Extreme Mechanics Letters 5 (2015) 37–46

Contents lists available at ScienceDirect

Extreme Mechanics Letters

journal homepage: www.elsevier.com/locate/eml

Acoustic field controlled patterning and assembly of anisotropic particles

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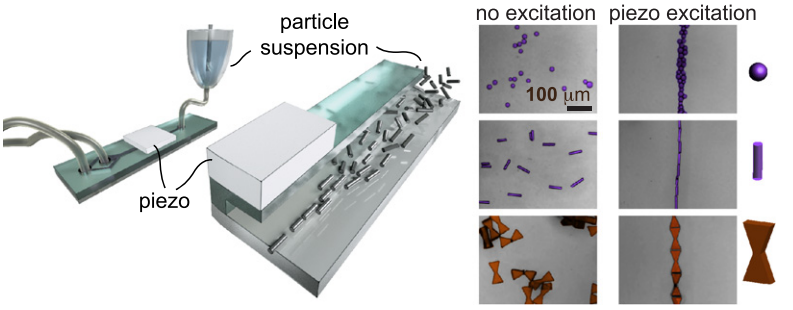
HIGHLIGHTS

- Acoustic waves are used to align and order anisotropic particles in suspensions.
- Microrods form isolated chains, parallel columns, and brick-and-mortar structures.
- Particle arrays are preserved in solid hydrogels and in solution *via* DNA-binding.
- Our acoustic assembly model accurately predicts column spacing and stability.
- This model is broadly applicable to various particle shapes and two-phase systems.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 8 July 2015
Received in revised form 11 September 2015
Accepted 13 September 2015
Available online 15 September 2015

Keywords:
Assembly
Anisotropy
Microparticles

ABSTRACT

Highly-organized particle assembly at the microscale offers the potential to transform fields ranging from medical diagnostics to materials-by-design. Here, we demonstrate tunable ordering and alignment of microscale particles, including ~10 × 50 μm glass rods and SU-8 brick and bowtie particles, *via* acoustic excitation, which produces columns with controlled spacing or highly-regular 'brick-and-mortar' packing. The method does not require surface functionalization and is broadly agnostic to colloidal chemistry or particle properties. We show that particle anisotropy promotes alignment and ordering even in the presence of flow, and can be used to increase the efficiency of particle delivery and sorting. Acoustically-assembled microstructures are preserved *via* photopolymerization of hydrogel matrices or binding with DNA 'mortar;' the latter provides sufficient stability

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http://dx.doi.org/10.1016/j.eml.2015.09.003

Microfluidics
Acoustic waves

and flexibility to fold ribbon-like structures in solution by tuning the direction of the acoustic waves. Significant insight into pattern spacing and stability is provided by models which incorporate primary acoustic focusing and secondary acoustic scattering forces. These results suggest that acoustic excitation may be a powerful complement to diffusion-controlled self-assembly and enable hierarchical fabrication; *e.g.*, micron-scale anisotropic crystals formed *via* self-assembly of smaller particles can be rapidly transported large distances, aligned and packed together to quickly assemble much larger objects.

1. Introduction

The assembly of ordered microstructures from colloids or colloidal suspensions is of interest for wide-ranging applications including novel materials [1], sensors [2], and diagnostics [3]. Many of these applications will require ordered microstructures with millimeter scale (or larger) domains. This is an inherent challenge for many self-assembly techniques, as thermodiffusive limits often inhibit the overall size-scale, time-scale, and stability of the assembly process [4]. While emerging techniques that exploit surface functionalization (*e.g.*, DNA interactions) continue to address the challenge of rapid assembly of stable structures at larger length-scales [5,6], such techniques still can require large time-scales to transport particles to growing domains. Thus, there remains a critical need for approaches that involve long-range assembly forces capable of transporting 'micro-domains' over large distances relative to their size and assimilate micro-domains into macro-domains.

Acoustic focusing presents a powerful opportunity in this regard; as it is relatively insensitive to solution chemistry and particle properties, it is a promising pathway to create hierarchical assembly techniques through combination with other assembly methods. For example, particles at one length-scale (*i.e.*, the nanoscale) could be assembled *via* chemical functionalization, and the resulting crystals (or disordered aggregates) could then be assembled at scales ranging from several microns to millimeters using acoustic forces which would not interfere with nanoscale assembly mechanisms. Alternatively, micron-scale particles could be formed *via* diffusion- or lithography-based techniques [1–3,7–9] and assembled downstream at the millimeter scale using acoustics. Since acoustic forces are active over large length-scales and provide rapid transport, continued assembly of microscale particles can be achieved over much larger length-scales and shorter time-scales (several seconds or less as shown here (*e.g.*, in Supplemental Video 2) and in other microchannel acoustic-focusing studies [10]) than those associated with diffusion.

In contrast to many existing chemical, magnetic, or rheological techniques that require either functionalized particles or a narrow range of particle/fluid properties, acoustic focusing (acoustophoresis) is label-free, relatively 'material agnostic,' and is effective for particles ranging from nanotubes [11] to hydrogel particles several hundred microns in size [12]. The utility of acoustics in controlling particle motion has been widely demonstrated in cell sorting applications (*e.g.*, cytometry [13]), which exploit the acoustic impedance mismatch between cells and the surrounding fluid as well as the scaling of acoustic forces with cell volume [14,15]. However, when the goal is close-packing with long-range ordering, the use of acoustophoresis to control particle agglomeration or patterning is far less developed. Notable examples include ordering of nanowires using surface acoustic waves [16], controlled rotation of various fiber types in aqueous [17,18] and polymeric [19,20] suspensions, the assembly of hydrogel particles in droplets [12], the patterning of spherical particles [21,22] and protein crystals [23] in two-dimensional patterns, and processing fibers in slurry for the manufacture of paper [24].

Here, we demonstrate that acoustically-driven assembly can be exploited to achieve *tunable* patterning of anisotropic particles. Fig. 1 shows the device concept and optical micrographs of assembled columns of glass microrods and spheres, as well as micro-fabricated bricks and bowties. Figs. 2 and 3 illustrate that stable columns of aligned particles and 2D arrays can also be achieved and preserved in the presence of flow (see Supplemental Videos 1–3), which has important implications for developing novel printheads to deposit ordered microstructures. ('Pearl-chains' [25] and aligned bands of spheres in acoustic fields have been previously observed and modeled [26–28]; our experiments revealed that spherical particle ordering is far less stable in the presence of flow than that of anisotropic particles).

As detailed below, both the formation and the stability of the structures seen in Figs. 1–3 are controlled by the competition of the primary focusing force that drives particles that are denser than the fluid to the pressure node of the standing wave, and secondary scattering forces [14,15,25–30]. These secondary scattering forces are attractive when the interacting particles are aligned parallel to the wave, causing particles to snap end-to-end in a given column (Fig. 1(c)), and repulsive when particles are aligned perpendicular to the wave propagation direction, keeping multiple assembled columns from collapsing into a single column (Figs. 2 and 5). At excitation energies large enough to overcome repulsive scattering forces, parallel columns collapse into highly-ordered arrays (Fig. 3). It should be emphasized that the particles have not been functionalized; further, average column spacing d is insensitive to anionic and non-ionic surfactants in solution (Supplemental Figure S2). Thus, acoustophoresis and surface functionalization are highly-complementary strategies that can be combined to increase pattern stability and/or enhance