This is an Accepted Manuscript, which has been through the Royal Society of Chemistry peer review process and has been accepted for publication.

Accepted Manuscripts are published online shortly after acceptance, before technical editing, formatting and proof reading. Using this free service, authors can make their results available to the community, in citable form, before we publish the edited article. This Accepted Manuscript will be replaced by the edited, formatted and paginated article as soon as this is available.

You can find more information about Accepted Manuscripts in the Information for Authors.

Please note that technical editing may introduce minor changes to the text and/or graphics, which may alter content. The journal’s standard Terms & Conditions and the Ethical guidelines still apply. In no event shall the Royal Society of Chemistry be held responsible for any errors or omissions in this Accepted Manuscript or any consequences arising from the use of any information it contains.
A method is presented for rapid, and scalable purification of graphene oxide. This method is useful for its industrial-scale production.
Rapid Method for the Purification of Graphene Oxide

Gabriel Ceriotti, a Anna Yu. Romanchuk, b Alexander S. Slesarev, a and Stepan N. Kalmykov†

A rapid and facile purification method for graphene oxide (GO) is important for its production above the gram scale. Such a method would allow for the development of GO’s large-scale industrial applications. Out of several protocols in this study, including centrifugation, filtration, precipitation and decantation, filtration using a gas-press proved to be the most effective. Gas-press filtration using filter beds of Celite, perlite, glass wool, ceramic tape, or woven glass fibre allowed for adequate purification of 1 g of crude large-flake (~30 μm flake diameter) GO in less than 60 min using a lab-scale set-up. The present technique could be easily scaled-up, it generates minimal waste, and can be tuned by changing the dimensions of the equipment, pressure, and filter bed. This would allow a user to obtain a higher work-up efficiency. The quickly purified product is called efficiently purified GO or EGO.

Introduction

Graphite oxide (GO) is made by oxidizing graphite in concentrated acid with strongly oxidizing agents to obtain single-atom-thick sheets of carbon with covalently attached oxygen functionalities. The most common methods used for its synthesis are those developed by Brodie,1 Hummers,2 Studenmaier,3 Hofmann,4 and others.5,6 This material was observed for the first time by Brodie in 1890 and denoted “graphitic oxide”.1 Further studies by Boehm et al. in 1961 revealed the single-layer nature of the material.7

With the increased popularity of graphene and its derivatives, GO has found much worth as a more water soluble and chemically active graphene-like material. GO can be used for making robust paper,8 membranes permeable to water but not other fluids including He gas,9 as a chelating agent to remove heavy metals10 and radionuclides11 in contaminated water, as an additive to polymer matrices,12 and as a pore-plugging additive in oil drilling fluids.13 GO has been shown to be a good candidate for environmentally friendly applications as it naturally reduces in the environment through the action of ubiquitous shewanella bacteria,14 or decomposes to humic acids.15 However, the use of GO at larger scales has been slowed by its difficult work-up after synthesis.

Separation and purification of GO from the reaction mixture are the steps in its synthesis incurring the most cost due to the waste produced and the time involved. There is also the risk of degrading the GO on extended exposure to water and light.15,16 Commonly used purification methods for GO and its derivatives may require organic solvents, be time consuming, and/or involve inconvenient processes such as repeated high-speed centrifugation, long hours of filtration, or dialysis.1,2,6,15 GO produced by a quick-washing method might be of lower purity than those produced by the usual methods, but be of sufficient quality for some bulk applications. This quickly processed GO is here referred to as “efficiently purified” GO (EGO). Several methods were considered in this study for quick-washing, each offering certain advantages and disadvantages. The procedures considered here are: 1.}

---

Electronic Supplementary Information (ESI) available: further discussion on the use of perlite, Celite, polyester fibre, and glass wool for gas-press filtration of crude LFGO, SEM images of EGO on Celite and glass wool (Figure SI-1), TGA and ATR-FTIR spectra for different EGOs (Figure SI-2), ATR-FTIR spectra of crude PGO purified with different solvents (Figure SI-3), discussion on the TGA and ATR-FTIR data obtained, experimentally determined naturally occurring concentrations of aqueous SO4-2 in GO suspensions (Table SI-1), computationally-derived speciation diagrams for aqueous U(VI), Am(III), and Ra(II) at different pH in the presence of sulphate ions (Figure SI-4), and a schematic of the custom made gas-press filtration assembly (Figure SI-5). See DOI: 10.1039/b000000x/
Centrifuging the crude reaction mixture to precipitate the GO, discarding the acidic supernatant and re-suspending the centrifuged GO in water and adjusting the pH to 5-7 (Figure 1a); 2. Using polyester fibre, glass wool, ceramic paper, woven glass fibre, perlite, or Celite (diatomaceous earths) to enhance filtration (Figure 1b); 3. Using mineral salts or reactive solids (calcium chloride, bentonite clay, kaolinite) to flocculate and precipitate the GO (Figure 1c); 4. Neutralize the GO by adding an agent such as CaCO$_3$ (Figure 1d); and/or 5. Using particular solvents or solvent combinations (diethyl ether, methanol, isopropyl alcohol, diethyl ether/acetone) to make the GO or GO-derived product precipitate and more easily filtered, flocculated, or otherwise separated from its impurities (Figure 1e).

Figure 1. Five general procedures considered for rapidly purifying GO were attempted. (a) Procedure 1: crude GO slurry was centrifuged; most of the supernatant acid was discarded. The precipitated GO was slurried with water and enough base was added dropwise until pH 5 – 7. Then, the slurry was centrifuged again. (b) Procedure 2: crude GO was filtered on a Büchner funnel using a solid support. Then, the filter cake was re-suspended in water and filtered again. (c) Procedure 3: crude GO was diluted with water (or another solvent), and a flocculant (such as CaCl$_2$, Celite or BaSO$_4$) was added to the suspension. Once the suspension settled, the supernatant was decanted, and the procedure repeated. (d) Procedure 4: crude GO was diluted with water and CaCO$_3$ was added until pH 7. CO$_2$ evolved and a thick suspension of GO and μm-sized CaSO$_4$ particles were formed. (e) Procedure 5: crude GO was shaken vigorously with at least 5 vol equiv of a volatile solvent (such as diethyl ether) capable of dissolving H$_2$SO$_4$, but not of suspending GO, solids were allowed to settle, and the supernatant was decanted. The procedure repeated as needed.

This article focuses on the development of procedure 2 (Figure 1b) using ceramic paper, glass wool, or woven glass fibre beds and the aid of a gas press filter to increase the rate of filtration (Figure 2). This method utilized no organic solvents and was found to be the fastest, most consistent, and resource-efficient. For comparison, the existing published method of purification by centrifugation (see the experimental section for more details) and a form of procedure 5 (Figure 1e) where GO was quenched and centrifuge-washed with methanol are discussed as well. In addition, each of these methods was tested with crude GO made from graphite powder (PGO), and crude GO made from large-flake graphite (LFGO). From here on “GO” is used to mean GO from either source (PGO or LFGO).
Figure 2. Simple set-ups for rapid filtration of crude GO using ceramic paper, glass wool, or woven glass fibre with (a) vacuum filtration and (b) gas press filtration. For filter pressing, a stainless steel lid fastened to the top of the funnel and connected to pressurized air was used.

Experimental

Preparation of Crude PGO and LFGO

Crude PGO and LFGO were synthesized using the same procedure, with the only difference being the starting carbonaceous material. Crude PGO was derived from microcrystalline synthetic graphite (< 20 µm diameter sheets) and crude LFGO from graphite flakes (+100 mesh), both from Sigma-Aldrich. In a typical synthesis, the carbon precursor (10 g each) was added to a 1 L Erlenmeyer flask. H_2SO_4 (540 mL, 96.6% w/w, Fischer Scientific) and H_3PO_4 (60 mL, ≥ 85%, Sigma-Aldrich) (i.e. 600 mL of 9:1 H_2SO_4:H_3PO_4) and the first of five portions of KMnO_4 (10 g each, 99%, J.T. Baker) was added to the flask and the mixture was stirred with a PTFE (Teflon) rod for 5 min. The remaining KMnO_4 portions were added approximately every 12 h until all of the KMnO_4 was added; each addition was accompanied by 5 min of stirring with a PTFE rod. As more KMnO_4 was added and the graphite exfoliated, the mixture thickened. The vessel was covered with a piece of PVDC foil (Saran Wrap™) in between additions and stirrings. The process described is similar to a previously published procedure for the synthesis of GO. The concentration of KMnO_4 added at any time to the H_2SO_4 solution was 5% wt/vol. A new portion of KMnO_4 was not added until the green MnSO_4 species was observed to have disappeared. Caution: Do not exceed ~ 5% wt/vol and do not apply heat; it is reported that at concentrations of 7% wt/vol KMnO_4 in H_2SO_4 the mixture can explode upon heating.  

Existing Published Method for the Purification of Crude GO

In accordance to an established method for the purification of GO, the solution was quenched by pouring the mixture into a beaker filled with an amount of ice equivalent to 0.5 g for every mL of crude to be purified. Then, H_2O_2 (30% w/w Fischer Scientific) was added 1 mL at a time, with stirring in between additions, until the colour of the quenched mixture turned a constant golden-yellow, and gas evolution ceased. The total H_2O_2 added was ~ 0.1 mL per mL of crude, and the resulting mixture was repeatedly centrifuge-washed. For this purpose, the quenched solution was evenly distributed among four 250 mL polypropylene centrifuge bottles (Nalgene, NY, U.S.A.) and centrifuged (Sorvall T1, Thermo Scientific) at 4000 rpm for 90 min for crude LFGO and 30 min for crude PGO. The supernatant was discarded, and the precipitate re-suspended by shaking with DI H_2O (200 mL) in each bottle. This process of precipitation and re-suspension was repeated again with DI H_2O, 10% HCl (made by diluting 37% HCl from Sigma-Aldrich with DI H_2O), twice with anhydrous ethanol (Sigma-Aldrich), and twice with methanol (Sigma-Aldrich.) The precipitate was placed under vacuum at 4.0 torr over night to evaporate the methanol, and ground into a fine powder with a mortar and pestle. The results of this process are referred to in this article as: modified Hummer’s LFGO (HLFGO) if done with crude LFGO, and modified Hummer’s PGO (HPGO) if done with crude PGO.

Methanol Quenching and Purification of Crude GO

To make methanol quenched GO, the crude GO mixture was quenched by cooling it to 0 °C and pouring the mixture into a beaker filled with 0.6 vol equiv of anhydrous methanol cooled in an ice bath. The crude was slowly but thoroughly stirred into the methanol until a homogenous dispersion was achieved. Then, the quenched solution was evenly distributed among four 250 mL polypropylene centrifuge bottles (Nalgene, NY, U.S.A.) and centrifuged (Sorvall T1, Thermo Scientific) at 2500 rpm for 30 min for crude LFGO or 10 min for crude PGO. The supernatant was discarded, and the precipitate re-suspended by shaking with more methanol (200 mL, at room temperature) in each bottle twice more. The resulting solid was placed under vacuum at 4.0 torr over night to evaporate the methanol, and ground into a fine powder with a mortar and pestle. It should be noted that in the case of crude PGO, the material can be seen to bottom flocculate spontaneously over time, making it possible to shake the product in methanol, wait 1 - 3 h, and then decant the supernatant instead of centrifuging it. This precipitation procedure might have to be repeated many times before a neutral pH is reached. The results of this process are referred to in this article as: methanol quenched LFGO (MLFGO) if done with crude LFGO, and methanol quenched PGO (MPGO) if done with crude PGO.
Gas-press Purification of Crude GO

Filtrations were done using a gas press on a 3.5 in diameter Büchner funnel with breathing air at 20 to 60 psi; using higher pressures, as expected, resulted in a faster filtration. The gas press apparatus (Figure 2b) was built-in-house consisted of a pressurized gas line connecting a compressed breathing air gas bottle to an orifice in a 3.5 in stainless steel lid that was placed on top of the Büchner funnel. The lid was held tightly in place by a custom-made screw clamp casing that compressed the funnel and the lid together. A schematic for the gas press apparatus can be found in Figure SI-5 of the supplemental section. Gas-press purification of GO was performed using filter beds of ceramic paper (ceramic tape, Wale Apparatus), woven glass fibre (1/2-in-thick woven glass tape, Wale Apparatus), and glass wool (Sigma-Aldrich). Attempts using polyester fibre (Carpenter Co.), perlite (Miracle-Gro), and Celite (Celite 521, Sigma Aldrich) are discussed in the supplemental section. The first press can be performed with crude GO that has been quenched or not quenched. Using quenched crude GO results in greater removal of acid and salts in fewer presses, but pressing the crude first before quenching allows separation of the acid to be used for recycling. Subsequent presses can be accomplished by washing or scrapping the filter cake onto the solvent to be used in the next press, resuspending the GO, reinstalling the cleaned filter bed, and pressing again. The solvents tested in this study for filter washing were: H₂O, 1 M NaOH, 0.1 M NaOH, and 10% HCl. Data on gas-press washings using NaOH or HCl solutions is provided in the supplemental section. The results of this process are referred to in this study as: efficiently purified LFGO (ELFGO) if done with crude LFGO, and efficiently purified PGO (EPGO) if done with crude PGO.

On a typical run, 150 mL of well-stirred crude GO (equivalent to 2.5 g of graphite precursor) were gas-press filtered using glass fibre paper if crude PGO was used, or glass wool or woven glass fibre if crude LFGO was used. The resulting filtercake was placed together with the filter in a 400 mL beaker with 100 mL of solvent (for example H₂O), and stirred with a magnetic stirrer for 5 min. The filter was recovered and placed back in the filter press. Then, the 100 mL of GO suspension were poured onto the funnel and pressed. Each press took 15 – 30 min for crude PGO suspensions and 30 – 60 min for crude LFGO suspensions at 60 psi.

Instrumental Analyses

X-ray photoelectron spectroscopy (XPS) was performed on a PHI Quantera SXM scanning X-ray microprobe with 26.00 eV passing energy, 45 °C take-off angle and 100 µm beam size. Samples were prepared by pressing the derived GO product on an indium film. Thermogravimetric analyses (TGA) were performed on a Q50 TA instrument under 98% purity Argon gas. The temperature was ramped from 30 to 950 °C at a rate of 5 °C/min except at 1 °C/min between 120 °C and 400 °C. Scanning electron microscopy (SEM) was performed using a FEI Quanta 400 high-resolution field emission scanning electron microscope in high vacuum mode. SEM samples were prepared by suspending the dry GO product in water and spin-coating the suspension on a SiO₂ substrate at 3,000 rpm.

Titration and Rheological Studies

150 mL samples of well-stirred crude GO (equivalent to 2.5 g of graphite precursor) were purified by the methanol quenching, gas-pressing, and existing published methods. The resulting products were suspended in 200 mL H₂O and sheared at 900 rpm. The suspensions were magnetically stirred while 50% w/w NaOH was added, and the pH change monitored using an electronic pH meter (Fisher Science Education) and pH strips (Hydron, range 1 – 11). At pH 7 and 9, titration was briefly stopped, and the suspensions transferred to a bob-and-cylinder rheometer (Fann Viscometer, 34A) where their viscoelastic response at 50 °C was analyzed.

Radionuclide Uptake Studies

U(VI), Am(III), and Ra(II) sorption experiments were performed in polypropylene conical centrifuge tubes (Starstedt AG & Co.) A solution of U(VI), Am(III) or Ra(II) nitrate was added to GO suspensions until the desired GO and radionuclide concentrations were achieved. Then, dilute HClO₄ or NaOH were added dropwise until the desired pH was reached. The change in pH was monitored using a pH-meter equipped with a glass electrode (InLab Expert Pro, Mettler Toledo). After equilibration to the desired pH, the suspension was centrifuged at 40,000 gₑ for 20 min (Allegra 64R, Beckman Coulter) to precipitate the GO/radionuclide complex. The ELFGO agglomerated and precipitated on its own with time. The total sorption was calculated from the difference between the initial activity of the radionuclides and that measured in solution after equilibration. The initial total concentrations of ²³⁵U(VI), ²⁴¹Am(III), and ²²³Ra(II) were 2.15·10⁻⁷ M, 3.94·10⁻¹⁰ M, and 4.01·10⁻¹³ M, respectively. The concentration of the GO suspension was 0.077 g/L in 0.01 M NaClO₄. The total ion concentration was much less than the solubility limit, and the GO/radionuclide ratio corresponded to a very high undersaturation of GO sorption sites. The natural, dissociated sulphur content in GO suspensions was measured using an ion chromatograph with electrogenerated eluent (Dionex ICS-3000). Results are shown in Table SI-1 and were used for the
pH-dependent speciation calculations shown in Figure SI-4 in the supporting information.

### Sample Descriptions Quick Reference Table

<table>
<thead>
<tr>
<th>Abbreviation</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>GO</td>
<td>Graphene oxide</td>
</tr>
<tr>
<td>PGO</td>
<td>Graphene oxide made with powder graphite</td>
</tr>
<tr>
<td>LFGO</td>
<td>Graphene oxide made with large-flake graphite</td>
</tr>
<tr>
<td>EPGO</td>
<td>PGO purified by the gas-press purification method</td>
</tr>
<tr>
<td>ELFGO</td>
<td>LFGO purified by the gas-press purification method</td>
</tr>
<tr>
<td>HPGO</td>
<td>PGO purified as in the existing published modified Hummer’s method</td>
</tr>
<tr>
<td>HLFGO</td>
<td>LFGO purified as in the existing published modified Hummer’s method</td>
</tr>
<tr>
<td>MPGO</td>
<td>PGO purified by the methanol quenching method</td>
</tr>
<tr>
<td>MLFGO</td>
<td>LFGO purified by the methanol quenching method</td>
</tr>
</tbody>
</table>

### Results and Discussion

Compared to typical methods for centrifugation gas press filtration took less time, produced less waste, and yielded a purified material using fewer steps. At 60 psi (the highest pressure used) the filtration time for 50 mL of crude PGO using a ceramic paper filter bed was approximately 15 min for the acid removal and each subsequent H₂O wash. Crude LFGO was usually impossible to filter with ceramic paper as used for crude PGO, but if the filter bed was changed to glass wool or woven glass fibre, 50 mL of crude LFGO were filtered at 60 psi in 30 to 60 min. As shown in Figure 3, EPGO made with two filter press washes of crude PGO (Figure 3d) is less acidic than HLFGO (Figure 3a) using the traditional purification procedure that required centrifugation in relatively large volumes of four different solvents. Titration data for MLFGO is provided as a reference (Figure 3b).

As all the GO had been oxidized using the same procedure, the degree of oxidation determined by XPS (Figure 4) had low variability. The atomic O content in all six samples varied from 32.4% to 40.4% (a 8% range), and the C/O varied from 1.4 to 2.1 (a 0.7 range).

Impurities in the materials were always <6%. MLFGO, HPGO and EPGO show no impurities by XPS (Figure 4e, d and f respectively). HLFGO (Figure 4a), shows a 1.5% atomic content of Si coming from point mineral impurities in the graphite precursor. ELFGO (Figure 4c), and MPGO (Figure 4e) show 3.6% and 5.9% atomic content of sulphur respectively, with MPGO also showing trace amounts of potassium (<0.1%). The presence of sulphur indicated in Figures 4c and 4e is not necessarily from H₂SO₄ residue, but is more likely from sulphate functionalities produced in the GO during its synthesis. The C₁s XPS analysis of these materials (Figure 5)
indicates that the samples showing higher S content (ELFGO and MPGO, Figure 4c and e respectively) also have significantly lower bands for C=O (Figure 5c and e), suggesting that the sulphate moieties in these samples have not yet been hydrolyzed to carboxyls and that these materials have more C-C basal plane connectivity. TGA and ATR-FTIR analysis (Figure SI-2 and SI-3) confirm and complement the observations derived from the analysis of XPS survey and C1s spectra. Further discussion of these observations follows in the supplemental section.

Figure 5. XPS carbon 1s peak deconvolution of a) HLFGO and b) MLFGO, c) ELFGO, d) HPGO, e) MPGO, and f) EPGO.

Although centrifuge-washing with H2O efficiently removes salt impurities in the reaction mixture, suspending GO in water produces a viscous mixture from which it is difficult to centrifuge, filter or otherwise separate the GO. This is particularly true with HLFGO, and might be due to its high aspect ratio; GO at high concentrations has been shown to form strong liquid crystalline domains with a highly stable aqueous suspension. In addition, prolonged exposure to water has been shown to damage the GO basal plane connectivity. Centrifuge washing crude GO with methanol significantly improves the ease with which the material precipitates, and does not damage the basal plane connectivity of GO as much. This is suggested by the lower C=O content in the MLFGO sample compared to HLFGO and ELFGO in C1s XPS analysis (Figure 5). The shorter exposure to H2O in ELFGO also produces a sample with lower C=O content than HLFGO (Figure 5c). But, as mentioned before, MLFGO and ELFGO show a higher content of atomic sulphur by XPS than HLFGO (Figure 5). Depending on the requirements of the application, crude LFGO can be filter pressed with H2O multiple times to remove sulphur moieties or filter-pressed with less or no H2O to produce samples with less C=O content.

Figure 6. SEM images of a) HLFGO, b) MLFGO c) ELFGO, d) HPGO, e) MPGO, and f) EPGO. The scale bar in all images is 10 μm.

SEM images (Figure 6) show that flakes from purified crude LFGO are larger than flakes from purified crude PGO, as expected due to the difference in the sizes of their graphite precursors (a <20 μm flake diameter for powder graphite, and a >150 μm flake diameter for flake graphite). The sizes of HLFGO, MLFGO, ELFGO are comparable (~30 μm in average), with the average flake of ELFGO being 2 – 5 μm larger than the other two. In turn, the sizes of HPGO and MPGO are also comparable (~10 μm average flake diameter), but EPGO flakes are 0 – 9 μm smaller and look less aggregated.
Two examples of industrial applications of GO where EGO could be applied are aqueous radionuclide uptake, and fluid-loss reduction/ rheological modification in oil drilling fluids. In a typical aqueous radionuclide uptake scheme, GO is mixed with the water sample to be cleaned. Radionuclides sorb onto the GO, and the sample is filtered to remove the GO + radionuclides from the treated water. In this application, residues from synthesis in the GO would be disposed of together with adsorbed radionuclides. As an additive to oil drilling formulations, GO is introduced in concentrations ≤4 g/L and the alkalinity of the suspension adjusted to pH 9. In this application, residual potassium, and the residual sulphate salts resulting from treating GO’s sulphate impurities with base, would be inconsequential. These substances are found in abundance in formation solids and mineral additives used in drilling fluids. Typical examples of these mineral additives containing potassium and sulphate salts are bentonite ([(K,Na)(Al,Fe,Mg)2(Si,Al)4O10(OH)2·(H2O)], used as a rheological modifier), and barite (BaSO4, used as a weighting agent).

The suitability of GO for some industrial applications might depend on the colloidal properties as well as the chemical composition of the GO. While the atomic C:O of the products studied was similar (1.4 – 2.1 range, 1.6 average) by XPS, the colloidal behaviour of different products varied more pronouncedly. For example, while the rheological behaviours of HLFGO and MLFGO suspensions (Figure 7) were similar to each other, ELFGO suspensions do not show the same increased viscosity and pronounced thixotropic response as HLFGO and MLFGO dispersions. While this might make ELFGO a poor candidate for viscosifying aqueous solutions, it could work well in a filtration assembly since ELFGO will not restrict water flow as much as its “non-dirty” counterparts, allowing the process to operate at higher speeds. In fact, this might be an asset for certain applications where the GO needs to be recovered from an aqueous suspension, such as when using GO to remove radionuclides and/or harmful metals from contaminated waters as mentioned before.

The radionuclide sorption experiments performed in this study show that even though the purity of ELFGO is lower than that of HLFGO, its performance as a radionuclide sorbent drops by only 20% at most for U(VI) and Am(III), and 40% for Ra(II) (as shown in figure 8.) This drop in sorption is small enough to propose that the amount of ELFGO used could be easily increased to match the performance of HLFGO. Thus, the lower price and higher flow afforded by ELFGO may result in an equally effective, but faster and less expensive process. It might be that the higher concentration of sulphates in ELFGO is the reason for this material’s decreased adsorption. In the presence of SO4 2-, some radionuclides are in the form of neutral or anionic complexes (see Figure SI-4 in the supporting information) that are not adsorbed by GO. The sorption performance of the different GO samples agrees well with the speciation diagrams calculated for experimentally determined concentrations of SO4 2- in GO suspensions (see Table SI-1 in the supporting information.) The observed drop in ELFGO performance corresponds to the increased fraction of neutral or anionic species found in solution. This finding allows for a simple characterization of the relative performance of each GO product as a radionuclide sorption agent by analyzing its sulphate content.

Conclusions
Removing potassium and sulphate impurities from EGO might not be necessary for applications such as water purification (e.g., filtration, radionuclide adsorption), and oil drilling (e.g., pore plugging, and/or rheological modification agents in oil drilling fluids). In filtration and adsorption applications, any impurities in the GO will be discarded at the end of the process together with any other adsorbed materials. Potassium, manganese and sulphate salts are already found in copious amounts from formation solids and clay-based additives in drilling formulations. The products resulting from methanol quenching and quick-filtering schemes are comparable to GO cleaned by published methods, but obtained faster, producing minimal waste, readily lending itself to scale-up, and using no centrifugation. In order to minimize the time of filtration while retaining the most product, it is necessary to use a filter bed adequate for the size of the GO being purified. Ceramic paper is preferred for crude PGO and woven glass fibre for crude LFGO. Because they can be purified using less time and resources, these products might be obtained with less cost than crude GO purified using other methods.

Acknowledgements
Part of the work on radionuclide sorption was supported by the Russian Science Foundation (project 14-13-01279). Thanks to Prof. James M. Tour for allowing us to use his laboratory and associated resources for some of the experiments in this article.

Notes and references
1. Brodie, Phil. Trans., 1869, 149, 249.