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(54) **METHODS FOR CHARGE-TITRATING PARTICLE ASSEMBLY, AND STRUCTURES PRODUCED THEREFROM**

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(57) **ABSTRACT**

Methods to fabricate tightly packed arrays of nanoparticles are disclosed, without relying on organic ligands or a substrate. In some variations, a method of assembling particles into an array comprises dispersing particles in a liquid solution; introducing a triggerable pH-control substance capable of generating an acid or a base; and triggering the pH-control substance to generate an acid or a base within the liquid solution, thereby titrating the pH. During pH titration, the particle-surface charge magnitude is reduced, causing the particles to assemble into a particle array. Other variations provide a device for assembling particles into particle arrays, comprising a droplet-generating microfluidic region; a first-fluid inlet port; a second-fluid inlet port; a reaction microfluidic region, disposed in fluid communication with the droplet-generating microfluidic region; and a trigger source configured to trigger generation of an acid or a base from at least one pH-control substance contained within the reaction microfluidic region.

31 Claims, 20 Drawing Sheets