

# A Review on Effects of Electrophoretic Deposition Parameters over Graphene Related Materials Coating

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**Abstract:** This review consists basic parameters of Electro Phonic Deposition that indulges in the graphene-related material coating over the base material. A detailed study on the deposition bath is done and its effect on the efficiency of the coating is established. Major material switch increase the mechanical property (corrosion resistance, wear rat infraction coefficient, interfacial shear strength) of base metal are Nickle, cobalt, copper, tungsten, HAP. Structural characterizes the composition of the bath, temperature, current density, time for deposition play a vital role in the performance of the GRM sover the basemetals. I norder to confirm their impact material characterization techniques like SEM, FESEM, AFMEDS, XRD, FTIR,Raman Spectro scopy,Vickers hardness test, Friction test using sliding ball. Generally, SEM is tested for surface morphology,EDS for checking the amount of Graphene platelets coated ,XRD view grain size and crystal orientation, Vickers hardness to measure the hardness value after and before coating, the Tribological behavior are tested via friction test using sliding ball over the specimen.

**Keywords:** - EPD, Graphene, GRMs.

## I. INTRODUCTION

Graphene related materials, including graphene, few-layer edgraphene, graphene oxide and reduced graphene oxide are building blocks with outstanding properties which will contribute to researchers[1].Electrophoretic deposition (EPD) is developed by the Russian scientist Ruess observed an electric field induced movement of clay particles in water. EPD in its simplest form requires the application of electrical potential to electro demerged in nano particle dispersion. Two sub-processes take place: (1) electrophoresis movement of suspended charged particles in a liquid phase towards an electrode under the effect of an electric field; (2) collection of the particles at the electrode surface and formation of a deposit [3].

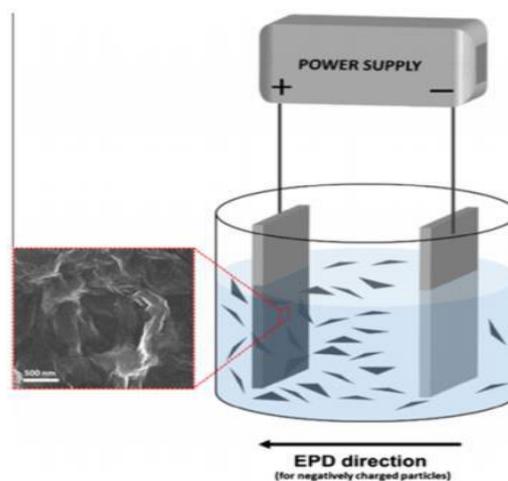


Fig. 1.

### A. Advantages

- Cost-effective[2]
- Simple equipment[2]
- Size-scalability [3-5]
- Uniformity of deposit[6]
- Dense packing of deposit[7]
- High and controlled deposition rate[8]
- Predictable deposition kinetics [7]
- Controlled thickness of deposits [6]
- Deposition at room temperature [4,7]
- Possibility of sequential- and co-deposition of different particles[9-11]
- Possibility of continuous production [4,12,13]
- Possibility to produce porous graphene-based deposits[14]

### B. Limitations

- Need for a stable suspension with sufficient particle surface charge[6]

- Need for electrically conductive substrate [6]
- Limited thickness of insulating deposits (e.g. GO)[12]
- Possibility of side electrochemical reactions [7]

Some of the Side Reactions Which Would take Place are

- Dissolution of electrodes
- Reduction of GO
- Degradation of suspension media (bubble formation)
- Electrochemical reactions of additives.

## II. PREPARATION METHODS OF GRAPHENE OR GRAPHENE OXIDE

GO was synthesized from natural graphite by the Hummers method or else with the new modified Hummers method. Once graphene oxides are generated from graphite, Graphene was synthesized via chemical reduction of GO by using hydrazine hydrate as a reducing agent. GPLs are also directly procured from the certain organization.

### A. Effects of EPD Parameters

In addition to the basic role of EPD parameters in determining EPD kinetics and deposition yield (film thickness), they can also affect other micro structural characteristics of GRM deposits:

### B. Effects of Voltage

Generally, increasing EPD voltage is reported to increase the extent of wrinkling and the film roughness [13], as well as generating porosity and heterogeneity through bubble formation via electrolysis in water [7]. Non-uniformity of deposits at high voltages may also originate from the anisotropy of the electric field on the substrate, leading to preferential deposition at the electrode edges [14], the possible formation of aggregates in suspension, or Joule heating effects on relatively insulating deposits [15]. On the other hand, too low a voltage will not generate a high-quality film; so in some ranges, increasing the EPD voltage leads to a reduced porosity. For example, one study [16] reported reduced porosity of GRM deposits when the EPD voltage was increased from 5 to 20 V. This phenomenon was attributed to the enhanced deposition rate of GRM sheets at the higher voltage leading to a greater packing between depositing particles and consequent void filling between them.

### C. Effects of Deposition Time

Another parameter that can affect the microstructure of the EPD GRM films is the deposition time [17,18,19]. Different types of GRM sheets have been reported to show different size-dependent electrophoretic mobility, leading to graded deposition [18]. Longer deposition times have been also reported [7,17] to lead to the higher surface roughness of GRM films, possibly due to agglomeration within the suspension medium during EPD. For similar reasons, an increase of deposition time above a certain level can lead to deposition of non-uniform thick coatings with insufficient attachment to the substrate [7,20], or with varying porosity [21].

### D. Effects of Particle Concentration

Increasing the particle concentration in suspension has been reported to enhance the surface roughness of GRM films [7]. Highly concentrated suspensions of GRM sheets tend to suffer from low stability and poor dispersion, leading to greater roughness. For example, one study [14] reported that increasing the concentration of GO sheets in suspension (1 mg mL<sup>-1</sup>) negatively affected the quality of films; the agglomerates in solution disturbed the substrate surface coverage and introduced lumps in the deposit.

### E. Effects of Suspension PH

Suspension pH can modify dispersion state, side reactions, and deposition mechanisms. In a study by Hasan et al., the microstructure of GO films was modulated by using acidic or basic conditions [22]. When basic conditions (pH = 11.5) were used, GO sheets were deposited on the anode with a parallel alignment to the electrode's surface. The films had a rug-like structure consisting of mainly flat but partially wrinkled GO sheets. On the other hand, in acidic conditions (pH = 2.8), the GO sheets formed face-to-face stacked multilayers in the suspension and were mainly deposited on the cathode with a brick-like morphology. When the pH is decreased prior to EPD, the net surface charge of the GO sheets decreases leading to an inferior interparticle electrostatic repulsion. In this condition, a face-to-face restacking of GO sheets is favored due to their shape. During EPD experiments, hydrolysis occurs which further decreases the pH of suspensions. The GO sheets in the basic suspension have a negative net charge, even during EPD, leading to anodic deposition. On the other hand, GO sheets in the acidic suspension will experience a charge reversal during EPD and therefore the deposition occurs mainly on the cathode. The pH-dependent face-to-face stacking of GO sheets has been

also reported for GO–chitosan mixed suspensions, leading to the formation of rougher deposits during EPD [23].

#### F. Effects of Graphene Functionalization and Treatment

In an EPD process, the alignment of GRM sheets within a deposit can be altered by fictionalization of sheet surfaces[24,25]. Different fictionalization strategies can lead to different characteristics of edge or basal planes of GRM sheets. Yang et al[25].compared the effects of metal ion and polyelectrolyte fictionalizations' on the EPD of rGO sheets. During EPD, the positively charged edges of the Mg<sup>2+</sup>-decorated rGO sheets preferentially moved to and deposited on the cathode. This process led to the vertical alignment of the rGO sheets in the deposit and production of a porous deposit with a relatively rough surface. Similarly, other studies[26,27]reported that vertically aligned sheets were observed in the EPD of Mg<sup>2+</sup>-modified GO. In contrast, when the rGO sheets were decorated with a polyelectrolyte like PDDA, an in-plane deposition of the rGO sheets and subsequent production of a relatively smooth deposit were observed due to the presence of PDDA on the basal plane. In addition to the effects of chemical decoration already mentioned, other types of treatments can also affect the microstructure. For example, in one study[28], pre-EPD ozonization of GO was reported to change the interlayer distance between the deposited GO sheets by varying the GO oxygen functionalities. The results suggest that the types of GO functional groups may play a more important role in determining the interlayer distance than the quantity of oxygen groups.

#### G. Effects of Incorporation of Other Nanoparticles

In order to produce porous GRM structures, and to limit layer–layer restacking, several studies have used nanoparticles (i.e. carbon black[29]), CNTs [30], partially-unzipped carbon fibres[31] , ZnS[32] , CoFe<sub>2</sub>O<sub>4</sub>[33] , Ni[34] as inter-layer spacers between deposited GRM sheets. Lee et al. reported that EPD using mixed suspensions of rGO and layered metal oxide nanosheets led to the formation of porous morphologies via a house-of-cards-type stacking. In general, the additional nanoparticles in GRM films also increase the films' surface roughness, since GRMs are characteristically thinner/flatter than most other materials, at least if folding, restacking and agglomeration are avoided. It is worth noting that the presence and interaction of secondary non-GRM particles in a suspension can affect the deposition rate. For example, Bae et al[28].reported that enhanced interaction of functionalized GO with Sn nanoparticles decreased the GO electrophoresis mobility and the deposition yield.

#### H. Effects of Post-EPD Treatment

Different results have been reported for post-EPD treatment of GRM structures. On the one hand, several studies [7]have reported that post-EPD reduction or thermal annealing of GRM films leads to compaction, surface smoothing and a decrease of the interlayer spacing of the deposited GRM films and fibers through the removal of trapped water molecules and/or oxygen-containing functional groups. Formation of crevices[34], protruding GRM sheets edges, and flaky surface shave been also reported as the result of post-EPD treatment. On the other hand, some studies have not observed significant micro structural changes after post-EPD treatment of GRM films. For example, Akhavan et al[35].reported that post-EPD reduction of GRM films with vertically aligned sheets does not alter the films' microstructure. Therefore, it can be concluded that the effects of any post-EPD treatment on the deposits' microstructure depend on the initial deposit morphology. For example, if the initial film morphology is porous, gases produced by a post-EPD reduction, or simply a rapid drying process, may readily escape without affecting the films' microstructure. In addition,the amount of water (or other solvent/thermally volatile) molecules trapped between the deposited GRM layers, or the rate of heating during annealing, can determine the degree of micro structural changes induced by post-EPD treatments [8].

#### I. Other Factors

The EPD mode and type of electrodes used are also important factors. Firstly, in constant-current EPD, the film surface was reported to be significantly smoother compared to constant-voltage EPD[15]. However, further study would be necessary to deduce the difference in mechanisms involved. Secondly, the type of working electrodes will control electrical conductivity, microstructure and surface roughness[8]which affect the deposition process. These factors can alter the deposition rate and gas evolution at the substrates' surface and therefore affect the deposits' microstructure and thickness. 108 M. Diba et al. / Progress in Materials Science 82 (2016) 83–117 In recent studies, mechanical properties of carbon fiber-reinforced composites were improved by electrophoretically depositing GO onto carbon fibers. It was also found that ultrasound-assisted EPD produces thicker (160 nm vs 70 nm) and rougher (97.3% vs 37.4%) coatings of GO on de-sized carbon fibers[36] as compared to conventional EPD. In this case, a continuous 60 W at 40 kHz ultrasound was applied onto the EPD bath during the deposition process.[36]

### III. CONCLUSION

Relatively few studies have compared the microstructures of GRM films prepared by EPD to films prepared by other methods. Some researchers have reported that films prepared by EPD have a rougher surface [17] and a lower degree of binding [12], compared to the films prepared by vacuum filtration, whilst others report a similar morphology for the two techniques [8]. On the other hand, EPD deposits were found to be more uniform than rGO films made by drop-casting, which consisted of large clusters possibly due to Marangoni effects during drying, or by an AC discharge method [37].

### REFERENCES

- [1]. Huang X, Yin Z, Wu S, Qi X, He Q, Zhang Q, et al. Graphene-based materials: synthesis, characterization, properties, and applications. *Small* 2011;7:1876–902.
- [2]. Besra L, Liu M. A review on fundamentals and applications of electrophoretic deposition (EPD). *Prog Mater Sci* 2007;52:1–61.
- [3]. Van der Biest OO, Vandeperre LJ. Electrophoretic deposition of materials. *Annu Rev Mater Sci* 1999;29:327–52.
- [4]. Oakes L, Hanken T, Carter R, Yates W, Pint CL. Roll-to-roll nanomanufacturing of hybrid nanostructures for energy storage device design. *ACS Appl Mater Interfaces* 2015;7:14201–10.
- [5]. Shang Y, Zhang D, Liu Y, Liu Y. Simultaneous synthesis of diverse graphene via electrochemical reduction of graphene oxide. *J Appl Electrochem* 2015;45:453–62.
- [6]. Chavez-Valdez A, Shaffer MSP, Boccaccini AR. Applications of graphene electrophoretic deposition. A review. *J Phys Chem B* 2013;117:1502–15.
- [7]. Diba M, García-Gallastegui A, Klupp Taylor RN, Pishbin F, Ryan MP, Shaffer MSP, et al. Quantitative evaluation of electrophoretic deposition kinetics of graphene oxide. *Carbon* 2014;67:656–61.
- [8]. An SJ, Zhu YW, Lee SH, Stoller MD, Emilsson T, Park S, et al. Thin film fabrication and simultaneous anodic reduction of deposited graphene oxide platelets by electrophoretic deposition. *J Phys Chem Lett* 2010;1:1259–63.
- [9]. Suryanto BHR, Lu X, Zhao C. Layer-by-layer assembly of transparent amorphous Co<sub>3</sub>O<sub>4</sub> nanoparticles/graphene composite electrodes for sustained oxygen evolution reaction. *J Mater Chem A* 2013;1:12726–31.
- [10]. Jankovic A, Erakovic S, Mitric M, Matic IZ, Juranic ZD, Tsui GCP, et al. Bioactive hydroxyapatite/graphene composite coating and its corrosion stability in simulated body fluid. *J Alloys Compd* 2015;624:148–57.
- [11]. Hong X, Chen Y, Wu PZ, Zheng H. Simple, effective fabrication of layered carbon nanotube/graphene hybrid field emitters by electrophoretic deposition. *J Vac Sci Technol B* 2015;33:011802.
- [12]. Zhang S, Li Y, Pan N. Graphene-based supercapacitor fabricated by vacuum filtration deposition. *J Power Sources* 2012;206:476–82.
- [13]. Liang H, Bu Y, Zhang J, Cao Z, Liang A. Graphene oxide film as a solid lubricant. *ACS Appl Mater Interfaces* 2013;5:6369–75.
- [14]. Sansone L, Malachovska V, La Manna P, Musto P, Borriello A, De Luca G, et al. Nanochemical fabrication of a graphene oxide-based nanohybrid for label-free optical sensing with fiber optics. *Sens Actuators B – Chem* 2014;202:523–6.
- [15]. Lee V, Whittaker L, Jaye C, Baroudi KM, Fischer DA, Banerjee S. Large-area chemically modified graphene films: electrophoretic deposition and characterization by soft X-ray absorption spectroscopy. *Chem Mater* 2009;21:3905–16.
- [16]. Su Y, Zhitomirsky I. Electrophoretic deposition of graphene, carbon nanotubes and composite films using methyl violet dye as a dispersing agent. *Colloid Surf A* 2013;436:97–103.
- [17]. Koh ATT, Chen T, Pan L, Sun Z, Chua DHC. Effective hybrid graphene/carbon nanotubes field emitters by electrophoretic deposition. *J Appl Phys* 2013;113:174909.
- [18]. Du J, Zhang Y, Deng S, Xu N, Xiao Z, She J, et al. Correlation between topographic structures and local field emission characteristics of graphene-sheet films. *Carbon* 2013;61:507–14.
- [19]. Liu Y, Zhang D, Pang S, Liu Y, Shang Y. Size separation of graphene oxide using preparative free-flow electrophoresis. *J Sep Sci* 2015;38:157–63.
- [20]. He W, Zhu L, Chen H, Nan H, Li W, Liu H, et al. Electrophoretic deposition of graphene oxide as a corrosion inhibitor for sintered NdFeB. *Appl Surf Sci* 2013;279:416–23.
- [21]. Subramanian P, Niedziolka-Jonsson J, Lesniewski A, Wang Q, Li MS, Boukherroub R, et al. Preparation of reduced graphene oxide–Ni(OH)<sub>2</sub> composites by electrophoretic deposition: application for non-enzymatic glucose sensing. *J Mater Chem A* 2014;2:5525–33.
- [22]. Hasan SA, Rigueur JL, Harl RR, Krejci AJ, Gonzalo-Juan I, Rogers BR, et al. Transferable graphene oxide films with tunable microstructures. *ACS Nano* 2010;4:7367–72.
- [23]. Ordikhani F, Ramezani Farani M, Dehghani M, Tamjid E, Simchi A. Physicochemical and biological properties of electrodeposited graphene oxide/chitosan films with drug-eluting capacity. *Carbon* 2015;84:91–102.
- [24]. Srivastava S, Kumar V, Ali MA, Solanki PR, Srivastava A, Sumana G, et al. Electrophoretically deposited reduced graphene oxide platform for food toxin detection. *Nanoscale* 2013;5:3043–51.

- [25]. Yang J, Yan X, Chen J, Ma H, Sun D, Xue Q. Comparison between the metal ion and polyelectrolyte functionalization for electrophoretic deposition of graphene nanosheet films. *RSC Adv* 2012;2:9665–70
- [26]. Akhavan O, Ghaderi E, Rahighi R. Toward single-DNA electrochemical biosensing by graphene nanowalls. *ACS Nano* 2012;6:2904–16.
- [27]. Chen JT, Cui LF, Sun DF, Yang BJ, Yang J, Yan XB. Enhanced field emission properties from aligned graphenes fabricated on micro-hole patterned stainless steel. *Appl Phys Lett* 2014;105.
- [28]. Bae EG, Hwang YH, Pyo M. Binder-free Sn/graphene nanocomposites prepared by electrophoretic deposition of anode materials in lithium-ion batteries. *Bull Kor Chem Soc* 2013;34:1199–204.
- [29]. Wang M, Oh J, Ghosh T, Hong S, Nam G, Hwang T, et al. An interleaved porous laminate composed of reduced graphene oxide sheets and carbon black spacers by in situ electrophoretic deposition. *RSC Adv* 2014;4:3284–92.
- [30]. Hung CJ, Lin P, Tseng TY. Electrophoretic fabrication and pseudocapacitive properties of graphene/manganese oxide/carbon nanotube nanocomposites. *J Power Sources* 2013;243:594–602.
- [31]. Wu M-S, Fu Y-H. Tubular graphene nanoribbons with attached manganese oxide nanoparticles for use as electrodes in high-performance supercapacitors. *Carbon* 2013;60:236–45.
- [32]. Sookhakian M, Amin YM, Baradaran S, Tajabadi MT, Golsheikh AM, Basirun WJ. A layer-by-layer assembled graphene/zinc sulfide/polypyrrole thin-film electrode via electrophoretic deposition for solar cells. *Thin Solid Films* 2014;552:204–11.
- [33]. Wang B, Li SM, Liu JH, Yu M, Li B, Wu XY. An efficient route to a hierarchical CoFe<sub>2</sub>O<sub>4</sub>@graphene hybrid films with superior cycling stability and rate capability for lithium storage. *Electrochim Acta* 2014;146:679–87.
- [34]. Wu M-S, Lin Y-P, Lin C-H, Lee J-T. Formation of nano-scaled crevices and spacers in NiO-attached graphene oxide nanosheets for supercapacitors. *J Mater Chem* 2012;22:2442–8.
- [35]. Akhavan O, Ghaderi E. Toxicity of graphene and graphene oxide nanowalls against bacteria. *ACS Nano* 2010;4:5731–6
- [36]. Hassan S, Suzuki M, Abd El-Moneim A. Facile synthesis of MnO<sub>2</sub>/graphene electrode by two-steps electrodeposition for energy storage application. *Int J Electrochem Sci* 2014;9:8340–54.
- [37]. Ding J, Yan X, Li J, Shen B, Yang J, Chen J, et al. Enhancement of field emission and photoluminescence properties of graphene–SnO<sub>2</sub> composite nanostructures. *ACS Appl Mater Interfaces* 2011;3:4299–305.