Frequency dependence and assembly characteristics of silver nanomaterials trapped by dielectrophoresis

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Abstract. For rapid and accurate fabrication of industrial products, the development of effective technique to manipulate and assemble nanomaterials is essential. In the present work, we investigated the dielectrophoretic behavior of silver nanoparticles and nanowires at various driving frequencies and voltage amplitudes, and examined three-dimensional assembly of the nanomaterials using dielectrophoretic devices with microstructures. The collectable range in frequency for nanoparticles and nanowires was from 10 kHz to 1 MHz. In particular, the aggregation shape of nanowires was changed against driving frequency. The bunch of nanowires was bridged between pillars of dielectrophoretic device. When the applied voltage was turned off, the trapped nanowires were entirely released. In pit type device, trapping of nanoparticles was observed in outermost pits of array. The aggregates of nanoparticles were directly fixed to the bottom electrode.

1. Introduction

Nanomaterials have high reactivity and applicability to various objects, since their specific interfacial area is large. The characteristics have been used to improve the performance of coloring materials and cosmetics. Nanomaterials have been also applied to new electronic devices such as high-functioning sensors and specific-shape microelectrodes [1-3]. Furthermore, nanowires and carbon nanotubes were examined for wiring parts of nano-circuits [4]. For rapid and accurate fabrications of above industrial products, the establishment of effective scheme to manipulate and assemble nanomaterials is essential.

Electrical handling by dielectrophoresis (DEP) is a powerful technique of bottom-up process for particulate manipulation. DEP is a phenomenon that the particles under non-uniform electric field migrate along the gradient of electric field. It is possible to control the particle behavior easily and rapidly, since DEP force strongly depends on the electric field gradient and driving frequency [5]. In fact, TiO_2 NWs were collected on the gold plate by DEP, and the formation products were available for sensor electrodes to detect organic vapors [6]. We fabricated three-dimensional DEP (3D-DEP) devices with dielectric pillars by proton beam writing [7] and succeeded in high concentration of bacteria around pillars. However, the suitable conditions of DEP trapping have not been understood well for variously-shaped nanomaterials.

In the present work, we investigated the frequency dependence of the trapping number and collected configuration for silver (Ag) nanoparticles (NPs) and nanowires (NWs). Moreover, we examined the trapping and release operation of Ag NWs around pillar array of DEP device. We also discussed the influence of voltage amplitude and polarity on the assembly state of Ag NPs using a pit type DEP device.


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2. Materials and Method

We prepared Ag NPs of 100 nm in diameter, and NWs of 60 nm in diameter and 10 μm in length (Sigma-Aldrich). 20 mg of NPs and 2-20 μl of NWs were suspended each by distilled water. The measurements of conductivities of NPs and NWs were 0.41 mS/m and 0.19 mS/m, respectively.

The DEP device consists of basal plate with microelectrodes, cover plate with microchannel and device holder. Figure 1 shows photographs of the electrode configurations. Interdigitated electrodes were fabricated by deposition of indium tin oxide (ITO) on silica substrate. The electrode width is 110 μm and gap length is 10 μm. With respect to 3D-DEP devices, dielectric pillars were formed in a 200 μm of gap area between electrodes. The diameter, height and pitch of pillar were 3 μm, 10 μm and 24 μm, respectively. The pit type DEP device has ITO plate electrodes and silicon spacer of 500 μm in thickness. Polymethyl methacrylate pits were deposited on the bottom electrode. The diameter, depth and pitch of pit were 4 μm, 10 μm and 10 μm, respectively.

The suspension was delivered into the channel using a peristaltic pump (PSM050DA, Advantec) for interdigitated plate and pillar type DEP devices. Only the suspension in pit type DEP device was filled using micropipettes. The applied voltage was supplied to the electrodes by a function generator (AFG3022, Tektronix). The collected state of nanomaterials was observed with a microscope (LV-100DA, Nikon) with CCD camera (DXM1200C, Nikon).

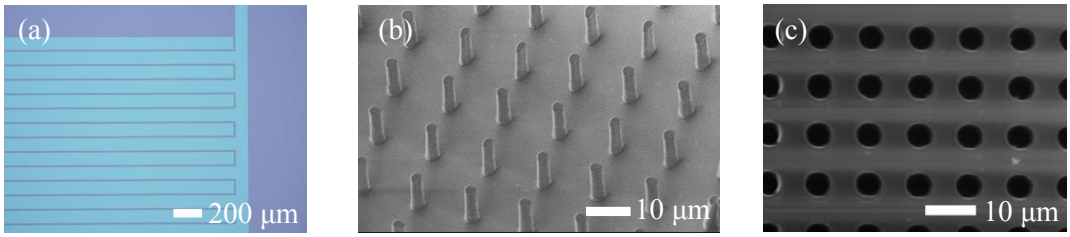

Figure 1. Photograph of electrode configurations. (a) interdigitated plate, (b) pillar and (c) pit.

3. Results and discussion

3.1. Frequency dependence of dielectrophoretic trapping for nanomaterials

We investigated collecting characteristics of Ag NPs at various driving frequencies f from 1 kHz to 10 MHz as is shown in figure 2. Here, the voltage amplitude V_a was 15 V_{pp} and flow rate ϕ was 10 ml/h. Trapping of the particles did not occur at 1 kHz and 10 MHz. In collectable frequency region between 10 kHz and 1 MHz, the NPs were firstly trapped in the gap and then were chained each other. This reason is that electric field was generated locally at trapped particles in upstream part. The thickness of chain gradually increased with frequency.

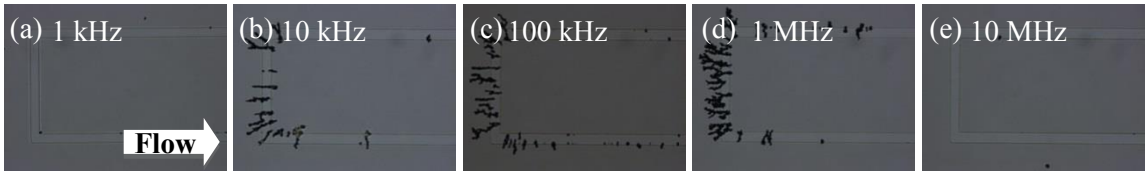

Figure 2. Dielectrophoretic trapping of Ag nanoparticles at various frequencies.

In the case of NWs (figure 3), the collectable frequency region was same as that of NPs. However, the frequency dependency of collected shapes was different from that of NPs. At a 10 kHz of frequency, NWs are collected in a straight line on the gap. In addition, small aggregates were observed in the center of electrode. The wires were arranged as a netlike appearance on overall electrode at 100 kHz. Up to 1 MHz, they were parallelized along the gaps. Gierhart *et al*. showed that the collection amount and shape of NPs depend on the frequency change of direct and mutual DEP forces F_{DEP} [8]. In addition to the above reason, the balance of AC electroosmosis between microelectrodes [9] and external flow would influence the distribution of Ag NWs in the present system. The numerical

simulation of particle dynamics, however, should be required for quantitative and morphological assessment.

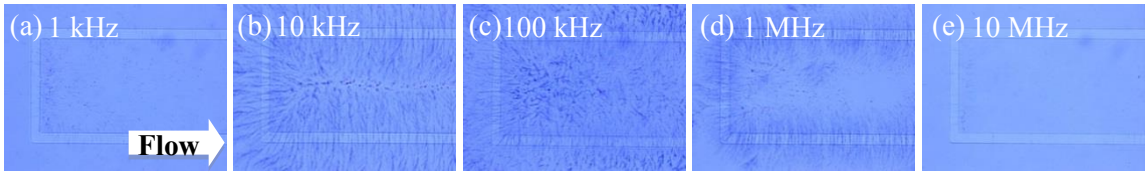

Figure 3. Dielectrophoretic trapping of Ag nanowires at various frequencies.

3.2. Numerical analysis of DEP force in 3D-DEP devices

With an electromagnetic field solver (COMSOL Multiphysics 4.4), we calculated the special distribution of electric field E (figure 4). Here, V_a was normalized to be 1 V. In the pillar type, E at 5 μm height distorts at boundary of pillar and medium. E around nearest pillars from the electrodes was about 1.6 times as large as that around central pillars. F_{DEP} at center and edge were 3.7×10^{-17} N and 2.9×10^{-15} N, respectively. These results suggested that bridging of NWs would occur from pillars near the electrodes. In the case of pit type at 12 μm height from the bottom, outer pits had stronger E than inner pits, although the direction of E was parallel to pits. Since the present pitch of pits was narrow, the influence of nearest pits on distortion of E became large. F_{DEP} of inner and outer pits were 2.2×10^{-16} N and 2.5×10^{-15} N, respectively. The force ratio of inner and outer was one tenth compared with pillar type. The present numerical evaluations should be useful for voltage control of assembly process in 3D-DEP devices.

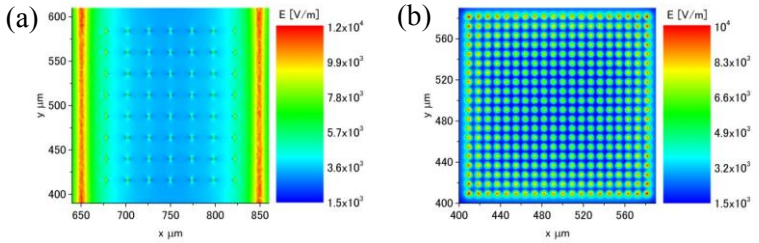

Figure 4. Spatial distribution of electric field. (a) pillar type and (b) pit type.

3.3. Trapping and release operations of nanowires using pillar type DEP device

On the basis of above findings, we tried the condensation and release operations of NWs using pillar type DEP device. Figure 5 shows the temporal behavior of Ag NWs between pillars. V_a was 10 V_{pp}, f was 100 kHz and ϕ was 10 ml/h. NWs were collected only around pillars. After 30 min, the wires were bridged between the pillars and weakly-bind chains were formed. When the applied voltage was turned off, the NWs were rapidly released from pillars. In the case of over 10 V_{pp}, the collected wires aggregated each other and the large aggregates broke some pillars.

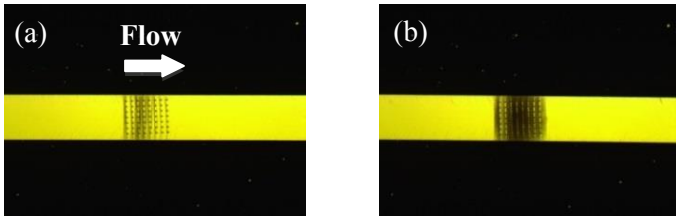

Figure 5. Temporal behavior of trapped nanowires in pillar type DEP device. (a) 5 min and (b) 30 min.

3.4. Assembly of nanoparticles using pit type DEP device

To explore effective assembly of nanomaterials, we investigated the voltage dependency of Ag NP trapping in pit type DEP device. Figure 6 shows the collecting states of NPs at 100 kHz. When V_a was

10 V_{pp}, the particles were not collected anywhere. At a voltage of 20 V_{pp}, they were focused in outermost pits at 30 min. However, the bunch of particles grew up to dendritic outside the pit within 1 min, if V_a was raised up to 30 V_{pp}. The suitable threshold of V_a was 20 V under the present condition.

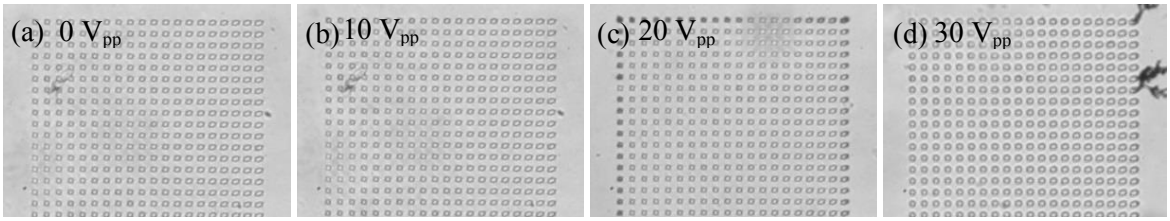

Figure 6. Voltage dependency of Ag nanoparticle trapping in pit type DEP device.

Moreover, we examined superposition of DC offset bias on AC driving voltage to transport NPs equally around every pit. As is shown in figure 7, the collectable points at 1 V bias were randomly observed in inner pits, although the total collection amount was rarely different from that without bias voltage. On the other hand, we obtained thin film of Ag particles on overall of pit array within 1 min by changing the polarity of offset bias. This film was not dispersed even if the applied voltage was turned off. They was then retained its shape even if explored in air.

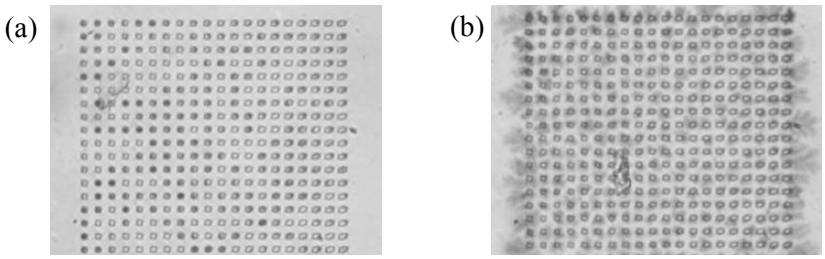

Figure 7. Influence of offset voltage on DEP assembly. (a) 1.0 V at 90 min and (b) -1.5 V at 1 min.

4. Conclusion

We investigated DEP characteristics and effective assembly of Ag nanomaterials using various microelectrodes. The trapping number of NPs depended on applied frequency. In particular, the collecting shape of NWs changed because of their large dipole moment. Using pillar type device, we succeeded in high concentration and rapid release of NWs around pillar. Also, we evaluated the voltage dependency for NP assembly and specified effective value of AC applied and DC bias voltages in pit type device. In the nearest future, we will fabricate functional nanostructures by DEP assembly on the basis of above information of control parameters.

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