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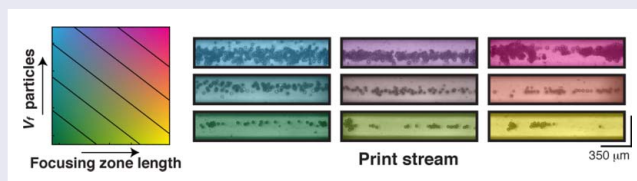
Scaling relationships for acoustic control of two-phase microstructures during direct-write printing

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ABSTRACT

Acoustic forces can align and consolidate particles in fluids, enabling microstructural control of two-phase materials at time-scales compatible with direct-write printing of composites. This paper presents key scaling relationships for acoustically-assisted direct-write printing that describe characteristic time-scales for assembly and alignment of particles during printing. Critical combinations of system parameters (including particle and nozzle dimensions, acoustic excitation amplitude, viscosity, and flow rate) are defined that govern particle focusing and assembly in the print stream. The results can be used to identify combinations of printing protocols and nozzle configurations that control particle packing parallel and transverse to the print direction.



IMPACT STATEMENT

We present theory and experiments demonstrating acoustic focusing in conjunction with direct-write printing for 'on-the-fly' control of two-phase microstructures, and a design framework for printing arbitrary material combinations.

ARTICLE HISTORY

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KEYWORDS

Additive manufacturing; composites; direct-write; acoustic waves

1. Introduction

The impact of 3D printing can be enhanced considerably by developing techniques to deposit multiphase composites with control over phase alignment and density. Such control would enable novel optical, thermal, and electrical properties by achieving particle distributions that introduce functionalities such as photonic band-gaps [1], coupled electro-mechanical behaviors [2], and coupled thermo-mechanical behaviors [3]. The control of particle arrangements within a single print stream can significantly decrease the complexity and cost of printing such functional materials. For example, one could continuously switch from conductors to insulators with conductive particles in an insulating matrix by toggling the microstructure above and below a percolation limit.

Acoustic forces on particles, arising from standing pressure waves in a fluid-filled channel, are an attractive pathway to controlling the density and alignment of particles in a printed composite. These forces are active over large distances and are relatively material agnostic compared to other assembly techniques, as they do not require specific surface chemistries, solution chemistries, or electromagnetic particles. In many of the applications described above, particles are micron-sized, such that primary acoustic radiation forces are much greater than those associated with fluid drag, and particles can be transported over millimeters on the order of seconds or less. These benefits create new opportunities to incorporate acoustic excitation into nozzles used for 3D printing (e.g. by direct-write [4,5] or stereolithography [6]),

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potentially increasing the density and alignment of particles in the printed material significantly relative to that of the ‘base ink’ [5].

Perhaps even more enticingly, these properties create opportunities to achieve hierarchical assembly of bulk specimens using sub-micron particles. Diffusion-controlled self-assembly typically scales with the cube of particle size [7]; aggregates of $O[1\ \mu\text{m}]$ form in seconds, however, aggregates of $O[100\ \mu\text{m}]$ require times on the order of 10^6 seconds (10 days) [7,8]. In contrast, acoustic forces offer a powerful route to commercial-scale fabrication by subverting the limitations of diffusion. While acoustic forces are not typically large enough to quickly assemble sub-micron particles, they can be combined with other techniques to promote rapid assembly of ‘meso-particles’ (e.g. aggregates of sub-micron particles formed *via* self-assembly). Previous studies of the manipulation of sub-micron particles in acoustic waves [9,10] strongly suggest such hierarchical frameworks can be developed to integrate a wide range of time and length-scales.

The aim here is to identify the time- and length-scales associated with acoustic assembly in *continuous flows in channels*, motivated by the development of direct-writing techniques for multiphase inks. This is distinct from previous demonstrations of static or quasi-static acoustic assembly; direct-write printing must appropriately couple the time- and length-scales of both acoustic assembly and fluid transport. This work elucidates the connections between channel dimensions, particle dimensions, acoustic pressures, focusing zone size, and flow characteristics needed to design effective nozzles for acoustic control of microstructures during continuous material deposition.

2. Review of focusing forces

In this section, we present a summary of acoustic forces for particles in standing pressure waves; complete descriptions are available elsewhere [11–15]. The theory summarized here assumes that the particle size is much smaller than the standing wavelength, and that wave behavior is unaffected by viscosity. Using these solutions, others have considered time scales associated with assembly in stationary fluids [16,17]. The results presented here are new in that they extend these efforts to quantify conditions for microstructural control during direct write of two-phase materials.

Consider a standing half-wave in pressure in the x -direction (with $x=0$ at the channel center) and flow in the positive z -direction; a schematic of the device geometry used in this work is presented in Figure 1(A). The *time-average* of the primary radiation force in the

x -direction, $\bar{F}_p$, is given by [11,12,14,15,18]:

$$\bar{F}_p \equiv \frac{F_p}{F_o} = -\sin(2\pi\bar{x}); \quad F_o = \frac{\pi^2 a^3 \beta_f p_o^2 \cdot \phi}{3W};$$

$$\phi(\beta, \rho) = \frac{5\rho_p - 2\rho_f}{2\rho_p + \rho_f} - \frac{\beta_p}{\beta_f} \quad (1)$$

where F_o is the peak focusing force, $\bar{x} = x/W$ is the particle position, a is the particle radius, p_o is the acoustic pressure amplitude, and W is the channel width. ρ and β are, respectively, the density and compressibility, where subscripts denote the fluid (f) and particle (p). In the present study, we consider only particles with a positive contrast factor ϕ , such that the primary radiation force transports particles to the pressure node at the channel center. However, these results can easily be extended to scenarios with multiple nodes or particle types.

In addition to the primary radiation force, there are secondary scattering forces associated with particle interactions, given by [13,14]:

$$F_s(x, t) = 4\pi a^6 \left[\frac{(\rho_p - \rho_f)^2 (3 \cos^2 \theta_{ij} - 1)}{6\rho_f d_{ij}^4} v(x, t)^2 - \frac{\omega^2 \rho_f (\beta_p - \beta_f)^2}{9d_{ij}^2} p(x, t)^2 \right] \quad (2)$$

where d_{ij} is the distance between particle i and particle j (Figure 1(B)), θ_{ij} is the angle formed between the vector passing from particle i to particle j and the x -axis, and ω is the frequency of the pressure and velocity waves. The first term depends on local wave velocity, $v(x, t)$, while the second term depends on local wave pressure, $p(x, t)$. We utilize the time-average of this scattering force, and corresponding time-average of the squares of the pressure and velocity fields:

$$\overline{\langle v(x, t)^2 \rangle} = \frac{p_o^2}{2} \cos^2(\pi x); \quad \overline{\langle p(x, t)^2 \rangle} = \frac{p_o^2 \beta_f}{2\rho_f} \sin^2(\pi x) \quad (3)$$

These expressions are very similar to solutions presented elsewhere, e.g. Bruus [15], but utilize a coordinate system where $x=0$ corresponds to the channel center (Figure 1(A)). Substituting the time-average fields and normalizing by F_o produces:

$$\bar{F}_s \equiv \frac{F_s}{F_o} = \bar{a}^3 \left[\frac{\tilde{\alpha}_\rho}{\bar{d}_{ij}^4} (3 \cos^2 \theta_{ij} - 1) \cos^2 \pi \bar{x}_{ij} - \frac{\tilde{\alpha}_\beta}{\bar{d}_{ij}^2} \sin^2 \pi \bar{x}_{ij} \right] \quad (4)$$

where $\bar{a} = a/W$ is the normalized particle size, $\bar{d}_{ij} = d_{ij}/W$ is the normalized distance between particles i and

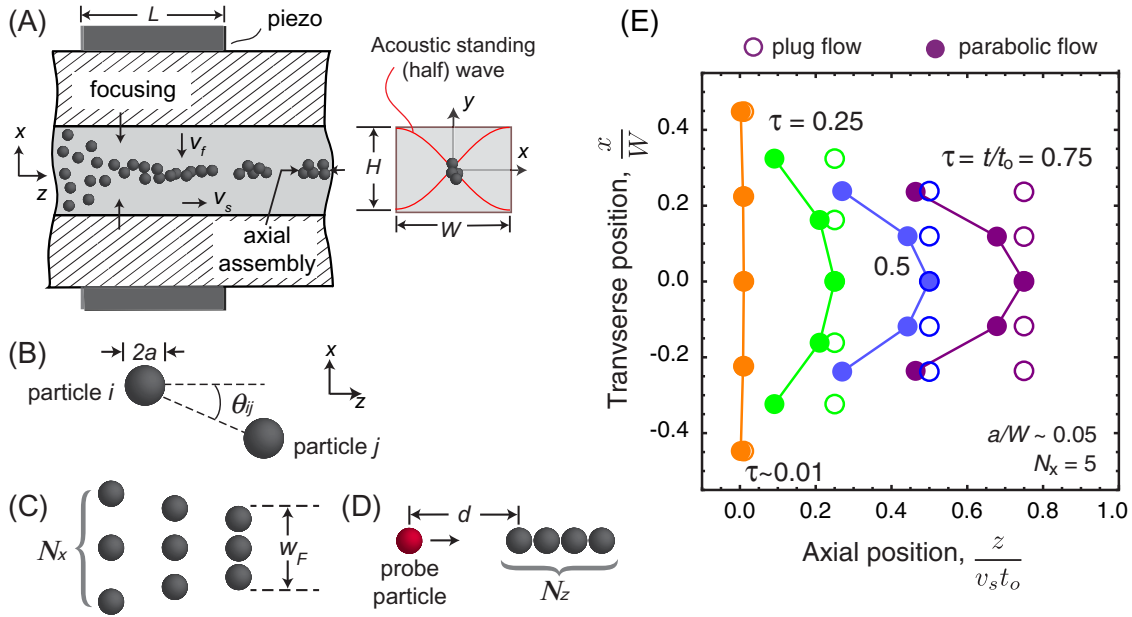


Figure 1. (A) Acoustic nozzle geometry and coordinate system. The piezoelectric element is excited at a frequency corresponding to a half-standing-wave in pressure, such that particles with positive contrast factor ϕ focus to the pressure node due to the primary acoustic radiation force (PARF). Secondary scattering forces lead to particle assembly in the axial direction. (B) Particle geometry and (C) idealized row and (D) column formation behavior as a result of the PARF and scattering forces (E). Particle focusing as a function of time τ in parabolic and plug flows.

j , and the pre-factors $\tilde{\alpha}_\rho$ and $\tilde{\alpha}_\beta$ are given by:

$$\begin{aligned}\tilde{\alpha}_\rho &= \frac{6}{\pi\phi}\alpha_\rho = \frac{1}{\pi\phi}\left(\frac{\rho_p}{\rho_f} - 1\right)^2; \\ \tilde{\alpha}_\beta &= \frac{24}{\pi\phi}\alpha_\beta = \frac{8\pi}{3\phi}\left(\frac{\beta_p}{\beta_f} - 1\right)^2\end{aligned}\quad (5)$$

Finally, moving particles are subjected to drag and inertial forces, given by:

$$(\bar{F}_d^x, \bar{F}_d^z) \equiv \frac{(F_d^x, F_d^z)}{F_o} = \frac{6\pi\eta a}{F_o}(v_x^p, v_z^p) = (\dot{\bar{x}}, \dot{\bar{z}}) \quad (6)$$

where $\dot{\bar{x}}$ and $\dot{\bar{z}}$ are the normalized velocities of the particle relative to the local fluid velocity. Dots denote derivatives with respect to the dimensionless time $\tau = t/t_o$. t_o is defined as the time required to drive a particle from the wall ($x = \pm W/2$ or $\bar{x} = \pm 1/2$) to the center of the channel ($\bar{x} = 0$), under the spatial average of the primary acoustic focusing force, $\langle F_o \sin(2\pi\bar{x}) \rangle = 2F_o/\pi$:

$$t_o = \frac{3\pi^2\eta a W}{2F_o} = \frac{9\eta}{2\bar{a}^2\beta_f p_o^2\phi} \quad (7)$$

This assumes the fluid velocity in the x -direction is zero, for laminar flow conditions.

The inertial force acting on a particle in the x -direction, using the same normalizations as above, is

given by:

$$\begin{aligned}\bar{F}_I &\equiv \frac{F_I}{F_o} = \frac{4\pi\rho_p a^3}{3F_o}\dot{v}_x^p = \bar{m}\ddot{\bar{x}}; \\ \bar{m} &= \frac{4\pi\rho_p a^3 W}{3F_o t_o^2} = \frac{16a^4\rho_p\phi\beta_f p_o^2}{81\pi\eta^2 W^2}\end{aligned}\quad (8)$$

For most microparticles in fluids with viscosities equal to or greater than that of water, inertial forces are negligible in comparison to drag and acoustic forces, and particle transport is dictated by the viscous time scale. Example calculations are provided in Experimental Parameters, in SI.

3. Time scales associated with focusing and printing

Prior to addressing non-uniform flow profiles and scattering between particles, it is instructive to consider simple estimates of the time scales associated with transport in the printing (flow) direction and focusing (transverse to flow) directions. Consider flow in a rectangular channel with a standing acoustic wave (Figure 1(A)). For simplicity, assume that both the fluid velocity v_s and the focusing force F_o are uniform, and the focusing time is t_o . The transport time for a particle to pass through the length of the channel coupled to the piezoelectric, L , is $t_T = L/v_s$. The ratio of these times provides an insightful

dimensionless measure of the nozzle length, given by:

$$\tilde{L} \equiv \frac{t_T}{t_o} = \frac{2La^2\beta_f\phi p_o^2}{9W^2\eta v_s} \quad (9)$$

For $\tilde{L} > 1$, the focusing zone is long enough for complete focusing, whereas little focusing occurs if $\tilde{L} \ll 1$. Alternatively, $\tilde{v}_s = 1/\tilde{L}$ can be thought of as a dimensionless flow velocity, where complete focusing only occurs for $\tilde{v}_s < 1$; otherwise, particles move through the focusing zone too quickly.

In the next section, a more detailed examination of particle motions illustrates that this elementary picture (single particle, uniform field) captures the essential behaviors for transport and focusing time: spatial variations in velocity and focusing force do not significantly change the relevant time scales, nor does acoustic scattering. Section 3.2 introduces the time required to aggregate particles due to scattering forces in the flow direction. The balance of these three time scales (focusing, transport, and aggregation) is discussed in Section 3.3.

3.1. Time scales for focusing accounting for non-uniform fields and scattering

To gain insight into the impact of acoustic scattering and spatial variations in focusing force, consider a row of N_x equally-spaced particles (Figure 1(C)). For simplicity, we neglect any rearrangement of the particles in the axial (flow) direction. The sum of forces in the x -direction on each particle leads to N_x coupled non-linear, first-order differential equations for particle position.

Previous quasi-static solutions [18] reveal that the equilibrium particle spacing (balancing focusing and scattering forces) is only slightly non-uniform. Therefore, we impose uniform spacing at all times: $\bar{d}_x = 2\bar{x}_o/(N_x - 1)$, where $\bar{x}_o$ is the position of the outermost particle. The governing equation is:

$$\begin{aligned} \dot{\bar{x}}_o + \sin 2\pi \bar{x}_o - \frac{(N_x - 1)^4 \tilde{\alpha}_\rho \bar{a}^3 \cos^2 c_N \pi \bar{x}_o}{16\bar{x}_o^4} \\ + \frac{(N_x - 1)^2 \tilde{\alpha}_\beta \bar{a}^3 \sin^2 c_N \pi \bar{x}_o}{4\bar{x}_o^2} = 0 \end{aligned} \quad (10)$$

where $c_N = 2(N_x - 2)/(N_x - 1)$ defines the average position between the outermost particle and its neighbor. We neglect scattering between non-immediately-adjacent particles, as scattering forces decrease rapidly with distance.

Single particle without scattering: As expected, the scattering forces represented by the last two terms in Equation (10) vanish for a single particle ($N_x = 1$), yielding an analytical solution (similar to those presented in

Refs. [19,20]):

$$\bar{x}_o(\tau) = \frac{1}{\pi} \cot^{-1} [\exp(2\pi\tau) \cot(\pi\bar{x}_o^i)] \quad (11)$$

where $\bar{x}_o^i$ is the initial position of the particle. This can be inverted analytically to predict the time to move a particle from one position to another:

$$\tau_F = \frac{1}{2\pi} \ln \left[\cot(\pi\bar{x}_o^f) \tan(\pi\bar{x}_o^i) \right] \quad (12)$$

where $\bar{x}_o^f$ is the final position of the particle.

Multiple particles with scattering: Figure 1(E) illustrates the focusing of an idealized array of particles for both plug flow and parabolic flow, where the transverse position is determined by numerical solution of Equation (10). For plug flow, indicated by open circles in Figure 1(E) and dashed lines in Figure 2(A,B), the fluid's axial velocity is constant across ($\bar{x}$), such that the axial position of particles is $\bar{z} = z/(v_s t_o)$. Thus, axial position is synonymous with time, since $\bar{z} = \tau$. For parabolic flows, the axial velocity of each particle is $\dot{\bar{z}}_i(\bar{x}_i) = v_s(1 - 4\bar{x}_i(t)^2)$, where v_s now represents the fluid velocity at the centerline. To compute the axial positions shown in Figure 2, this velocity profile is integrated with $\bar{x}_i(\tau)$ determined by the solution to Equation (10).

Figure 2(A) shows the focusing width, $\bar{w}_F$, as a function of time and as a function of the distance traveled. The distance traveled by the trailing particle is the inferred axial length of the focusing zone, $\tilde{L}_z = L_z/(v_s t_o)$, since it is assumed that motion in the x -direction arrests after exiting the focusing zone. For plug flow, the normalized length of the focusing zone is identical to the normalized time, i.e. $\tilde{L}_z = \tau_F$. For parabolic flow, the required focusing zone length is $\tilde{L}_z < \tau_F$ since particles near the wall have smaller velocities and therefore spend more time in the focusing zone. The distance implied by the difference between the $\tilde{L}_z$ and τ curves in Figure 2(A) represents the axial spread of the particles in parabolic flows. This is also illustrated in Figure 2(B), which depicts both the time spent in the focusing zone and the axial spread of the particles as a function of focusing zone length (for parabolic flows).

It is clear that the number of particles in the row affects the final focusing width, due to inter-particle scattering. However, the effect of scattering on the focusing time is rather minimal. Furthermore, the time required to reach the minimum focusing width decreases with increasing particle density, since particles travel shorter distances before they are arrested by scattering. As shown in Figure 2, the final focusing state for multiple particles occurs for $\tau_F \simeq 0.5$, or focusing zone lengths of $\tilde{L}_z \simeq 0.3$. The practical implication of this is simply that shorter

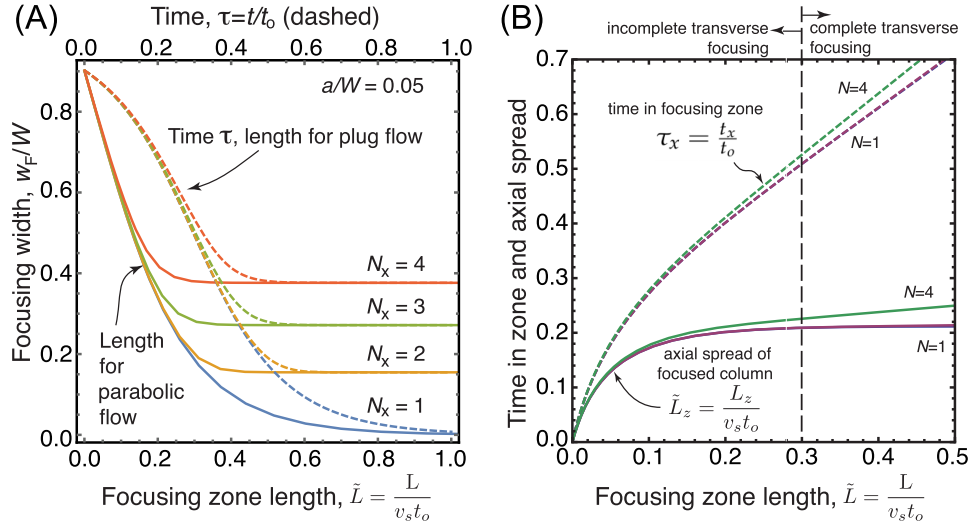


Figure 2. (A) Width of the focused particle array as a function of the traversed distance and time, for various initial numbers of particles in the wave direction (N_x). Solid and dashed lines represent values for parabolic and plug flows, respectively. (B) Particle focusing behavior for parabolic flows. Time in the focusing zone (dashed lines) and axial spread (solid lines) for arrays of 1 and 4 columns of initially equally-spaced particles in the transverse (wave) direction.

nozzles than those inferred from isolated particles are likely to be sufficient, since the time to reach the ‘scattered steady-state’ is smaller.

3.2. Time scale for axial aggregation due to attractive scattering forces

For particles aligned with the flow such that $\theta_{ij} = \pi/2$, scattering forces are attractive and lead to particle aggregation. Consider a column of N_z particles near the center of the channel (such that the $1/\tilde{d}_{ij}^4$ term dominates) and whose particles are in contact (Figure 1(D)). For a ‘probe’ particle a distance $\tilde{d}$ away from the first particle in the column, the governing equation is:

$$\dot{\tilde{d}}(\tau) = - \sum_{k=1}^{N_z} \frac{2\tilde{\alpha}_\rho \tilde{a}^3}{(\tilde{d}(\tau) + (k-1)2\tilde{a})^4} \quad (13)$$

The summation reflects the attractive force exerted on the probe particle by each particle in the column, i.e. the distance to the k^{th} particle in the assembled column is $\tilde{d} + 2\tilde{a}(k-1)$. This can be simplified by defining $\tau^* = \tilde{\alpha}_\rho \tau / (16\tilde{a}^2)$ and $\tilde{D} = \tilde{d}/(2\tilde{a})$, to describe spacing in terms of the number of particle diameters:

$$\tilde{D}'(\tau^*) = - \sum_{k=1}^{N_z} \frac{1}{(\tilde{D}(\tau^*) + (k-1))^4} \quad (14)$$

For $N_z = 2$, i.e. for two particles to form a couplet, this can be integrated in closed form. In this case, the time-to-contact $\tilde{D}(\tau^*) = 1$ in terms of the initial spacing

$\tilde{D}_o \equiv \tilde{D}(0)$ is:

$$\tau_A^* = \frac{1}{5} (\tilde{D}_o^5 - 1); \quad \tau_A = \frac{16\tilde{a}^2}{5\tilde{\alpha}_\rho} (\tilde{D}_o^5 - 1) \quad (15)$$

where τ_A is an estimate of the time to aggregate particles in the axial direction. The time to form a triplet (by combining a couplet with the probe particle) and so forth can be found by numerically integrating the full equation and solving for the time at which $\tilde{D}(\tau^*) = 1$, starting from $\tilde{D}_o$. Additional discussion of steady-state aggregation is provided in SI, with the conclusion that τ_A provides a reasonable time estimate for modest volume fractions.

3.3. Balancing focusing and axial assembly: ideal focusing zone size

For printing of lines with uniform, focused microstructures, the times required for focusing (transverse to flow) and aggregation (aligned with flow) should be balanced. If the time required for aggregation is too large, the printed line will have greater transverse particle density than axial particle density. Conversely, if the time for aggregation is too small, particle clusters may lead to axial variations in printed microstructures, or even worse, jamming. Here, we define an assembly parameter P_A as:

$$P_A \equiv \frac{\tau_A}{\tau_F} = \frac{32\pi\tilde{a}^2}{5f_V^{5/3}\tilde{\alpha}_\rho \ln [\cot(\pi\tilde{w}_F) \tan(\pi(1/2 - \tilde{a}))]} \quad (16)$$

where the relation for modest volume fractions, $\tilde{D}_o \simeq f_V^{-1/3} \gg 1$, has been substituted. This represents the

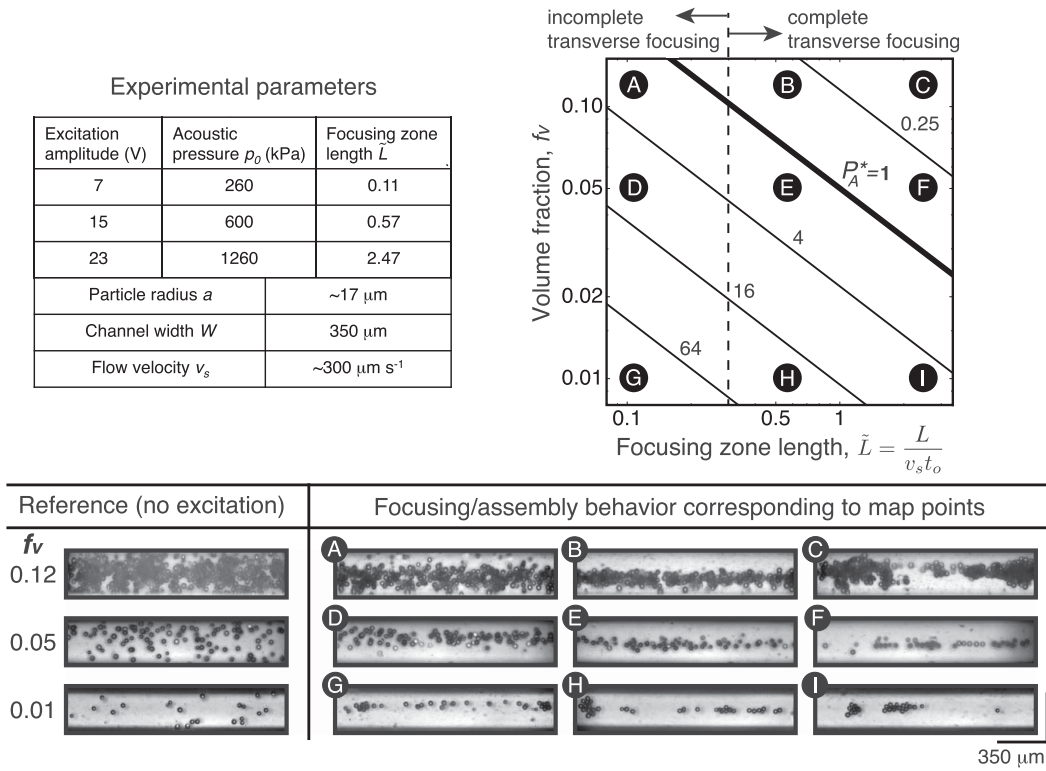


Figure 3. (Top) Experimental parameters and contour plot of focusing and assembly behavior of BaTiO₃ spheres in epoxy resin. The plot shows contours of the critical assembly parameter P_A^* as a function of the focusing zone length $\tilde{L}$. For $P_A^* > 1$, axial assembly occurs faster than focusing; the opposite is true for $P_A^* < 1$. $\tilde{L} \sim 0.3$ (dashed line) delineates the approximate transition between incomplete and complete focusing. (Bottom) Images at various excitation conditions and suspension concentrations, corresponding to map points (A)–(I), for a qualitative illustration of steady-state focusing and assembly behaviors.

balance of aggregation and focusing times: we can expect axial aggregation much faster than transverse focusing when $P_A < 1$.

Note that the dependence on focusing zone length and flow stream velocity is embedded in $\tilde{w}_F$, the desired width of the focusing zone. Supposing we want significant focusing ($\tilde{w}_F \ll 1$), then we require $\tilde{L}_z \approx \tau_F$; in this case, the expression in eqn. (16) is greatly simplified and we define a ‘critical’ assembly parameter P_A^* :

$$P_A^* \equiv \frac{16\tilde{a}^2}{5\tilde{\alpha}_\rho \tilde{L}_z f_V^{5/3}} \quad (17)$$

or $P_A^* \equiv \tau_z/\tilde{L}$. When $P_A^* \simeq 1$, the time scales for axial aggregation and transverse focusing are in balance. Substituting $\tilde{L}_z = L_s/(v_s t_o)$, an ‘ideal’ length of the focusing zone as a function of nozzle and solution properties can be approximated by:

$$\frac{L_z}{W} = \frac{9\eta v_s}{2\tilde{a}^2 \beta_f \phi p_o^2} \frac{16\tilde{a}^2}{5W\tilde{\alpha}_\rho f_V^{5/3}} \simeq 14 \frac{\eta v_s}{W\phi \beta_f p_o^2 \tilde{\alpha}_\rho f_V^{5/3}} \quad (18)$$

which interestingly, is independent of particle size and yields $L_z/W \simeq 15$ for flow rates equal to one channel

width per second (similar to the current experiments). This key result provides guidance as to the desired length of the nozzle as a function of the acoustic excitation parameters, channel geometry, transport speed, and initial particle density. We note that the spatial and temporal estimates provided here are in good qualitative agreement with observed particle packing but do not encompass factors such as particle functionalization or particle/solution chemistry that will affect short-range interactions (e.g. Debye screening). The estimates provided here address the assembly response over the meso- to macro-scale; however, it is likely that short-range interactions will affect assembly dynamics, and to a larger extent, the stability and packing arrangements of assembled particles. Furthermore, in scenarios with multiple nodes, the resolution of particle patterns will likely be a strong function of not only the particle and wavelength dimensions [13], but possibly also thermal and viscous boundary layer effects [21].

The boundaries defined by $P_A^* \simeq 1$ and $\tilde{L}_z \simeq 0.3$ delineate four regimes of behavior for each combination of focusing and aggregation. Qualitative experimental validation of these behaviors is shown in Figure 3, which provide images of steady-state particle focusing and

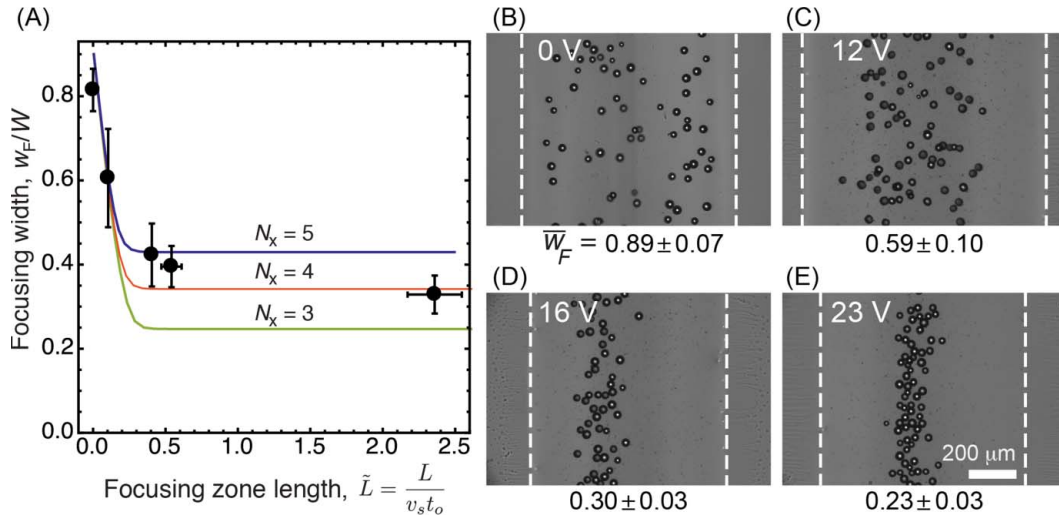


Figure 4. (A) Focusing width w_F vs. focusing zone length, for $34.5 \mu\text{m}$ BaTiO₃ particles in resin. (B,C,D,E) Printed, cured epoxy lines (edges indicated by dashes). w_F values represent standard deviation from the mean of ≥ 15 measurements for ((B) zero voltage, (C) 12 V, (D) 16 V, and (E) 23 V amplitudes.)

assembly behaviors in a print nozzle, corresponding to varying ink suspension and nozzle excitation parameters marked as (A)–(I) in the plot of volume fraction v_s vs. focusing zone lengths. Reference images of particles in suspension are provided in control conditions (same flow velocity but zero piezoelectric excitation). The contours represent values of the critical assembly parameter P_A^* . For sub-critical focusing zone lengths in Figure 3 particles become more focused to the channel centerline as volume fraction decreases (in (A), (D), (G), respectively). Conversely, for a given particle volume fraction, as the focusing zone length is increased beyond the critical value (e.g. from (A)–(C) or (G)–(I)), axial aggregation behavior dominates.

4. Printed microstructures

To validate the focusing zone model, the focusing width of BaTiO₃ spheres in a Newtonian epoxy resin (viscosity of $\sim 10 \text{ Pa s}$) [4], was measured for varying focusing zone lengths and is in good agreement with predictions in a parabolic flow profile (Figure 4(A)). Excitation voltage was varied to induce $\tilde{L} = 0$ to ~ 2.5 , (from no-focusing to well beyond steady-state focusing). In practice, the microstructure of printed lines will be a function of not only particle manipulation in-channel, but also exit effects at the nozzle including hydrodynamic and surface-wetting effects. In the above analysis, we consider a Newtonian ink matrix, although for printing viscous media it may be preferable to select a shear-thinning matrix to lower required flow pressure while retaining shape at rest, post-deposition [5]. Figure 4(B) shows that tunable focusing behavior is retained in cured lines printed from

a shear-thinning ink comprised of the same epoxy resin and particles, but with added silica filler and acetone [4].

For this particle volume fraction, the focused column reaches a steady-state width of ~ 3 – 5 particles, as suggested in Figure 4(A). This two-dimensional model assumes particles remain in-plane, whereas in the nozzle particles can stack, likely leading to better agreement with smaller N_x curves at larger $\tilde{L}$. In all cases, lines were printed at flow rates similar to those considered above (measured particle velocities $\sim 300 \mu\text{m s}^{-1}$).

5. Conclusions

In this work, we demonstrate that a ‘focusing zone’ model that considers time scales for acoustic focusing, axial assembly, and transport, paired with deposition, can be used to tailor the microstructure of two-component composite inks, with the ability to manipulate particle assembly both parallel and transverse to the print direction. The ability to control particle behavior within a single ink stream has implications for many technologies, for example, optimizing particle volume fractions for the casting of high-density green form ceramics, or the deposition of highly-spatially-controlled networks for local tuning of conductivity of heat or electricity (e.g. by toggling above or below a percolation limit). This framework can be used to guide the design of both the nozzle geometry and ink component properties, including critical assembly sizes for nano-to-meso-scale manufacturing. This technique can provide an additional level of particle control when coupled with other extrusion-based techniques, and presents a powerful complement to other hydrodynamic effects: for example, particle alignment in shear

flows [22] and in inertial flows [23]. It is also orthogonal to other physical field-based assembly methods (e.g. electrical and magnetic) and can thus be leveraged in a hybrid printing scheme for additional levels of particle control.

Disclosure statement

No potential conflict of interest was reported by the authors.

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