Voltage and current conditions for nanoparticle chain formation using dielectrophoresis

Tomoya Koshi, Yuta Nakajima, Eiji Iwase ✉

Department of Applied Mechanics, Waseda University, 3-4-1 Okubo, Shinjuku-ku, Tokyo 169-8555, Japan
✉ *E-mail: iwase@waseda.jp*

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Here, the voltage and current conditions required to form a nanoparticle chain bridging a gap between electrodes that is several tens of micrometres wide have been examined. When a voltage is applied to a gap between electrodes covered with a dispersion of nanoparticles, the nanoparticles are trapped in the gap by dielectrophoresis, forming a nanoparticle chain. It has been considered that a nanoparticle chain is formed in a gap only when a voltage higher than a certain value with a current lower than a certain value is applied to the gap. In this work, certain voltages were first applied with changing current to a 10 μm-wide gap between electrodes covered with a dispersion of 150 nm-diameter gold nanoparticles, and they found that a nanoparticle chain was formed only when 6.1 V_{rms} or more with 15 mA_{rms} or less was applied. Next, the process of nanoparticle chain formation in a 10 μm-wide gap was directly observed with high-magnification and high-speed microscopy, and they clarified the behaviour of nanoparticle chain formation when a low voltage, high voltage with high current, and high voltage with low current were applied. Finally, they measured the critical value of current for nanoparticle chain formation in a gap several tens of micrometres wide.

1. Introduction: Nanoparticle trapping in a gap between electrodes through dielectrophoresis is used for the formation of micro- and nanostructures of nanoparticles [1–3] or the property measurement of nanoparticles [4–8]. In particular, the formation of a nanoparticle chain structure has been of great interest because of its application to micro- and nanodevices [9–12]. When a certain voltage is applied across a gap between electrodes covered with a nanoparticle dispersion, an electric field is generated around the gap. Subsequently, the nanoparticles are trapped in the gap because of dielectrophoresis force on the nanoparticles (Fig. 1*a*). Consequently, the nanoparticles bridge the gap by forming a chain. In this phenomenon, the gap width is one of the important parameters for nanoparticle chain formation. A previous study [12] reported that a gold nanoparticle chain was successfully formed in a several-μm-wide gap between gold electrodes and was not formed in a wider gap. On the other hand, other studies [13–15] reported that a gold nanoparticle chain was formed even in a wider gap between gold electrodes (width of several tens of micrometres or more). The conditions enabling nanoparticle chain formation in such a wide gap remains unclear.

In previous studies, dielectrophoresis force is usually controlled by a voltage applied across a gap between electrodes. Previous studies [12, 16] indicated that an applied voltage higher than a certain value can form a nanoparticle chain in a wide gap because a higher applied voltage causes a stronger dielectrophoresis force on the nanoparticles. If the voltage is lower than a certain value, nanoparticles are not trapped, as shown in Fig. 1*b*. However, in a previous study, a wide gap was not bridged even with a higher applied voltage [12]. Therefore, we believe that the current flowing when nanoparticles are assembled to bridge a gap may play an important role. That is, Joule heating due to the current under a high applied voltage might break the nanoparticle chain (Fig. 1*c*). Joule heating is proportional to both the resistance of the nanoparticle chain and square of the current. The resistance depends on the gap width, which determines the length of the nanoparticle chain. Therefore, the current must have a critical value for nanoparticle chain formation. We considered that a nanoparticle chain can be formed in a gap several tens of micrometres wide only when a voltage higher than a certain value with current lower than a certain value is applied (Fig. 1*d*).

In this Letter, we first applied certain voltages with changing current to a wide gap between electrodes in order to confirm that a nanoparticle chain is formed only when a high voltage with low current is applied. Next, to clarify the behaviour of nanoparticle chain formation when a lower voltage, higher voltage with higher current, and higher voltage with lower current are applied, we directly observed the process of nanoparticle chain formation through high-magnification and high-speed microscopy. Finally, we measured the maximum value of current that allows nanoparticle chain formation, I_{max}, by applying increasing currents to a nanoparticle chain.

2. Measurement of conditions for nanoparticle chain formation: To confirm that a nanoparticle chain is formed in a wide gap when a high voltage with low current is applied, we fabricated a 10 μm-wide gap between gold electrodes on a glass substrate, and applied certain voltages with changing current to the gap, as shown in Fig. 2. A 0.01 μm-thick chromium layer as an adhesion layer and a 0.5 μm-thick gold layer were deposited on a glass substrate. The deposited metal layers were patterned to an electrode shape (10 μm gap width and 25 μm electrode width). The gap between the electrodes was covered with an aqueous dispersion of gold nanoparticles 150 nm in diameter (Au colloid solution-SC, Tanaka). A resistor was connected to the electrodes in series, and an AC voltage was applied across both the gap and resistor by a waveform generator (33500B, Agilent) and an amplifier (HAS 4101, NF) for ∼3 min. The frequency was chosen as 100 kHz, because the number of trapped gold nanoparticles was determined by frequency and 100 kHz was suitable for nanoparticle chain formation [17]. The voltage applied across the gap between the electrodes, V_{gap}, and current flowing in the assembled nanoparticles were changed by changing both the resistance of the resistor, R_{res}, and voltage applied to the circuit, V_{all}. A spark was not observed during the measurement. The values of R_{res} were 4.63 kΩ, 1.78 kΩ, 985 Ω, 462 Ω, and 177 Ω. When a nanoparticle chain bridges the gap, V_{gap} changes to V'_{gap} because the impedance of the gap between the electrodes is sharply decreased. V_{gap} was monitored using an oscilloscope (DSO-X 2024A, Agilent). We used V_{all}/R_{res} as the representative value of current flowing in a nanoparticle chain. Once a

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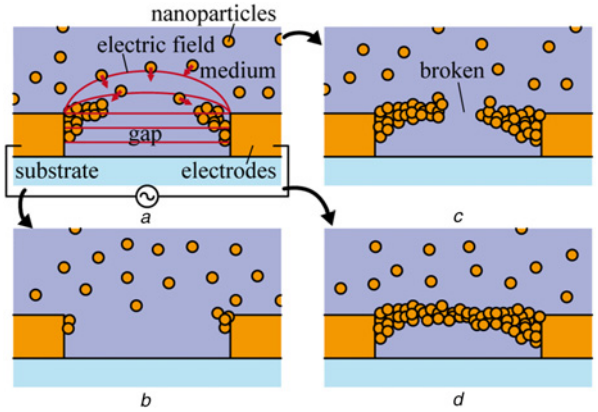


Fig. 1 *Schematic illustration of nanoparticle chain formation through dielectrophoresis in a wide crack*
a Trapping of nanoparticles in a gap by dielectrophoresis
b Nanoparticles are not trapped when the voltage is low
c Nanoparticle chain is broken when a high voltage with high current is applied
d Nanoparticle chain is formed when a high voltage with low current is applied

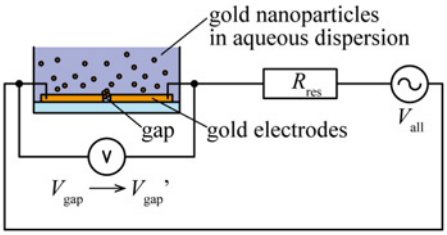


Fig. 2 *Measurement setup for gold nanoparticle chain formation*

nanoparticle chain was formed in the gap, the nanoparticle chain keeps its formation even after the voltage applying was stopped. After the measurement, the gap between electrodes was washed with pure water and dried for scanning electron microscope (SEM) observation.

Fig. 3 shows a condition plot for nanoparticle chain formation in the case of a 10 µm gap width. When a voltage of 4.1 V_{rms} or less was applied across the gap, a nanoparticle chain was not observed, as shown in Fig. 3*b*. This indicates that the nanoparticles were not trapped, because the dielectrophoresis force on the nanoparticles was weak owing to the low applied voltage. When a voltage ≥6.1 V_{rms} with a current ≥19 mA_{rms} was applied, an imperfect nanoparticle chain was observed, as shown in Fig. 3*c*. A micrometre-sized lump of gold was observed in a part of the imperfect nanoparticle chain, which indicates that some of the nanoparticles in the nanoparticle chain were melted. On the other hand, a nanoparticle chain was observed in the gap when a voltage ≥6.1 V_{rms} with a current ≤15 mA_{rms} was applied across a gap between electrodes, as shown in Fig. 3*d*. This indicates that a nanoparticle chain can be formed in a wide gap when a high voltage with low current is applied. In this case, a nanoparticle chain was often formed at the edges of electrodes because nanoparticles were trapped more at edges because the gradient of the electric field was especially strong around the edges.

3. Direct observation of process of nanoparticle chain formation: To clarify the behaviour of nanoparticle chain formation when a lower voltage, higher voltage with higher current, and higher voltage with lower current is applied, we observed the process of nanoparticle chain formation directly with high-magnification and high-speed microscopy. We

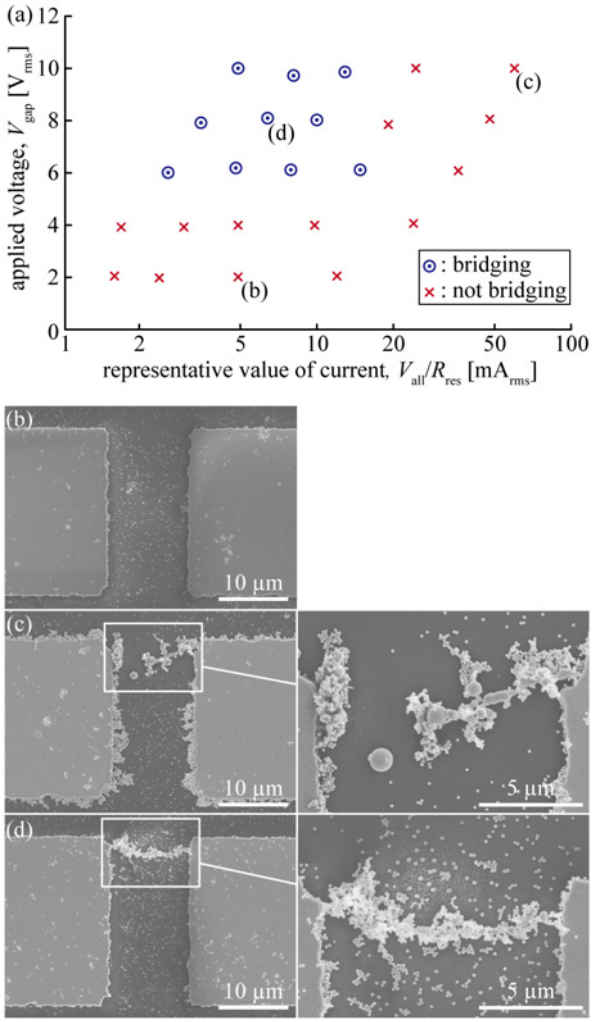


Fig. 3 *Measurement of conditions for gold nanoparticle chain formation in the case of a 10 µm-wide gap*
a Formation condition plot between applied voltage, V_{gap}, and representative value of current, V_{all}/R_{res} (the number of trials $n = 1$)
b SEM image in the case of 2.0 V_{rms} with 4.9 mA_{rms}
c SEM images in the case of 10.0 V_{rms} with 60 mA_{rms}
d SEM images in the case of 8.1 V_{rms} with 6.4 mA_{rms}

fabricated a 10 µm-wide gap of gold electrodes on a glass substrate in the same manner as for the measurement of conditions for nanoparticle chain formation. An aqueous dispersion of 150 nm-diameter gold nanoparticles was dropped on the gap between electrodes and covered with cover glass. Based on the above measurement, as shown in Fig. 3*a*, V_{gap} and V_{all}/R_{res} were set to 1.9 V_{rms} and 11 mA_{rms} to represent the case of low voltage, 10.3 V_{rms} and 60 mA_{rms} to represent the case of high voltage with high current, and 8.9 V_{rms} and 1.8 mA_{rms} to represent the case of high voltage with low current, respectively. We used a high-speed optical microscope (VW-9000, Keyence) with a magnification of 3000×. Both the top and bottom light sources were a metal halide lamp. The frame rate of microscopy was 4000 fps (0.25 ms/frame), and images were taken for 5.47 s several seconds after we started to apply the voltage to the gap between electrodes.

Figs. 4–6 show a series of optical images of nanoparticle chain formation for low voltage, high voltage with high current, and high voltage with low current, respectively. When a low voltage was applied, nanoparticles were not trapped in the gap between electrodes, as shown in Figs. 4*a* and *b*. On the other hand, when a high voltage with high current was applied, some nanoparticles were trapped at the edges of electrodes at the beginning of

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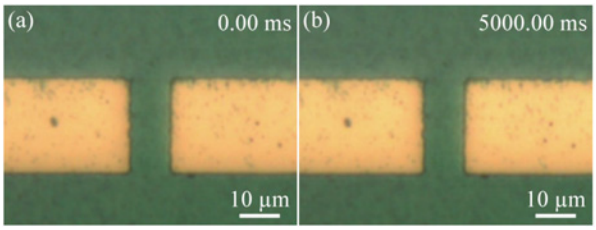


Fig. 4 *Series of optical images showing gold nanoparticle chain formation in the case of a low voltage applied to a gap between gold electrodes*
a, b Nanoparticle chain was not observed clearly

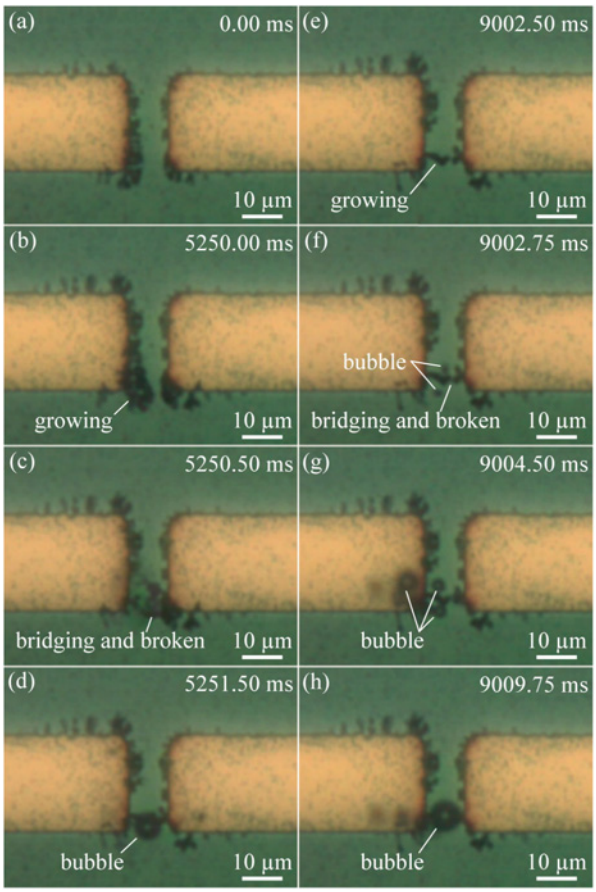


Fig. 5 *Series of optical images showing gold nanoparticle chain formation in the case of a high voltage with high current*
a, b Nanoparticle chains are growing
c, d One of the nanoparticle chains is bridging and being broken
e Another nanoparticle chain is re-growing
f, g, h Nanoparticle chain is being broken again

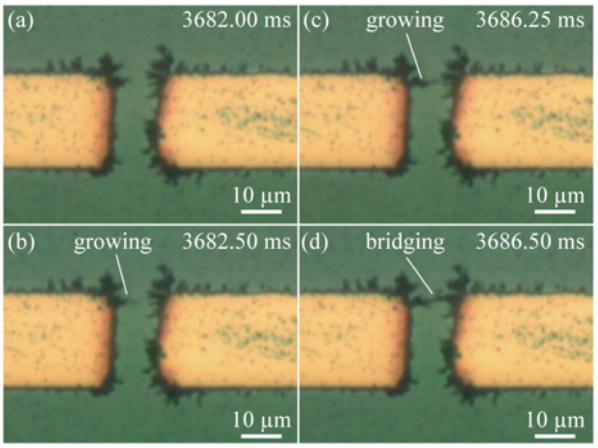


Fig. 6 *Series of optical images showing gold nanoparticle chain formation in the case of a high voltage with low current*
a Nanoparticles are being trapped in the gap
b, c Nanoparticle chain is growing
d Nanoparticle chain is bridging a gap

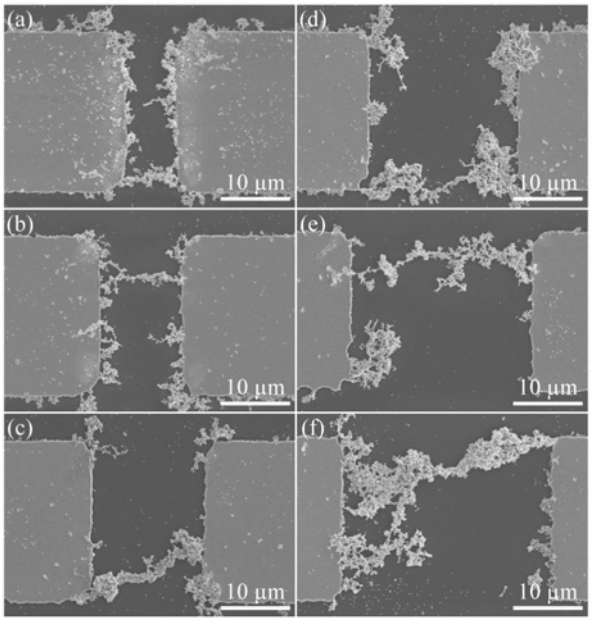


Fig. 7 *SEM images of gold nanoparticle chain in a gap between gold electrodes on a glass substrate*
a, b, c, d, e, f correspond to gap widths of 5, 10, 15, 20, 25, and 30 μm, respectively

microscopy (Fig. 5*a*). Approximately 5 s later, the number of the trapped nanoparticles was increased and nanoparticle chains were growing, as shown in Fig. 5*b*. Subsequently, one of the nanoparticle chains started to break just when the nanoparticle chain bridged the gap (Fig. 5*c*), and some micro bubbles were observed around the gap later (Fig. 5*d*). This indicates that a part of the water around the nanoparticle chain was suddenly heated and evaporated by Joule heating, breaking the nanoparticle chain. Approximately 4 s later, another nanoparticle chain was growing (Fig. 5*e*), and subsequently, it started to break again just when the nanoparticle chain bridged the gap (Fig. 5*f*). Some micro bubbles were observed again, as shown in Figs. 5*g* and *h*. This breaking process was repeatedly observed while voltage was applied across the gap. When a high voltage with low current was applied, some nanoparticles were also trapped at the edges of electrodes, as shown Fig. 6*a*. Subsequently, one of the nanoparticle chains rapidly grew (Figs. 6*b–d*), bridging a gap in ~4 ms. In this case, micro bubbles were not observed around the nanoparticle chain.

4. Measurement of maximum allowable current: We measured I_{max} for nanoparticle chain formation in a gap several tens of micrometres wide by applying an increasing current to a nanoparticle chain. We fabricated a 5–30 μm-wide gap between gold electrodes on a glass substrate, and each gap was covered with an aqueous dispersion of 150 nm-diameter gold nanoparticles. We applied an AC voltage (V_{gap} and V_{all}/R_{res} were 15 V_{rms} and 4.9 mA_{rms}, respectively) to each gap between electrodes in the same manner as shown in Fig. 2. We confirmed that a nanoparticle chain was formed in each gap, as shown in Fig. 7. Subsequently, the electrodes were connected to a source

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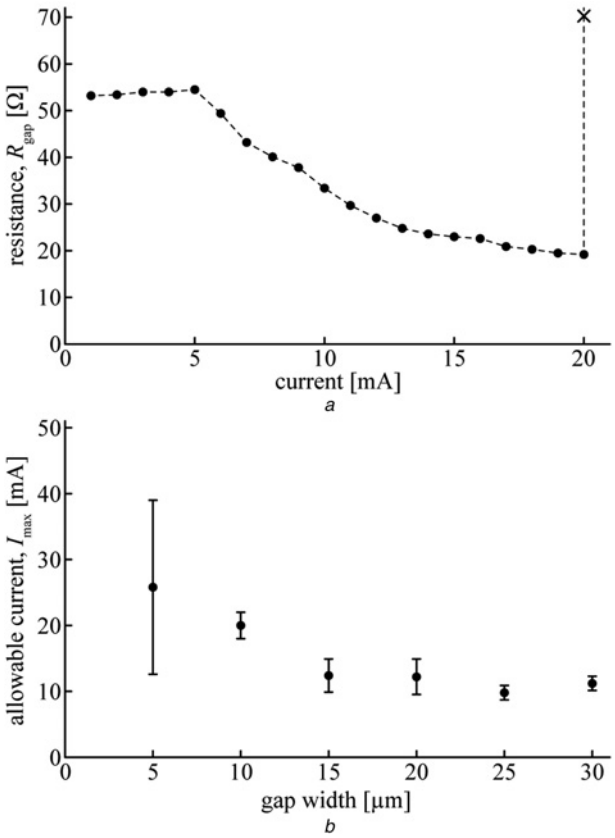


Fig. 8 *Measurement of maximum allowable current, I_{max}*
a Resistance of electrodes and nanoparticle chain, R_{gap}, changes on increasing current in the case of a 10 μm-wide gap
b I_{max} determined by gap width (the number of trials $n = 5$)

meter (2614B, Keithley), and we applied increasing current to each nanoparticle chain while measuring the resistance of the electrodes and nanoparticle chain, R_{gap}. During the measurement, each gap was covered with the gold nanoparticle dispersion, and the current was increased gradually in steps of 1.0 mA. During each steps, the current was applied in several seconds until the value of the resistance became stable, because the value of the resistance gradually decreased in each steps. The number of trials was five for each gap width.

Fig. 8*a* shows the resistance change of R_{gap} in the case of 10 μm gap width. R_{gap} increased slightly from 1.0 to 5.0 mA. On the other hand, R_{gap} decreased as current increased beyond 5.0 mA, which indicates that a part of the nanoparticle chain was gradually melted and the contact resistance between nanoparticles decreased. R_{gap} increased sharply when the current increased from 20.0 to 21.0 mA, which indicates that a nanoparticle chain was broken by excessive current. After the measurement, we confirmed that a nanoparticle chain was broken as shown in Fig. 3*c*. Therefore, I_{max} is 20 mA in Fig. 8*a*. Fig. 8*b* shows the measurement result of I_{max} determined by gap width. I_{max} decreased as gap width increased from 5 to 15 μm. This is because the resistance of a nanoparticle chain increased as the gap width increased, which implies an increase in the length of the nanoparticle chain, and Joule heating increases even when the current is low. I_{max} was ∼11 mA from 15 to 30 μm, which indicates that I_{max} must be ≥11 mA for a gap width >30 μm. Therefore, when an aqueous dispersion of 150 nm-diameter gold nanoparticles is used, a nanoparticle chain must be formed in a gap several tens of micrometres wide when a high voltage (e.g. 15 V_{rm} or more) with current <11 mA is applied.

5. Conclusion: We examined the voltage and current conditions for nanoparticle chain formation in a gap between electrodes that is several tens of micrometres wide. In this study, an aqueous dispersion of 150 nm-diameter gold nanoparticles was used. We applied certain voltages with changing current to a 10 μm-wide gap between gold electrodes, and found that a nanoparticle chain is observed in a gap only when high voltage (6.1 V_{rms} or more) with low current (15 mA_{rms} or less) was applied. We also measured I_{max} by applying increasing current to the nanoparticle chain. The value of I_{max} decreased as gap width increased, and I_{max} was ∼11 mA for gap width in the range of 15–30 μm. This result indicates that a nanoparticle chain must be formed in a gap several tens of micrometres wide when a high voltage with current <11 mA is applied.

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