain and constant electric field, respectively. Here, we assume the electric field is *quasi-static* and hence $\partial D_i/\partial t \approx 0$. Together with the infinitesimal strain-displacement relationship,

$$\frac{\partial S_{kl}}{\partial t} = \frac{1}{2}\left(\frac{\partial^2 \xi_k}{\partial x_l \partial t} + \frac{\partial^2 \xi_l}{\partial x_k \partial t}\right), \qquad (S3)$$

Eqns. (S1) and (S2) can be simplified to

$$\frac{\partial E_k}{\partial t} = \frac{e_{ikl}}{\varepsilon_{ik}^{S}}\left[\frac{1}{2}\left(\frac{\partial v_k}{\partial x_l} + \frac{\partial v_l}{\partial x_k}\right)\right], \qquad (S4)$$

$$\frac{\partial T_{ij}}{\partial t} = c_{ijkl}^{E}\left[\frac{1}{2}\left(\frac{\partial v_k}{\partial x_l} + \frac{\partial v_l}{\partial x_k}\right)\right] - e_{kij}\frac{\partial E_k}{\partial t}, \qquad (S5)$$

where ξ is the particle displacement and v is the velocity of the solid.

Equations (S4) and (S5) are solved together with Newton's second law of motion

$$\rho_s\frac{\partial v_j}{\partial t} = \frac{\partial T_{ij}}{\partial t}, \qquad (S6)$$

simultaneously using a finite difference time-domain method; in the above, ρ_s being the mass density of the substrate. To minimize wave reflection from the boundaries, split-field perfectly matched layers are adopted along the left, right, and bottom boundaries of the piezoelectric domain (see Fig. 2 in the manuscript).[6,7] To generate the SAW, a sinusoidal electric potential, $\varphi = \varphi_{p-p}\sin\left(2\pi x/\lambda_{SAW}\right)\sin\omega t$, in which φ_{p-p} is the peak-to-peak voltage and $\omega = 2\pi f_{SAW}$ is the angular frequency with $f_{SAW} = 30$ MHz, is imposed on the surface of the piezoelectric substrate. The dimensions of the piezoelectric substrate are $L_{LN} \approx 15\lambda_{SAW}$ and $H_{LN} \approx 4\lambda_{SAW}$; the distance between computational nodes is $\Delta x_{LN} = \Delta y_{LN} = 0.468\ \mu$m.

To model the acoustic wave propagation and the subsequent acoustic streaming in the liquid atop the LN substrate, we employ a lattice Boltzmann model based on the single-relaxation-time Bhatnagar-Gross-Krook scheme on a two-dimensional square lattice with nine velocities (D2Q9). The macroscopic fluid velocity is defined by[8-11]

$$u(x,t) = \frac{1}{\rho}\sum_{i=0}^{8} f_i e_i, \qquad (S7)$$

and the fluid density is defined by

$$\rho(x,t) = \frac{1}{\rho}\sum_{i=0}^{8} f_i, \qquad (S8)$$

where x denotes the position on the lattice; for D2Q9 (see Fig. w in the manuscript), the lattice vectors are $e_0 = (0,0)$, $e_1 = (1,0)$, $e_2 = (-1,0)$, $e_3 = (0,1)$, $e_4 = (0,-1)$, $e_5 = (1,1)$, $e_6 = (-1,1)$, $e_7 = (-1,-1)$ and $e_8 = (1,-1)$. The particle distribution function f_i spatiotemporally evolves as

$$f_i(x + e_i\Delta t, t + \Delta t) = f_i(x,t) - \frac{1}{\tau}\left[f_i(x,t) - f_i^{eq}(x,t)\right], \qquad (S9)$$

where Δt is the time step and τ is the relaxation time, which is related to the kinematic shear viscosity $\nu = c_f^2(\tau - 1/2)\left(\Delta^2/\Delta t\right)$, where for D2Q9 $c_f = 1/\sqrt{3}$ is the sound speed in the lattice unit.

$$f_i^{eq} = w_i\rho\left(1 + \frac{e_i \cdot u}{c_f^2}\right), \qquad (S10)$$

is the local equilibrium distribution function, whose weight factors w_i are $w_1 = w_2 = w_3 = w_4 = 1/9$, $w_5 = w_6 = w_7 = w_8 = 1/36$ and $w_0 = 4/9$ for the D2Q9 lattice.

We assume the top boundary of the liquid film ($y_f = H_f$) is non-deformable ($u = 0$), i.e., a bounce-back boundary condition applies; periodic boundary conditions, on the other hand, are assumed at both ends of the fluid domain, i.e., at $x_f = 0$ and $x_f = L_f$. As noted earlier, the aim of this simplified calculation is to approximate the ion distribution within the liquid film atop the LN substrate, rather than an accurate hydrodynamic quantification of the flow; thus, we adopt a simplified assumption of a bounce-back boundary condition is employed on the top surface. The total number of cycles is $t = 300f_{SAW}^{-1}$ and the streaming velocities u_{dc} are computed from $t = 20f_{SAW}^{-1}$to $t = 300f_{SAW}^{-1}$. The dimensions for the liquid domain are $L_f \approx 22\lambda_f$ and $H_f \approx 0.5\lambda_f$ whereas the distance between computational nodes is $\Delta x_f = \Delta y_f = 0.117\ \mu$m. At the interface between the piezoelectric substrate and the liquid, i.e., $y_{LN} = H_{LN}$ and $y_f = 0$, the domains are coupled through continuity in the velocities and stresses.

In the absence of any ion generation reactions, the equation governing the transport of ionic species in the liquid is:[12-14]

$$\frac{\partial C_i}{\partial t} + u_{dc} \cdot \nabla C_i = \frac{z_i F D_i}{RT} \nabla \cdot \left(C_i \nabla \varphi_f \right) + D_i \nabla^2 C_i , \qquad (S11)$$

where C_i denotes the ionic concentration of species i, z_i is the ion valency, F is the Faraday constant, D_i is the diffusion coefficient of ionic species i, $R = k_B N_A$ is the molar gas constant in which k_B is the Boltzmann constant, N_A is Avogadro's constant and T is the temperature of the liquid. Additionally, we solve the Poisson equation

$$\nabla^2 \varphi_f = -\frac{\rho_e}{\varepsilon_r \varepsilon_0}, \qquad (S12)$$

to obtain the potential field φ_f; $\rho_e = F \sum_i z_i C_i$ is the net charge density, ε_r is the relative permittivity of the medium, and ε_0 is the permittivity of free space. At the interface between the piezoelectric substrate and the liquid, i.e., $y_{LN} = H_{LN}$ and $y_f = 0$, the boundary conditions $\varphi_{LN} = \varphi_f$ and $\varepsilon_{33} \partial \varphi_{LN} / \partial n = \varepsilon_f \partial \varphi_f / \partial n$ (n being the normal coordinate) are imposed, whereas far from the LN substrate, $\varphi_f(y_f \to \infty) = 0$. For simplicity, we assume that $|z_1 : z_2| = 1:1$ wherein z_1 and z_2 denote the valence for the positive and negative ions, respectively, the bulk ionic concentration $C_\infty = 10^{-6}$ M, the temperature $T = 293$ K, the dielectric constant $\varepsilon_f = \varepsilon_r \varepsilon_0 = 2.6 \times 10^{-10}$ $C^2/J \cdot m$, and the diffusion coefficients for both ions $D_1 = D_2 = 1 \times 10^{-8}$ m^2/s.[13] Equation (S11) is solved using the finite difference time-domain method, whereas Eq. (S12) is solved using the Gauss-Seidel method.[15]

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