J Materiomics 4 (2018) 108–120

Contents lists available at ScienceDirect

J Materiomics

journal homepage: www.journals.elsevier.com/journal-of-materiomics/

Electrophoretic deposition of graphene-based materials: A review of materials and their applications

Yifei Ma [a], Jiemin Han [a], Mei Wang [a,*], Xuyuan Chen [a,b], Suotang Jia [a]

[a] *State Key Laboratory of Quantum Optics and Quantum Optics Devices, Institute of Laser Spectroscopy, Collaborative Innovation Center of Extreme Optics, Shanxi University, Taiyuan, Shanxi 030006, China*

[b] *Department of Micro- and Nanosystem Technology, Faculty of Technology and Maritime Sciences, University College of Southeast Norway, 3184 Borre, Norway*

A R T I C L E I N F O

Article history:
Received 21 January 2018
Received in revised form
7 February 2018
Accepted 12 February 2018
Available online 17 February 2018

Keywords:
Electrophoretic deposition
Graphene
Graphene-based composite materials
Applications

A B S T R A C T

Recently, graphene-based materials have been successfully fabricated by the electrophoretic deposition (EPD) technique and exhibited various extraordinary properties. Here, research progress of the field of graphene-based materials prepared by the EPD process in recent 5 years is reviewed, including graphene films, graphene/non-metal composites, graphene/metal-based nanoparticles composites, graphene/polymer composites. We also summarize the experimental deposition conditions and the applications of the deposited graphene-based materials that have been reported. It can be concluded that EPD is a simple and reliable manipulation technique and promises a bright future for the production of graphene-based materials in the field of advanced nanocomposite materials. Finally the current issues and outlook of the development direction of EPD in future are also proposed.

Contents

1. Introduction

Electrophoretic deposition (EPD) is a colloidal process where the suspended particles are impelled from the suspension medium to the substrate by an electric field. EPD was discovered by Ruess in

---

* Corresponding author.
E-mail address: wangmei@sxu.edu.cn (M. Wang).

Peer review under responsibility of The Chinese Ceramic Society.

https://doi.org/10.1016/j.jmat.2018.02.004

1808 and was practically applied to deposit thoria particles on a Pt cathode as an emitter for electron tube in 1933 [1]. Afterward, EPD was evolved from being a technique restricted only to traditional ceramics to become an important tool in the processing of advanced materials, such as metals, polymers, carbides, oxides [2,3]. EPD can meet many extreme requirements for the substrates and has plenty of advantages over other membrane fabrication techniques such as moldable nature, uniform and controllable thickness, smooth surface, etc [4]. In recent years, EPD has been widely employed to produce composite materials for coatings, shaping monolithic, laminated and graded free-standing objectives, infiltration of porous materials and woven fiber preforms, and so on [3].

Especially, EPD has been shown to be an effective technique for manipulating graphene layers in liquid suspensions with the aim to produce graphene-related materials including graphene films and graphene-based composite materials [5–9]. Recently, there has been increasing number of publications reporting the research progress of the EPD of graphene and graphene-based composite materials, in which the advantages of EPD is utilized for manipulating graphene to satisfy a variety of applications. The mechanically robust graphene-based nanocomposite coatings, as well as functional nanostructured graphene-based films obtained by the EPD technique, anticipate a promising future for electronic [10], sensing [11–13], biomedical [14], energy harvesting [15], catalytic [16], energy storage [17,18], and environmental applications [19,20].

The intention of this review is to present a comprehensive summary of relevant previous work and describe the application of the EPD technique in the processing of graphene-based materials. The mechanisms and kinetics of graphene-based EPD technique are discussed, followed by a summary of the important progress made in recent 5 years. Furthermore, we sum up the graphene-based materials prepared by EPD, the corresponding EPD conditions, as well as their applications such as supercapacitors, solar cells, sensors, coatings, etc.

2. EPD mechanisms and kinetics

2.1. EPD mechanisms

EPD is usually carried out in a two-electrode cell, where the electric field can be either in a direct electric current mode or in a modulated electric current mode (Fig. 1) [21]. EPD can be applied to any colloidal system with the suspended particles size <30 μm. The EPD of graphene-based materials consists of two steps, electrophoresis and deposition [22]. Electrophoresis happens when the electric field is applied to the graphene suspension, the charged graphene flakes move toward the oppositely charged electrode driven by the electric force, subsequently, the deposition process occurs on the electrode surface where the graphene flakes accumulate under the electric force.

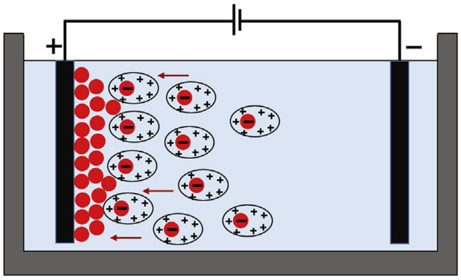

Fig. 1. Schematic diagram of EPD of charged graphene flakes on the anode of an EPD cell with planar electrodes.

Theoretical and modeling studies are being carried out to clarify the mechanisms of EPD, including the EPD of graphene. The affections of electrochemical parameters such as conductivity, solvents, zeta potential, electric field, concentration, etc. on the EPD of graphene-based materials are also being studied. EPD relies on the capability of the graphene sheets to acquire an electric charge in the solvent of suspension [2]. A stable graphene suspension is the prerequisite of the EPD of graphene, which means the graphene flakes have to keep dispersed in the solvent and move towards the electrode independently of each other so that the graphene sheets can be deposited without agglomeration and keep opening the possibility of rearrangement of graphene sheets during packing [23].

2.2. EPD kinetics

In order to predict the kinetics of EPD for particulate materials, Hamaker proposed the Hamaker's law by simply applying the principle of conservation of mass, as shown in the following equation [24,25]:

$$dm/dt = f\mu ESC \tag{1}$$

where m is the mass of deposition (g) and t is the deposition time (s). f is a factor taking into account that only a fraction of the particles brought to the electrode by electrophoresis is incorporated in the deposit ($f \le 1$). μ and E represent the electrophoretic mobility ($m^2/V \cdot s$) and the strength of electric field, respectively. S is the surface area of the electrode (m^2) and C is the concentration of the colloidal suspension (g/m^3) [24,25]. The Hamaker's law indicates a way to predict the deposition yield from the strength of electric field. However, regarding to the strength of electric field, it is subject to the EPD conditions, such as the applied voltage, the distance of the electrodes, resistances of the deposit and the suspension, thickness of the deposit, etc.

In spite of the deposition yield, if other charged powders are deposited with graphene simultaneously, the Hamaker's equation can also be used to predict the mass ratio of the graphene-based composite deposits. For example, a method has been reported to calculate the mass ratio of reduced graphene oxide (RGO) and carbon black (CB) in an interleaved RGO/CB film prepared by EPD [26]. Based on the Hamaker's law, the μ can be expressed by the permittivity of the free space and the suspension medium (ε_0 and ε_r), the zeta potential of colloidal particles (ξ), and the viscosity of the suspension medium (η):

$$\mu = \varepsilon_0 \varepsilon_r \xi / \eta \tag{2}$$

Therefore, the mass of the deposit can be calculated as follow:

$$m = f C \varepsilon_0 \varepsilon_r \xi S E t / \eta \tag{3}$$

Assuming the RGO and CB have the same f, the weight ratio of RGO and CB in the deposited film can be estimated only by the concentrations of the RGO and CB suspensions and the zeta potentials of RGO and CB:

$$m_{RGO}/m_{CB} = C_{RGO}\xi_{RGO}/C_{CB}\xi_{CB} \tag{4}$$

In addition, it also indicates that the EPD membrane yield or thickness can be easily controlled by varying the deposition conditions, such as the suspension concentration, pH of the dispersion (or zeta potential), applied voltage, and deposition time. This strategy can be also employed in the other systems, in which the simultaneous deposition of graphene and other nanoparticles is achieved.

2.3. EPD equipment

Fig. 2(a) represents a typical EPD equipment for graphene deposition. A stable colloidal suspension was prepared and two electrodes are immersed in the suspension in parallel. When deposition on both side of the plate (working electrode) is needed, two counter electrodes can be used, where the two counter electrodes and one working electrode are aligned in parallel with the working electrode in the middle [18]. The substrate can be in an arbitrary shape or be patterned to a certain morphology [12,27,28]. However, this setting has the disadvantage of low yield that can only produce one piece of product at once. Kwon et al. developed an EPD setting with several working and counter electrodes alternately aligned, which greatly increase the yield of EPD (Fig. 2(b)) [29]. In addition, shorten the deposition time can also reduce the side reaction of graphene agglomeration. EPD technique has also been widely used to enhance the mechanical properties of carbon fibers, where the EPD is mostly carried out on the carbon fiber fabrics. However, this technique is limited to the deposition area. Wang et al. proposed an EPD setting, which can achieve the continuous EPD of carbon fibers as presented in Fig. 2(c) [30]. In addition, ultrasonication is applied to the GO suspension during the EPD process to avoid the aggregation of the GO under the loaded voltage. The proposed EPD equipment shows a great potential for the scalable production of graphene-based materials by EPD.

3. Graphene films fabricated by the EPD and their applications

3.1. EPD conditions for depositing graphene-based materials

EPD can be applied to any solid with certain particle surface charges in a stable colloidal suspension. Since from the scientists developed the way to exfoliate the graphite (or graphite oxide) layers and disperse graphene (or GO) in an aqueous, an organic or a mixer solution stably, the EPD of graphene had become possible. Table 1 presents a summary of the studies reviewed on the graphene materials prepared by EPD, collating the relevant parameters on EPD, including the suspension medium, EPD voltage, EPD time, and applications.

GO and RGO are mostly used as graphene precursor for EPD due to the easy preparation of graphene dispersion derived from the oxygen-containing functional groups. Among them, RGO is mostly reduced from the GO in different approaches: chemically reduced before the EPD process [17], electrochemically reduced during the EPD process [4,5], and post-reduced after the EPD process [11,31–33]. As listed in Table 1, several types of solvents have been used to disperse GO, RGO or modified graphene flakes for EPD, including DI water [34], isopropyl alcohol (IPA) [30], ethanol [35], dimethylformamide (DMF) [36], N-Methyl-2-pyrrolidone (NMP) [13], and acetone/ethanol mixture [37]. Aqueous solutions are more widely used for the EPD of graphene than organic solutions because it has the advantages that lower EPD voltage can be used in an aqueous system and it is more environmentally friendly. Moreover, aqueous solvents also have a faster kinetics and are higher

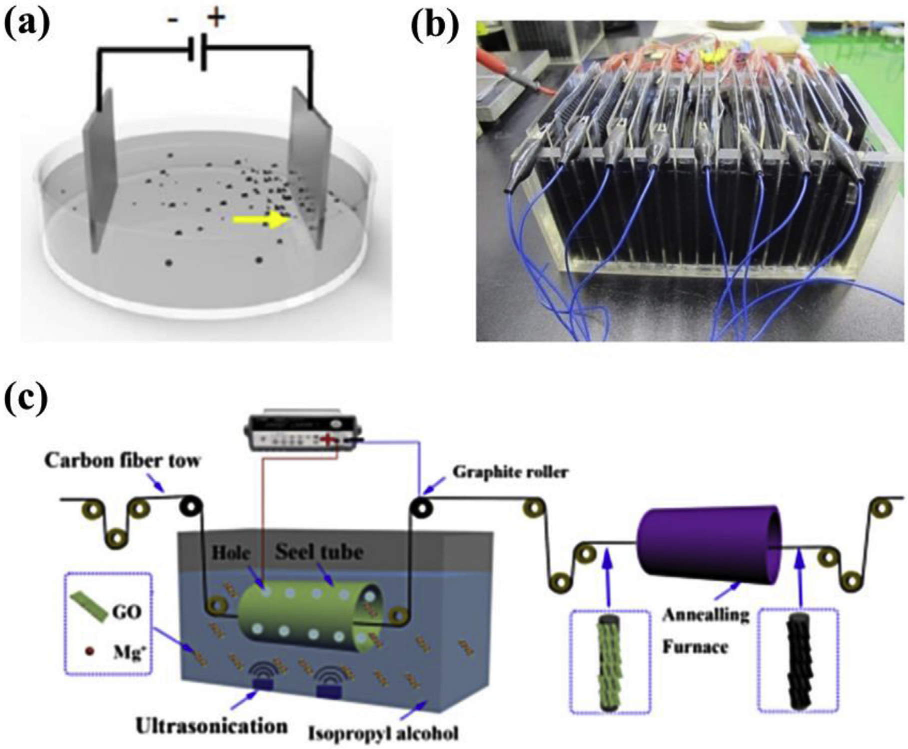

Fig. 2. (a), Typical EPD equipment for deposition of GO with a positive and a negative electrodes aligned in parallel [31]. Copyright 2014, American Society of Chemistry. (b), EPD equipment that can produce 16 pieces of GO/CNT coated carbon fabrics simultaneously [29]. Copyright 2017, Elsevier. (c), Equipment of continuous EPD of graphene on carbon fibers [30]. Copyright 2016, Elsevier.

Table 1
Overview of graphene films prepared by EPD process in recent 5 years.

Graphene precursor	EPD substrate	Suspension medium	Voltage	Time	Application	Year of Publication	Ref.
GO nanowalls	graphite rod	aqueous $Mg(NO_3)_2$ solution	30 V	10 min	single-DNA electrochemical biosensing	2012	[11]
GO	Au	aqueous $LiClO_4$ suspension	−1.2 V	5–60 s	supercapacitor	2012	[5]
GO	Si wafer	ultrapure water	20-45 V	1 h	solid lubricant for MEMS/NEMS devices	2013	[39]
Graphene	stainless steel foil	aqueous methyl violet solution	30 V	2 min	supercapacitors	2013	[45]
GO	carbon cloth	water	6 V	10 h	solid-state supercapacitor	2013	[44]
Graphene quantum dots (GQDs)	Au	DMF with $Mg(NO_3)_2$	80 V	30 min	micro-supercapacitors	2013	[36,42]
RGO	ITO glass	aqueous $Mg(NO_3)_2$ solution	70 V	2 min	food toxin detection	2013	[46]
GO	carbon fibers	water	5 V	1 min	sizing agent	2013	[32]
GO	graphite rod	distilled water	30 V	10 min	electrode for electrochemical detection	2014	[12]
GO	Ag	NMP suspension	3 V	–	gas sensing	2014	[13]
GO	carbon steel	water	4 V	10 s	anti-corrosion	2014	[20]
RGO	TiO_2 nanotube	water	4 V	30 min	Li-ion battery	2014	[28]
Sulphonated RGO	carbon fiber cloth	ethanol/acetone mixture	20 V	30 min	capacitive deionization	2014	[37]
RGO	SS	DI water	3 V	5 min	supercapacitor	2015	[17]
GO	carbon felt	water	1.5 mA/cm^2	10 min	dye pollutants removal	2015	[19]
GO	SS	DI water	4 V	5 min	supercapacitor	2015	[47]
GO	glass fibers	water	10 V/cm	5 min	fiber/matrix bond	2016	[48]
GO	carbon fibers	IPA	160 V	1 min	mechanical strength enhancement	2016	[30]
GO	carbon fibers	aqueous NaOH solution	20 V	20 min	interfacial strength enhancement	2016	[43]
RGO	carbon fibers	NH_3HCO_3 solution	15 V	–	electromagnetic interference shielding	2016	[10]
GO	Ti foil	water	10 V	10 s	photocatalyst	2017	[41]
GO	steel	DI water	3–4 V	4–10 min	corrosion protection coating	2017	[34]
GO	carbon steel	aqueous $CaCl_2$ solution	2.3 V	90 min	anticorrosive coating	2017	[7]
GO	copper	DI water	10 V	1 s	corrosion prevention	2017	[49]
RGO-Mg^{2+}	micro-crystalline diamond	ethanol	15 V	20 min	tribological enhancement coating	2017	[35]
GO	carbon fiber	water	15 V	30-150 min	in-tube solid-phase microextraction	2017	[50]
GO	copper	DI water	5 V	10 s	anti-corrosive coating	2017	[51]

temperature applicable and low cost. Nevertheless, the aqueous suspension also causes problems to the EPD efficiency and the uniformity of the deposit because the electrochemical side reactions often happen along with the EPD such as the electrolysis of water, oxidation of the metal electrodes, etc.

The EPD of graphene can be divided into two types: cathodic EPD and anionic EPD. When graphene sheets are positively charged, the EPD happens on the cathode and the process is cathodic EPD. The EPD of negatively charged graphene sheets on the anode is called anodic EPD. Due to the negatively charged nature of GO and RGO, EPD of GO (or RGO) is mostly an anodic process. However, during the EPD, some metal ions are introduced in the suspension through the addition of salts such as $LiClO_4$, $Mg(NO_3)_2$, $La(NO_3)_3$, $Y(NO_3)_3$, $MgCl_2$, $AlCl_3$ [3,5,36,38]. The graphene flakes are charged positively by adsorption of metal ions on their surface. For example, Mg^{2+} has been used to modify the negatively charged graphene flakes to positively charged Mg^{2+}-graphene for a cathodic EPD (Fig. 3(a)) [36,38]. When a current is passed through solutions of these salts, the formation of a hydroxide has been observed [3]. However, it has also been disclosed that the additive Mg salts may break the stability of the electrolyte and even weaken the tribological performance of the EPD graphene film [39]. In addition, polymer has also been used to modify the surface charge of the graphene flakes owing to their abundant positively charged functional groups (Fig. 3(b)) [40].

3.2. EPD prepared graphene materials

Owing to the advantages of EPD technique that the deposit can occur on substrate with arbitrary shape and surface, the graphene deposit can be in different forms, including continuous in-planar film on the plate substrate [17], fibers or other irregular substrates [10], porous deposits, vertical-aligned graphene deposits [12], non-continuous decoration on the electrodes [41], patterned graphene deposits [42], etc. Hence, the EPD graphene sheets can exhibit different morphologies depends on the deposition conditions such as substrate morphology, graphene precursor for EPD, post-treatment techniques, etc. Mostly, the graphene deposits obtained from EPD are layer-by-layer aligned graphene film (Fig. 4(a)) [17,31,43], while in some cases the modified graphene sheets can have a vertically aligned morphology due to the charge modification on the GO surface (Fig. 4(b)) [11,12]. Other morphologies of graphene deposits with porous nanostructure have also been reported with a freeze-drying process after the EPD of graphene (Fig. 4(c)) [5]. Besides, Dryfe et al. revealed that the morphology and porosity of the EPD graphene also depend on the size of the graphene sheets [44]. When the fine-size graphene is applied as the precursor for the EPD, a highly porous deposit layer can be obtained while a non-porous surface is obtained using the large-size graphene precursor (Fig. 4(d) and (e)). By a detachment process, the deposited graphene film can become a freestanding and flexible membrane. A chemical and an electrochemical methods have been developed to detach the graphene deposit from the substrate and obtain the RGO free-standing membrane with large-area and good electrical conductivity [31].

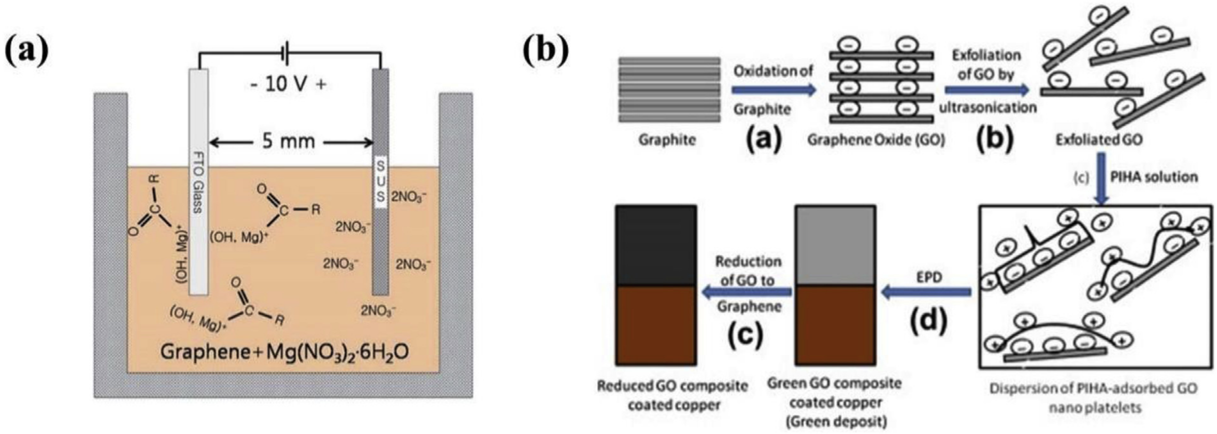

Fig. 3. (a), Set of cathodic EPD of Mg^{2+}-graphene solution [38]. Copyright 2011, Royal Society of Chemistry. (b), Charge modification of GO flakes by adsorption of positively charged PIHA on exfoliated highly negatively charged GO [40]. Copyright 2013, Elsevier.

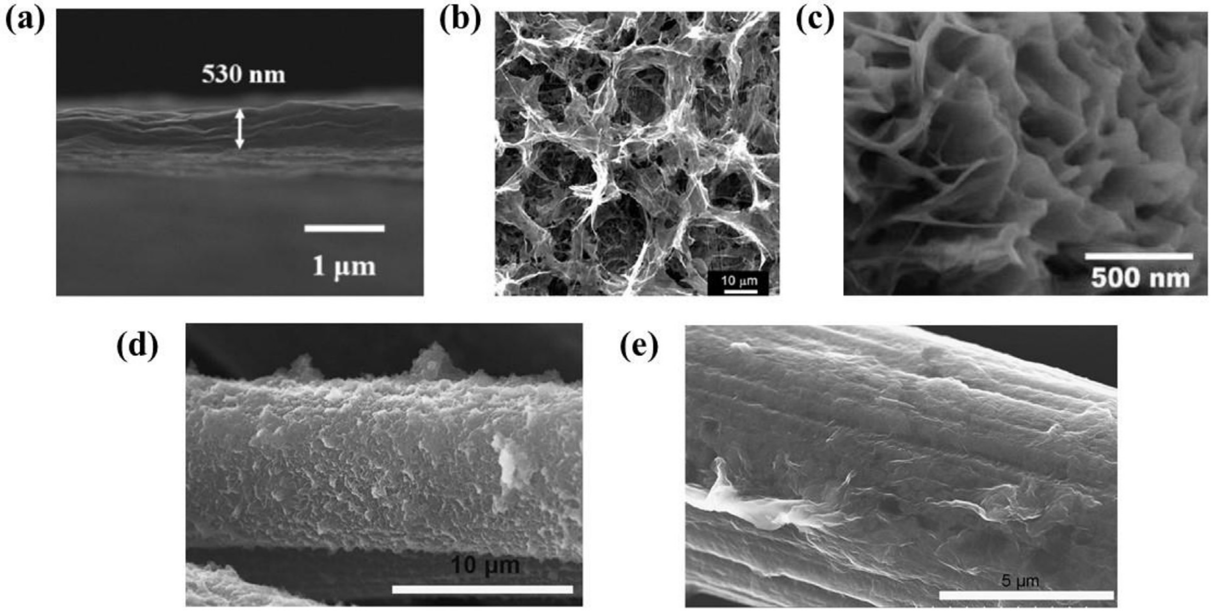

Fig. 4. Different forms of graphene deposits obtained by EPD. (a), An EPD RGO membrane with a layer-by-layer morphology [31]. Copyright 2014, American Chemical Society. (b), The Porous morphology of EPD graphene with post-treatment of freeze-drying [5]. Copyright 2012, Springer Nature. (c), GO nanowalls on a graphite rod electrode with the GO flakes vertically aligned [11]. Copyright 2012, American Chemical Society. (d), EPD of graphene with fine-size graphene as precursor. (e), EPD of graphene with large-size graphene as precursor [44]. Copyright 2013, Elsevier.

3.3. *Applications of EPD prepared graphene materials*

On account of the excellent electrical conductivity, optical transparency, large specific surface area and desirable mechanical properties of graphene, EPD graphene has been increasingly employed as the material to various applications such as supercapacitors, sensors, anti-corrosive coatings, mechanical enhancement agent, and so on, as listed in Table 1. Among the applications, supercapacitors and anti-corrosive coatings are more reported than others according to the publication records in recent 5 years. Graphene has been confirmed to be a desirable material to be used in supercapacitor electrode [52–54]. By the EPD process, it is reported that the RGO electrode fabricated by EPD contains an in-plane layer-by-layer alignment, desirable electrical conductivity, and a moderate porosity that accommodate the aqueous electrolyte ions [17]. Based on the EPD graphene electrode, the all-solid-state supercapacitor exhibits high specific volumetric capacitance (108 F/cm^3) and excellent energy and power densities (7.5 Wh/cm^3 and 2.9 W/cm^3, respectively) (Fig. 5(a)). Impressively, the supercapacitor is also demonstrated to have a long cyclic stability for as long as 180 days (335,000 cycles) (Fig. 5(b)). The simple fabrication and the excellent performance of the device support the application of EPD graphene as large-area, portable, and long-life supercapacitors.

Furthermore, EPD has become one of the most used techniques to produce anti-corrosive coatings onto metals [20,34,49,51]. However, for some specific case of carbon steel's protection, EPD graphene cannot always achieve desirable results. Rangel-Mendez et al. revealed that the reason is the defects (vacancies) involved during the anodic oxidation process and thus a cathodic EPD of GO with the aid of Ca^{2+} was been developed to improve the anti-corrosion of carbon steel [7]. The results show that the cathodic

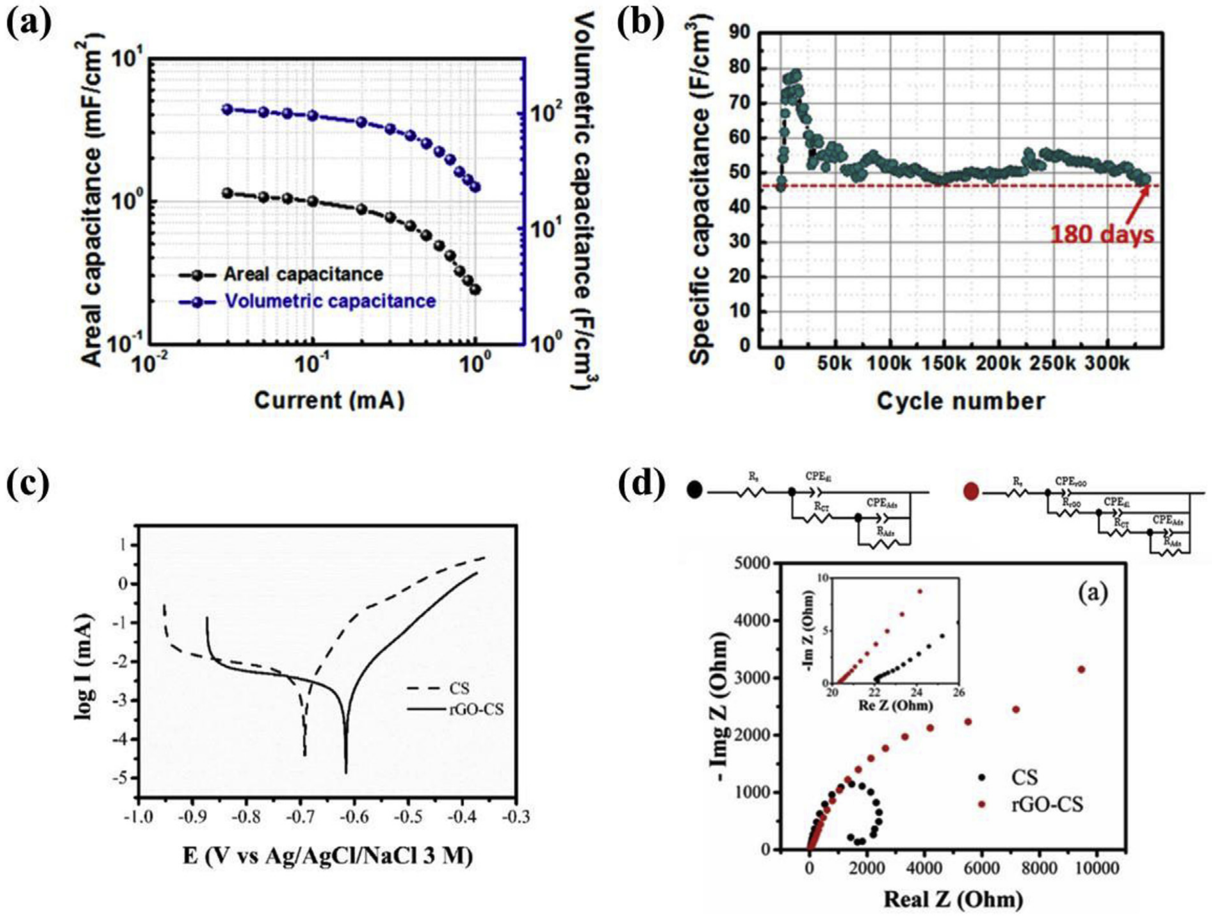

Fig. 5. High specific volumetric capacitance (a) and long cyclic stability (b) obtained from an all-solid-state supercapacitor based on layer-by-layer graphene electrodes fabricated by EPD [17]. Copyright 2015, American Chemical Society. Tafel plot of the polarization curves of carbon steel and RGO-coated carbon steel (c) and electrochemical impedance spectroscopy data for carbon steel and RGO-coated carbon steel (d) [7]. Copyright 2017, Elsevier.

EPD RGO film could reduce up to three times the corrosion rate of carbon steel, which is proved by the decrease of I_{cor}, the shifting of E_{or} to more positive values, and the increase of R_{CT} of carbon steel, as shown in Fig. 5(c) and (d).

4. Graphene-based composites prepared by EPD

Out of the preparation of graphene materials such as GO, RGO, and modified GO by the EPD, there have been growing interests in employing EPD to fabricate the graphene-based composite materials, including: (i), Graphene/non-metal nanoparticle composites such as graphene/CNT, graphene/carbon black, graphene/Si; (ii), Graphene/metal-based nanoparticle composites such as graphene/metal, graphene/metal oxide, graphene/mineral, graphene/metal hydroxide; (iii), Graphene/polymer materials.

With the aim of fabricating graphene reinforced composite materials, interleaved porous structures, and nanoparticle spaced graphene films, the co-EPD strategies to fabricate graphene/nanoparticle composite materials can mainly be divided into three types, as presented in Fig. 6. The EPD suspension consists of graphene and one or more other components, which are stably co-dispersed in three types: (I), simultaneous deposition of the separately dispersed graphene flakes and nanoparticles; (II), graphene flakes are dispersed and the nanoparticles with the opposite charges self-assembled on the graphene surface, the overall charge of the colloid depends on which component possesses higher zeta-potential; (III), graphene is compounded with big molecules (polymer chains) before EPD.

4.1. Graphene/non-metal nanoparticle composites and their applications

The reported works based on graphene/non-metal nanoparticle composites are summarized in Table 2. As listed in Table 2, the applications of the graphene/non-metal nanoparticle composites are mainly targeted at supercapacitors [45,55], dye-sensitized solar cells (DSSC) [15,56], and Li-ion batteries [57,58]. As discussed in session 3.2, the EPD graphene has good electrical conductivity and porous structure, however, when the deposited layers are thick or large electrolyte ions such as organic electrolytes are used, the existed pores in the EPD graphene are not enough to penetrate the electrolyte ions and thus the surface area of the EPD graphene cannot be fully utilized. On the other respect, the in-plane alignment of GO or RGO flakes also affects the interlayer electrical conductivity. It has been proven to be effective to combine graphene with the spacers such as carbon black to expose more surface area and increase the interlayer conductivity as well [26]. Therefore, carbon nanoparticles, including carbon blacks (CB) and carbon nanotubes (CNT) are introduced during the EPD process to enhance the interlayer electrical conductivity and improve the porosity and surface areas.

For example, an EPD dispersion of graphene and CB has been

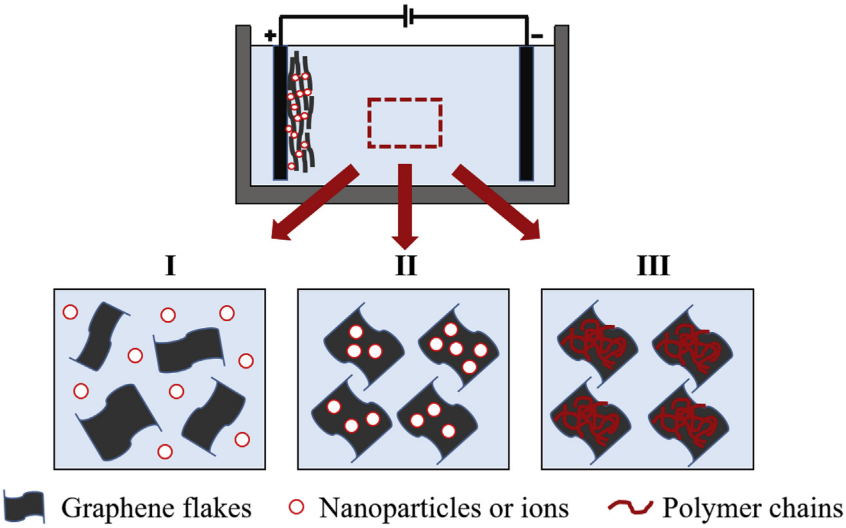

Fig. 6. Three different strategies to fabricate graphene-based composite materials by co-EPD: co-dispersion of nanoparticles and graphene flakes (I), self-assembled nanoparticles and graphene flakes (II), and dispersion of graphene/polymer composite particles (III).

Table 2
Overview of the graphene/non-metal composites prepared by EPD.

Graphene/carbon composites	EPD substrate	Suspension medium	Voltage	Time	Application	Year of Publication	Ref.
GO/SWNTs	FTO substrate	$Mg(NO_3)_2$ in ethanol	30 V	–	DSSC	2012	[56]
Exfoliated graphite/MWCNT	Ni foil	IPA	100 V	10 min	Li-ion battery	2012	[57]
Graphene/Si	Cu foil	water	–	–	lithium ion batteries	2012	[59]
Graphene/activated carbon	FTO glass	IPA	−100 V	–	DSSC	2013	[15]
GO/CNT	carbon cloth	water	6 V	10 h	supercapacitor	2013	[33]
Graphene/MWCNT	stainless steel foil	aqueous methyl violet solution	20 V	2 min	supercapacitors	2013	[45]
RGO/MWNTs	glassy carbon plates	DI water	4 V	90 s	supercapacitor	2013	[60]
RGO/carbon black	SS	water	6 V	10 min	supercapacitor	2014	[26]
Graphene/CNT	Ni substrate	HCl/IPA mixture	50 V	1.5 min	supercapacitor	2014	[61]
Graphene/MWCNT	Si wafer	IPA with $Mg(NO_3)_2$	100 V	15 min	electron field emission	2014	[62]
Graphene/MWCNT	SS	IPA	45-80 V	2-10 min	–	2015	[63]
GO/MWCNT	SS	water	50 V	–	supercapacitors	2015	[64]
RGO/CNT	SS	water/ethanol	0.5–1.5 V	–	supercapacitors	2015	[55]
GO/Si@polyethylene glycol	Cu foil	PEG containing acetone	100 V	10 s - 1 min	lithium ion battery	2016	[58]
GO/CNT	carbon fabrics	DI-water	5-10 V	1-10 min	interfacial reinforcement	2017	[29]

prepared with the assistance of anionic surfactant sodium dodecylbenzenesulfonate (SDBS), following the dispersion strategy type I. RGO and CB are stably dispersed with negative zeta-potentials and an anionic EPD process occurs with the simultaneous co-deposition of RGO and CB when an electric field applies to the solution. As shown in Fig. 7, the CB particles are successfully inserted into the interlayer spaces of RGO layers, obviously increasing the interlayer distance and facilitate the diffusion of electrolyte ions. The results indicate that the spontaneous co-deposition of graphene and charged nanoparticles can be achieved by EPD and the contents of the additive nanoparticles can be controlled depending on the target applications.

4.2. Graphene/metal-based nanoparticle composites and their applications

Recent research progress on the EPD of the graphene/metal based nanoparticle composites are listed in Table 3. Similar with the co-deposition of non-metal nanoparticles, by adjusting the charge of the nanoparticles to be coherent with graphene, some of the metal-based nanoparticles are simultaneously deposited onto the substrate with graphene flakes by the EPD technique [16], following the dispersion strategy type I. For example, graphene and Co_3O_4 nanoparticles are co-dispersed in acetone solvent and migrates together to the Cu foil substrate under the electric field, depositing in a sandwich-like structure, as shown in Fig. 8(a). Contributed from the flexibility of the graphene, the RGO/Co_3O_4 hybrid films can show excellent flexibility (Fig. 8(b)-(c)). In addition, the RGO/Co_3O_4 hybrid films can be deposited to the irregular substrate, owing to the advantage of EPD technique (Fig. 8(d)). Applied as a Li-ion battery electrode, the structural integrity and unobstructed conductive network of the RGO/Co_3O_4 hybrid film can be maintained during cycling, owing to the excellent flexibility of graphene and a large number of voids in this sandwich-like structure.

However, in some cases, an electrochemical deposition process simultaneously happens along with the EPD process [65]. When GO sheets are dispersed in a solution containing metal ions, such as Cu^{2+}, the positively charged Cu^{2+} ions adsorb on the surface of the negatively charged GO sheets automatically and the GO/Cu^{2+} sheets become negatively charged. Followed by a cathodic EPD process, the positive GO/Cu^{2+} sheets deposit onto the cathode and simultaneously, the Cu^{2+} and GO are reduced into Cu nanoparticles

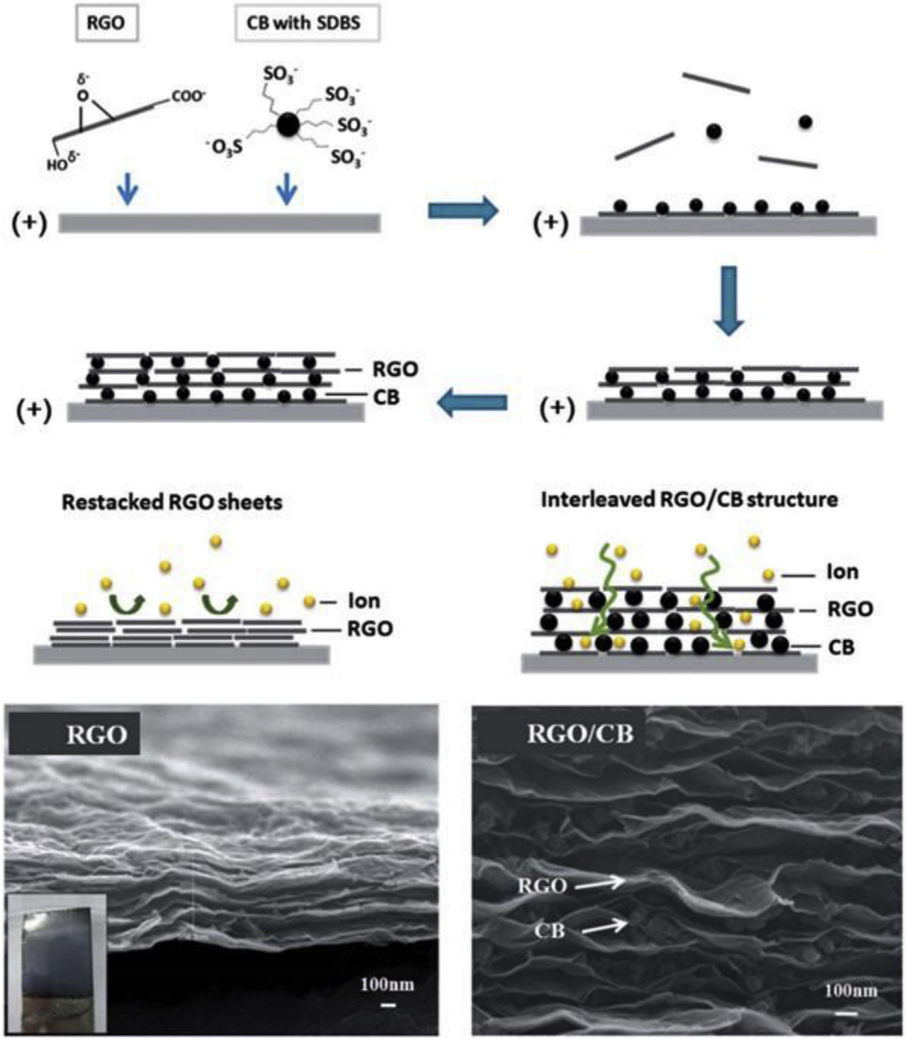

Fig. 7. Schematic representation of the EPD process of graphene and carbon black nanoparticles [26]. Copyright 2014, Royal Society of Chemistry.

and RGO, respectively, forming the RGO/Cu nanoparticles composite, as illustrated in Fig. 8(e). The resultant product shows desirable sensitivity and selectivity toward nonenzymatic glucose sensing.

4.3. Graphene/polymer composite materials prepared by EPD

EPD has also become an effective approach to fabricate the graphene/polymer composite materials, as listed in Table 4. The methods are mostly in two ways: one-step EPD of pre-prepared graphene/polymer composite particles [9,87] and successional EPD of each component [6,88]. The EPD dispersion for preparing graphene/polymer composites follows the strategy III, which indicates that the graphene flakes and polymer chains are compounded and dispersed in the solvents before EPD. However, in some specific cases, the graphene flakes are compounded with the monomers by chemical bonding or hydrogen bonding, subsequently, the in-situ polymerization happens on the surface of graphene flakes, as presented in Fig. 9. After the polymerization step, the GO is reduced into RGO and the polymer chains are grown all over the surface of the RGO sheets. The RGO/polyaniline composites prepared by EPD have several advantages working as electrode materials for pseudocapacitors, including short ionic diffusion and full utilization of polyaniline due to the thin polymer layer, good electrical conductivity derived from the graphene conducting backbone, etc. The EPD of graphene/polymer composite materials may also avoid the problems of graphene agglomeration and difficulty in uniform dispersion of graphene during the fabrication of graphene/polymer composites in traditional processing techniques.

5. Conclusions and outlook

In this review, the fundamentals and research progress on the EPD of graphene-based materials in recent 5 years are comprehensively summarized. EPD has been attracting increasing interests in the research area of graphene processing and applications of graphene-based materials, therefore, a review article is necessary for summarizing the newest progress in the area of graphene EPD. This review literature has indicated that EPD is an effective and versatile technique for the production of graphene and its composite materials for a variety of applications. Especially in the EPD process of graphene-based composite materials, EPD provides a facile and effective way to fabricate the uniform and well-connected composites in one-step, and impressively, the content

Table 3
Summary of the graphene/metal-based nanoparticle composites prepared by the EPD.

Graphene/metal-based nanoparticle composites	EPD substrate	Suspension medium	Voltage	Time	Application	Year of Publication	Ref.
Graphene/Pt nanoparticles	ITO coated glass	DMF	5 V	10/30/ 60 s	electro-catalytic electrodes	2012	[16]
Graphene/ZnO	Si wafer	IPA	300 V	3 min	field emission	2012	[67]
GO/Ni	SS	IPA with NiNO3	60 V	–	supercapacitor	2012	[68]
RGO/Ni(OH)$_2$ composite film	Ni foam, ITO, SS, and Pt	water	2-10 V	30-600 s	supercapacitor	2013	[69]
GO/MnO$_2$/CNTs	Ni substrate	IPA	50 V	2 min	supercapacitor	2013	[70]
Ag/hydroxyapatite/graphene	Ti plate	absolute ethanol	60 V	2 min	antibacterial coating	2015	[71]
Graphene nanosheets/Co(OH)$_2$	TCO	anhydrous IPA	50 V	10 min	dye-sensitized solar cell (DSSC)	2014	[72]
RGO/Ni(OH)$_2$	gold substrate	ethanol	50 V	20 s	glucose sensing	2014	[73]
GO-hydroxyapatite	Ti sheets	ethanol	30 V	1-5 min	biological applications	2014	[74]
GO/Si-CuO quantum dots	Cu electrodes	DI water	10 V	60 s	Li-ion battery	2014	[75]
Co(OH)$_2$/Fe(OH)$_3$@GO films	Cu foils	absolute ethanol	60 V	400 s	lithium storage	2014	[76]
GO-Sn	Ni foam	water	5 V	30 s	lithium ion storage	2014	[77]
GO-hydroxyapatite	Ti plate	absolute ethanol	60 V	2 min	bioactive coating	2015	[78]
RGO/MoS$_2$/CNT	FTO glass	acetone/ethanol mixture	80 V	–	DSSC	2014	[79]
RGO/CoS hybrid film	FTO glass	water	3 V	5 s	DSSC	2015	[80]
GO/lithium iron phosphate	carbon cloth	IPA with Mg(NO$_3$)$_2$	90 V	–	lithium ion batteries	2015	[81]
RGO/Cu	gold	ethanol	50 V	2 min	glucose sensing	2015	[65]
Co(OH)$_2$@graphene hybrid film	coppor foil	absolute ethanol	60 V	200 s	lithium-ion batteries	2015	[82]
graphene-iron oxide-chitosan hybrid nanocomposite	ITO	water/ethanol mixture with acetic acid	10 V	60 s	pathogen detection	2015	[83]
CaSiO$_3$/RGO	Ti substrate	IPA	60 V	5 min	coatings	2016	[84]
GO/NiO	nickel foam	IPA	200 V	10 min	supercapacitor	2016	[18]
GO/TiO$_2$ hierarchical spheres	Ti threads	acetone	7-20 V	2 min	DSSC	2016	[85]
Ammonia-doped-porous RGO/CuO	gold electrode	ethanol	30 V	30 s	glucose sensing	2017	[8]
RGO/MnO$_2$	ITO substrate	water and acetonitrile mixture	25 V	–	supercapacitor	2017	[86]
RGO/Co$_3$O$_4$ nanocubes	Cu foil	acetone	100 V	25 s	lithium ion battery	2017	[66]

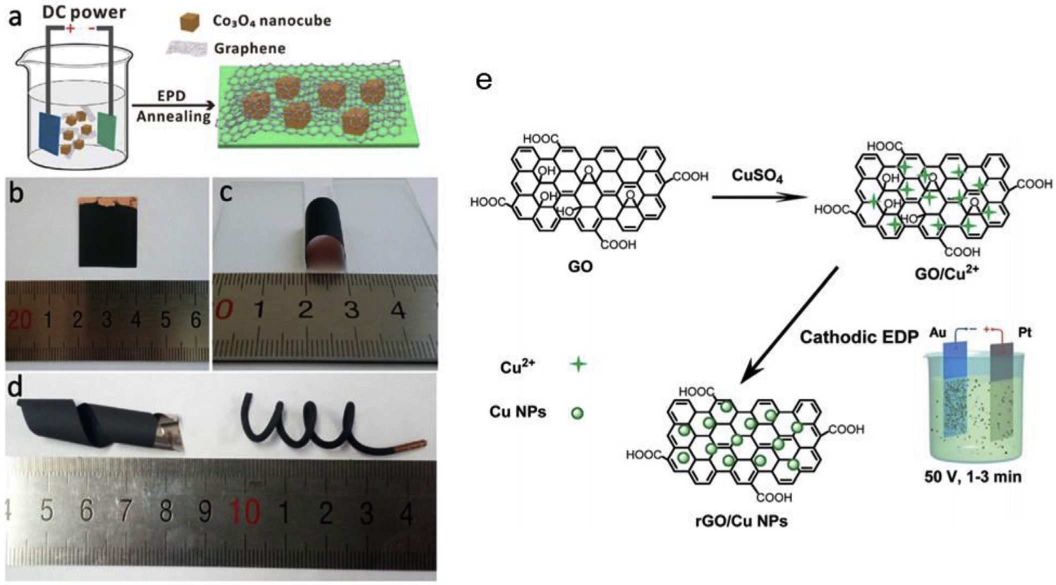

Fig. 8. (a), EPD fabrication procedures of the RGO/Co$_3$O$_4$ hybrid materials. (b), Photographs of RGO/Co$_3$O$_4$ hybrid electrode on the surface. (c), Curved RGO/Co$_3$O$_4$ hybrid electrode. (d), RGO/Co$_3$O$_4$ hybrid film deposited on the irregular substrates [66]. Copyright 2017, American Chemistry Society. (e), Schematic illustration of the preparation of RGO/Cu nanoparticles (Cu NPs) using electrophoretic deposition/reduction [65]. Copyright 2015, Royal Society of Chemistry.

of each component is possible to be controlled by controlling the EPD parameters. However, in some specific cases such as EPD of graphene/metal hydroxides, the EPD process happens along with an electrochemical deposition process, which facilitates the processing but also makes the mechanism unclear and the process complicated. In addition, it is hard to control the morphology, contents ratio of each component, etc. Even though the EPD has been used in many graphene-based materials and applications, the underlying mechanisms of the EPD of graphene are still unclear. Deeper investigation of the mechanisms of graphene EPD should be carried out in future, which will provide a strong guidance in controlling the parameters of EPD graphene, realize the EPD of more materials, and eliminate the unbeneficial side reactions.

Table 4
Summary of graphene/polymer composite materials fabricated by the EPD.

Graphene/polymer composites	EPD substrate	Suspension medium	Voltage	Time	Application	Year of Publication	Ref.
Graphene nanosheet/polydiallyldimethylammonium chloride	silicon wafer	methanol	300 V	10 min	field emission and biocompatibility application	2012	[89]
RGO/polypyrrole	glassy carbon electrode	ultrapure water	0.8 V	–	supercapacitor	2012	[90]
GO-polymetric isocyanate crosslinked with hydroxyl functional acrylic adhesive	Cu plates	water	10 V	30 s	composite coating	2013	[40]
GO-hydroxyapatite-hyaluronic acid	Ti substrate	ethanol-water mixture	30 V	1-5 min	anti-corrosive coating	2013	[14]
RGO/polyethylenimine	SS	water	2-7 V	5-10 min	electromagnetic interference shielding	2014	[6]
Graphene/polyaniline composite film	nickel alloy plate	DI water	−20 V	20 min	supercapacitors	2014	[87]
Graphene/ZnS/polypyrrole	ITO glass	IPA	30 V/ 60 V	10 min/ 5 min	solar cells	2014	[88]
GO/chitosan films	Ti foils	aqueous acetic acid solution	10 V	10 min	drug-eluting	2015	[91]
RGO/polypyrrole	titanium	water	30 V	60 s	supercapacitor	2015	[9]
silk fibroin/GO/hydroxyapatite	Ti sheet	water/ethanol mixture	10 V	30-45 s	orthopedic applications	2016	[92]
GO reinforced chitosan-hydroxyapatite	Ti substrate	ethanol and water mixture	20 V	3 min	coatings	2016	[93]
GO/polyethylenimine	glass/Ti/Au substrate	Milli-Q water	15 V	2 min	lysozyme sensing in serum	2017	[94]

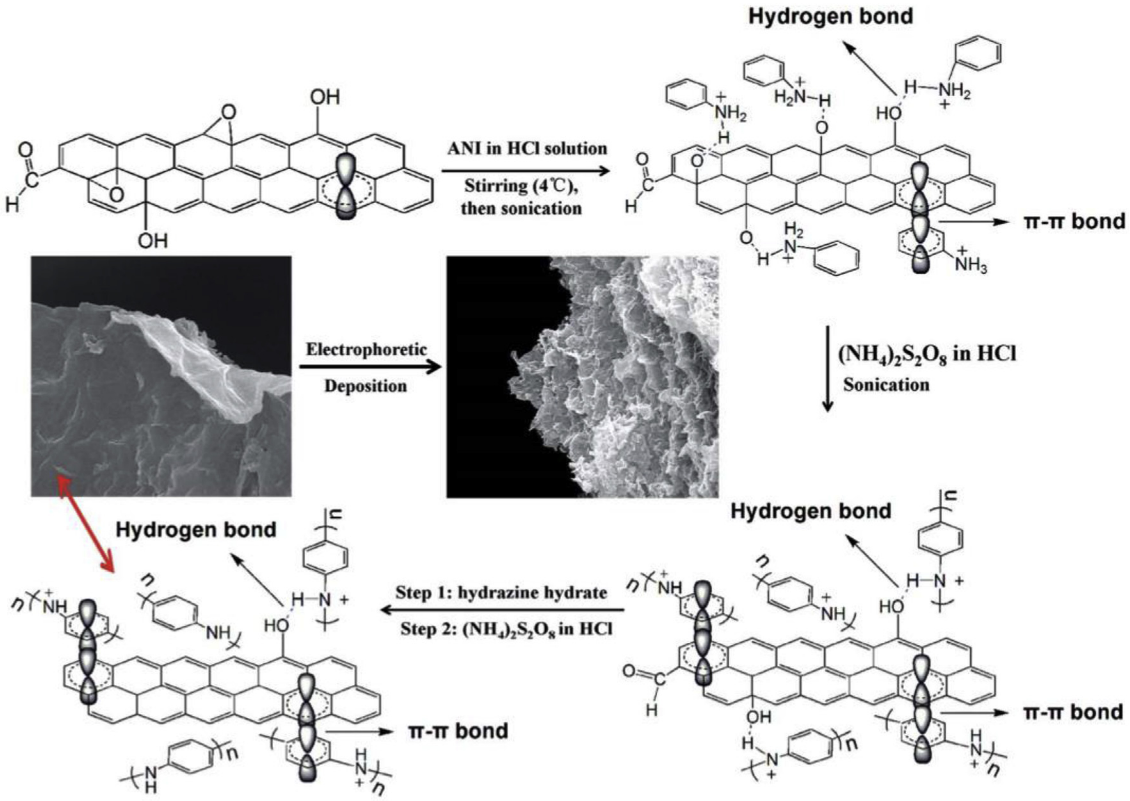

Fig. 9. Schematics of the EPD fabrication process of graphene/polyaniline composite electrodes for pseudocapacitors [87]. Copyright 2014, Royal Society of Chemistry.

References

[1] Corni I, Ryan MP, Boccaccini AR. Electrophoretic deposition: from traditional ceramics to nanotechnology. J Eur Ceram Soc 2008;28(7):1353–67.

[2] Besra L, Liu M. A review on fundamentals and applications of electrophoretic deposition (EPD). Prog Mater Sci 2007;52(1):1–61.

[3] Van der Biest OO, Vandeperre LJ. Electrophoretic deposition of materials. Annu Rev Mater Sci 1999;29(1):327–52.

[4] An SJ, Zhu Y, Lee SH, Stoller MD, Emilsson T, Park S, Velamakanni A, An J, Ruoff RS. Thin film fabrication and simultaneous anodic reduction of deposited graphene oxide platelets by electrophoretic deposition. J Phys Chem Lett 2010;1(8):1259–63.

[5] Sheng K, Sun Y, Li C, Yuan W, Shi G. Ultrahigh-rate supercapacitors based on electrochemically reduced graphene oxide for ac line-filtering. Sci Rep 2012;2:247.

[6] Kim S, Oh JS, Kim MG, Jang W, Wang M, Kim Y, Seo HW, Kim YC, Lee JH, Lee YK, Nam JD. Electromagnetic interference (EMI) transparent shielding of

reduced graphene oxide (RGO) interleaved structure fabricated by electrophoretic deposition. ACS Appl Mater Interfaces 2014;6(20):17647–53.

[7] Quezada-Rentería JA, Cházaro-Ruiz LF, Rangel-Mendez JR. Synthesis of reduced graphene oxide (rGO) films onto carbon steel by cathodic electrophoretic deposition: anticorrosive coating. Carbon 2017;122:266–75.

[8] Maaoui H, Singh SK, Teodorescu F, Coffinier Y, Barras A, Chtourou R, Kurungot S, Szunerits S, Boukherroub R. Copper oxide supported on three-dimensional ammonia-doped porous reduced graphene oxide prepared through electrophoretic deposition for non-enzymatic glucose sensing. Electrochim Acta 2017;224:346–54.

[9] Liu X, Qian T, Xu N, Zhou J, Guo J, Yan C. Preparation of on chip, flexible supercapacitor with high performance based on electrophoretic deposition of reduced graphene oxide/polypyrrole composites. Carbon 2015;92:348–53.

[10] Chen J, Wu J, Ge H, Zhao D, Liu C, Hong X. Reduced graphene oxide deposited carbon fiber reinforced polymer composites for electromagnetic interference shielding. Compos Part A-Appl S 2016;82:141–50.

[11] Akhavan O, Ghaderi E, Rahighi R. Toward single-DNA electrochemical biosensing by graphene nanowalls. ACS Nano 2012;6(4):2904–16.

[12] Akhavan O, Ghaderi E, Rahighi R, Abdolahad M. Spongy graphene electrode in electrochemical detection of leukemia at single-cell levels. Carbon 2014;79:654–63.

[13] Sansone L, Malachovska V, La Manna P, Musto P, Borriello A, De Luca G, Giordano M. Nanochemical fabrication of a graphene oxide-based nanohybrid for label-free optical sensing with fiber optics. Sensor Actuator B Chem 2014;202:523–6.

[14] Li M, Liu Q, Jia Z, Xu X, Shi Y, Cheng Y, Zheng YF, Xi TF, Wei SC. Electrophoretic deposition and electrochemical behavior of novel graphene oxide-hyaluronic acid-hydroxyapatite nanocomposite coatings. Appl Surf Sci 2013;284:804–10.

[15] Wu MS, Zheng YJ. Electrophoresis of randomly and vertically embedded graphene nanosheets in activated carbon film as a counter electrode for dye-sensitized solar cells. Phys Chem Chem Phys 2013;15(6):1782–7.

[16] Chartarrayawadee W, Moulton SE, Li D, Too CO, Wallace GG. Novel composite graphene/platinum electro-catalytic electrodes prepared by electrophoretic deposition from colloidal solutions. Electrochim Acta 2012;60:213–23.

[17] Wang M, Duong LD, Mai NT, Kim S, Kim Y, Seo H, Kim YC, Jang WJ, Lee YK, Suhr JW, Nam JD. All-solid-state reduced graphene oxide supercapacitor with large volumetric capacitance and ultralong stability prepared by electrophoretic deposition method. ACS Appl Mater Interfaces 2015;7(2):1348–54.

[18] Hui X, Qian L, Harris G, Wang T, Che J. Fast fabrication of NiO@graphene composites for supercapacitor electrodes: combination of reduction and deposition. Mater Des 2016;109:242–50.

[19] Le TXH, Bechelany M, Lacour S, Oturan N, Oturan MA, Cretin M. High removal efficiency of dye pollutants by electron-Fenton process using a graphene based cathode. Carbon 2015;94:1003–11.

[20] Park JH, Park JM. Electrophoretic deposition of graphene oxide on mild carbon steel for anti-corrosion application. Surf Coating Technol 2014;254:167–74.

[21] Diba M, Fam DWH, Boccaccini AR, Shaffer MSP. Electrophoretic deposition of graphene-related materials: a review of the fundamentals. Prog Mater Sci 2016;82:83–117.

[22] Sarkar P, Nicholson PS. Electrophoretic deposition (EPD): mechanisms, kinetics, and application to ceramics. J Am Ceram Soc 1996;79(8):1987–2002.

[23] Boccaccini AR, Cho J, Roether JA, Thomas BJC, Jane Minay E, Shaffer MSP. Electrophoretic deposition of carbon nanotubes. Carbon 2006;44(15):3149–60.

[24] Hamaker HC. Formation of a deposit by electrophoresis. Trans Faraday Soc 1940;35(0):279–87.

[25] Hamaker HC, Verwey EJW. Part II.-(C) Colloid stability. The role of the forces between the particles in electrodeposition and other phenomena. Trans Faraday Soc 1940;35(0):180–5.

[26] Wang M, Oh J, Ghosh T, Hong S, Nam G, Hwang T, Nam JD. An interleaved porous laminate composed of reduced graphene oxide sheets and carbon black spacers by in situ electrophoretic deposition. RSC Adv 2014;4(7):3284–92.

[27] Deng C, Jiang J, Liu F, Fang L, Wang J, Li D, Wu J. Effects of electrophoretically deposited graphene oxide coatings on interfacial properties of carbon fiber composite. J Mater Sci 2015;50(17):5886–92.

[28] Menéndez R, Alvarez P, Botas C, Nacimiento F, Alcántara R, Tirado JL, Ortiz GF. Self-organized amorphous titania nanotubes with deposited graphene film like a new heterostructured electrode for lithium ion batteries. J Power Sources 2014;248:886–93.

[29] Kwon YJ, Kim Y, Jeon H, Cho S, Lee W, Lee JU. Graphene/carbon nanotube hybrid as a multi-functional interfacial reinforcement for carbon fiber-reinforced composites. Compos B Eng 2017;122:23–30.

[30] Wang C, Li J, Sun S, Li X, Zhao F, Jiang B, Huang Y. Electrophoretic deposition of graphene oxide on continuous carbon fibers for reinforcement of both tensile and interfacial strength. Compos Sci Technol 2016;135:46–53.

[31] Wang M, Duong le D, Oh JS, Mai NT, Kim S, Hong S, Hwang T, Lee Y, Nam JD. Large-area, conductive and flexible reduced graphene oxide (RGO) membrane fabricated by electrophoretic deposition (EPD). ACS Appl Mater Interfaces 2014;6(3):1747–53.

[32] Lee W, Lee JU, Cha H-J, Byun J-H. Partially reduced graphene oxide as a multi-functional sizing agent for carbon fiber composites by electrophoretic deposition. RSC Adv 2013;3(48):25609.

[33] Wang S, Dryfe RAW. Graphene oxide-assisted deposition of carbon nanotubes on carbon cloth as advanced binder-free electrodes for flexible supercapacitors. J Mater Chem C 2013;1(17):5279.

[34] Ho CY, Huang SM, Lee ST, Chang YJ. Evaluation of synthesized graphene oxide as corrosion protection film coating on steel substrate by electrophoretic deposition. Appl Surf Sci 2017. in press.

[35] Chen S, Shen B, Sun F. The influence of normal load on the tribological performance of electrophoretic deposition prepared graphene coating on microcrystalline diamond surface. Diam Relat Mater 2017;76:50–7.

[36] Liu W, Yan X, Chen J, Feng Y, Xue Q. Novel and high-performance asymmetric micro-supercapacitors based on graphene quantum dots and polyaniline nanofibers. Nanoscale 2013;5(13):6053–62.

[37] Li H, Zaviska F, Liang S, Li J, He L, Yang HY. A high charge efficiency electrode by self-assembling sulphonated reduced graphene oxide onto carbon fibre: towards enhanced capacitive deionization. J Mater Chem 2014;2(10):3484.

[38] Choi H, Kim H, Hwang S, Han Y, Jeon M. Graphene counter electrodes for dye-sensitized solar cells prepared by electrophoretic deposition. J Mater Chem 2011;21(21):7548.

[39] Liang H, Bu Y, Zhang J, Cao Z, Liang A. Graphene oxide film as solid lubricant. ACS Appl Mater Interfaces 2013;5(13):6369–75.

[40] Singh BP, Nayak S, Nanda KK, Jena BK, Bhattacharjee S, Besra L. The production of a corrosion resistant graphene reinforced composite coating on copper by electrophoretic deposition. Carbon 2013;61:47–56.

[41] Razzaq A, Grimes CA, In S-I. Facile fabrication of a noble metal-free photocatalyst: TiO2 nanotube arrays covered with reduced graphene oxide. Carbon 2016;98:537–44.

[42] Liu WW, Feng YQ, Yan XB, Chen JT, Xue QJ. Superior micro-supercapacitors based on graphene quantum dots. Adv Funct Mater 2013;23(33):4111–22.

[43] Jiang J, Yao X, Xu C, Su Y, Deng C, Liu F, Wu J. Preparation of graphene oxide coatings onto carbon fibers by electrophoretic deposition for enhancing interfacial strength in carbon fiber composites. J Electrochem Soc 2016;163(5):133–9.

[44] Wang S, Pei B, Zhao X, Dryfe RAW. Highly porous graphene on carbon cloth as advanced electrodes for flexible all-solid-state supercapacitors. Nanomater Energy 2013;2(4):530–6.

[45] Su Y, Zhitomirsky I. Electrophoretic deposition of graphene, carbon nanotubes and composite films using methyl violet dye as a dispersing agent. Colloid Surface 2013;436:97–103.

[46] Srivastava S, Kumar V, Ali MA, Solanki PR, Srivastava A, Sumana G, Saxena PS, Joshi AG, Malhotra BD. Electrophoretically deposited reduced graphene oxide platform for food toxin detection. Nanoscale 2013;5(7):3043–51.

[47] Ghasemi S, Hosseinzadeh R, Jafari M. MnO_2 nanoparticles decorated on electrophoretically deposited graphene nanosheets for high performance supercapacitor. Int J Hydrogen Energy 2015;40(2):1037–46.

[48] Mahmood H, Tripathi M, Pugno N, Pegoretti A. Enhancement of interfacial adhesion in glass fiber/epoxy composites by electrophoretic deposition of graphene oxide on glass fibers. Compos Sci Technol 2016;126:149–57.

[49] Usha Kiran N, Dey S, Singh B, Besra L. Graphene coating on copper by electrophoretic deposition for corrosion prevention. Coatings 2017;7(12):214.

[50] Feng J, Wang X, Tian Y, Bu Y, Luo C, Sun M. Electrophoretic deposition of graphene oxide onto carbon fibers for in-tube solid-phase microextraction. J Chromatogr A 2017;1517:209–14.

[51] Raza MA, Ali A, Ghauri FA, Aslam A, Yaqoob K, Wasay A, Raffi M. Electrochemical behavior of graphene coatings deposited on copper metal by electrophoretic deposition and chemical vapor deposition. Surf Coating Technol 2017;332:112–9.

[52] Ke Q, Wang J. Graphene-based materials for supercapacitor electrodes - a review. J Materiomics 2016;2(1):37–54.

[53] Gao Y. Graphene and polymer composites for supercapacitor applications: a review. Nanoscale Res Lett 2017;12(1):387.

[54] Chee WK, Lim HN, Zainal Z, Huang NM, Harrison I, Andou Y. Flexible graphene-based supercapacitors: a review. J Phys Chem C 2016:120.

[55] Wang YS, Li SM, Hsiao ST, Liao WH, Yang SY, Ma CCM, Hu CC. Electrochemical composite deposition of porous cactus-like manganese oxide/reduced graphene oxide–carbon nanotube hybrids for high-power asymmetric supercapacitors. J Mater Chem C 2015;3(19):4987–96.

[56] Kim H, Choi H, Hwang S, Kim Y, Jeon M. Fabrication and characterization of carbon-based counter electrodes prepared by electrophoretic deposition for dye-sensitized solar cells. Nanoscale Res Lett 2012;7(1):53.

[57] Seo SD, Hwang IS, Lee SH, Shim HW, Kim DW. 1D/2D carbon nanotube/graphene nanosheet composite anodes fabricated using electrophoretic assembly. Ceram Int 2012;38(4):3017–21.

[58] Yang Y, Li J, Chen D, Fu T, Sun D, Zhao J. Binder-free carbon-coated silicon-reduced graphene oxide nanocomposite electrode prepared by electrophoretic deposition as a high-performance anode for lithium-ion batteries. ChemElectroChem 2016;3(5):757–63.

[59] Zhang YQ, Xia XH, Wang XL, Mai YJ, Shi SJ, Tang YY, Li L, Tu JP. Silicon/graphene-sheet hybrid film as anode for lithium ion batteries. Electrochem Commun 2012;23:17–20.

[60] Chartarrayawadee W, Moulton SE, Too CO, Kim BC, Yepuri R, Romeo T, Wallace GG. Facile synthesis of reduced graphene oxide/MWNTs nanocomposite supercapacitor materials tested as electrophoretically deposited films on glassy carbon electrodes. J Appl Electrochem 2013;43(9):865–77.

[61] Hung CJ, Lin P, Tseng TY. High energy density asymmetric pseudocapacitors fabricated by graphene/carbon nanotube/MnO_2 plus carbon nanotubes nanocomposites electrode. J Power Sources 2014;259:145–53.

[62] Chen L, He H, Yu H, Cao Y, Lei D, Menggen Q, Wu C, Hu L. Electron field emission characteristics of graphene/carbon nanotubes hybrid field emitter. J Alloy Comp 2014;610:659–64.

[63] Talib E, Lau KT, Zaimi M, Bistamam MSA, Abdul Manaf NS, Raja Seman RNA, Zukapli NN, Azam MA. Electrochemical performance of multi walled carbon nanotube and graphene composite films using electrophoretic deposition technique. Appl Mech Mater 2015;761:468–72.

[64] Ajayi OA, Guitierrez DH, Peaslee D, Cheng A, Gao T, Wong CW, Chen B. Electrophoretically deposited graphene oxide and carbon nanotube composite for electrochemical capacitors. Nanotechnology 2015;26(41):415203.

[65] Wang Q, Wang Q, Li M, Szunerits S, Boukherroub R. Preparation of reduced graphene oxide/Cu nanoparticle composites through electrophoretic deposition: application for nonenzymatic glucose sensing. RSC Adv 2015;5(21):15861–9.

[66] Yang Y, Huang J, Zeng J, Xiong J, Zhao J. Direct electrophoretic deposition of binder-free Co_3O_4/graphene sandwich-like hybrid electrode as remarkable lithium ion battery anode. ACS Appl Mater Interfaces 2017;9(38):32801–11.

[67] Ding J, Yan X, Xue Q. Study on field emission and photoluminescence properties of ZnO/graphene hybrids grown on Si substrates. Mater Chem Phys 2012;133(1):405–9.

[68] Wu MS, Lin YP, Lin CH, Lee JT. Formation of nano-scaled crevices and spacers in NiO-attached graphene oxidenanosheets for supercapacitors. J Mater Chem 2012;22(6):2442–8.

[69] Zhang H, Zhang X, Zhang D, Sun X, Lin H, Wang C, Ma Y. One-step electrophoretic deposition of reduced graphene oxide and $Ni(OH)_2$ composite films for controlled syntheses supercapacitor electrodes. J Phys Chem B 2013;117(6):1616–27.

[70] 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.

[71] Janković A, Eraković S, Vukašinović-Sekulić M, Mišković-Stanković V, Park SJ, Rhee KY. Graphene-based antibacterial composite coatings electrodeposited on titanium for biomedical applications. Prog Org Coating 2015;83:1–10.

[72] Miao X, Pan K, Wang G, Liao Y, Wang L, Zhou W, Jiang B, Pan Q, Tian G. Well-dispersed CoS nanoparticles on a functionalized graphene nanosheet surface: a counter electrode of dye-sensitized solar cells. Chem Eur J 2014;20(2):474–82.

[73] Subramanian P, Niedziolka-Jonsson J, Lesniewski A, Wang Q, Li M, Boukherroub R, Szunerits S. Preparation of reduced graphene oxide–$Ni(OH)_2$ composites by electrophoretic deposition: application for non-enzymatic glucose sensing. J Mater Chem 2014;2(15):5525–33.

[74] Li M, Liu Q, Jia Z, Xu X, Cheng Y, Zheng Y, Xi T, Wei S. Graphene oxide/hydroxyapatite composite coatings fabricated by electrophoretic nanotechnology for biological applications. Carbon 2014;67:185–97.

[75] Rangasamy B, Hwang JY, Choi W. Multi layered Si–CuO quantum dots wrapped by graphene for high-performance anode material in lithium-ion battery. Carbon 2014;77:1065–72.

[76] Wang B, Li S, Liu J, Yu M, Li B, Wu X. An efficient route to a hierarchical $CoFe_2O_4$@graphene hybrid films with superior cycling stability and rate capability for lithium storage. Electrochim Acta 2014;146:679–87.

[77] Zhu J, Wang D, Cao L, Liu T. Ultrafast preparation of three-dimensional porous tin–graphene composites with superior lithium ion storage. J Mater Chem 2014;2(32):12918.

[78] Janković A, Eraković S, Mitrić M, Matić IZ, Juranić ZD, Tsui GCP, Tang C, Mišković-Stanković V, Rhee KY, Park SJ. Bioactive hydroxyapatite/graphene composite coating and its corrosion stability in simulated body fluid. J Alloy Comp 2015;624:148–57.

[79] Lin J-Y, Su A-L, Chang C-Y, Hung K-C, Lin T-W. Molybdenum disulfide/reduced graphene oxide-carbon nanotube hybrids as efficient catalytic materials in dye-sensitized solar cells. ChemElectroChem 2015;2(5):720–5.

[80] Huo J, Wu J, Zheng M, Tu Y, Lan Z. High performance sponge-like cobalt sulfide/reduced graphene oxide hybrid counter electrode for dye-sensitized solar cells. J Power Sources 2015;293:570–6.

[81] Huang Y, Liu H, Lu Y-C, Hou Y, Li Q. Electrophoretic lithium iron phosphate/reduced graphene oxide composite for lithium ion battery cathode application. J Power Sources 2015;284:236–44.

[82] Wu X, Wang B, Li S, Liu J, Yu M. Electrophoretic deposition of hierarchical Co_3O_4@graphene hybrid films as binder-free anodes for high-performance lithium-ion batteries. RSC Adv 2015;5(42):33438–44.

[83] Tiwari I, Singh M, Pandey CM, Sumana G. Electrochemical genosensor based on graphene oxide modified iron oxide–chitosan hybrid nanocomposite for pathogen detection. Sensor Actuator B Chem 2015;206:276–83.

[84] Mehrali M, Akhiani AR, Talebian S, Mehrali M, Latibari ST, Dolatshahi-Pirouz A, Metselaar HSC. Electrophoretic deposition of calcium silicate–reduced graphene oxide composites on titanium substrate. J Eur Ceram Soc 2016;36(2):319–32.

[85] Li Z, Zhou Y, Yang Y, Dai H. Electrophoretic deposition of graphene-TiO_2 hierarchical spheres onto Ti thread for flexible fiber-shaped dye-sensitized solar cells. Mater Des 2016;105:352–8.

[86] Lee YR, Kim IY, Kim TW, Lee JM, Hwang SJ. Mixed colloidal suspensions of reduced graphene oxide and layered metal oxide nanosheets: useful precursors for the porous nanocomposites and hybrid films of graphene/metal oxide. Chem Eur J 2012;18(8):2263–71.

[87] Tong Z, Yang Y, Wang J, Zhao J, Su BL, Li Y. Layered polyaniline/graphene film from sandwich-structured polyaniline/graphene/polyaniline nanosheets for high-performance pseudosupercapacitors. J Mater Chem 2014;2(13):4642–51.

[88] 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.

[89] Yang J, Yan X, Chen J, Ma H, Sun D, Xue Q. Comparison between metal ion and polyelectrolyte functionalization for electrophoretic deposition of graphene nanosheet films. RSC Adv 2012;2(25):9665.

[90] Zhu C, Zhai J, Wen D, Dong S. Graphene oxide/polypyrrole nanocomposites: one-step electrochemical doping, coating and synergistic effect for energy storage. J Mater Chem 2012;22(13):6300.

[91] 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.

[92] Li M, Xiong P, Mo M, Cheng Y, Zheng Y. Electrophoretic-deposited novel ternary silk fibroin/graphene oxide/hydroxyapatite nanocomposite coatings on titanium substrate for orthopedic applications. Front Mater Sci 2016;10(3):270–80.

[93] Shi YY, Li M, Liu Q, Jia ZJ, Xu XC, Cheng Y, Zheng YF. Electrophoretic deposition of graphene oxide reinforced chitosan-hydroxyapatite nanocomposite coatings on Ti substrate. J Mater Sci Mater Med 2016;27(3):48.

[94] Wang Q, Vasilescu A, Wang Q, Coffinier Y, Li M, Boukherroub R, Szunerits S. Electrophoretic approach for the simultaneous deposition and functionalization of reduced graphene oxide nanosheets with diazonium compounds: application for lysozyme sensing in serum. ACS Appl Mater Interfaces 2017;9(14):12823–31.

Yifei Ma is currently an assistant professor at the State Key Laboratory of Quantum Optics and Quantum Optics Devices, Institute of Laser Spectroscopy, Shanxi University, China. He obtained his Ph.D. degree in 2016 from Sungkyunkwan University, South Korea, and joined Shanxi University afterward. His research interests include synthesis and functionalization of low-dimensional materials and the applications.

Jiemin Han is a master candidate under the supervision of Prof. Xuyuan Chen and Prof. Yifei Ma at Shanxi University. He earned his Bachelor's degree from Shanxi Normal University (China) in 2017. His current research interests are 2D materials and their electrochemical properties.

Mei Wang is currently a professor at the State Key Laboratory of Quantum Optics and Quantum Optics Devices, Institute of Laser Spectroscopy, Shanxi University. She received her Ph.D. degree in 2014 from the Sungkyunkwan University, South Korea, and worked at Sungkyunkwan University as a post-doctoral researcher until 2017. Her research interests include graphene-based materials, electrophoretic deposition, electrode materials for energy storage devices, electrochemical sensing, etc.

Xuyuan Chen is a professor at the State Key Laboratory of Quantum Optics and Quantum Optics Devices, Institute of Laser Spectroscopy, Shanxi University, China. He also works at Department of Micro- and Nanosystem Technology, University College of Southeast Norway. He was awarded the “Thousand Talent Program” from China in 2010. His research focuses on low dimensional materials, supercapacitors, Micro- and Nano-technology and systems, laser displays and speckle reduction, etc.

Suotang Jia is a professor at the State Key Laboratory of Quantum Optics and Quantum Optics Devices, Institute of Laser Spectroscopy, Shanxi University, China. He earned his Ph.D. degree in 1994, from East China Normal University, China. His research fields include functional materials, low-dimensional materials, laser spectroscopy, quantum optics, etc.