Video Article

Preparation of Janus Particles and Alternating Current Electrokinetic Measurements with a Rapidly Fabricated Indium Tin Oxide Electrode Array

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Abstract

This article provides a simple method to prepare partially or fully coated metallic particles and to perform the rapid fabrication of electrode arrays, which can facilitate electrical experiments in microfluidic devices. Janus particles are asymmetric particles that contain two different surface properties on their two sides. To prepare Janus particles, a monolayer of silica particles is prepared by a drying process. Gold (Au) is deposited on one side of each particle using a sputtering device. The fully coated metallic particles are completed after the second coating process. To analyze the electrical surface properties of Janus particles, alternating current (AC) electrokinetic measurements, such as dielectrophoresis (DEP) and electrorotation (EROT)- which require specifically designed electrode arrays in the experimental device- are performed. However, traditional methods to fabricate electrode arrays, such as the photolithographic technique, require a series of complicated procedures. Here, we introduce a flexible method to fabricate a designed electrode array. An indium tin oxide (ITO) glass is patterned by a fiber laser marking machine (1,064 nm, 20 W, 90 to 120 ns pulse-width, and 20 to 80 kHz pulse repetition frequency) to create a four-phase electrode array. To generate the four-phase electric field, the electrodes are connected to a 2-channel function generator and to two invertors. The phase shift between the adjacent electrodes is set at either 90° (for EROT) or 180° (for DEP). Representative results of AC electrokinetic measurements with a four-phase ITO electrode array are presented.

Video Link

The video component of this article can be found at https://www.jove.com/video/55950/

Introduction

Janus particles, named after the Roman god with a double face, are asymmetric particles whose two sides have physically or chemically different surface properties[1,2]. Due to this asymmetric feature, Janus particles exhibit special responses under electric fields, such as DEP[3,4,5,6], EROT[2], and induced-charge electrophoresis (ICEP)[7,8,9]. Recently, several methods to prepare Janus particles have been reported, including the Pickering emulsion method[10], the electrohydrodynamic co-jetting method[11], and the microfluidic photopolymerization method[12]. However, these methods require a series of complicated apparatus and procedures. This article introduces a simple method to prepare Janus particles and fully coated metallic particles. A monolayer of micro-scaled silica particles is prepared in a drying process and is put into a sputtering device to be coated with Au. One hemisphere of the particle is shaded, and only the other hemisphere is coated with Au[2,13]. The monolayer of the Janus particle is stamped with a polydimethylsiloxane (PDMS) stamp and then treated with a second coating process to prepare fully coated metallic particles[14].

To characterize the electrical properties of a Janus particle, different AC electrokinetic responses, such as DEP, EROT, and electro-orientation, are widely used[9,15,16,17,18,19]. For example, EROT is the steady-state rotational response of a particle under an externally imposed rotating electric field[2,9,15,16]. By measuring the EROT, the interaction between the induced dipole of the particles and the electric fields can be obtained. DEP, which arises from the interaction between the induced dipoles and a non-uniform electric field, is capable of leading to particle movement[3,4,5,9,15]. Different kinds of particles can be attracted to (positive DEP) or repelled from (negative DEP) the electrode edges, which serves as a general method for manipulating and characterizing particles in the microfluidic device. The translational (DEP) and rotational (EROT) characteristics of the particle under the electric field are dominated by the real and imaginary part of the Clausius-Mossotti (CM) factor, respectively. The CM factor depends on the electrical properties of the particles and the surrounding liquid, which are revealed from the characteristic frequency, $\omega_c = 2\sigma / aC_{DL}$, of DEP and EROT, where σ is the liquid conductivity, a is the particle radius, and C_{DL} is the capacitance of the electrical double layer[15,16]. To measure the EROT and DEP of particles, specially designed electrode array patterns are needed. Traditionally, a photolithographic technique is used to create electrode arrays and requires a series of complicated procedures, including photoresist spin-coating, mask alignment, exposure, and development[15,18,19,20].

In this article, the rapid fabrication of electrode arrays is demonstrated by direct optical patterning. A transparent thin-film ITO layer, which is coated on the glass substrate, is partially removed by a fiber laser marking machine (1,064 nm, 20 W, 90 to 120 ns pulse width, and 20 to 80 kHz pulse repetition frequency) to form a four-phase electrode array. The distance between the diagonal electrodes is 150-800 μm, which can be

adjusted to suit the experiments. The four-phase electrode array can be used to characterize and concentrate particles in different microfluidic devices[15,16,18]. To generate the four-phase electric field, the electrode array is connected to a 2-channel function generator and to two invertors. The phase shift between the adjacent electrodes is set at either 90° (for EROT) or 180° (for DEP)[15]. The AC signal is applied at a 0.5 to 4 V_{pp} voltage amplitude, and the frequency ranges from 100 Hz to 5 MHz during the operation process. Janus particles, metallic particles, and silica particles are used as samples to measure their AC electrokinetic properties. Suspensions of the particles are placed on the center region of the electrode array and are observed under an inverted optical microscope with a 40X, NA 0.6 objective. Particle motion and rotation are recorded with a digital camera. The DEP movement is recorded at the annular region, between 40 and 65 µm radially away from the array center, and EROT is recorded at the circular region, 65 µm radially away from the array center. Particle velocity and angular velocity are measured by the particle-tracking method. The particle centroids are distinguished by gray scale or geometry of particles using software. The particle velocity and angular velocity are obtained by measuring the movements of the particle centroids.

This article provides a simple method to rapidly fabricate arbitrarily patterned electrode arrays. It introduces the preparation of fully or partially coated metallic particles, which can be used in different fields, with uses ranging from biology to industry applications.

Protocol

1. Fabrication of the Microchip

1. **Preparation of the ITO electrode**
    1. Use commercial illustration software to draw a cross pattern. Set the distance between the diagonal electrodes to 160 µm and make the arms of the cross pattern 30 mm wide and 55 mm long, as shown in **Figure 1**. Save the illustration file as a DXF file.
    2. Use a glass cutter to trim the ITO glass to a size of 25 mm x 50 mm (width x length). Use 75% ethanol and DI water to rinse the ITO glass several times.
    3. Put the ITO glass onto the pulsed-fiber laser marking machine. Focus the laser on the surface of the ITO glass by adjusting the distance between the ITO glass and the laser to 279.5 mm.
    NOTE: The laser used here has the following specifications: 1,064 nm, 20 W, 90 to 120 ns pulse width, and 20 to 80-kHz pulse repetition frequency, with a pulsed-light intensity at about 5 x 10^5 W/cm^2).
    4. Directly input the illustration file (DXF file) onto the computer of the laser marking machine. Click the "Mark Parameter" button and input the following parameters: speed, "800 mm/s;" power, "60%;" and frequency, "40 kHz." Tick the "Frame," "Fill," and "Fill First" terms.
        1. Click the "Preview" button position the pattern at the ITO glass center. Click the "Mark Sample" button to pattern the ITO glass (**Figure 1A**).

2. **Setting up a four-phase generator and connecting the experimental microchip**
    1. Build the circuits of the invertors, as shown in **Figure 2A**.
    2. Connect the 4 wires on the four-phase electrode by direct contact with tape, as shown in **Figure 2C**. Divide the "Channel 1" of the function generator into two branches using a double BNC connector.
        1. Connect one branch to the wire (#1) attached to the ITO electrode and the other to the input of the invertor. Connect the output of the invertor to the wire (#3), as shown in **Figure 2B**.
    3. Connect "Channel 2" with the same procedure as in step 1.2.2, but connect to the wires (#2 and #4) as shown in **Figure 2B**.
    4. For the EROT experiments, set the phase shift between the two channels to 90°, directly on the function generator. Apply a sine wave at 0.5-4 V_{pp} voltage amplitude and a frequency ranging from 100 Hz to 5 MHz during the experiments, as shown in **Figure 2D**.
    5. For the DEP experiments, connect one branch of Channel 1 to the wire (#1) attached to the ITO electrode and the other to the input of the invertor. Connect the output of the invertor to the wire (#2). Connect Channel 2 using the same procedure, but connect to the wires (#3 and #4).
    6. Set the phase shift between the 2 channels at 0°, directly on the function generator. Apply a sine wave at 0.5-4 V_{pp} voltage amplitude and a frequency ranging from 100 Hz to 5 MHz during the experiments, as shown in **Figure 2D**.

2. Preparation of the Samples

1. **Preparation of Janus particles**
    1. Centrifuge the 2 µm silica particle aqueous suspension (10% w/w) at 2,200 x g for 1 min.
    2. Pipette 2 µL of sedimentary silica particles into a 1.5 mL microcentrifuge tube and add 500 µL of ethanol (99.5% v/v).
    NOTE: The supernatant does not need to discarded; just save it in the refrigerator at 4 °C. It does not need to be resuspended before pipetting.
    3. Sonicate the ethanol-silica particle suspension using an ultrasonicator (43 kHz, 50 W) for 1 min and then centrifuge it at 2,200 x g for 3 min.
    4. Substitute the supernatant with 500 µL of ethanol and repeat step 2.1.3 three times.
    5. Substitute the supernatant with 8 µL of ethanol and sonicate the ethanol-silica particle suspension using an ultrasonicator (43 kHz, 50 W) for 3 min.
    NOTE: About 10 µL of ethanol-silica particle suspension should remain in the tube at this step.
    6. Pipette 2 µL of ethanol-silica particle suspension and drop it on a normal glass slide (width: 25 mm, length: 75 mm, and thickness: 1.2 mm) to form a droplet.
    Note: This quantity of particle suspension is enough to prepare the monolayers for 5-6 slides (2 µL for each slide).
    7. Slowly drag the ethanol-silica particle droplet slightly with a cover glass to form a monolayer of silica particles (**Figure 3A**).
    8. Put the slide with the monolayer of silica particles into a sputtering device to be coated with Au.

1. Remove the air from the sputter chamber at 100 mTorr and inject argon for 10 min (replace the air with argon). Stop injecting argon and then remove the argon from the chamber at 70 mTorr.
2. Set the current to 15 mA for 200 s. (**Figure 3B**); Janus particles are already prepared at this step.

9. Drop 20 µL of DI water on the sputtered slide and scrape the Janus particles from the monolayer using a normal 200 µL pipette tip.
NOTE: The Janus particles scraped from the monolayer suspend in the DI water droplet at this step.
10. Pipette the Janus particle suspension droplet and drop it into another 1.5-mL centrifuge tube.
11. Use the Janus particle suspension to prepare the sample by diluting it with DI water to a suitable concentration for experiments.
NOTE: The concentration of the particle suspension in the experiments described here is about 2,000 counts/µL.

2. **Preparation of fully coated metallic particles**[14]
1. Mix the PDMS polymer base and the curing agent at a 10:1 weight ratio.
2. Tape around the glass slide to form the lateral walls of the container. Pour the mixture of PDMS onto a taped slide to achieve a PDMS layer of 2-3 mm, as shown in **Figure 4A**.
3. Put the taped slide (container) with the PDMS mixture in an airtight chamber and run the vacuum pump for 30 min to remove the bubbles in the PDMS mixture.
4. Put the taped slide (container) with the PDMS mixture (step 2.2.3) into the oven. Cure the PDMS mixture at 70 °C for 2 h to form a PDMS stamp.
5. After the PDMS stamp is cured, remove the slide and tape to get the PDMS stamp, the surface of which attached originally to the glass slide, forming a flat surface, as shown in **Figure 4B**.
6. Follow the steps from 2.1.1-2.1.8 to prepare a monolayer of Janus particles on a slide.
7. Use the flat surface of the PDMS stamp to stamp the monolayer with uniform pressure.
8. Put the PDMS stamp with the monolayer of Janus particles, which is inverted from the glass slide made in step 2.1.8, into the sputtering device to be coated with Au.
1. Remove air from the sputter chamber at 100 mTorr and inject argon for 10 min (replace the air with argon). Stop injecting argon and then remove the argon from the chamber at 70 mTorr.
2. Set the current to 15 mA for 200 s (**Figure 4C**); the fully coated metallic particles are already prepared at this step.

9. Follow the steps from 2.1.9-2.1.11 to prepare the sample for experiments.

3. Experiments for AC Electrokinetic Measurement

1. **Wrap 5 pieces of paraffin film to prepare a spacer. Combine the ITO electrode array with the film spacer of 500-µm thickness using a heat gun and place the electrode on the microscope stage.**
1. Drop 8 µL of particle suspension, which was prepared in steps 2.1 and 2.2, onto the center of the cross electrode array. Place a cover glass on the spacer.

2. **For the EROT experiments, on the function generator, set the phase shift between the 2 channels to 90°. Apply a sine wave at 0.5-4 V_{pp} voltage amplitude and a frequency ranging from 100 Hz to 5 MHz during the experiments (based on the connection in steps 1.2.2-1.2.3).**
1. Choose the wave form by clicking the "waveform" button on the function generator. Input the voltage and frequency value using the numbered buttons on the function generator and then turn on the AC signal by clicking "output" button.

3. For the DEP experiments, set the phase shift between the 2 channels to 0°. Apply a sine wave at 0.5-4 V_{pp} voltage amplitude and a frequency of 100 Hz to 5 MHz during the experiments (based on the connections in step 1.2.5) by setting up the function generator as in step 3.2.1.
4. Turn on the AC signal by clicking the "output" button and capture the images of particle motion and rotation under an inverted optical microscope with a 40X, NA 0.6 objective using a camera.
5. Input the images of particle motion and rotation to the software and analyze the trajectory of particles by particle tracking to obtain the particle and angular velocities.
Note: The "Image J" software and its "MultiTracker" plugin were used here for image binarization and particle tracking.

Representative Results

The four-phase electrode array is created by a fiber laser marking machine. The ITO conductive layer coated on the glass is removed by a focus laser to form a cross pattern with a gap of 160 µm, as shown in **Figure 1B**.

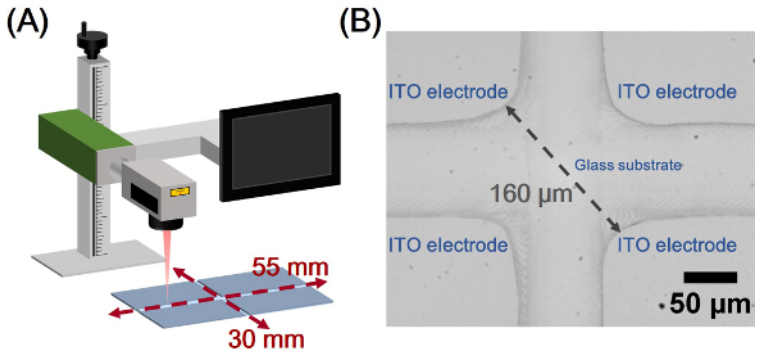

Figure 1: Preparation of the ITO Electrode. (**A**) Schematics of creating a four-phase ITO electrode with a fiber laser marking machine (1,064 nm, 20 W, 90- to 120 ns pulse-width, and 20 to 80 kHz pulse repetition frequency). (**B**) Bright-field image of the cross electrode array under a microscope. Please click here to view a larger version of this figure.

The circuit diagram of the invertor is shown in **Figure 2A**. To create the four-phase electric field, the electrode array is connected to a 2-channel function generator and to two invertors, as shown in **Figure 2B**.

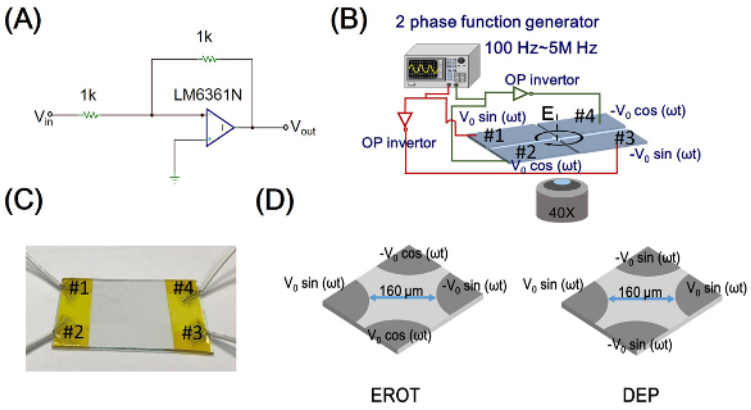

Figure 2: Setup of a Four-phase Generator and the Connection of the Experimental Microchip. (**A**) The circuit diagram of the invertor. (**B**) Schematics of the experimental microchip. (**C**) Connect the 4 wires on the four-phase electrode through direct contact with tape. (**D**) The phase shift between the adjacent electrodes is set at either 90° (for EROT) or 180° (for DEP). Please click here to view a larger version of this figure.

A monolayer of silica particles is prepared by slowly dragging the ethanol-silica particle droplet slightly using a cover glass, as shown in **Figure 3A**. The monolayer of silica particles is put into the sputtering device to be coated with Au. Finally, the Janus particles are prepared, as shown in **Figure 3C**.

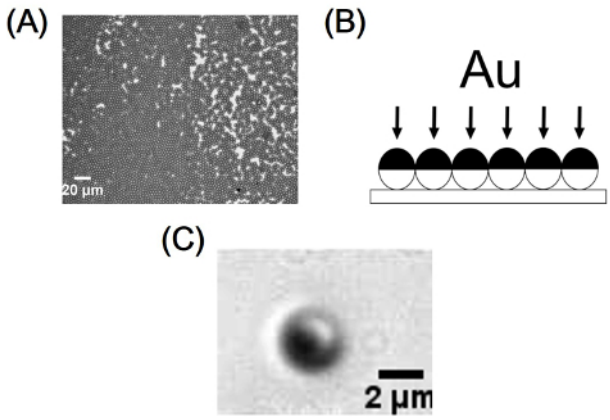

Figure 3: Preparation Procedures of Janus Particles. (**A**) The monolayer of silica particles under a microscope. (**B**) Schematic drawing of coating a thin layer of Au onto a monolayer of silica particles. (**C**) Bright-field image of a Janus particle under a microscope. The dark side of the particle is the Au coating. Please click here to view a larger version of this figure.

The preparation of fully coated metallic particles is shown in **Figure 4**. The container, which consists of a slide and tape, is loaded with the PDMS mixture to a 2 to 3 mm height, as shown in **Figure 4A**. The PDMS mixture is put into the oven at 70 °C for 2 h to form a PDMS stamp. The PDMS stamp, with a flat surface, is shown in **Figure 4B**. The preparation procedure of fully coated metallic particles is shown in **Figure 4C**. The fully coated metallic particle is shown in **Figure 4D**.

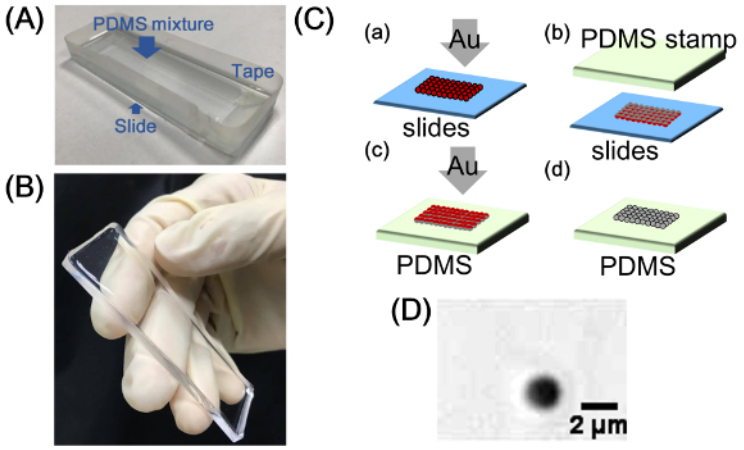

Figure 4: Preparation of Fully Coated Metallic Particles. (**A**) The PDMS mixture in the slide-tape container. (**B**) The PDMS stamp with a flat surface. (**C**) Preparation procedures of fully coated metallic particles. (**D**) Bright-field image of a fully coated metallic particle under a microscope. Please click here to view a larger version of this figure.

The representative results of particle EROT and DEP are shown in **Figure 5**. The EROT of Janus particles is generally in the opposite direction of the electric field (counter-field), with a maximum angular speed at a characteristic frequency, as shown in **Figure 5A**. The EROT of Janus particles at a low frequency reverses to the co-filed direction, which could be because of the more complicated polarization mechanism and the induced-charge electroosmotic flow around the metallic surface hemisphere[15,16]. The EROT of silica particles is co-field in all testing frequency ranges, and its characteristic frequency is at the lowest testing frequency (~500 Hz), as shown in **Figure 5B**. The EROT of metallic particles is counter-field in all testing frequency ranges, and its characteristic frequency is lower than that of Janus particles, as shown in **Figure 5C**. From **Figure 5A-5C**, we can see the change in the imaginary part of the CM factors, with the electric field frequencies between different kinds of particles. Besides, we can see that the EROT characteristic frequency of the Janus particles is higher than in fully coated metallic particles. This result suggests that the polarization of Janus particles cannot be directly explained by a simple superposition model of two-hemisphere structures. There is a more complicated mechanism for the polarization of Janus particles[2]. The DEP measurements of metallic particles are shown in **Figure 5D**.The results show that the DEP response of metallic particles is n-DEP at lower frequencies, but p-DEP at higher frequencies, with a crossover frequency that agrees with the characteristic frequency in the EROT measurement.

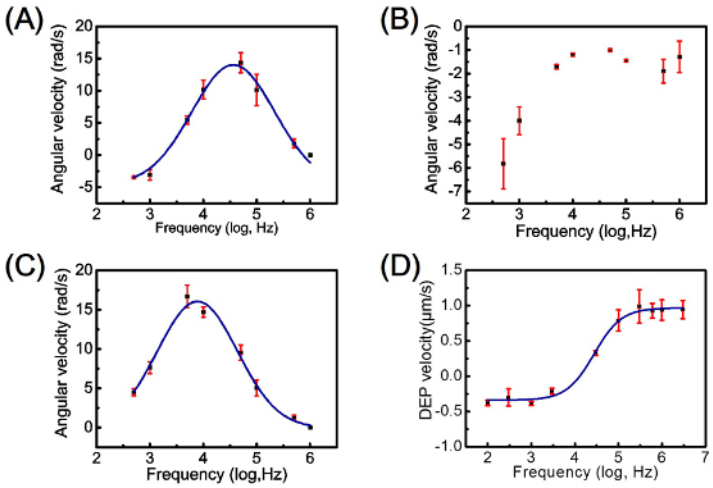

Figure 5: The Representative Results of Particle EROT and DEP. (**A**) EROT spectrum of Janus particles. (**B**) EROT spectrum of silica particles. (**C**) EROT spectrum of fully coated metallic particles. (**D**) DEP spectrum of fully coated metallic particles. Please click here to view a larger version of this figure.

Discussion

Fabricating ITO electrode arrays using the fiber laser marking machine provides a rapid method to prepare electrodes with arbitrary patterns. However, there are still some disadvantages to this method, such as fewer charge carriers and the lower fabrication accuracy of ITO electrodes compared to metal electrodes created by traditional methods. These disadvantages could limit some experiments. For example, fewer charges carriers could affect the distribution of the electric field when there is a large distance between electrodes. In addition, the adjustment of the

patterning parameters is a critical step in this method, which directly affects the quality of the ITO electrode arrays. For example, the laser power affects the removal of the ITO conductive layer from the glass substrate. The frequency and speed of the laser determine the smoothness of the ITO electrode edges. Usually, appropriate patterning parameters are found by trial and error. In short, this method is capable of rapidly and efficiently producing electrodes on ITO glass in arbitrary patterns, which can to applied to many kinds of electrical experiments, for research and other applications.

Preparing the Janus particles and metallic particles with the drying process is a simple and convenient method. In contrast to other methods, such as the Pickering emulsion method[10], the electrohydrodynamic co-jetting method[11], the microfluidic photopolymerization method[12], and the chemical synthesis method[15], this method is capable of preparing a large number of particles in a short time. However, the limitation of this method is that the metal deposition on the surfaces of the particles could be non-uniform, which could slightly change the particle shape. Although the drying process method has this limitation, it is still a useful method to prepare Janus particles and metallic particles.

In summary, this article provides functional methods to rapidly prepare electrode arrays in arbitrary patterns, as well as a large number of fully or partially coated metallic particles. This can facilitate the development and application of electrokinetics, including for the manipulation and characterization of particles in microfluidic devices.

Disclosures

The authors have nothing to disclose.

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