Gas sensors based on dielectrophoresis of graphene, and graphene oxide, and reduced graphene oxide

A Review

Queen's University

Shamim Azimi

Abstract

Dielectrophoresis (DEP) is a label-free, accurate, fast, and low-cost diagnostic technique that uses the principles of polarization and the motion of bioparticles in applied electric fields. DEP occurs when uncharged particles in the solution are subject to a spatially non-uniform alternating-current (AC) electric field, resulting in the motion of particles by creating a polarizability gradient between the particles and the suspending medium. The movement of particles in DEP is based on the difference in polarizability between the particles and the surrounding medium. If the particles move toward the electrode edge, the region of high electric field gradient, the response is called positive DEP (p-DEP). At the same time, if the particles move away from the electrode edge, the response is called negative DEP (n-DEP). This phenomenon provides a powerful and versatile tool for the non-destructive manipulation of nanoscale materials, allowing for the control of the resistance and the type of the assembly. This technique has been proven to be beneficial in various fields, including environmental research, polymer research, sensors, biosensors, microfluidics, medicine, and diagnostics. This paper reviews the fundamentals of DEP and its specific application in the incorporation of graphene, graphene oxide(GO), and reduced graphene oxide(RGO), enabling the assembly of individual two-dimensional nanostructures at predefined locations in microdevices for gas sensor applications. The review provides an essential framework for parallel fabrication approaches of graphene-based devices.

Keywords: Dielectrophoresis, Graphene materials, gas sensors.

Contents

Table of figures

1. introduction

1.1. DEP background

DEP is the force experienced by a particle in a solution due to the non-uniform electric field. This phenomenon provides a versatile and powerful tool for the manipulation of nanoscale materials such as GO, RGO, and single-walled carbon nanotubes (SWNTs). This force does not need the particle to be charged; all particles exhibit dielectrophoretic activity in the presence of electric fields. Because this process requires only mild voltages and electrodes, it may be performed on a wide variety of platforms ranging from flexible polymer substrates to lab-on-chip. The DEP force is a function of the applied field's frequency and strength, allowing for the control of the type of assembly and resistance. Thus, DEP is an ideal assembly method for the development of micro-assemblies of numerous nanomaterials of varying strength for a wide variety of resistive sensors. It allows controlled and uniform alignment of nanotubes, nanoparticles, and biomaterials for nano-device applications. In DEP, the particles carry electrical potential and respond uniquely to the different frequencies.

1.2. DEP theory

In DEP, when cells/particles are exposed to the non-uniform electric field, two different forces occur between the cells and surrounding medium. Depending on the relative polarizability between the cell and the suspending medium, the motion of the cells/particles can be in response to n-DEP or p-DEP effects. Figure 1 is an illustration of DEP phenomena. N-DEP effect occurs when the cells go toward the low electric field gradient, while the p-DEP effect occurs when the cells travel toward high electric field gradient; Both of these phenomena depend on Clausius-Mossotti (CM) factor.

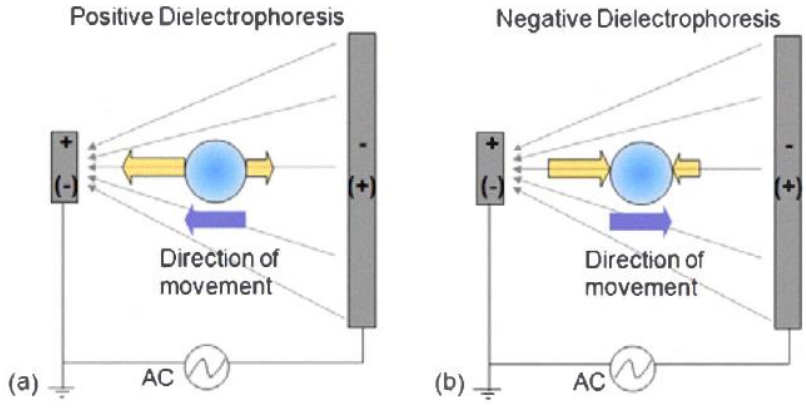

Figure 1. The principle of Dielectrophoresis[1]

DEP force applied to homogeneous sphere of a radius r in a suspension medium can be demonstrated by

Equation 1: $$<F_{DEP}> = 2\pi r^3 \varepsilon_m Re(K_{CM}) \nabla E^2$$

Where r is the radius of the particle, ε_m is the permittivity of the medium, and Re[K_{CM}] is the real part of the CM factor. According to this formula, DEP force is controlled by the CM factor, which is frequency-dependent. The CM factor can be calculated using Equation 2:

Equation 2: $$K_{CM} = \frac{\varepsilon_p^* - \varepsilon_m^*}{\varepsilon_p^* + 2\varepsilon_m^*}$$

Where ε_p^* is the complex permittivity of the particles and ε_m^* is the complex permittivity of the medium. The complex permittivity ε^* is given by the

Equation 3: $$\varepsilon^* = \varepsilon - j\frac{\sigma}{\omega}$$

Where j= $\sqrt{-1}$, σ is the material conductivity, and ω is the angular frequency. The direction of the DEP force is independent of the applied voltage and changing the voltage would not interfere with the direction of the DEP force. However, the relative polarizability of the molecules and suspending medium can be controlled by manipulating the frequency of the applied electric field.

By studying the CM factor, it was found that conductivity controls the low-frequency DEP behavior, while the permittivity controls the high-frequency behavior. Therefore, two main cases govern the relationship between $Re[K_{CM}]$ and applied signal frequency. The first case occurs when $\sigma_p < \sigma_m$ and $\varepsilon_p > \varepsilon_m$, making $Re[K_{CM}]$ negative at low frequencies and positive at high frequencies. The second case is when $\sigma_p > \sigma_m$ and $\varepsilon_p < \varepsilon_m$, then $Re[K_{CM}]$ becomes positive at low frequencies and negative at high frequencies. The point where particles do not move, and DEP force becomes zero is called zero-force frequency or cross-over frequency. This phenomenon occurs when the real part of the effective polarizabilities of the particles and the surrounding medium equal to each other ($i.e., Re[K_{CM}] = 0$). Unique characteristics of DEP make it an ideal electrical method for the deposition and manipulation of micrometer and nanometer-sized particles dispersed in a liquid. DEP has many advantages, such as controlled deposition at specific locations, low cost, the ability to be performed at room temperature, short deposition time (a few seconds), right thickness controllability, and excellent uniformity. This contrasts with other deposition approaches such as dip coating and drop-casting on pre-treated self-assembled monolayers (SAM), where it would take several hours to deposit, and it is difficult to maintain uniformity and deposition at predefined locations.[2]

1.3. Electronic properties of graphene, GO, RGO

Graphene, monolayers of sp^2 hybridized carbon atoms arranged in a honeycomb network, has recently attracted the attention of chemical sensor researchers owing to its exceptional mechanical, electrical, and chemical properties. The potentiality of ultrafast electron transport, excellent mechanical strength, along with the high surface to volume ratio has opened up the opportunities to use the material for vapor and gas sensors with long-term durability and ultrafast speed. Due to the two-dimensional structure, every atom of graphene may be considered a surface atom; as a result, every atom site may be involved in the gas interactions. This feature is responsible for its ultrasensitive sensor response with the lowest detection capability, sometimes approaching even a single molecule. Single and few layers graphene have numerous technological and scientific breakthroughs with nanodevice applications, such as chemical sensors and biosensors. The distinctive electronic structure of graphene, including π^* and π bands, has the potential to be used for the various domain of electronics. Moreover, graphene is conducting, bendable, transparent, and it is one of the most robust known materials. [3]

Every atom of graphene can be considered as a surface atom which can interact even with the molecules of the vapor or gas species; in this case, the smallest change in conduction can be measured in graphene, which eventually results in the ultrasensitive sensor response. Owing to change in free electron concentration, the electrical conductivity of graphene changes upon adsorption of the target chemical species (concentration decreases/increases if the target species act as acceptor/donor). Graphene has the advantage of low noise along with extremely high electrical conductivity, which makes the change in resistance detectable even to sub-ppm level.

On the other hand, GO is an engaging carbon nanostructure that has received attention for device applications owing to its exceptional optical and electrical properties. Its large surface-to-volume ratio, chemical stability, superior flexibility, and biocompatibility render GO an excellent material for electronic device applications. However, the controlled and precise integration of GO into a device has been challenging. Methods such as dip coating from GO suspensions, rapid freezing by spraying, and drop-casting have been employed to obtain thin films, multilayer sheets, or isolated individual films.

Due to its semi-conductive properties, RGO seems more promising as a practical gas sensor, in comparison to graphene. The function groups and active sites have been widely employed to detect NO_2, NH_3, H_2, and organic vapors.[3] Efforts have been ongoing to focus on the effect of reduction degree, chemical decoration, and device fabrication of RGO on recovery, sensitivity, and selectivity properties of detector devices. RGO is prepared in large quantities and is well-dispersed in aqueous solutions without additional surfactants. Figure 2 shows the schematic of the chemical construction of graphene, GO, and RGO.[4]

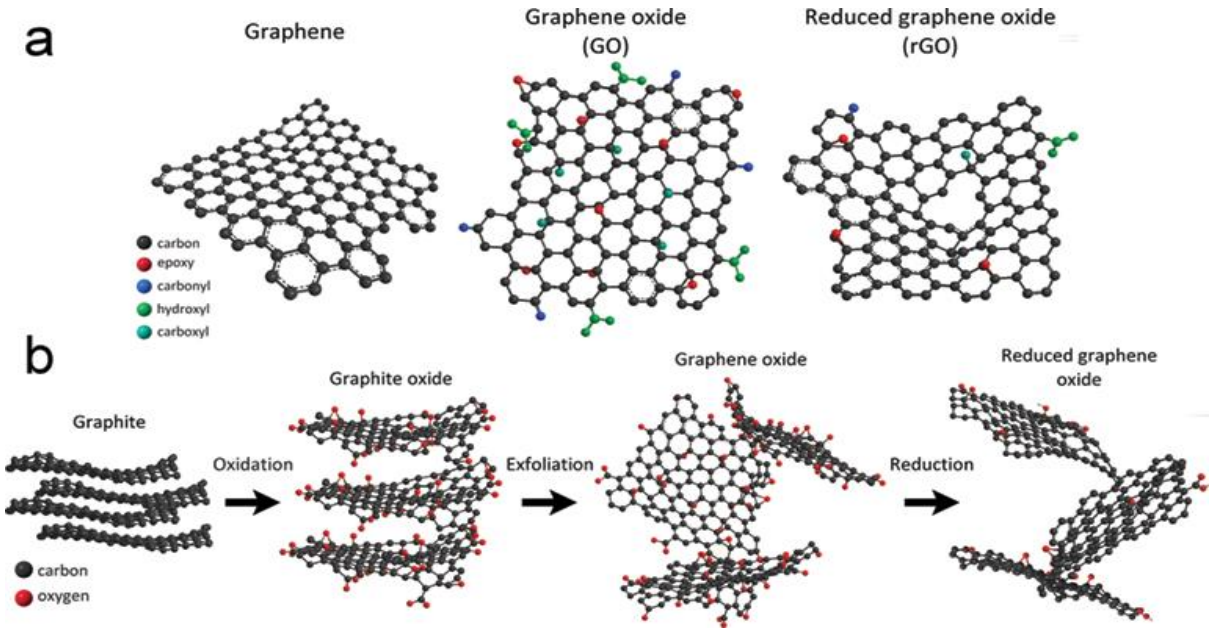

Figure 2. (a) Schematic chemical structures of graphene, graphene oxide, and reduced graphene oxide. (b) Route of graphite to reduce graphene oxide.[4]

2. Employing dielectrophoresis for graphene-based gas sensors

A major drawback in advanced processing technology is the absence of controllable, parallel assembly methods. Employing DEP, the large-scale high-yield integration and assembly of soluble ultrathin two-dimensional structures, such as dispersed graphene, is a significant step in this direction. DEP allows the directed movement or selective deposition of one-dimensional and spherical micro and nanoscale objects in nonuniform electric fields. DEP of graphene nanostructures allows the controlled manipulation of graphene nanostructure thin films over a large area with thickness ranging from a single monolayer to several layers with an excellent linking of the electrodes.[5]

Here, we describe DEP technique that allows uniform and controlled alignment of graphene, GO, and RGO for sensing applications.

2.1. Gas sensors based on GO

Singh *et al*.[6] aligned GO nanostructures between microgap electrodes via DEP for hydrogen gas sensing applications. The function generator connected with an oscilloscope was used to supply an ac voltage at a particular fixed frequency. The Ti/Au electrodes with 4μm gaps were fabricated on a SiO_2/Si substrate using photolithography and lift-off technique. Before the DEP process, the chip with Ti/Au numbering from

1 to 5 was passed through a cleaning process using acetone and ethanol and rinsed with deionized water. Afterward, the chip was dried with nitrogen gas. The GO nanostructures were synthesized by a modified Hummers method. During DEP, GO nanostructures with a concentration of 10 µg/ml in deionized water were used because the spherical droplets with GO nanostructures inside can be readily generated between the electrodes. The Vpp of 10V and 500 kHz frequency in 30s duration were the optimized DEP parameters. During DEP, the GO nanostructures were attracted randomly between the 4 µm electrode gaps and became immobilized before they were precisely aligned. After DEP processing, the sample was cleaned by isopropyl alcohol and gently dried by nitrogen flowing. Figure 3 shows the schematic representation of the experimental setup used for the DEP process.

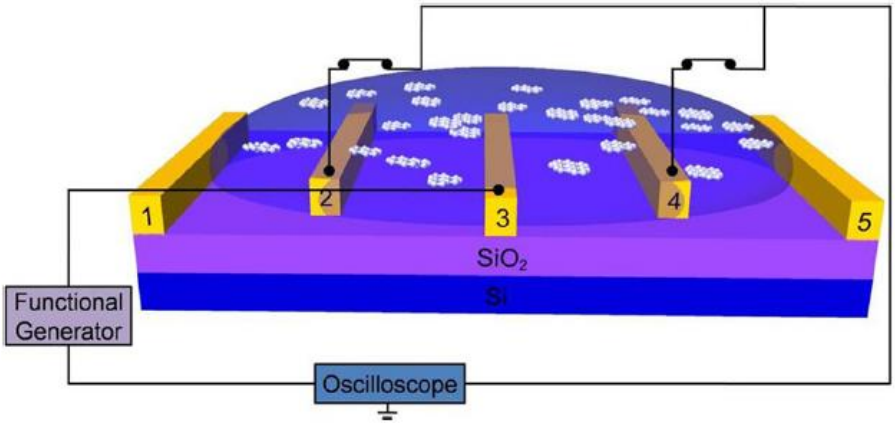

Figure 3. Schematic representation of the setup used for the DEP of GO nanostructures.[6]

The sensing behavior of the assembled GO nanostructures device was performed in a vacuum chamber by observing the change in resistance with and without gas flow. It was found that the sensitivity, response time, and recovery time for 800 ppm hydrogen gas at 300K were 6.0%, 270 s, and 306 s, respectively.

Wang *et al.*[7] fabricated a hydrogen gas sensor operating at room temperature. In this study, the GO nanostructure was assembled into microgap electrodes by controlling the DEP parameters, such as the applied frequency, applied voltage, and processing time. Fig. 4a,b, and c show the schematic representation of the experimental setup before, during, and after DEP, respectively. Fig. 4d shows a finite element method (FEM) simulated electric field profile of GO nanostructures on Si/SiO_2 substrates, where spatial electric field increases from top to bottom of the solution drop, resulting in the DEP force as shown by red arrows in Fig. 4b. A solution of 0.1 µL GO was dripped onto the Au electrodes using a micropipette, and an AC voltage using a function generator connected with an oscilloscope was applied to begin the DEP process. This field gradient was responsible for pulling the GO nanostructures from the solution towards the SiO_2 plane bounded by the electrodes, resulting in the fine GO-assembly between the electrode pair. The

optimum DEP parameters that manipulate GO nanostructures in a precise manner for hydrogen gas sensing were Vpp = 10 V, frequency = 500 kHz, and t = 30 s. The optimized device was proved to be an effective and better hydrogen gas sensor over a typical drop-dried device with a good sensing response of 5%, fast response time (<90 s), and fast recovery time (<60 s) for 100 ppm hydrogen gas concentration at room temperature.

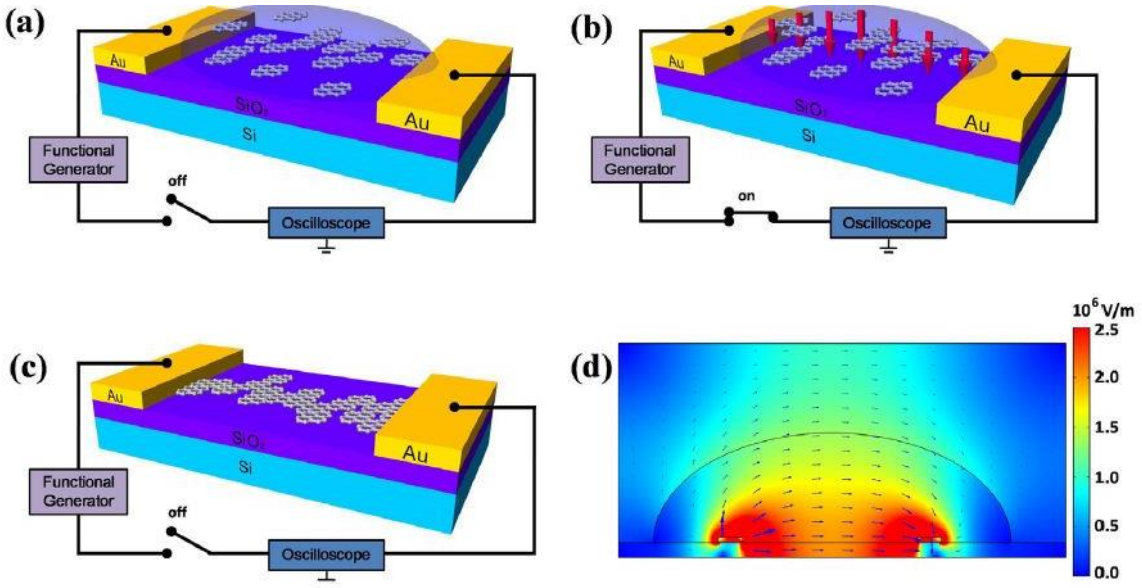

Figure 4. Schematic representation of the setup used for the DEP of GO nanostructures in a microgap of an Au electrode (a) before, (b) during, (c) after DEP. The bold red arrows in (b) shows the direction of the DEP force acting on the suspended GO nanostructures, under the influence of which the floating nanostructures got assembled between the electrodes. (d) FEM simulated electric field between a pair of coplanar Au electrodes with a solution drop showing the variation in the electric field intensity.[7]

In another study, Wang *et al*.[8] assembled GO nanostructures mixed with platinum (Pt) nanoparticles by employing DEP between the micro gap electrodes for a proficient hydrogen gas sensor. The GO nanostructures were synthesized using a modified Hummer's method. The concentration of the GO nanostructures solution was ~1 mg/mL, and the thickness of nanostructures was~ 1nm. Further, Pt nanoparticles and GO nanostructures hybrid solution was prepared by mixing 1 mg/mL GO nanostructures solution with 0.01 mmol/L Pt nanoparticle solution, in a ratio of 1:1 followed by ultrasonication. The reduction of GO (making RGO) was carried out by annealing the Pt-GO nanohybrid at 400 °C for 30 min. The gas sensor device was fabricated using the DEP technique to assemble the Pt-GO nanostructures into 4 μm gap Ti/Au electrodes. Figure 5A shows the schematic of the experimental setup for the DEP processing, with the use of a function generator and an oscilloscope. The function generator and the oscilloscope were connected to the chip by a series connection. Subsequently, 0.1 μL of the mixed solution was dropped on to the electrode area. As shown by arrows in Fig. 5A, during the DEP process, a gradient

in the electrical field was established toward the micro gap electrodes. The device fabricated with an applied frequency of 500 kHz, a processing time of 30 s, and a Vpp of 5V exhibit the best sensitivity to hydrogen gas.

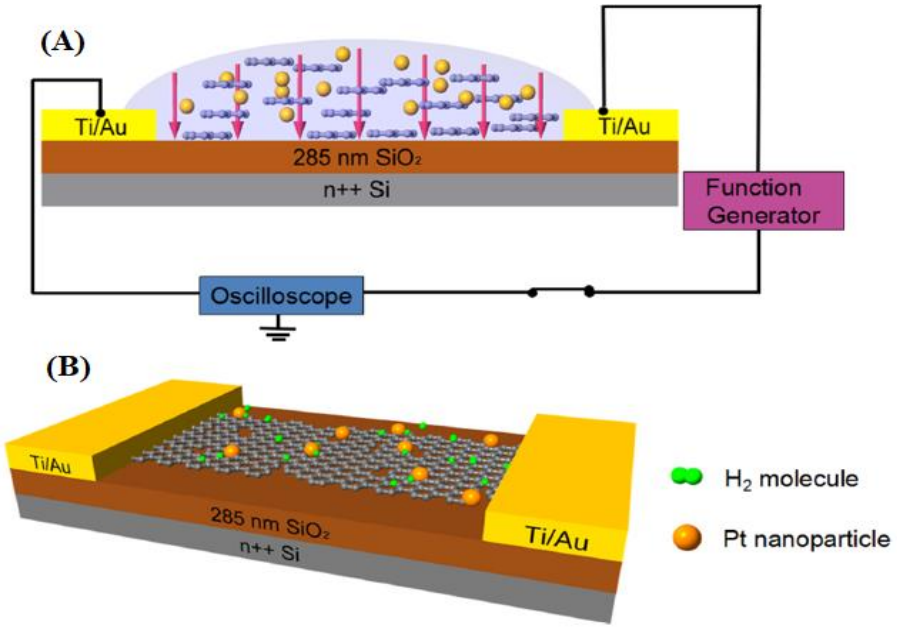

Figure 5. A) Schematic of the experimental setup for DEP processing. B) Schematic showing assembled GO nanostructures and Pt nanoparticles between microgap electrodes. The catalyst action of Pt nanoparticles also results in enhanced sensing response.[8]

2.2. Gas Sensor based on RGO

Improved gas sensing behavior was recently observed in RGO-based gas sensors decorated with rare metals. Li *et al*.[9] developed a NO sensor device using DEP of the palladium-decorated RGO (Pd-RGO) nanosheets on chemical vapor deposition (CVD)-grown graphene electrodes. The advantages of Pd decoration and CVD-grown graphene contacts of RGO in improving the sensitivity and stability of NO sensors have been revealed by performing comparison experiments of various devices. Figure 6 shows the schematic illustrations of the process of Pd-RGO composite preparations. (a) the device of Pd-RGO with CVD-grown graphene contacts (graphene-Pd-RGO) fabrication and sensor measurement. (b) RGO dispersion samples were synthesized through the reduction of GO, and GO was prepared by a modified Hummer's method. Sensor devices were fabricated by conventional photolithography and lift-off techniques on SiO_2/Si substrates. The nickel electrode patterns were deposited by magnetron sputtering. The growth of graphene on Ni electrodes was implemented by means of the CVD process using methane as a carbon source. DEP was employed with a voltage of 10 Vpp, frequency of 10 kHz, and t=30s. The highly sensitive,

recoverable sensor was able to detect NO gas ranging from 2 to 420 ppb with the response time of several hundred seconds at room temperature.

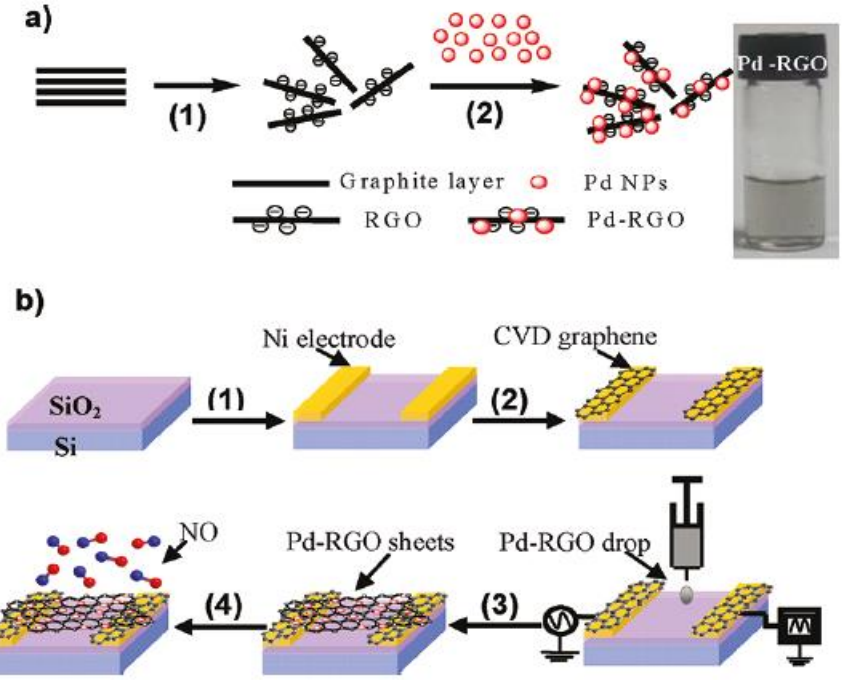

Figure 6. (a) Schematic illustration of the process for preparation of Pd-rGO composites: (1) RGO synthesis and (2) Pd decoration of RGO. The inset photograph is the diluted Pd-RGO nanosheet suspension used for ac-DEP. (b) Schematics of graphene-Pd-RGO device fabrication and gas sensing test: (1) Ni electrode fabrication, (2) chemical vapor deposition (CVD) growth of graphene, (3) ac-DEP of Pd-RGO nanosheets, and (4) sensor measurement.[9]

In another study, Wang *et al.*[10] fabricated RGO gas sensors functionalized with platinum (Pt) nanoparticles. A DEP technique was used for the precise alignment of the Pt-RGO nanohybrid between microgap electrodes, proceeded by the mid-temperature thermal annealing. GO nanostructures were synthesized using a modified Hummer's method. Prior to the DEP process, the GO solution was ultra-sonicated for 24 h to reduce the size of GO flakes and then filtered. Approximately, 1nm-thick, few microns-wide GO nanostructures were obtained by using the method. Then, 1mL of Pt nanoparticle was mixed into 5mL GO nanostructure solution. The mixed solution was ultrasonicated for ~1 h, and 0.1 µl of the resulting well-mixed solution was dropped on the microgap electrode. For the DEP process, the optimum parameters of 5V, 500 kHz, and 30s were chosen as the controlling parameters for the DEP process. Fig. 7a–c shows the schematic of the sample setup before, during, and after the DEP process, respectively. The red arrows in Fig. 7b show the downward exertion of the DEP force resulting from changes in the electric field. To explain these changes, the finite element method (FEM) was used to simulate the electric field between the

Au electrodes. Fig. 7d shows the result of this FEM simulation. The tested device exhibited sensitivities of 14%, 8%, and 10%, for 1000 ppm hydrogen, ammonia, and nitric oxide gases, respectively, at room temperature.

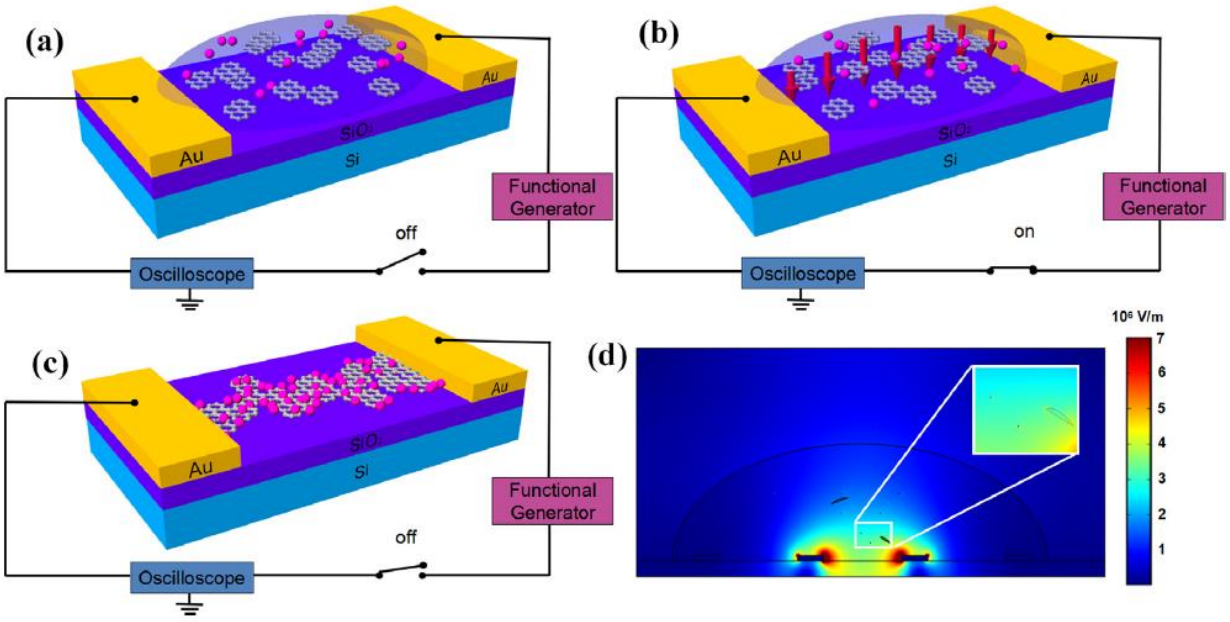

Figure 7. Schematic of the GO nanostructure assembly process (a) before, (b) during, and (c) after DEP; the pink balls represent the Pt nanoparticles. (d) FEM simulation result of the electric field between the Au electrodes.[10]

In another work, Wang *et al.*[11] developed a chemo-resistive hydrogen gas sensor based on thermally reduced GO on a micro-hotplate by positive DEP. The functionalized GO nanostructures were synthesized by a modified Hummers method. Figure 8(a) shows the schematic representation of the experimental setup used for a DEP assembly, which consists of a function generator connected directly through a chip to the oscilloscope. The micro hotplate was composed of 150 µm diameter integrated electronic devices (IEDs) and heaters, which can attain a maximum temperature of 600 °C. The operating temperature of the RGO sensor varied using the heater and recorded by a thermocouple connected to a digital display multimeter. The optimized DEP parameters for manipulating RGO nanostructures into Au electrodes for hydrogen sensing were applied frequency=1 MHz, peak-to-peak voltage= 5V, and DEP time= 30 s. Figure 8(b) shows the schematic representation of the experimental setup for the sensing measurement. A driving circuit was used to maintain the temperature of the micro-hotplate. The device exhibited good sensitivity (~6%) with fast response time (~11 s) and recovery time (~36 s) for 200 ppm hydrogen gas at room temperature.

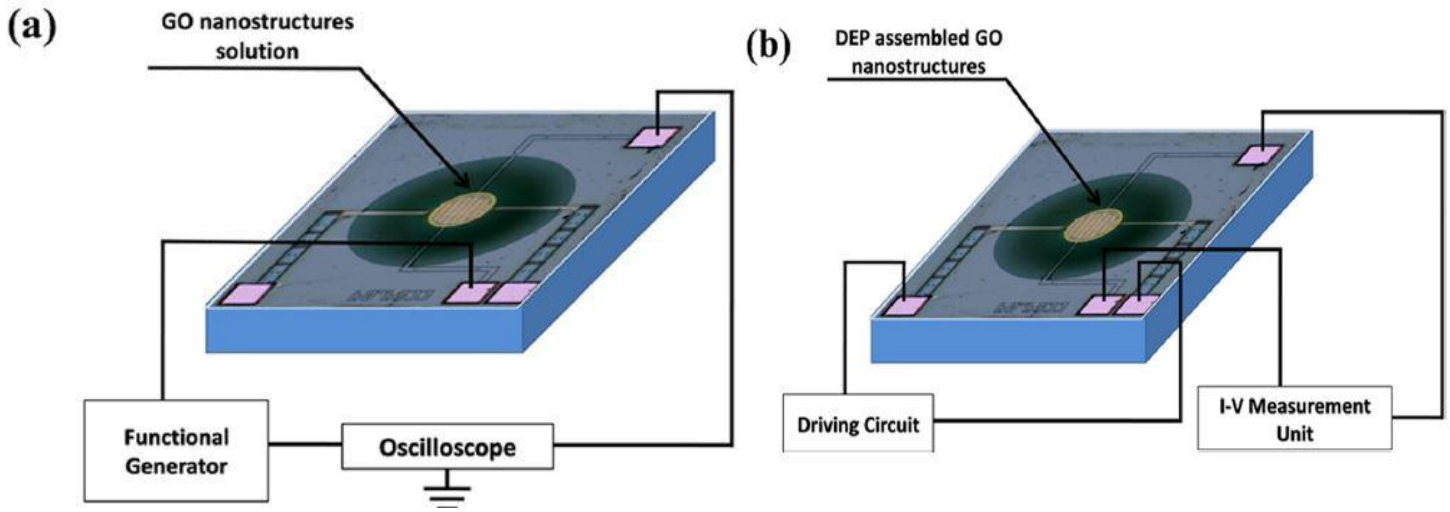

Figure 8. Schematic representation of experimental setup used for (a) DEP of rGO nanostructures into Au electrodes, and (b) hydrogen-gas sensing at different operating temperatures. Driving circuit maintains the temperature of the micro-hotplate.[11]

Hassan *et al*.[12] reported a GO-based gas sensor for sensing fast pulses of volatile organic compounds with a high signal-to-noise ratio. The device was fabricated on a silicon wafer (525 µm thick, 1–10 Ω.m) with a 280 nm-thick thermal oxide. The interdigitated electrode (IDE) layer is composed of a sputter-deposited 25 nm-thick Cr adhesion layer and a 200 nm-thick Au layer, patterned via lift-off, as shown in Figure 9a,b. An insulating oxide layer was deposited by plasma-enhanced chemical vapor deposition. The IDEs were then exposed by opening circular windows (1.3 mm diameter) in the insulation layer using photolithography and buffered oxide etching. An aqueous GO dispersion (500 mg/L) was produced via Hummer's method. GO was deposited across IDEs via the solvent evaporation method or the DEP method. For the DEP deposition of GO film, a 20 V peak-to-peak and 1 MHz square wave signal were applied for 10 min using a signal generator. GO reduction was carried out by placing GO-deposited chips next to a vial containing 50 µL liquid hydrazine hydrate. To investigate the impact of DEP, the sensor was also provided with solvent evaporation of GO, and the results were compared in terms of morphology and their sensing application. Solvent evaporation results in a discontinuous and random deposition, while film deposited by DEP looks continuous and uniform. GO coated with DEP led to a higher response(%), sensitivity, and better repeatability for the detection of acetone. Thus, it could be an appropriate and simple method to improve the signal-to-noise ratio in RGO sensors.

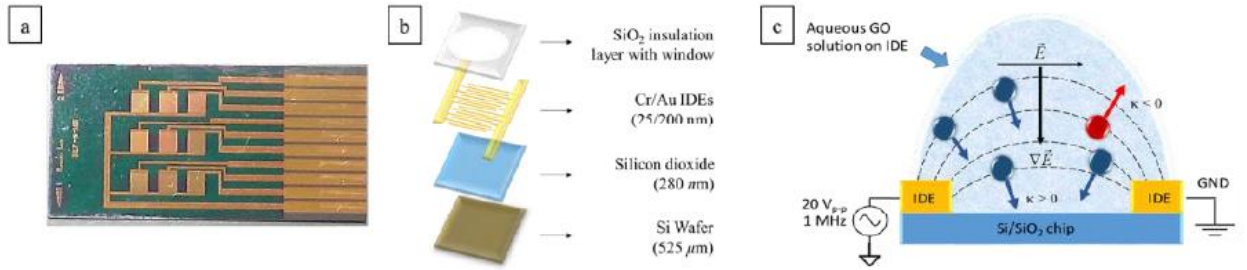

Figure 9. Figure 9. (a) Image of the chip with a 3 3 array of IDEs. The chip is 2.8 cm in length and 1.4 cm in width. Circular windows of 1.3 mm diameter were opened in the insulator layer over each IDE to expose electrodes for GO deposition. (b) Exploded view of the IDE fabricated on a silicon/silicon dioxide substrate consisting of a Cr/Au IDE pair coated with an oxide layer. (c) Depiction of the non-uniform electric field applied horizontally during DEP deposition of GO.[12]

Surwade *et al*.[13] developed a gas sensor based upon an array of RGO and SWNT micro-assemblies. Due to the nature of their structure and exceptional surface-to-volume ratio, graphene sheets and carbon nanotubes demonstrate unique chemisorption properties, providing higher sensitivities than bulk material. Micro assemblies of RGO platelets and SWNT's were created on lithographically patterned electrode arrays utilizing DEP. Both types of assemblies (RGO and SWNT) demonstrated unique and repeatable responses to various organic compounds and other vapors. The DEP-based assembly process was used for the integration of RGO and SWNTs on silicon. First, the RGO and SWNT dispersions were probe sonicated for two minutes. A small drop of the desired dispersion was placed on the chip, covering the assembly site. A function generator was used to supply the electric field necessary for DEP. SWNTs were assembled at frequencies ranging from 100kHz to 10 MHz; RGO was assembled in the range of 50kHz to 500kHz. The applied voltage varied from 2.5-10 V peak-to-peak for both materials. After approximately five minutes of assembly, the drop of suspended RGO or SWNTs was blown off using compressed dry air. DEP successfully created assemblies of SWNTs or RGO across 100% of all electrodes attempted. Figure 10 shows the schematic of the process to develop the micro-assembly sensor. The sensors responded quickly: a steep step in resistance was observed within seconds after the introduction of the gas vapor. The sensor showed consistent changes to gases when introduced into a vapor environment. Fractional changes in resistance in the range of 5-40% were observed depending on the gas environment. These responses were repeatable and consistent between chips.

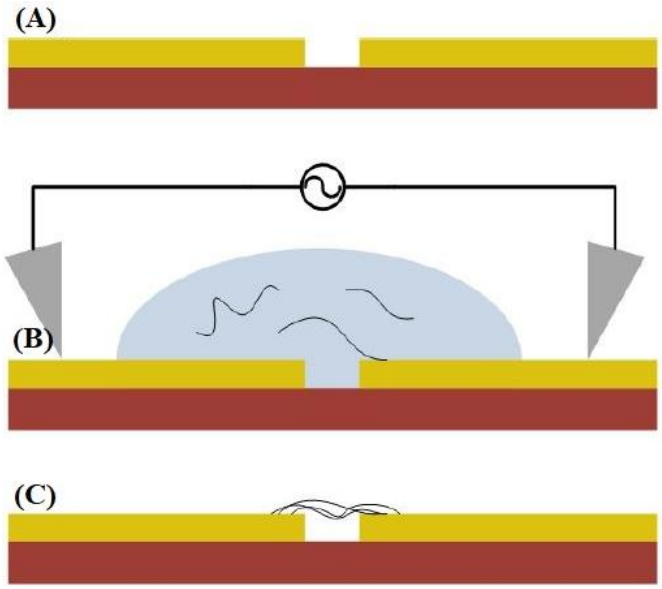

Figure 10. Process to create micro-assembly sensors. Gold electrodes patterned on silicon (A) are submerged in a dispersion of either SWNT's or RGO. An AC voltage is applied (B), and the drop is blown off to produce micro-assemblies (C).[13]

Conclusion

DEP is a fast, accurate, robust, cost-effective, and label-free assembly technique, allowing for the development of micro-assemblies of numerous nanomaterials and the control over the type of assembly and resistance. Moreover, this technique offers selectivity according to the size, dielectric properties, and surface electrical charge of the particle. This makes DEP and ideal tool for the manipulation of nanotubes, nanoparticles, and biomaterials for nanodevice applications. This study demonstrates the advantages of controlled alignment of graphene, GO, and RGO by employing DEP for the development of gas sensors with a high level of selectivity and low detection limit. It is expected that using the potentials of DEP can make progress in diagnostics and monitoring scenarios to develop accurate, robust, and more convenient nanodevices.

References

[1] Yue, T., Nakajima, M., Kojima, M. and Fukuda, T., 2011, November. Arbitrary microstructure fabrication embedding yeast cells patterned by dielectrophoresis. In *2011 International Symposium on Micro-NanoMechatronics and Human Science* (pp. 198-203). IEEE.

[2] Sharma, A. and Jang, J., 2019. Flexible electrical aptasensor using dielectrophoretic assembly of graphene oxide and its subsequent reduction for cardiac biomarker detection. *Scientific reports*, *9*(1), pp.1-10.

[3] Pearce, R., Iakimov, T., Andersson, M., Hultman, L., Spetz, A.L. and Yakimova, R., 2011. Epitaxially grown graphene based gas sensors for ultra sensitive NO2 detection. *Sensors and Actuators B: Chemical*, *155*(2), pp.451-455.

[4] Amieva, E.J.C., López-Barroso, J., Martínez-Hernández, A.L. and Velasco-Santos, C., 2016. Graphene-Based Materials Functionalization with Natural Polymeric Biomolecules. *Recent Advances in Graphene Research*, pp.257-298.

[5] Burg, B.R., Lütolf, F., Schneider, J., Schirmer, N.C., Schwamb, T. and Poulikakos, D., 2009. High-yield dielectrophoretic assembly of two-dimensional graphene nanostructures. *Applied Physics Letters*, *94*(5), p.053110.

[6] Singh, B., Wang, J., Rathi, S. and Kim, G.H., 2015. Alignment of graphene oxide nanostructures between microgap electrodes via dielectrophoresis for hydrogen gas sensing applications. *Applied Physics Letters*, *106*(20), p.203106.

[7] Farahani, A., Sereshti, H., Tashakori, C., & Azimi, S. 7th Iranian Biennial Chemometrics Seminar, 30-31 October 2019.

[8] Wang, J., Rathi, S., Singh, B., Lee, I., Joh, H.I. and Kim, G.H., 2015. Alternating current dielectrophoresis optimization of Pt-decorated graphene oxide nanostructures for proficient hydrogen gas sensor. *ACS applied materials & interfaces*, *7*(25), pp.13768-13775.

[9] Li, W., Geng, X., Guo, Y., Rong, J., Gong, Y., Wu, L., Zhang, X., Li, P., Xu, J., Cheng, G. and Sun, M., 2011. Reduced graphene oxide electrically contacted graphene sensor for highly sensitive nitric oxide detection. *ACS nano*, *5*(9), pp.6955-6961.

[10] Wang, J., Rathi, S., Singh, B., Lee, I., Maeng, S., Joh, H.I. and Kim, G.H., 2015. Dielectrophoretic assembly of Pt nanoparticle-reduced graphene oxide nanohybrid for highly-sensitive multiple gas sensor. *Sensors and Actuators B: Chemical*, *220*, pp.755-761.

[11] S. Azimi, A. Farahani, H. Sereshti, Plasma-functionalized Highly Aligned CNT-based Biosensor for Point of Care Determination of Glucose in Human Blood Plasma, Electroanalysis 32 (2) (2019) 394–403..

[12] Hasan, N., Zhang, W. and Radadia, A.D., 2017. Few-flakes reduced graphene oxide sensors for organic vapors with a high signal-to-noise ratio. *Nanomaterials*, *7*(10), p.339.

[13] MacNaughton, S., Sonkusale, S., Surwade, S., Ammu, S. and Manohar, S., 2010, November. Carbon nanotube and graphene based gas micro-sensors fabricated by dielectrophoresis on silicon. In *SENSORS, 2010 IEEE* (pp. 894-897). IEEE.