
Supplementary information

**Optofluidic three-dimensional
microfabrication and nanofabrication**

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Supplementary Materials for

Optofluidic three-dimensional microfabrication and nanofabrication

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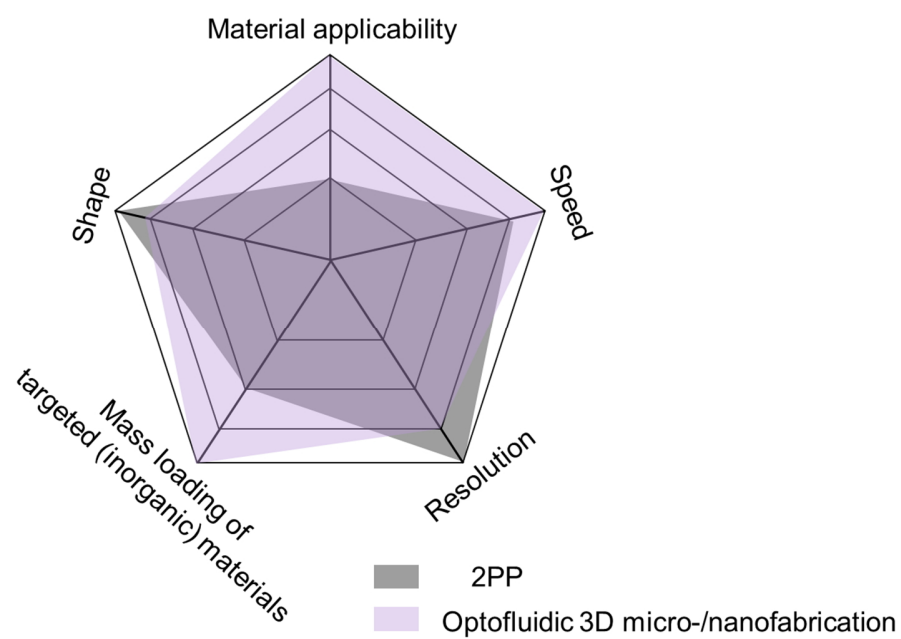
Other Supplementary Materials for this manuscript include the following:

Supplementary Videos 1 to 17

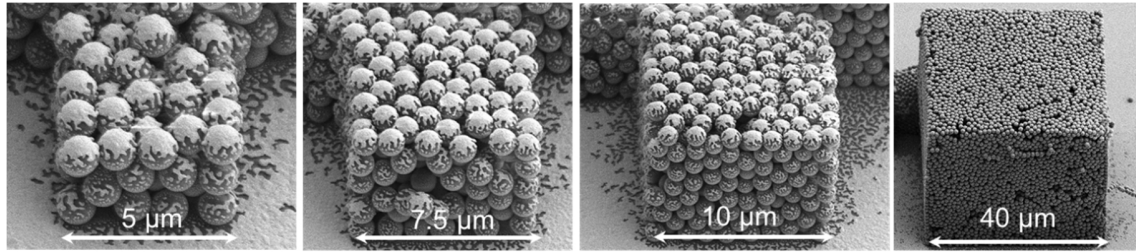
Supplementary Discussion 1

To successfully fabricate diverse 3D microstructures, it is crucial to ensure the continuous accumulation of micro-/nanomaterials within a 2PP-printed hollow template. As discussed in the assembly mechanism section, achieving the condition $\Delta U_{total} < 0$ requires the inter-particle attractive interaction to be slightly stronger than the particle-fluid interaction. Under this condition, a particle cluster forms at the laser-scanned area and is continuously transported into the hollow template. However, when the inter-particle interaction significantly exceeds the particle-fluid interaction, the resulting cluster can clog the template's opening, preventing further particle transport and disrupting the 3D assembly process. This issue is particularly pronounced for micrometer-sized particles, which have a larger volume, making it difficult to enter and fill the template, and smaller particles with diameters near or below 100 nm (Supplementary Video 15), which experience strong adhesion interactions, leading to aggregation and blocking.

To successfully assemble such materials, two strategies can be employed: (1) increasing the opening size, and (2) redesigning the 3D template with a suitable hole configuration to prevent clogging. For instance, as illustrated in Supplementary Figs. 16, 17, microtemplates with different opening sizes can be employed for the assembly of SiO₂ particles of different sizes. Additionally, as shown in Supplementary Figs. 10, 18, Supplementary Video 9, and Figs. 4s–z of the main text, microcubes with multiple holes were designed to facilitate the 3D assembly of various micro-/nanomaterials while preventing clogging. These modifications ensure that particle clusters can be efficiently transported into the template, enabling the successful formation of volumetric 3D structures.

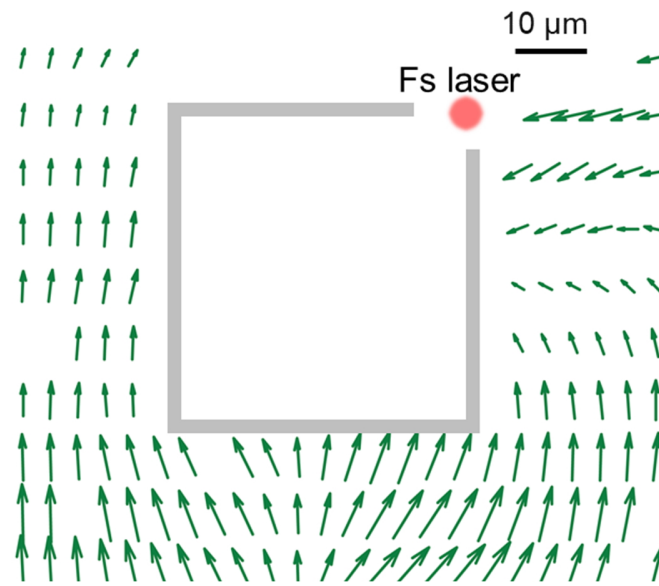


Supplementary Fig. 1 Radar comparison of the common 2PP technique and our optofluidic 3D micro-/nanofabrication approach.

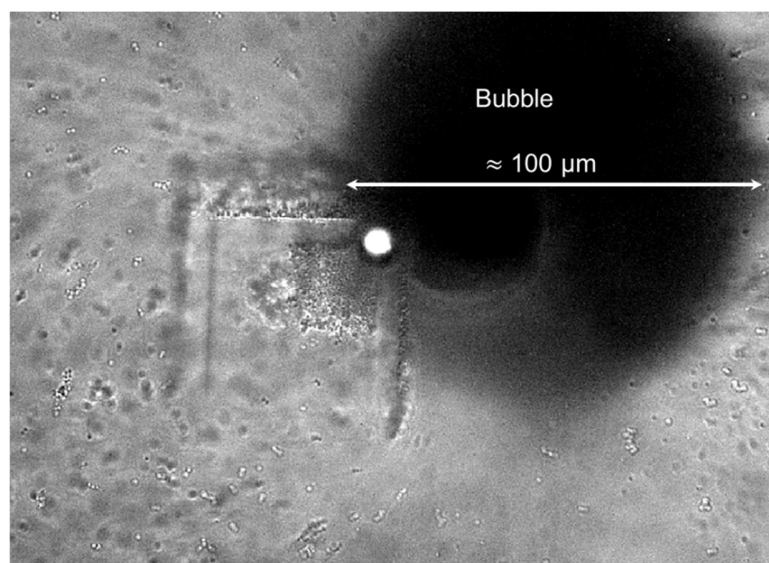


Supplementary Fig. 2 SEM images of SiO₂ colloidal-assembled microcubes of different sizes.

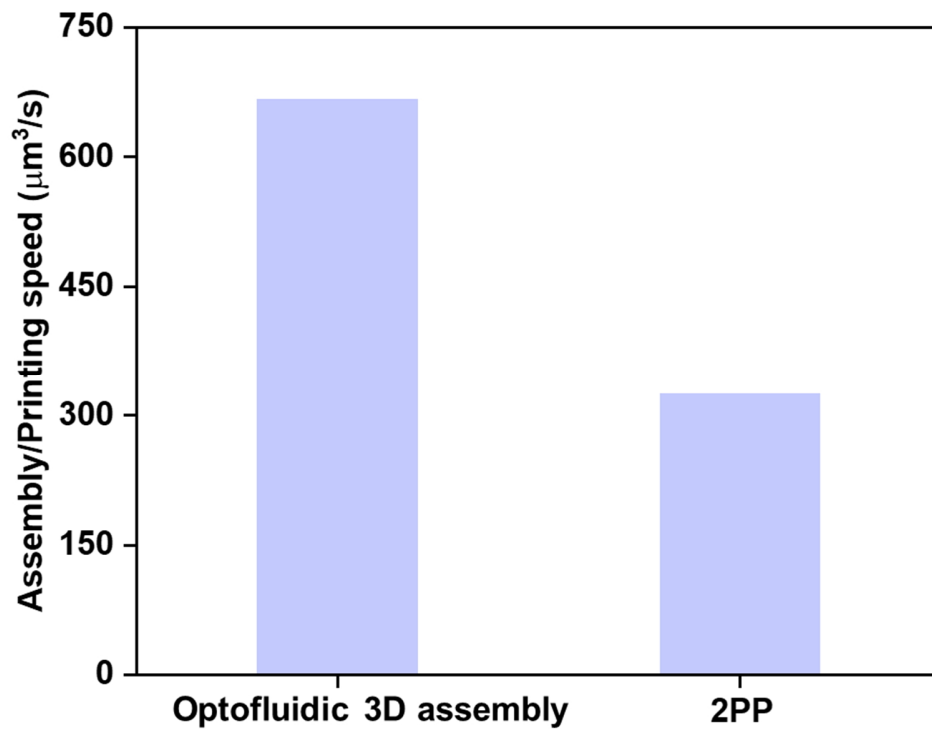
The white layer on the particle surface is Au sputtered for SEM characterization. Microstructures with feature sizes below 100 μm can be readily fabricated. However, for structures larger than 100 μm, a much stronger flow field than that employed in this work is required to provide sufficient driving force for continuous particle transport into the template and to ensure complete filling of the hollow cavity.



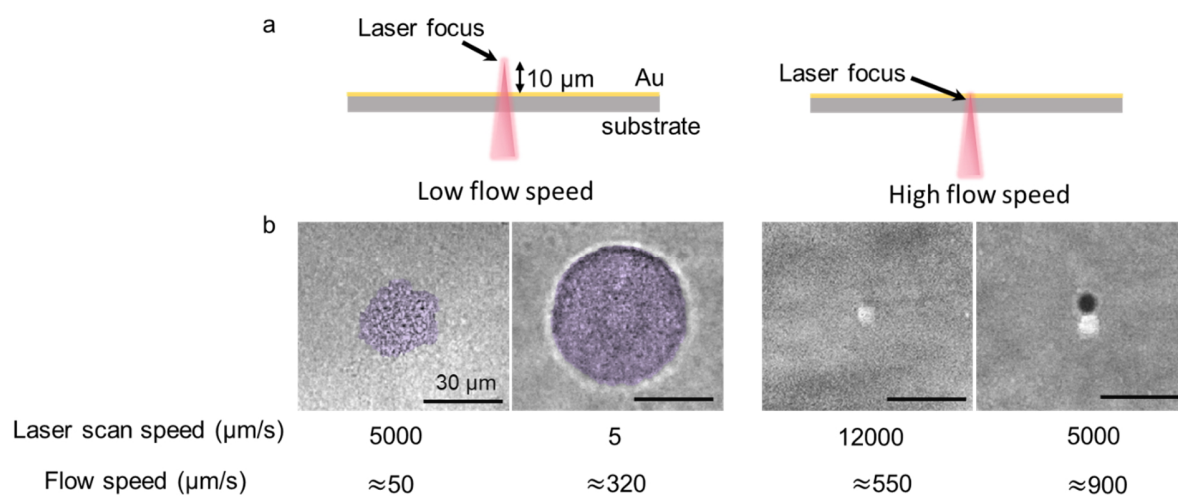
Supplementary Fig. 3 The Particle Image Velocimetry (PIV) result of the fluid flow field surrounding a hollow microcube under the laser illumination, showing the fluid flowing toward the inside of the cube.



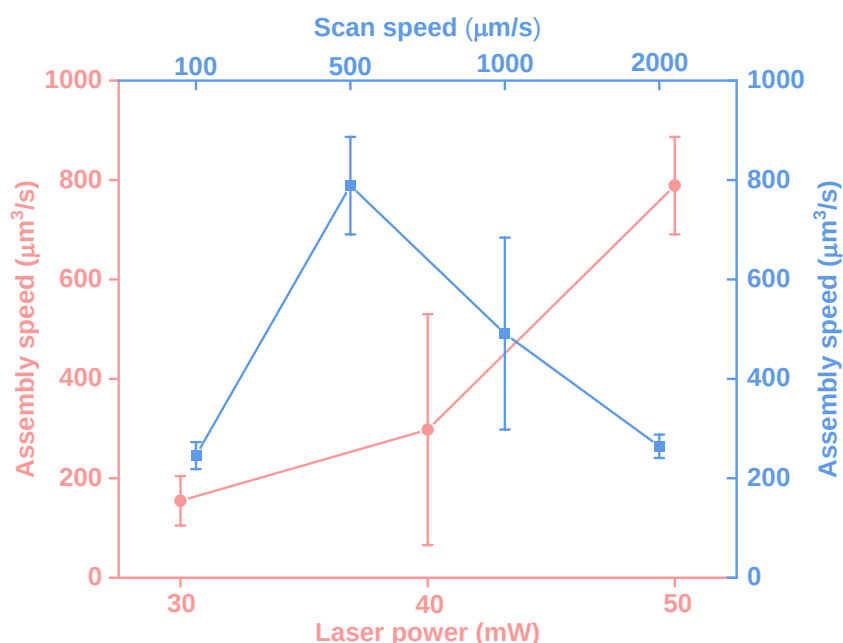
Supplementary Fig. 4 Optical image of bubble formation during the assembly of SiO₂ particles in immersion oil. The bubble appears at a laser power of 50 mW (maximum output of the instrument) with a scanning speed of 300 μm/s.



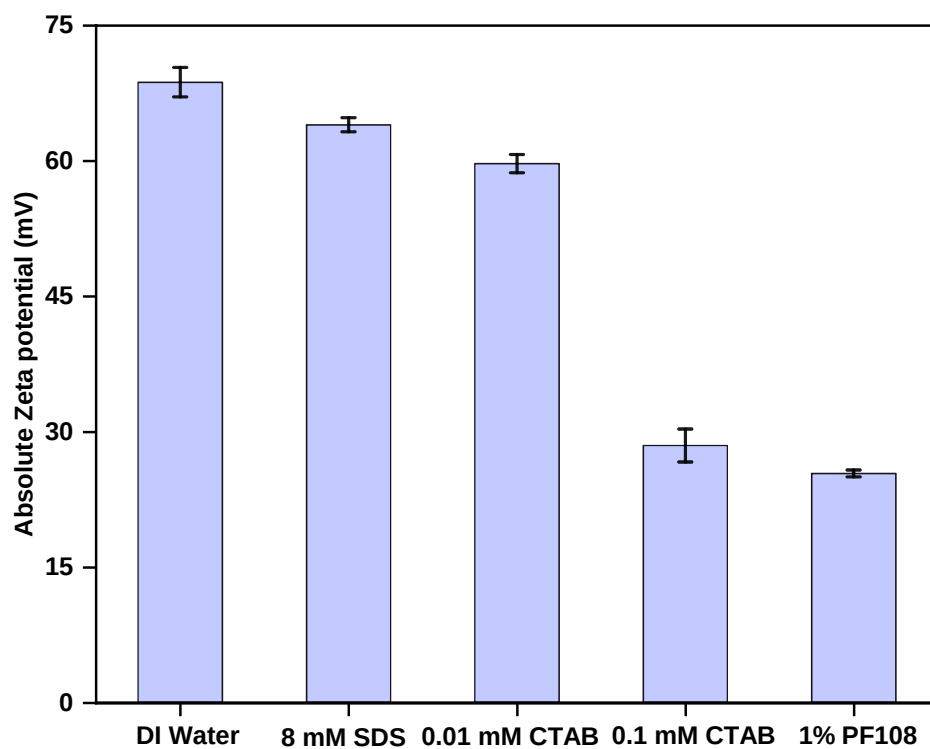
Supplementary Fig. 5 Comparison of the assembly/printing speed between our 3D OFCA approach and the 2PP technique. The scan speed in 2PP was set as 10,000 $\mu\text{m}/\text{s}$.



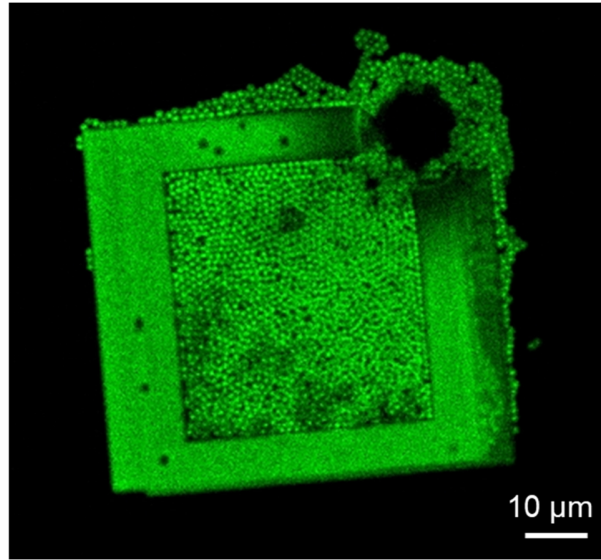
Supplementary Fig. 6 Clustering of SiO₂ particles (150 nm) at different flow speeds. a, Schematic illustration demonstrating how flow speed can be tuned by adjusting the laser focus position. Since most of the fs laser energy is concentrated at its focal point, positioning the laser focus on the surface of the sputtered Au layer results in a much faster flow speed. **b,** Optical images showing SiO₂ particle clustering after 60 s of laser treatment at 50 mW laser power in a 1 M NaCl solution.



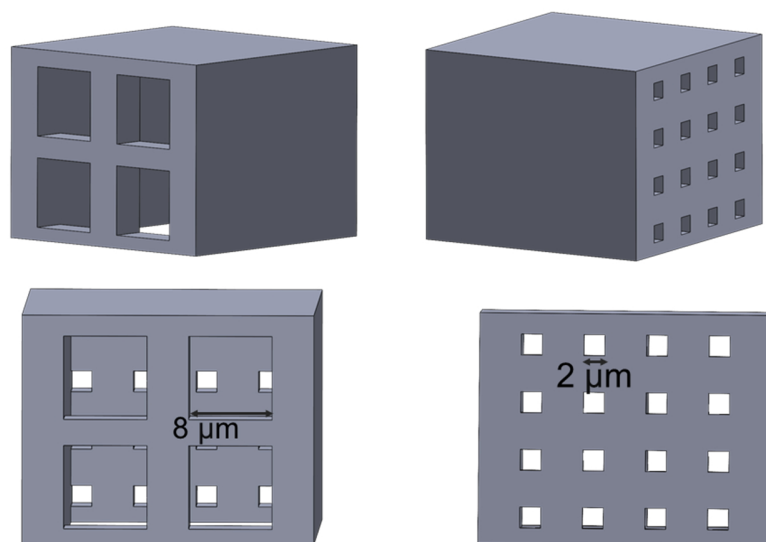
Supplementary Fig. 7 Assembly speed of SiO₂ particles as a function of the laser power (red spheres) and scan speed (blue squares). The assembly experiments were conducted in immersion oil, with SiO₂ particle dispersions at a concentration of around 0.5 wt%. The laser power was set to 50 mW for the scan speed series. The scan speed was set at 500 μm/s for the laser power series. It is also noted that a lower laser power of 20 mW (with a scan speed of 500 μm/s) would lead to an extremely weak flow field with a flow speed of around 40 μm/s, which cannot provide sufficient driving force to ensure particles fill up the entire template (Supplementary Video 4).



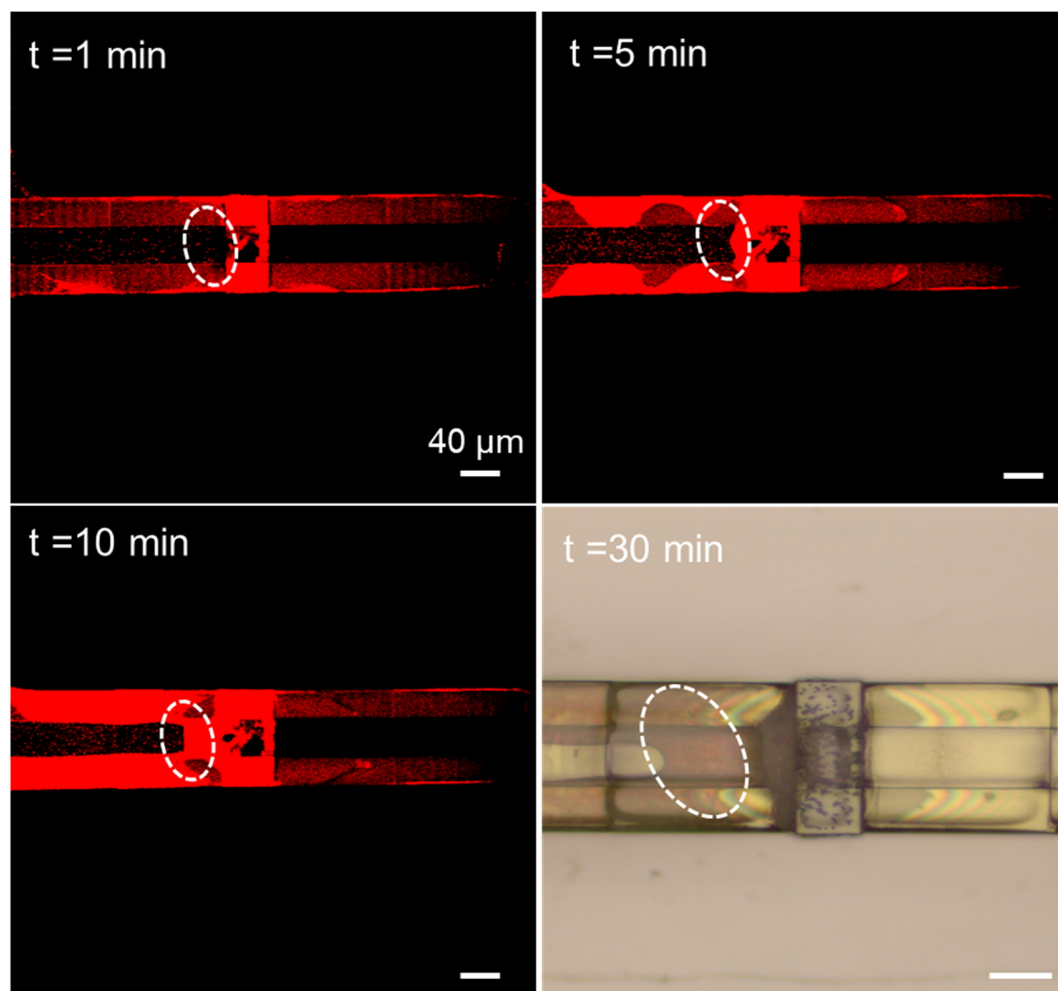
Supplementary Fig. 8 Zeta potential of SiO₂ colloidal particles (1 μm in diameter) in various solutions.



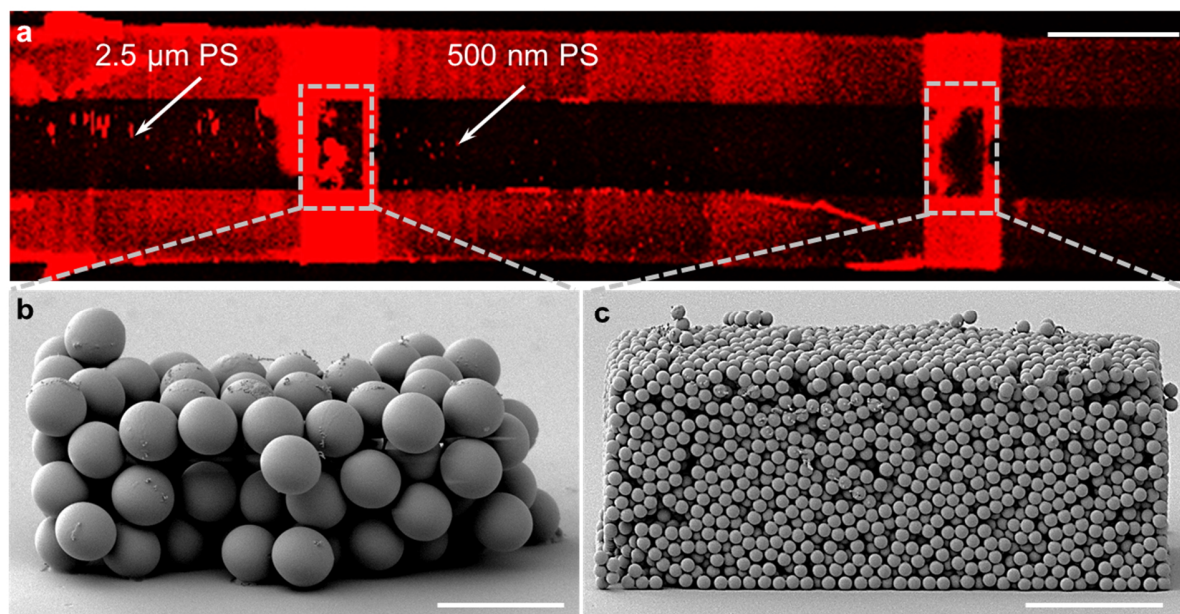
Supplementary Fig. 9 Bottom-view fluorescent image showing 1 μm SiO₂ particles assembled within a microcube.



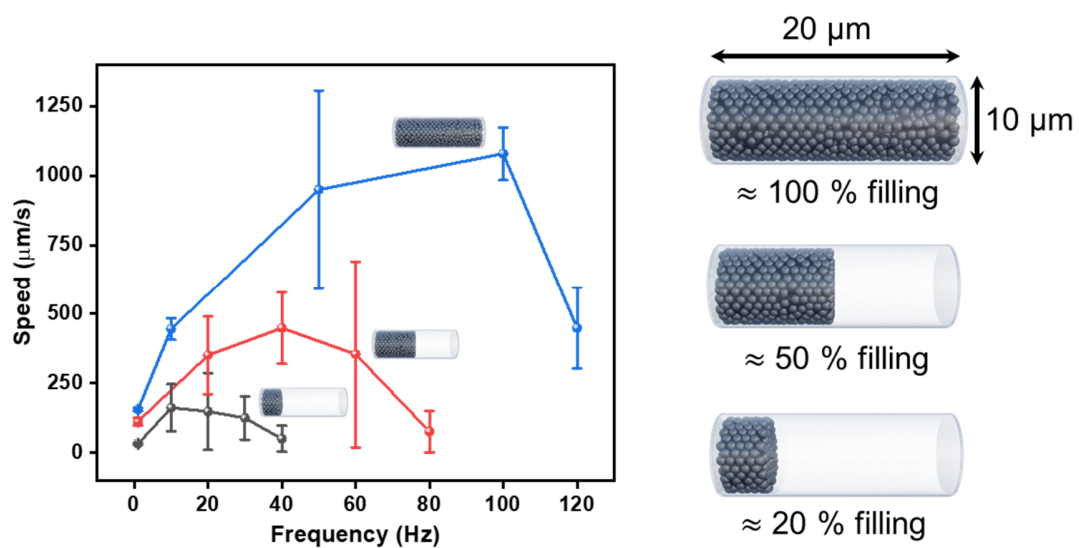
Supplementary Fig. 10 3D model of a cube template for 3D assembly of various materials, including TiO_2 NWs, diamond NPs, Fe_3O_4 NPs, WO_3 NWs, Al_2O_3 NWs, and Ag NPs.



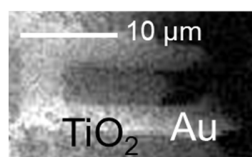
Supplementary Fig. 11 Fluorescent and optical images showing the enrichment of PS particles within the microfluidic chip.



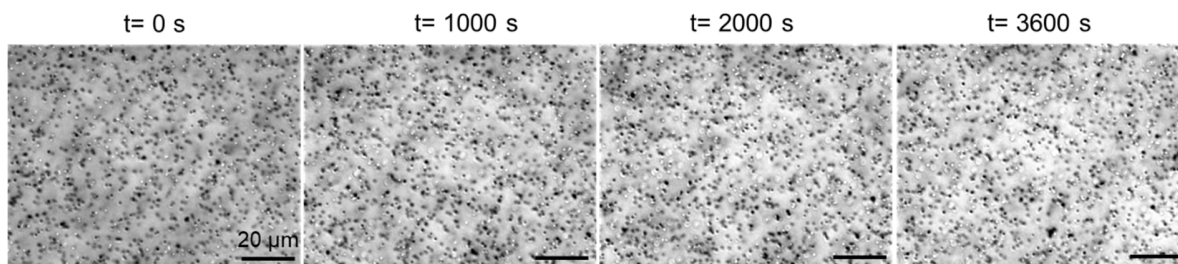
Supplementary Fig. 12 Size-selective sorting enabled by a microfluidic chip with two concatenated microvalves. **a**, Fluorescent image showing the size-selective sieving of 2.5 μm and 500 nm polystyrene (PS) colloidal particles. **b**, **c**, SEM images of microvalves composed of 5 μm (**b**) and 1 μm (**c**) SiO₂ particles. Scale bars are 50 μm in **a** and 10 μm in (**b**, **c**). This microfluidic chip enables the sequential separation of mixed micro-/nanoparticles, e.g., 2.5 μm and 500 nm PS particles. The 500 nm PS particles successfully pass through the first valve (5 μm SiO₂ assembly) but are retained by the second microvalve (1 μm SiO₂ assembly). In contrast, 2.5 μm PS spheres are rejected by the first valve (5 μm SiO₂ assembly).



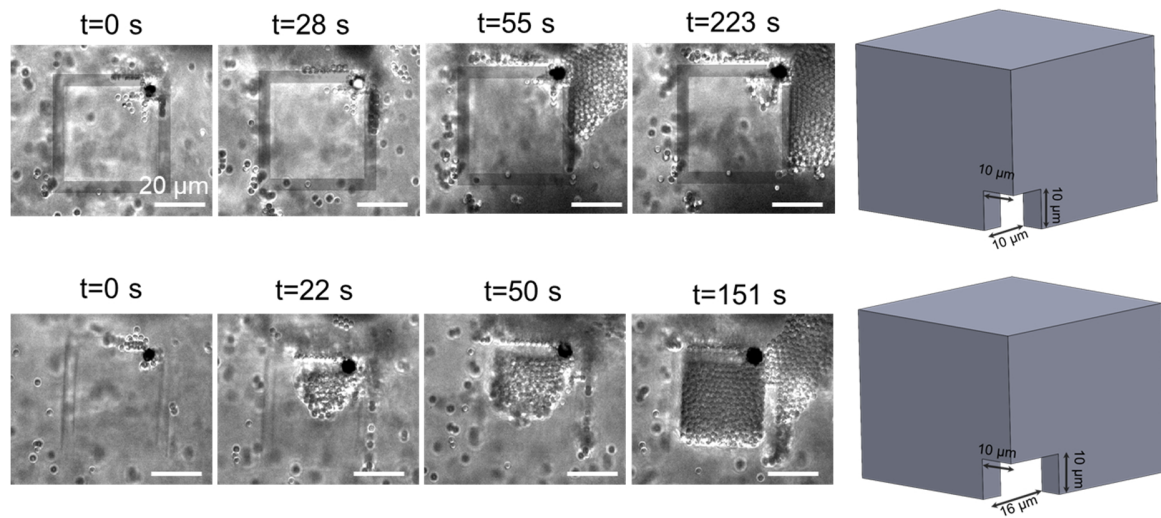
Supplementary Fig. 13 Averaged speeds (left) and schematic (right) of cylinder microrobots assembled with various contents of Fe_3O_4 NP. The magnetic strength is 10 mT.



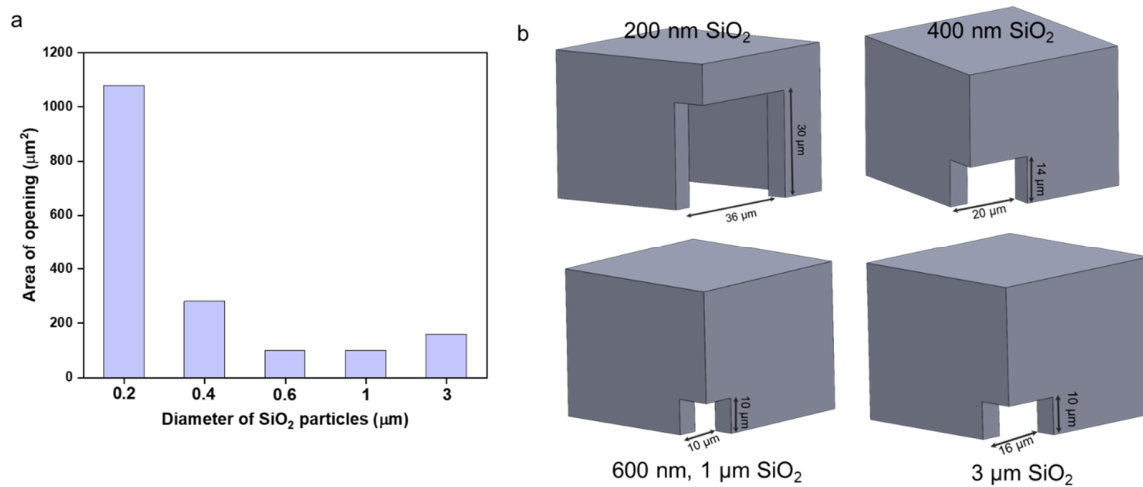
Supplementary Fig. 14 Optical image of a TiO₂-Au heterojunction within a cylinder microtemplate.



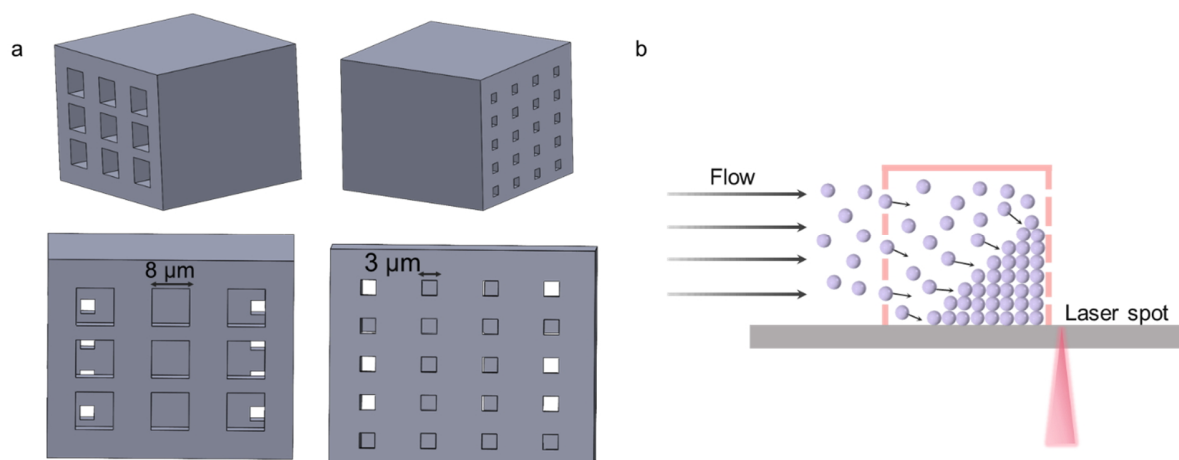
Supplementary Fig. 15 Time-lapse optical images showing that SiO₂ particles remain dispersed in immersion oil for at least 1 h. The highly viscous solvent (around 500 mPs·s) suppresses Brownian motion, thereby preventing particle agglomeration.



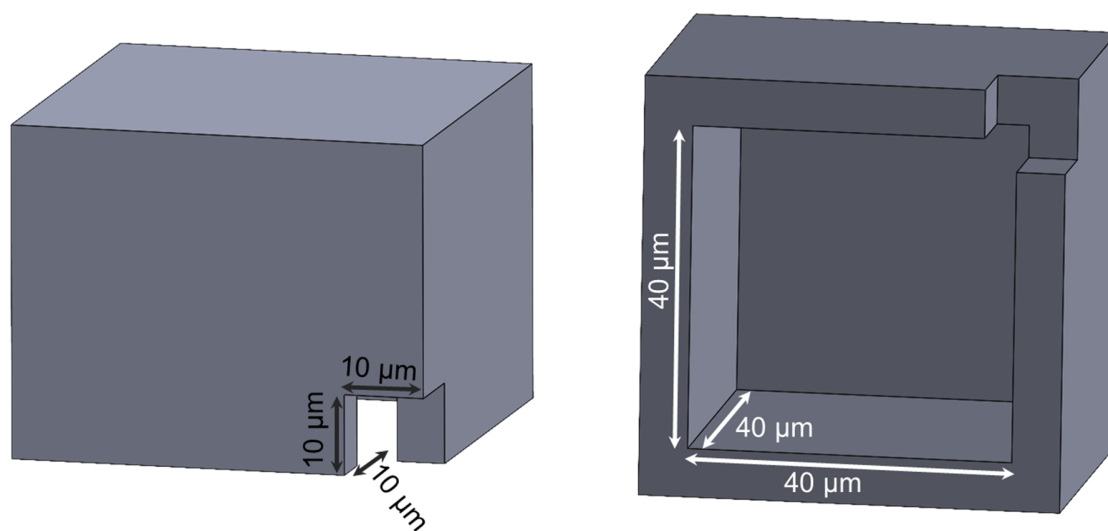
Supplementary Fig. 16 Time-lapse optical images of the assembly process of 3 μm SiO_2 nanoparticles into hollow microcubes with varying opening sizes.



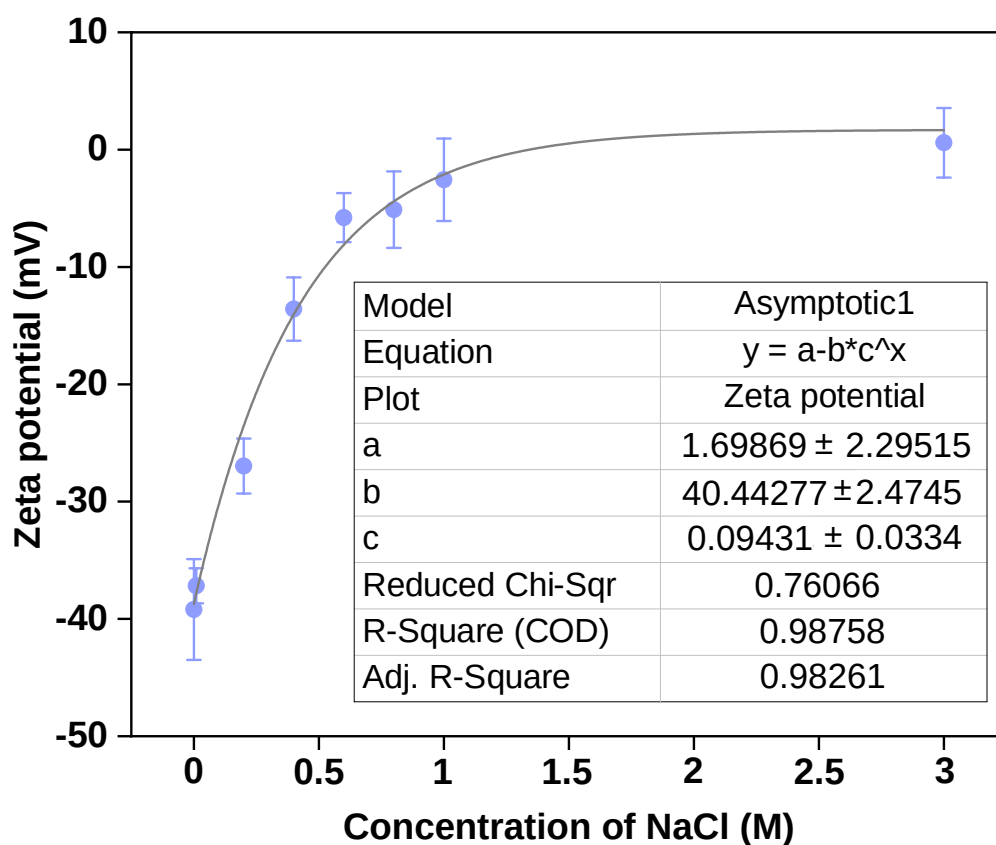
Supplementary Fig. 17 a, b, Critical opening sizes (a) for SiO₂ particles of different diameters during assembly in immersion oil and the corresponding 3D models (b).



Supplementary Fig. 18 Assembly of SiO₂ particles in a microcube template with multiple openings. **a**, 3D models of the microcube templates, featuring holes of two different sizes (8 μm and 3 μm) distributed on opposite sides of the cube. **b**, Schematic illustration of assembly process. When a laser beam is applied to the side with 3 μm holes (right), a directional fluid flow is generated from right to left, propelling dispersed SiO₂ particles through the 8 μm openings into the cube. The particles are then blocked by the smaller 3 μm holes, leading to continuous accumulation inside the cube until the template is fully filled.



Supplementary Fig. 19 3D model of a hollow cube template used in the simulation.



Supplementary Fig. 20 Zeta potential of SiO₂ particles dispersed in solutions of various NaCl concentrations and their fitting results.

Supplementary Table 1 Comparison among different assembly technologies.

Assembly techniques	Mechanism	Resolution	Material applicability	Structural complexity	Number of building blocks in assembled structures	Typical time scale or key parameters for assembly	Ref.
Capillary assembly	Capillary flows induced by the evaporation of colloidal suspensions transport dispersed particles toward the triple-phase contact line, guiding their assembly at predetermined substrate locations. 3D assembly configurations can be achieved with the assistance of a template.	Sub-1 μm	Accommodating a variety of micro-/nanoparticles, including polymer, metal, metal oxide, MOF, COF, etc.	2.5D or limited 3D geometries, e.g., cylinder and star-shaped Au NP superstructure	around 10^2 - 10^4	Upon evaporation, the receding speed of the triple-phase contact line is usually below 5 $\mu\text{m/s}$ to ensure effective particle filling of the template.	51,52
Interfacial assembly	Particles spontaneously adsorb at the liquid-liquid interface of two immiscible liquids (e.g., water and oil), where they assemble into organized interfacial patterns.	Sub-10 nm		2D lattice patterns or 3D structures are limited to spherical geometries.		Minutes	53-56
Electric field assembly	Charged metallic nanoparticles are directed onto the substrate using an electric field or a combination of electric and flow fields. By precisely moving the substrate in 3D space, complex 3D micro-/nanostructures can then be fabricated through the layer-by-layer assembly of particles.	Sub-100 nm	Metals, alloys	Free-standing complex 3D structures, e.g., helical or flower-shaped micro-/nanostructures.	/	To ensure the continuous growth of assembled structures, the moving speed of the substrate is usually below 100 nm s^{-1} .	57,58
Magnetic field assembly	Magnetic particles self-assemble into ordered superstructures through dipole-dipole interaction under an external magnetic field. By tuning the applied field, different assembly behaviours can be induced.		Magnetic particles	1D chain, 2D lattice, or limited 3D configurations, e.g., twisted ribbon.	around 10^1 - 10^3	Milliseconds to seconds	59-61
DNA-based assembly	DNA strands and DNA-functionalized colloidal particles self-assemble through the deterministic base-pairing interactions (A-T, C-G), analogous to the modular assembly of LEGO bricks.	Sub-10 nm (assembly of DNA molecules)	DNA molecules and DNA-functionalized colloidal particles	Complex DNA 3D nanostructures, such as monolith, railed bridge, and icosahedron. Colloidal superstructures with simple 3D configuration, for instance, analogous to AB-type molecules.	around 10^1 - 10^2	Hours to days	62-64
Polyphenol-based assembly	Polyphenol-functionalized particles first assemble through interfacial molecular interactions, and the resulting configurations are then stabilized via metal-ligand coordination.	around 1 μm	Polyphenol-functionalized colloidal particles	Limited 3D geometries, for instance, spherical and core-satellite configurations.	around 10^2 - 10^3	Seconds.	65

Optical assembly	Optical tweezer	An optical gradient force from a tightly focused laser beam is used to trap individual colloidal particles. By scanning the laser beam, the trapped particle can be transported and positioned at desired locations, enabling one-by-one assembly.	Single-particle resolution (Sub-1 μm)	Colloidal particles with appropriate size (e.g., 0.1-10 μm in diameter) ^{66,67} .	Limited 3D geometries, e.g., a cube-shaped superstructure.	around 10 ¹ -10 ³	The assembly efficiency is estimated to be around 10 ⁰ -10 ¹ particles/min.	68,69
	Thermophoretic assembly	Suspended particles in solution undergo directed thermophoretic migration toward either the hot or cold region when a temperature gradient is generated by laser irradiation. By tuning particle surface charge, solvent composition, and environmental temperature, particles can be trapped at the hot region (laser spot), enabling thermophoretic tweezer for assembly.		Colloidal particles with appropriate surface charges.	2D micro-/nanopatterns. 3D structures with fairly limited geometries, e.g., out-of-plane 1D chain made of 500 nm PS sphere ²⁰ .		The migration speed of particles is normally below 100 μm/s . Otherwise, the drag force exerted by the fluid will release the trapped particle from the laser beam, hindering the assembly. The assembly efficiency is estimated to be around 10 ¹ -10 ² particles/min.	70,71
	Thermoelectric assembly	In an electrolyte solution, various ions migrate at varying velocities and directions under a temperature gradient via thermophoresis. This imbalance generates a spatial distribution of ion species, giving rise to a thermoelectric field in the solution. The resulting electric field drives the directed migration of charged colloidal particles. By tuning experimental parameters, such as ion type and particle surface charge, colloidal particles can be selectively trapped at the laser spot, enabling an opto-thermoelectric tweezer for particle assembly.						72,73
	Convective flow-based assembly	A thermal gradient induces variations in density, pressure, and surface tension within the solution, generating strong convective flows. Suspended particles are carried along the streamlines, and ultimately assemble at the laser spot through van der Waals interactions.	Sub-1 μm	Broader material applicability irrespective of particle size, shape, or surface charge—for example, diverse micro-/nanoparticles and nanowires, including metal, metal oxide, carbon materials, and quantum dots.	2D micro-/nanostructures	around 10 ² -10 ⁴	The assembly efficiency is estimated to be around 10 ² -10 ³ particles/min.	74,75
	Optofluidic 3D assembly (our work)	By harnessing strong convective flows induced by steep temperature gradients within a confined 3D space, various micro-/nanoparticles can be volumetrically assembled inside micro-confinements, yielding freeform 3D architectures entirely composed of micro-/nanoparticles.			Highly freeform 3D architectures, e.g., a dangling croissant-shaped superstructure	around 10 ² -10 ⁵	Assembly can occur at flow speeds of up to several mm/s with an estimated assembly efficiency of up to ≈10 ⁵ particles/min.	

Supplementary Table 2 Comparison among different 3D micro-/nanofabrication techniques.

3D micro-/nanofabrication techniques	Mechanism	Resolution	Material applicability	Structural complexity	Ref.
Physical vapor deposition (PVD)	By evaporating a target source and condensing the vaporized species onto a substrate, 3D structures can be fabricated using glancing angle deposition (GLAD) through controlled “shadowing effect”.	Sub-100 nm	Metals, metal oxides, ceramics, semiconductors, and others.	Limited 3D geometries, e.g., pillars, helical structures, and nanotubes.	76,77
Lithographic methods (e.g., UV lithography, e-beam lithography)	A pattern is generated from a mask by selectively exposing a photoresist film, which is then transferred to the targeted substrate. 3D structures can be created either through repeated exposures or by inducing 3D photoresist exposure via spatially modulated light pattern.		Polymers, metals, metal oxides, semiconductors, and others.		78-80
Mechanically guided 3D assembly	2D structures can be transformed into 3D configurations through mechanical deformation process such as buckling, folding, and rolling.				81,82
Focused electron/ion beam-induced deposition (FEBID/FIBID)	Focused electron or ion beams locally decompose organometallic precursors, enabling the maskless direct writing of 3D nanostructures.	Sub-10 nm	Applicable materials are limited to metals, e.g., Au, Pt	Highly freeform 3D structures, e.g., tetragonal-bipyramid, icosahedron	83
Two-photon polymerization (2PP)	Nonlinear two-photon absorption (2PA) of a photoinitiator generates free radicals, triggering the chemical cross-linking of monomer molecules (photoresists) at the focal point of a tightly focused femtosecond laser beam. Scanning the laser beam in 3D space enables the direct laser writing (DLW) of 3D micro-/nanostructures.	Sub-1 μm	Printable materials can be expanded to metal, metal oxides, glass, quantum dots and more through the development of advanced photoresists with tailored chemistries. However, a general route applicable across diverse material systems remains challenging, and the mass loading of targeted materials in printed structures is typically limited to an upper bound of around 90%.	Highly freeform 3D micro-/nanoarchitectures, e.g., 3D lattice microstructures.	84-95
	Nanomaterials can be deposited or adsorbed onto 2PP-printed polymer templates through chemical coating, PVD, or weak interaction-based absorption (e.g., electrostatic interaction, hydrogen bonding, and van der Waals force).		Broad material applicability, including metal, metal oxides, carbon materials, QDs, ceramics, and others. However, the resulting structures are not volumetric. Desired materials are merely deposited onto the template surface with low mass loadings (< 60%).		96
Optofluidic 3D micro-/nanofabrication (our work)	By harnessing the strong convective flow induced by fs laser heating within a confined 3D space, various micro-/nanoparticles can be volumetrically assembled inside the confinement, enabling the creation of freeform 3D architectures entirely composed of micro-/nanoparticles.			Broader material applicability irrespective of their size, shape, or surface charge—for example, diverse micro-/nanoparticles and nanowires, including metal, metal oxide, carbon materials, and quantum dots. The fabricated architectures are fully composed of the targeted materials, with mass loading approaching ≈100%.	Highly freeform 3D volumetric structures, e.g., a dangling croissant-shaped SiO ₂ superstructure.

Supplementary Table 3 The physicochemical properties of micro-/nanomaterials used in this work.

Materials	Size	Concentration used for assembly	Solvent used for assembly
SiO ₂	150 nm - 10 μm	1 wt% for 150 nm SiO ₂ , and 0.5 wt% for others	Immersion oil, mineral oil, oleic acid, or 1 mM CTAB
WO ₃ NWs	50 nm × 10 μm	around 2 mg/mL	Mineral oil
Ag NPs	<100 nm		Mineral oil
Al ₂ O ₃ NWs	2-6 nm × 200-400 nm		Mineral oil
Fe ₃ O ₄ NPs	around 200 nm		Mineral oil
TiO ₂ NWs	100 nm × 10 μm		Mineral oil
diamond	around 10 nm		Immersion oil
CdTe Quantum dots	/		1 mM CTAB
Au NPs	around 50 nm		50 mM CTAB
Pt NPs	around 50 nm		50 mM CTAB
TiO ₂ NPs	around 100 nm		DMSO
Fe ₃ O ₄ NPs	around 20 nm		10 mM CTAB
SiO ₂ mixtures	600 nm, 1 μm	1 wt%	Immersion oil

Supplementary Table 4 Physical parameters used in the theoretical analysis.

Parameter name	Symbol	Unit	Value
Viscosity	μ	Pa.s	0.001
Radius of SiO ₂ particles	R	m	1.5×10^{-7}
Velocity	v	m/s	0 - 1×10^{-3} (vertical axis in Fig. 2e in the main text)
Dielectric permittivity of vacuum	ϵ_0	C/V/m	8.85×10^{-12}
Dielectric permittivity of water	ϵ_l	C/V/m	78.3
Temperature	T	K	298.15 (25 °C)
Boltzmann constant	K_B	J/K	1.38×10^{-23}
Charge of an electron	e	C	1.602×10^{-19}
Avogadro's number	N_A	mol ⁻¹	6.022×10^{23}
Ionic strength	I	mol/m ³	$0.5 \times 2 \times c$ (c: concentration of NaCl, horizontal axis in Fig. 2e in the main text)
The inverse of the Debye length	κ	m ⁻¹	$\kappa^{-1} = \sqrt{\frac{\epsilon_0 \epsilon_l K_B T}{2 N_A e^2 I}}$
Zeta potential	ζ	mV	$\zeta = 1.7 - 40.4 \times 0.094^c$ (Obtained by fitting the Zeta potential in Supplementary Fig. 20)

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