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Preparation of graphene thin films for radioactive samples

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HIGHLIGHTS

- Graphene thin films prepared for radioactive samples.
- Analysis of thin films.
- Films are used for $4\pi\beta$ - γ coincidence measurements.

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ABSTRACT

A new method for the preparation of conductive thin films is presented. The metallization of VYNS films guarantees the electrical conductivity but it results in the breaking of a high proportion of them. Graphene, a two-dimensional nanostructure of monolayer or few layers graphite has attracted a great deal of attention because of its excellent properties such as a good chemical stability, mechanical resistance and extraordinary electronic transport properties. In this work, the possibilities of graphene have been explored as a way to produce electrical conductive thin films without an extra metallization process. The procedure starts with preparing homogenous suspensions of reduced graphene oxide (rGO) in conventional VYNS solutions. Ultra-sonication is used to ensure a good dispersibility of rGO. Graphene oxide (GO) is prepared via oxidation of graphite and subsequent exfoliation by sonication. Different chemically rGO were obtained by reaction with hydrazine sulfate, sodium borohydride, ascorbic acid and hydroiodic acid as reducing agents. The preparation of the thin graphene films is done in a similar way as the conventional VYNS foil preparation procedure. Drops of the solution are deposited onto water.

The graphene films have been used to prepare sources containing some electron capture radionuclides (^{109}Cd , ^{55}Fe , ^{139}Ce) with an activity in the order of 3 kBq. The samples have been measured to test the attainable low energy electron efficiency and the energy resolution of Auger and conversion electrons by 4π (electron capture)- γ coincidence measurements. The 4π (electron capture)- γ coincidence setup includes a pressurized proportional counter and a NaI(Tl) detector. Tests with different pressures up to 1000 kPa were carried out. All these tests show similar values in both parameters (efficiency and resolution) as those obtained by using the conventional metallized films without the drawback of the high percentage of broken films.

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1. Introduction

The backing of radioactive sources used to measure charged particles must be homogeneous, chemically and mechanically stable and electrically conductive. Common applications use gold coated VYNS or collodion films, others use polypyrroles (Los Arcos et al., 1992) or conducting composite polymers (de Sanoit et al., 1997).

Graphene has attracted a great deal of attention since it was

discovered by Novoselov et al. (2004). Graphene is a two dimensional (2D) crystal made of a single layer of carbon atoms that are bonded together in a repeating flat honeycomb pattern. It possesses many interesting properties such as high specific surface area and excellent mechanical, electrical, thermal and optical properties (Geim and Novoselov, 2007). Several approaches have been developed for producing graphene, such as micromechanical exfoliation (Novoselov et al., 2004), chemically modified graphene (Stankovich et al., 2007) also named reduced graphene oxide (rGO), epitaxial growth (Winterlin and Bocquet, 2009) and chemical vapor deposition (CVD) (Kim et al., 2009). Among them, the chemical production is the most promising route for the production of graphene in large quantities because of the relatively low

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cost since it requires inexpensive equipment and because graphene-based materials can form stable colloidal suspensions which are important to facilitate the assembly of macroscopic structures by simple and cheap solution processes. Potential applications of rGO include electronic and optoelectronic devices (Avouris, 2010), sensors and biosensors (Shao et al., 2010), polymer nanocomposites (Potts et al., 2011) and energy storage and production (Brownson et al., 2011). In the chemical production of graphene route, rGO is finally obtained through the oxidation of graphite in presence of strong acids and oxidants, mechanical exfoliation to form graphene oxide (GO) and reduction to rGO. Reduction strategies include thermal reduction, chemical reduction or combined methods. Chemical reduction of graphene is the most widely applied technique to synthesize rGO. Several reducing agents have been used to reduce GO, such as hydrazine (Stankovich et al., 2007), hydroiodic acid (Moon et al., 2010), ascorbic acid (Zhang et al., 2010) or sodium borohydride (Murat et al., 2011). In addition, solvothermal treatment can enhance reduction efficiency of chemical procedures (Wang et al., 2009).

2. Synthesis and chemical preparations

We have prepared six different solutions with graphene: four different rGO suspensions in 10% v/v VYNS/cyclohexanone, one rGO aqueous suspension and one rGO dimethylformamide/water suspension. The detailed methods of synthesis are described in this section:

2.1. Synthesis of graphene oxide (GO)

GO was prepared following the Hummers method (Hummers and Offeman, 1958): in detail, 3 g of graphite powder and 1.5 g of NaNO_3 were mixed with 70 mL of concentrated H_2SO_4 and put into an ice bath. The mixture was stirred on a magnetic plate and 9 g of KMnO_4 were slowly added in small amounts to keep the reaction temperature below 10 °C. After stirring for 1 h, the ice bath was removed and the temperature increased up to (35 ± 5) °C by applying external heating keeping the stirring for another 2 h. Subsequently, 230 mL of ultrapure water of a resistivity of up to 18.2 M Ω were added producing a highly exothermic reaction. The temperature was kept below 70 °C during the addition of water. Then, external heating was applied to reach and maintain the reaction temperature at (90 ± 5) °C for 1 h. Shortly, 50 mL of H_2O_2 and 200 mL ultrapure water were added resulting in a brilliant yellow color along with bubbling. The obtained yellow-brown suspension was then cooled down to room temperature, allowed to settle overnight. The clear supernatant was decanted and the remaining mixture was filtered and washed with 500 mL ultrapure water. Finally the solid graphene oxide was dried under vacuum at 40 °C for two days.

2.2. Chemical reduction of graphene oxide by different reductants

2.2.1. Reduction of GO by hydroiodic acid (rGO-HI)

Synthesis of rGO-HI was carried out following the method proposed by Moon et al. (2010). Briefly, 0.5 g of GO were dispersed in 187 mL of acetic acid and sonicated using a probe sonicator (Bandelin Sonoplus HD 2070) for 2 h until a clear suspension was obtained. Then, 10 mL of concentrated HI were added and the mixture was stirred on a hot plate at (40 ± 5) °C for 40 h. The color changed from brown to black. The suspension was filtrated, washed five times with 50 mL of a saturated sodium bicarbonate solution followed by 250 mL of ultrapure water and 100 mL of acetone. Finally, it was vacuum dried at 40 °C for 24 h.

2.2.2. Reduction of GO by hydrazine sulfate (rGO-Hz)

0.5 g of GO were accurately weighed in a beaker and 170 mL of ultrapure water were added. The mixture was dispersed by probe sonication (Bandelin Sonoplus HD 2070) for 2 h obtaining a clear solution. Then, 450 mg of hydrazine sulfate were added and the mixture was heated at (80 ± 10) °C and stirred for 24 h. The remaining black suspension was filtered, washed twice with 250 mL ultrapure water and 100 mL of acetone. Finally, the solid was dried overnight under vacuum at 40 °C.

2.2.3. Reduction of GO by sodium borohydride (rGO-BH)

rGO-BH was prepared according to Murat et al. (2011) with slight modifications: 0.5 g of GO were suspended in 170 mL of ultrapure water and probe sonicated (Bandelin Sonoplus HD 2070) for 2 h. Then, a saturated sodium carbonate solution was added until the pH reached a value between 9 and 10. Next, 4 g of sodium borohydride were added and the suspension was stirred at (50 ± 5) °C for 24 h. The resulting liquid was filtered, washed twice with 250 mL ultrapure water and 100 mL of acetone. Finally, the solid rGO-BH was dried overnight under vacuum at 40 °C.

2.2.4. Reduction of GO by ascorbic acid (rGO-Asc)

This reduction was performed following the method by Zhang et al. (2010): Briefly, 0.5 g of GO were dispersed in 170 mL of ultrapure water and sonicated using a probe sonicator (Bandelin Sonoplus HD 2070) for 2 h. 2.5 g of L-ascorbic acid were added to the resulting clear solution and stirred at room temperature for 90 h. The obtained suspension was filtered, washed twice with 250 mL ultrapure water and 100 mL of acetone. Finally, the solid rGO-Asc was dried overnight under vacuum at 40 °C.

2.2.5. Solvothermal-assisted reduction of GO by hydroiodic acid (rGO-HI-ST)

GO (65 mg) was accurately weighed in a beaker and 23.4 mL of acetic acid were added. The suspension was sonicated (Bandelin Sonoplus HD 2070) obtaining a homogeneous dispersion as explained in Section 2.2.1. Then, the dispersion was transferred to a PTFE-lined autoclave and 1.25 mL of concentrated HI were added. The vessel was sealed and heated at 180 °C for 48 h. The remaining black suspension was filtered, washed with 25 mL of a saturated sodium bicarbonate solution followed by 100 mL of ultrapure water and 25 mL of acetone. Finally, the solid was dried under vacuum at 125 °C for 48 h.

2.3. Preparation of rGO suspensions in 10% v/v VYNS/cyclohexanone solutions

Stable suspensions of the different rGOs were prepared according to Secor et al. (2013). 70 mg of the corresponding rGO were weighed and dispersed in 20 mL of VYNS/cyclohexanone solution. Probe sonication (Bandelin Sonoplus HD 2070) was applied for 2 h obtaining a homogeneous suspension at a concentration of about 3.5 mg mL⁻¹.

2.4. Preparation of aqueous rGO-HI

An aqueous dispersion of rGO-HI (1 mg mL⁻¹) with added non-ionic surfactant Triton X-100 as stabilizing agent was prepared. Homogenization was carried out by probe sonication using the conditions described above.

2.5. Preparation of rGO-HI-ST suspensions in DMF/H₂O solutions

Since the high dispersibility of rGO in organic solvents such as DMF has been reported in the literature (Dang et al., 2012) rGO suspensions in DMF/H₂O solutions were tested. Then, two

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