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Dielectrophoretic Assembly of Single Gold Nanoparticle into Nanogap Electrodes

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We report the optimization study of assembling single 20 nm gold nano-particle in 20 nm spaced electrode gap via ac dielectrophoresis (DEP) technique. It was observed that time, voltage, and frequency variations influenced significantly the assembly of gold nano-particle in the nano-gap electrodes. Frequency variation study revealed that at lower frequencies (<1 MHz) the assembling was observed in low field regions; however, at a moderate frequency of 1 MHz, minimum number of nano-particles was assembled in high field region. Trapping of single 20 nm nano-particle in 20 nm spaced electrodes was successfully achieved under the optimized DEP parameters i.e., frequency, applied voltage and time of values corresponding to 1 MHz, 2 V, and 1 sec, respectively, with the yield of almost 66%. Our results show the promise of optimized dielectrophoresis in the future nano-engineering.

Keywords: Dielectrophoresis, Gold Nanoparticles, Nanogap Electrodes, Self Assembly.

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1. INTRODUCTION

In recent years, molecular electronics has gained considerable attention due to its unprecedented abilities which can be implemented for designing next generation of devices by controlling the passage of current through molecules.[1–8] The basis of these devices is the electrical transportation through single molecules. However, due to their natural small size (nm), the electrical contacts to the molecules still remain a challenge. The alternative approach is to assemble the molecules or colloidal nano-particles on a substrate pre-fabricated with electrodes with gap size varying from 20 nm to few mm using external electric field.[4–8] Tuning the microscopic particles assembly with external electric field has generated considerable interest due to applications in nanotechnology and biomedical research.[9] One of the most promising methods is dielectrophoresis (DEP), where the motion of polarizable particles is controlled by the externally applied non-uniform electric field.[10–18] DEP has recorded a promising success in micro- and nano-particles assembling of, for example, gold (Au) nano-colloids,[2–8] DNA,[19] micro-organism (biological particles like cell),[20] nanowires,[21–23] and carbon nano-tubes.[24] DEP offers the unique opportunity of selectively placing nano-objects inside electrically defined gaps with several potential advantages, mostly versatility and quick assembly time.[2–8] Amongst them, the assembly of colloidal nano-particles into functional structures is emerging as an exciting area of research for potential nanotechnology applications.[7,21] Colloidal metal nano-particles such as gold have unusual chemical and physical properties which make them attractive for applications such as catalysis, electronics, optics, and biotechnology.[25]

DEP has been extensively used for trapping Au nano-particles by many research groups.[2–8] Bezryadin et al.[1] produced some encouraging results in 1997 when they assembled metal nano-particles such as Palladium (Pd) in the Pt electrodes gap down to 4 nm using electrostatic trapping technique, and subsequently attempted to measure

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the transport property through single Pd nano-particle. Khondaker and Yao[5] reported exciting results of assembling Au nano-particles into nanogap electrodes using DEP technique. They reported successful assembly of 50 nm Au nano-particles in to 50 nm to 1 μm spaced electrodes.[5, 8] Hong et al.[6] studied the characteristics of ac and dc DEP techniques by assembling Au nano-particles (20–50 nm) in a system comprised of self assembled monolayers (SAM) between the gold electrodes (30 nm). Recently Stellacci group[7] demonstrated the optimized assembly of Au nano-particles (20 nm) in the electrode gaps (15–150 nm), created by localized melting technique,[11] using DEP technique. Inspite of several studies on gold nanoparticle sassembly, detail optimization of the controlled assembly of single nanosized Au nano-particle (20 nm) into nanogap is missing.

In this paper, we report the controlled assembly of 20 nm sized Au nano-particle in to 20 nm gap gold electrodes via ac DEP technique. We have investigated and optimized the experimental key parameters like applied voltage, frequency, and time, which are found to be crucial, for trapping such nano-particles into nanogap electrodes during DEP.

2. EXPERIMENTAL DETAILS

The Au electrodes used in the present work were fabricated on oxide-coated silicon substrate using standard electron-beam lithography (EBL) and lift-off technique. The separation between the Au electrodes was 20 nm, and such ten pair of electrodes was comprised in one chip (Fig. 1). Figure 1 depicts the scanning electron microscopy (SEM) images of the chip, showing the topography of the chip (Fig. 1(a)), and the enlarged view of the chip showing ten pairs of electrodes (Fig. 1(b)) and each pair of electrodes with a gap of 20 nm (Figs. 1(c–e)). The experimental set-up used for DEP assembly consists of a function generator connected directly, and through the chip, to the oscilloscope (Fig. 2). Au nano-particles of 20 nm diameter were purchased from British Bio Cell International and used at the as-purchased concentration (7×10^{11} particles per ml). Prior to DEP assembling, the chip was passed through a cleaning process such as O_2 plasma cleaning and rising in ethanol (Fig. 3). Figure 3 shows the flowchart of various steps followed in DEP assembling of Au nano-particles. A micropipette was employed to take an approximately one micro liter drop of aqueous nano-particles solution for the DEP processing. Finally, the trapping of Au nano-particles in the nanogap electrodes was examined by SEM after DEP processing.

3. DIELECTROPHORESIS

Dielectrophoresis (DEP) is used to describe the motion induced by a non-uniform electric field on polarized, but

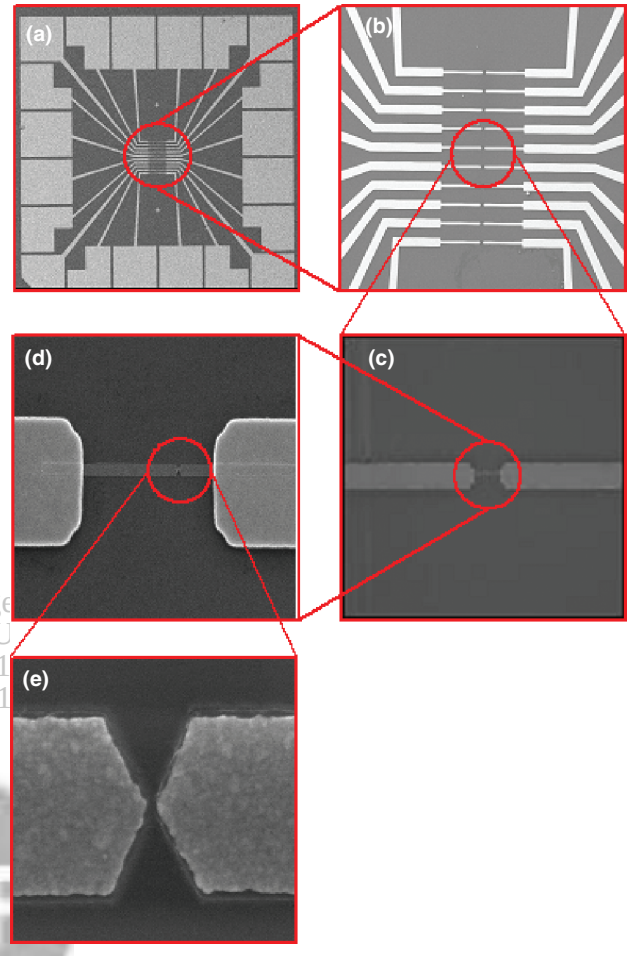

Fig. 1. SEM images of chip: (a) full image of chip showing ten parallel electrodes with individual contact pads; (b) magnified view of ten electrodes; (c and d) magnified views of one pair of electrodes; (e) magnified view showing a nanogap of 20 nm between one pair of electrodes.

electrically uncharged particles.[12, 13] The pioneering work by Pohl[10] provided the basic theory of DEP and its early applications. DEP can be understood by considering the simple picture of a dipole in a non-uniform electric field. The dipole will be at least partly oriented in the direction of the field gradient and hence one end of the dipole will experience stronger electric field strength than the other. This results in a non-zero net force and the dipole is set in motion. This has allowed the assembling of different species like nano-particles,[4, 7] nanowires[22] and manipulation of living cells[20] using DEP. Due to its diverse applications, DEP has drawn the attention from diverse fields of research including medicine, colloidal science and nanotechnology.[13]

In the case of DEP, the force acting on the particle known as DEP force (F_{DEP}) is dependent on the frequency of the applied external ac electric field as well as on the magnitude and complex dielectric constant of the particles under consideration. Normally in dilute form, the interaction between the individual particles is

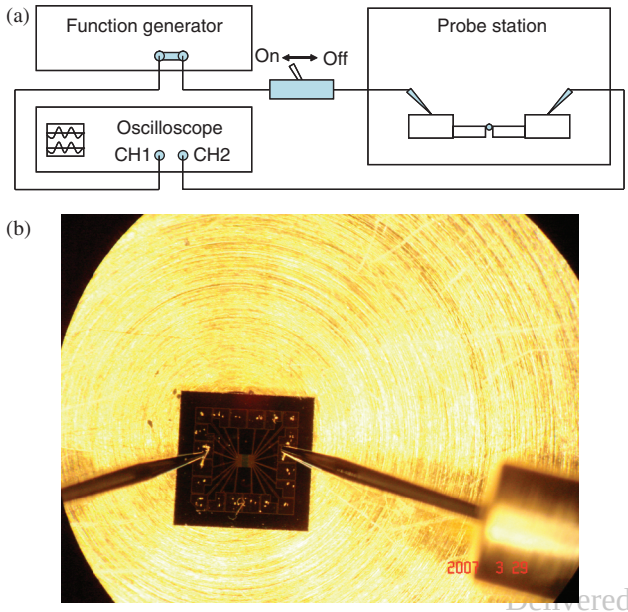

Fig. 2. (a) Block diagram of the experimental setup used for DEP; (b) photographic image showing a chip placed under a pair of probes in probe station.

ignored, and the DEP spectrum of individual particle can be considered.[9, 13]

The DEP force, F_{DEP}, experienced by the nano-particles is given by[12]

$$\vec{F}_{DEP} = \alpha(\vec{E} \cdot \nabla)\vec{E} \quad (1)$$

Where α is a spatially independent polarizability parameter whose magnitude and polarity are the function of the permittivity and polarity of the particle and suspending medium, and E is the applied electric field. At a frequency and the medium conductivity for which α attains a positive value, the nano-particles are directed to the strong field regions. The frequency and the conductivity values for which α is negative, the nano-particles are dragged to the regions of low strength of electric field.

4. RESULTS AND DISCUSSION

For the controlled assembly of Au nano-particle (20 nm) into nanogap (20 nm) electrodes, the experiments were carried out by varying the key experimental parameters such as the magnitude of applied ac frequency (f), applied peak to peak voltage (V_{pp}), and the time (t) during which non-uniform field is active. The optimization was carried out by varying one parameter and keeping the other two parameters fixed. For each variation of parameters, the number of nano-particles assembled in the nano-gap electrodes was counted.

4.1. Influence of Applied AC Voltage

To investigate the assembly under varying applied ac voltage (V_{pp}), the frequency of applied ac signal was kept at

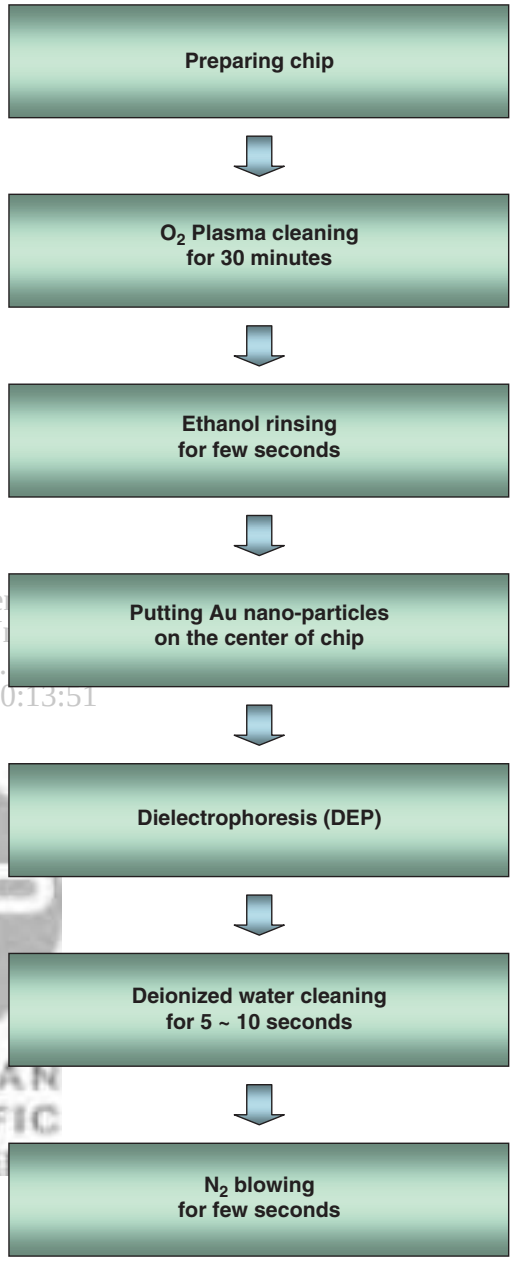

Fig. 3. Flow chat showing the various steps followed for the assembly of nano-particles in nanogap electrodes.

1 MHz, and the DEP time was restricted to 1 sec. Figure 4 shows the SEM images of assembled Au nano-particles under varying applied ac voltage ($V_{pp} = 1.5$ V, 2 V, 3 V and 4 V), and at fixed frequency of 1 MHz, and time of 1 sec. It can be inferred that as the applied voltage is increased, the number of nano-particles assembled in the nanogap is also found to increase. However, at an applied voltage of 2 V, on an average 1–2 nano-particles were found to be trapped in the nanogap. Figure 5 shows the variation of number of trapped nano-particles as a function of applied ac voltage. It may be noted that 2 V is the threshold voltage for the assembly of minimum number of nano-particles in the nanogap.

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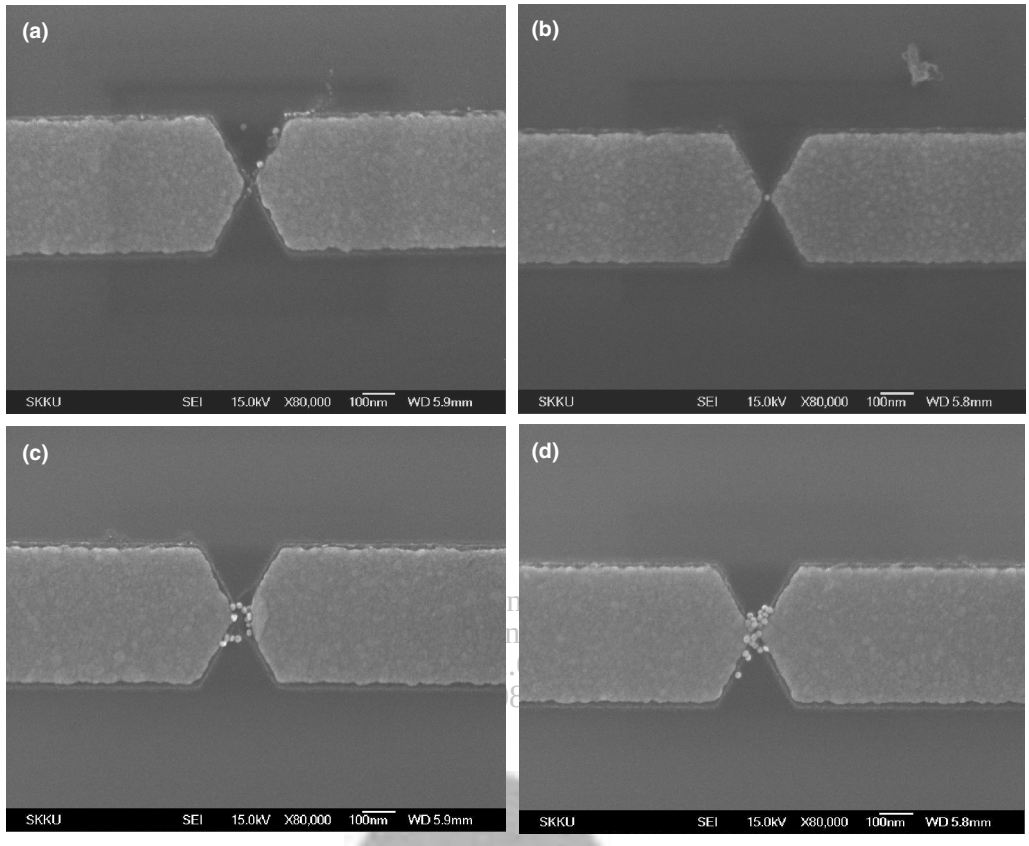

Fig. 4. SEM images of the assembly of nano-particles in nanogap electrodes when DEP was performed as function of voltage at a fixed frequency and time of 1 MHz and 1 sec, respectively. The images correspond to applied peak to peak voltage (V_{pp}) of (a) 1.5 V; (b) 2 V; (c) 3 V; and (d) 4 V.

4.2. Influence of Applied Frequency

To study the effect of frequency on nano-particle assembly, the applied ac peak to peak voltage was maintained at 2 V (optimized value), and the DEP time to 1 sec. Figure 6 shows the SEM images of the assembly of nano-particles at varying applied ac frequency. It can be seen that assembling of nano-particles strongly depends on the applied ac frequency. At lower frequency value of 0.1 MHz a large number of nano-particles assembled in the region between the electrodes. However, with an increase in frequency less number of particles assembled in the region between the electrodes. It was observed that at a frequency value of 1 MHz, a minimum number of nano-particles were trapped in the gap, and gave a clear idea that the DEP force becomes ineffective beyond this frequency. Since it is well known that DEP force depends on the applied ac frequency and assembling of nano-particles largely depends on this value.[7, 13] Therefore, we can conclude that applied frequency of 1 MHz is the optimized frequency to achieve assembly of minimum number of nano-particles into nanogap electrodes.

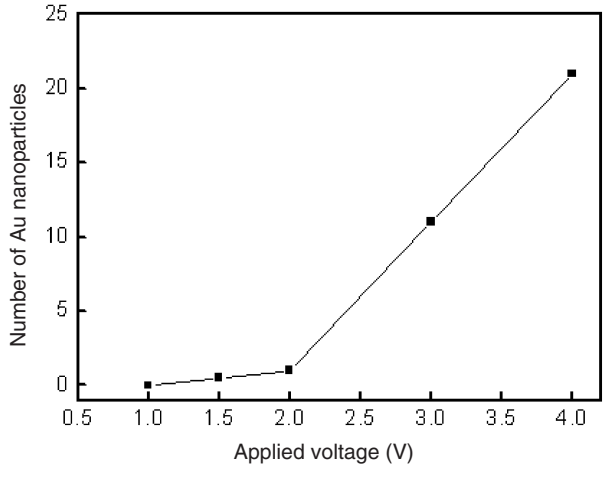

Fig. 5. The variation of number of nano-particles assembled in the vicinity of tip of electrodes as a function of applied peak to peak voltage (V_{pp}).

4.3. Influence of Time

To achieve the assembly of very few isolated particles in the gap, it was necessary to limit the assembly time. Therefore, we decided to perform DEP at optimized voltage and frequency values of 2 V and 1 MHz, respectively, whereas time was kept as variable parameter. Figure 7 shows that there is an increase in number of nano-particles assembly when DEP was operated for a longer time, hence it was necessary to limit the time to achieve the assembly of

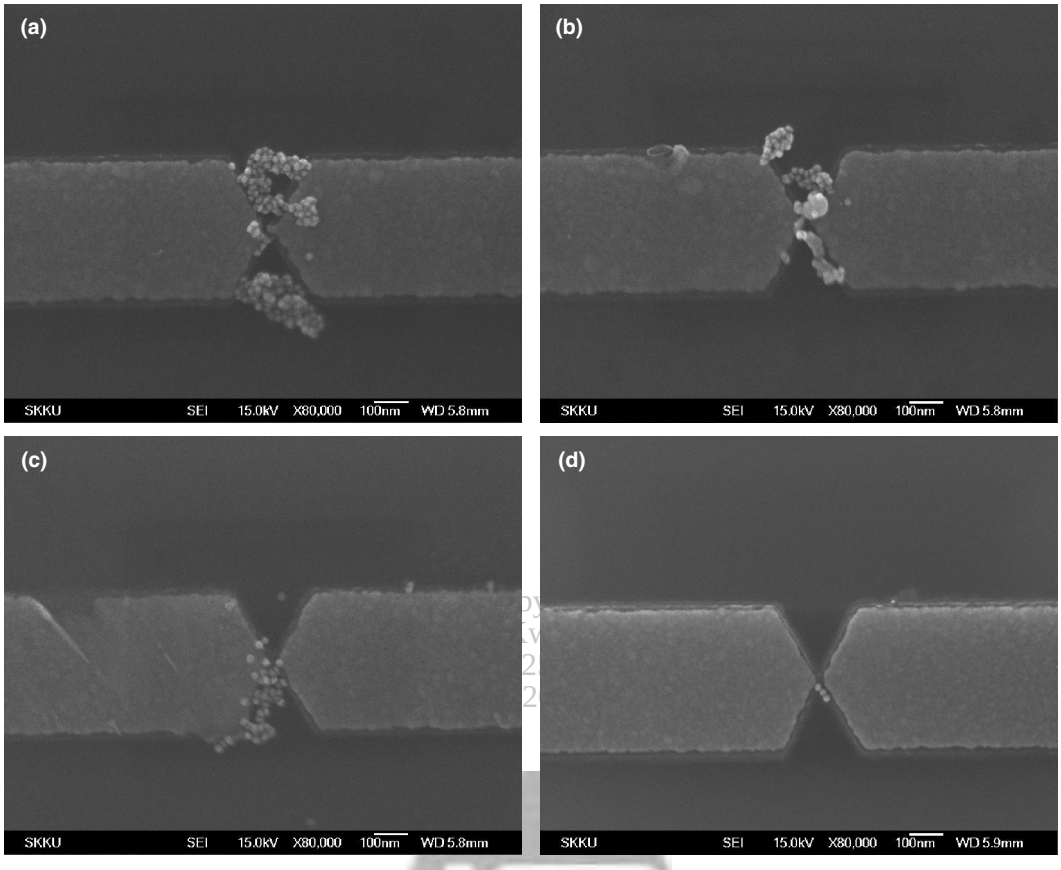

Fig. 6. SEM images of the assembly of nano-particles in nanogap electrodes when DEP was performed as function of frequency at a fixed applied voltage and time of 2 V and 1 sec, respectively. The images correspond to applied frequency values of (a) 0.1 MHz; (b) 0.2 MHz; (c) 0.5 MHz; and (d) 1.0 MHz.

single nano-particle into nanogap. Figure 8 shows the variation of the number of particles assembled in the region between the electrodes as a function of time. It is clearly seen that time, during which DEP is done, is very important, and number of nano-particles trapped varies linearly with time. It is to be noted that DEP time of 1 sec was found sufficient for trapping minimum number of nano-particles in between the electrodes gap (Fig. 8).

Under the optimized condition i.e., applied frequency of 1 MHz, applied ac voltage of 2 V, and DEP time of 2 sec, trapping of single gold nano-particle was successfully achieved, and the yield was approximately 66%. Figure 9 shows the assembly of single Au nano-particle into 20 nm gap electrodes.

4.4. Discussion

In the optimization of voltage it was observed that with an increase in voltage the assembly of nano-particles was found to increase with time. An increase in voltage results an increase in melting of nano-particles due to Joule heating. Once a significant amount of nano-particles is assembled near the tip of the electrodes, a possibility of flow of large amount of current causes melting and aggregation of nano-particles. In some instance, due to accumulation of nano-particles and due to localized melting, nano-bridges are seen to form, as has been observed in present case (Fig. 4(c)). It may be concluded from Figure 4 that positive DEP force is acting on the particles (Eq. (1)). It is important to note from Figure 4 that all the particles assemble (independent of any applied voltage value) in the regions of high electric field or close to the tip of the electrodes, which is the characteristic of positive DEP. However, at lower applied voltage ($V_{pp} = 1$ V), even though nano-particles are attracted to the point of high electric field, the current is not large enough to cause melting, and hence assembling. When the applied voltage reaches a threshold value and is moderate enough (in the present case, 2 V) to cause mild melting of the nano-particle or nano-particles, assembly of few nano-particles may be expected as has been observed in the present case (Fig. 4 (b)). A similar observation of voltage dependent particle assembly is also reported by Barsotti et al.[7]

In the second case, when the nano-particles assembly was optimized for frequency, some interesting observations were made. As we have seen earlier, we could achieve single or minimum particle assembly at a frequency value of 1 MHz, and at lower frequencies

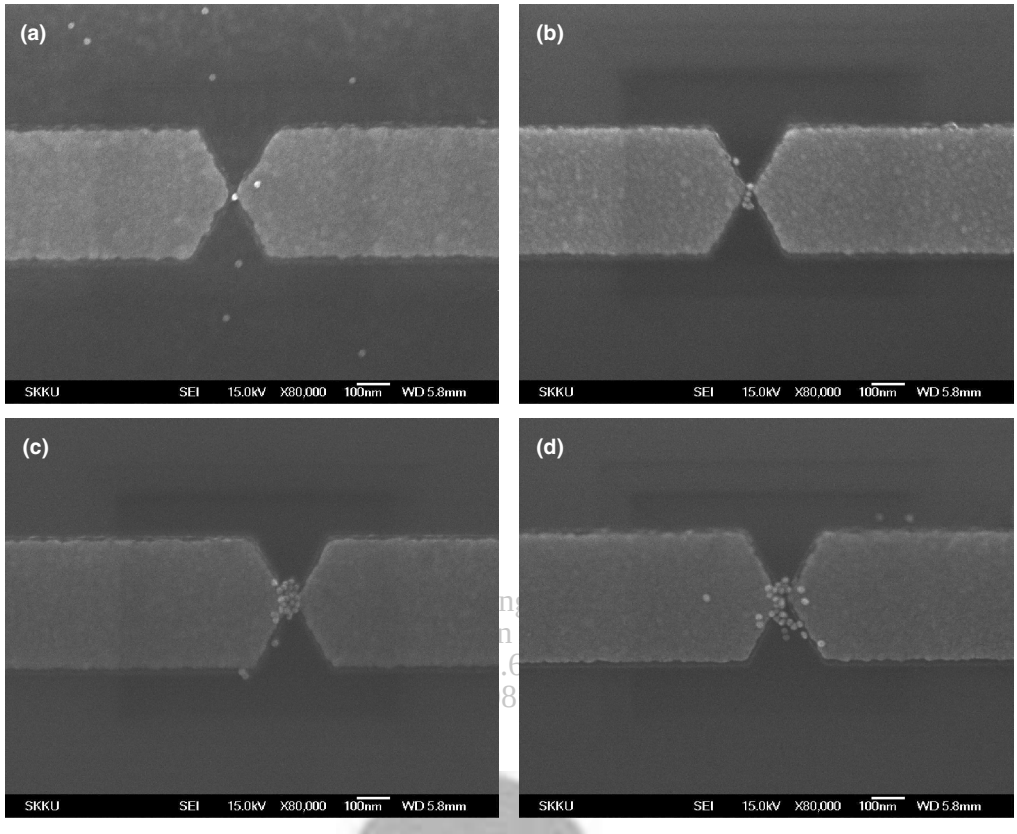

Fig. 7. SEM images of the assembly of nano-particles in nanogap electrodes when DEP was performed as function of time at a fixed applied voltage and frequency of 2 V and 1 MHz, respectively. The images correspond to time values of (a) 1 sec; (b) 2 sec; (c) 5 sec; and (d) 10 sec.

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(<1 MHz) aggregation of nano-particles was seen in the region between the electrodes. At higher frequencies no assembly was observed, and threshold frequency of 1 MHz provides the assembly of minimum number of nano-particles. At lower frequencies, large assembly of nano-particles was observed and could be attributed to surrounding of large counterion cloud around the nano-particles, which is susceptible to changes in electrical field (of non-uniform ac signal). This counterion could increase the effective radius of the nano-particles, thereby increases the DEP force and hence the particles assembly.[7] It is important to note that at lower frequencies nano-particles were assembled away from the region of strong electric field (towards the edges of the electrodes). This manifests that negative DEP is responsible for such assembling of particles (Eq. (1)), and is entirely dependent on the applied frequency and characteristics of nano-particles

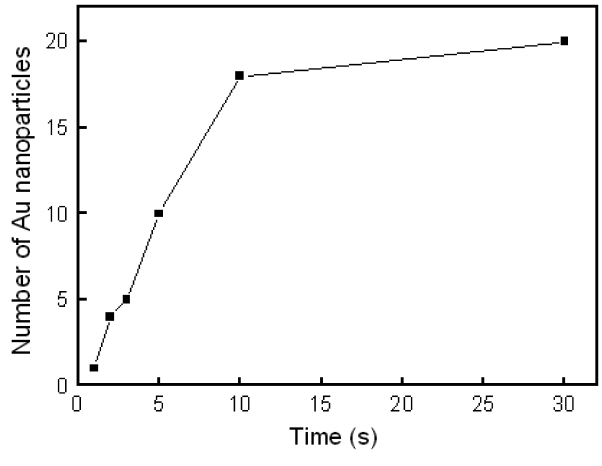

Fig. 8. The variation of number of nano-particles assembled in the vicinity of tip of electrodes as a function of time.

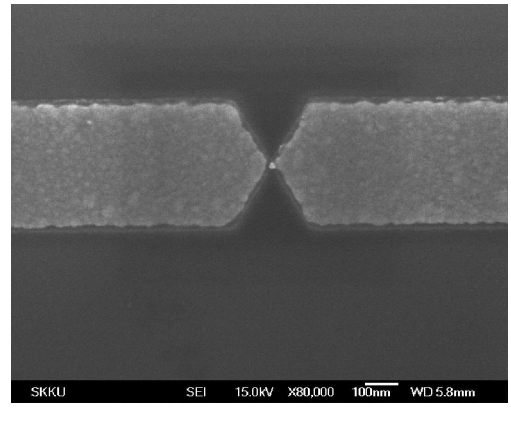

Fig. 9. SEM image showing the assembly of single 20 nm Au nano-particle in 20 nm gap electrodes under the optimised DEP parameters ($f = 1$ MHz, $V_{pp} = 2$ V, $t = 1$ sec).

and suspension medium. Moreover, at threshold frequency (1 MHz), single particle assembly is observed, which is nothing but the effect of positive DEP. These conclusions are in agreement with the results of Pethig et al.,[12] who assembled yeast cells in interdigital castellated microelectrode geometry, at varying frequency of the applied ac signal. Therefore, we conclude from our present observations that the type of DEP (positive or negative) not only depends on the characteristics of assembled nano-particle and medium, but is dependent on applied ac frequency also.

In the final case, when the DEP time was varied, it was observed that with an increase in DEP time large number of nano-particles aggregate near the tip of the electrodes. Previously we have seen that under the optimized conditions of frequency and applied voltage, a minimum number of nano-particles was seen to assembly in the nanogap. With an increase in DEP time, there is possibility of aggregation of nano-particles in the region of high electric field (positive DEP), which would cause the flow of large current through these nano-particles, and ultimately their melting and aggregation. Therefore, with increase in DEP time, a large assembly of nano-particles is seen in the region near the tip of electrodes.

5. CONCLUSIONS

We have demonstrated the assembly of single Au particle (20 nm) into 20 nm nanogap electrodes (1:1 ratio of particle size and gap) with the yield of approximately 66%. It was observed that assembly of nano-particles into nanogap strongly depends on DEP parameters like applied voltage, frequency and time. Assembly of single Au nanoparticle has been demonstrated under the optimized DEP conditions of frequency of 1 MHz, applied ac voltage of 2 V, and DEP time of 1 sec. Frequency dispersion study revealed that positive DEP is required for the assembly of single nano-particle into nanogap electrodes. Such assembly of single nano-particle demands potential application in studying the transport characteristics of metallic as well as biological particles such as DNA.

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