Graphene Oxide Dispersions in Organic Solvents

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The dispersion behavior of graphene oxide in different organic solvents has been investigated. As-prepared graphite oxide could be dispersed in *N*,*N*-dimethylformamide, *N*-methyl-2-pyrrolidone, tetrahydrofuran, and ethylene glycol. In all of these solvents, full exfoliation of the graphite oxide material into individual, single-layer graphene oxide sheets was achieved by sonication. The graphene oxide dispersions exhibited long-term stability and were made of sheets between a few hundred nanometers and a few micrometers large, similar to the case of graphene oxide dispersions in water. These results should facilitate the manipulation and processing of graphene-based materials for different applications.

1. Introduction

Graphene, the 2D lattice of sp^2-bonded carbon atoms from which graphite, carbon nanotubes, and fullerenes are derived, has emerged in recent years as a novel and important class of materials on its own merit. This is due not only to the plethora of new fundamental science that it has displayed[1] but also to the prospect of a variety of applications that span graphene-based nanoelectronic devices,[2,3] composite materials,[4,5] and gas sensors.[6]

As with any new material that is intended for large-scale applications, the development of methods that allow the mass production and processing of graphene sheets has become a top priority. In this regard, although the originally reported approach to the preparation of graphene (i.e., micromechanical cleavage of bulk graphite) leads to high-quality 2D crystals and is suitable for fundamental studies, it suffers from extremely low productivity and is therefore inadequate for large-scale use.[1,2] At present, the only route that affords graphene-based sheets in considerable quantities relies on the chemical conversion of graphite to graphite oxide.[4,5,7−13] Graphite oxide is a strongly oxygenated, highly hydrophilic layered material that can be readily exfoliated in water to yield stable dispersions consisting mostly of single-layer sheets, which are referred to as graphene oxide sheets.[4,9,11,12,14] This provides a very convenient setting for the implementation of solution-phase techniques toward the conversion of graphene oxide back to graphene by way of chemical reduction[10,11] or the processing of the graphene oxide sheets into films and paperlike materials.[3,10−13,15]

So far, most of the work reported on the solution-phase manipulation of graphene oxide has been carried out in aqueous media. However, the preparation of graphene oxide dispersions in other solvents, particularly organic solvents, is highly desirable because it may significantly facilitate the practical use of this material.[14,16] To date, the dispersion of graphene oxide in organic solvents has been accomplished via covalent functionalization of the graphene oxide sheets with different molecules and polymers,[17−19] but the presence of such stabilizers is not desirable for most applications.[10] By contrast, the dispersion behavior of as-prepared graphene oxide has remained largely unexplored. Recently, Cai and Song have reported that as-prepared graphite oxide nanoplatelets can be dispersed in *N*,*N*-dimethylformamide (DMF) without the assistance of chemical functionalization,[16,20] but exfoliation of the nanoplatelets down to the single-layer level was not demonstrated and dispersions in additional common solvents were not investigated.

With the aim of expanding its processability and future practical uses, we have investigated the dispersion behavior of graphene oxide in different organic solvents. Significantly, we identify several solvents in which as-prepared graphite oxide can be exfoliated into individual graphene oxide sheets or at most sheets composed of a few layers, forming dispersions with long-term stability in a way comparable to that of graphene oxide dispersions in water. This result should facilitate the preparation of graphene−polymer composites[20] or the development of graphene-based hybrid materials.[16]

2. Experimental Section

Graphite oxide was prepared from natural graphite powder (Fluka 50870) by the Hummers method using $NaNO_3$, H_2SO_4, and $KMnO_4$.[21]

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The oxidation product was purified by rinsing with a 10% HCl solution, repeatedly washing with copious amounts of Milli-Q water, and filtering through standard filter paper with a Büchner funnel. The filtered material was dried under vacuum (80 °C, 3 h) and finally peeled off of the filter paper in the form of an ~0.5-mm-thick film. For the preparation of graphite oxide dispersions in different solvents, the dried product was first ground with a mortar and pestle and then added to the solvent and sonicated in an ultrasound bath cleaner (J. P. Selecta Ultrasons system, 40 kHz) for 1 h. To allow direct comparisons between the dispersing behavior of the different solvents, a certain amount of graphite oxide powder (~5 mg) was added to a given volume of solvent (~10 mL) in such a way that the resulting nominal concentration was adjusted to 0.5 mg mL^{-1} for all of the solvents. Graphite oxide dispersions were tested in the following organic solvents: acetone, methanol, ethanol, 1-propanol, ethylene glycol, dimethyl sulfoxide (DMSO), *N*,*N*-dimethylformamide (DMF), *N*-methyl-2-pyrrolidone (NMP), pyridine, tetrahydrofuran (THF), dichloromethane, *o*-xylene, and *n*-hexane. In all of the solvents, the water content was below 0.1%. Because the common solvent for the preparation of graphite oxide dispersions is water, aqueous dispersions of our as-prepared graphite oxide material were also made under exactly the same conditions as those used in the case of the organic solvents. Such water dispersions served as a reference against which the organic solvent dispersions were compared.

A general characterization of the graphite oxide product was carried out by means of thermogravimetric analysis (TGA), X-ray photoelectron spectroscopy (XPS), and Fourier transform infrared (FTIR) spectroscopy. TGA was carried out in an SDT Q600 thermobalance (TA Instruments) under Ar gas flow (100 mL min^{-1}) and at a heating rate of 10 °C min^{-1}. XPS measurements were made in a SPECS spectrometer under 10^{-7} Pa with a monochromatic Al Kα X-ray source using a power of 100 W. Because graphite oxide is an electrically insulating solid, there was a significant surface charging effect that distorted the recorded spectra. Such an effect was counteracted by the use of an electron flood gun operating at 0.4 eV and 0.10 mA. The atomic percentages (atom %) of the different elements present in the ~10 nm upper layer probed by XPS were calculated from the survey spectra by considering the integrated areas of the main XPS peaks of the elements that were found. FTIR spectra were recorded with a Nicolet 8700 spectrometer (Thermo Scientific) using pellets in KBr with a sample concentration of ~0.1 wt %. The recorded spectra were the result of coadding 64 interferograms obtained at a resolution of 4 cm^{-1}.

The graphite oxide dispersions were characterized by UV−vis absorption spectroscopy and atomic force microscopy (AFM). UV−vis absorption spectra were recorded in a double-beam Heλios α spectrophotometer (Thermo Spectronic). AFM images were obtained under ambient conditions (relative humidity ~40%, temperature ~22−24 °C) with a Nanoscope IIIa Multimode apparatus (Veeco Instruments) in tapping mode. Rectangular Si cantilevers with a spring constant of ~40 N m^{-1} and a resonance frequency of ~250−300 kHz were employed. Samples for AFM imaging were prepared by drop-casting the dispersions onto freshly cleaved mica substrates (grade V-1, Electron Microscopy Sciences), which were then allowed to dry in air. In some cases, the dried mica substrates were gently rinsed with acetone or ethanol to remove the remnants of solvent that were not completely evaporated.

3. Results and Discussion

3.1. Graphite Oxide. Figure 1 shows the thermogravimetric (TG) plot of our graphite oxide material, together with its derivative (i.e., the so-called differential thermogravimetry (DTG) plot). In agreement with previous reports in the literature for graphite oxide,[4,12,14] the main mass loss (~30%) takes place around 200 °C and is ascribed to the decomposition of labile oxygen functional groups present in the material. There is also a mass loss (~15%) below 100 °C attributed to the removal of adsorbed water and a slower, steady mass loss (~20%) over the whole temperature range above 300 °C, which can be assigned to the removal of more stable oxygen functionalities.

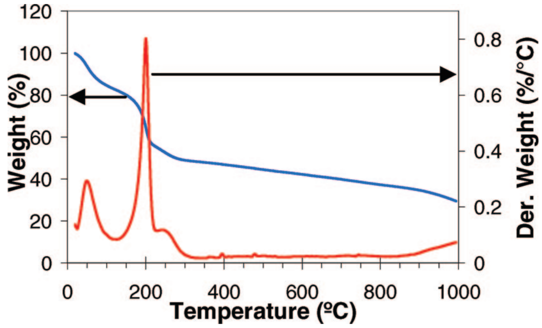

Figure 1. TG (blue) and DTG (red) plots of graphite oxide.

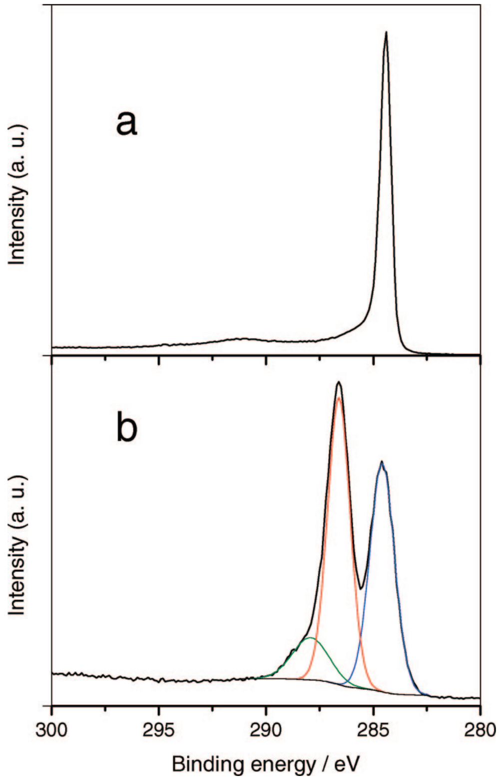

Figure 2. High-resolution core-level C 1s XPS spectra for graphite (a) and graphite oxide (b).

The strong degree of oxidation of graphite oxide was also confirmed by XPS: the survey spectra for pristine graphite and graphite oxide (not shown) yielded C/O atomic ratios of 99 and 2.3, respectively. The high-resolution C 1s core-level spectra of both samples are presented in Figure 2. The narrow, asymmetric C 1s band characteristic of pristine graphite (a) transforms into a complex band showing two maxima for graphite oxide (b). A review of the XPS data available in the literature for graphite oxide reveals that two types of C 1s band shapes are usually reported: (1) a wide C 1s band, markedly asymmetric on the high-binding-energy side,[14,22−24] and (2) a relatively narrow band with two clear maxima.[3,7,11,20,25] In principle, this could be attributed to differences in the proportions of chemical functionalities introduced onto the graphene sheets because a variety

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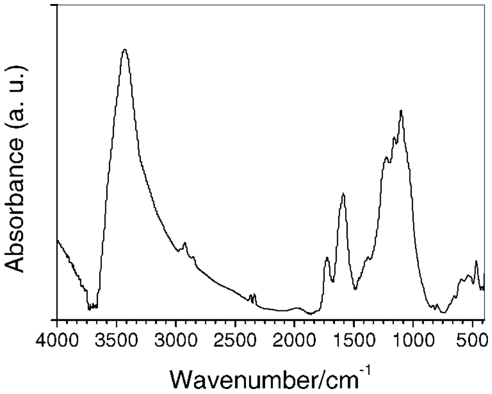

Figure 3. FTIR spectrum of graphite oxide.

of somewhat different procedures are used in the literature for the preparation of graphite oxide.[3−5,7] However, another possibility for the discrepancy could be the surface charging effect of graphite oxide during XPS measurement, a well-known problem in electrically insulating samples that is difficult to deal with.[26] Such an effect generally leads to a shift in the XPS bands toward higher binding energies. Furthermore, if the sample is electrically heterogeneous on a local scale, as graphite oxide is thought to be,[9] then local variations in the electrical conductivity will lead to differential charging. As a result, the individual components of a given XPS band arising from different chemical environments will shift to different extents so that an artificial distortion (widening) is introduced into the band. Such a distortion can be corrected through charge compensation with an electron gun.

We have observed both types of C 1s band shapes in our graphite oxide samples depending on whether the surface charging effect was corrected with an electron gun (Figure 2b) or not (spectrum not shown). The two maxima in the C 1s band of Figure 2b are separated by ~2 eV, in agreement with the results in the literature for similar C 1s band shapes.[7,20,25] The C 1s band can be fitted to three components, located at 284.6 (fwhm = 1.4 eV), 286.6 (fwhm = 1.2 eV), and 287.9 eV (fwhm = 2.0 eV). These components can be assigned to graphitic C=C, C−O, and C=O species, respectively.[27] Nevertheless, the component at 286.6 eV probably also has a contribution from defect (sp^3) C−C species, which are known to give rise to a component very close to this position,[28,29] but in the present case, it was not possible to discriminate clearly between a C−O component and a defect C−C component.

Figure 3 shows the typical FTIR spectrum obtained for our graphite oxide material. The most characteristic features are the broad, intense band at 3430 cm^{-1} (O−H stretching vibrations) and the bands at 1726 cm^{-1} (C=O stretching vibrations from carbonyl and carboxylic groups), 1588 cm^{-1} (skeletal vibrations from unoxidized graphitic domains), 1226 cm^{-1} (C−OH stretching vibrations), and 1103 cm^{-1} (C−O stretching vibrations).[11,18,30] Thus, similar to XPS, FTIR spectroscopy provided evidence of the presence of different types of oxygen functionalities on the graphite oxide material.

3.2. Graphene Oxide Dispersions. As mentioned in the Experimental Section, the as-prepared graphite oxide material was dispersed in water and 13 organic solvents to a nominal concentration of 0.5 mg mL^{-1} with the aid of bath ultrasonication, and the dispersions were then allowed to settle for several weeks. Figure 4 shows digital pictures of all of the dispersions immediately after sonication (top) and 3 weeks after sonication (bottom). For the just sonicated samples, it can be noticed that graphite oxide could be dispersed in almost all of the solvents, except dichloromethane, *n*-hexane, and, to a lesser extent, methanol and *o*-xylene. However, many of these dispersions displayed only short-term stability and precipitated completely in a matter of hours to a few days. This was the case for acetone, ethanol, 1-propanol, DMSO, and pyridine. By contrast, as-prepared graphite oxide dispersions in four organic solvents (ethylene glycol, DMF, NMP, and THF) were seen to exhibit long-term stability comparable to that observed for the dispersion of the same material in water. In the case of water and the four mentioned organic solvents, a small amount of precipitate was seen to develop only within the first few days after sonication (no additional precipitation was observed after such time), which we attribute to graphite oxide material that could not be sufficiently exfoliated during the 1 h sonication period. In fact, longer sonication times tended to decrease the amount of precipitate. Under identical preparation conditions, it was observed that ethylene glycol and THF dispersions yielded somewhat larger amounts of precipitate in relation to those of water, DMF, and NMP dispersions, suggesting that the former solvents possess a comparatively smaller dispersing ability.

UV−vis absorption spectroscopy was employed to gain further insight into the capability of the solvents to disperse graphite oxide. The UV−vis spectra in the different solvents were obtained under identical conditions (i.e., they were all recorded 3 weeks after the dispersions were prepared (stabilized dispersions) and from dispersions that were diluted by the same factor (5) so that qualitative comparisons between the different solvents could be made). Figure 5 shows UV−vis absorption spectra for the five stable graphite oxide dispersions (water, ethylene glycol, DMF, NMP, and THF), together with the spectrum from the dispersion in ethanol to provide a negative case. The spectra are plotted in the wavelength range from 200 to 1000 nm, except for DMF and NMP, for which data appear at ≥265 nm as a result of the impossibility of properly compensating for the strong absorption of both solvents at smaller wavelengths. The UV−vis spectrum of graphite oxide/graphene oxide exhibits two characteristic features that can be used as a means of identification: a maximum at 231 nm, corresponding to $\pi \rightarrow \pi^*$ transitions of aromatic C−C bonds, and a shoulder at ~300 nm, which can be attributed to $n \rightarrow \pi^*$ transitions of C=O bonds;[31] both are bathochromically shifted by conjugation. The spectra recorded in water, ethylene glycol, DMF, NMP, and THF confirm that the as-prepared graphite oxide material was successfully dispersed in these solvents (for DMF and NMP, the shoulder at ~300 nm is clearly observed). By contrast, no absorption was detected in ethanol, corroborating the previous observation that graphite oxide dispersions in such a solvent are not stable. For the five successful solvents, we notice from Figure 5 that water displays the best dispersing ability because it provides the highest absorption intensity and therefore the largest amount of suspended graphite oxide, followed closely by DMF and NMP. Ethylene glycol and THF exhibit very similar dispersing abilities toward as-prepared graphite oxide, although they are noticeably

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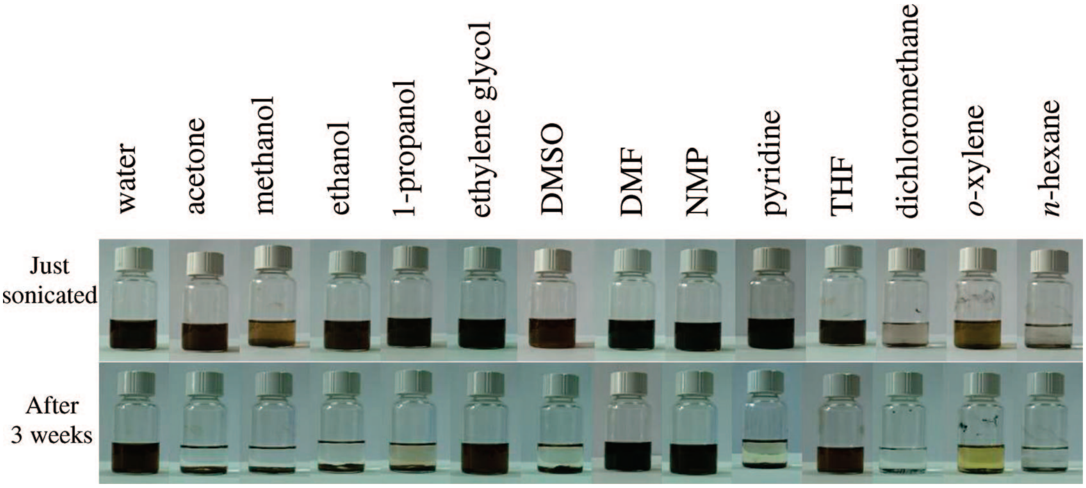

Figure 4. Digital pictures of as-prepared graphite oxide dispersed in water and 13 organic solvents through bath ultrasonication (1 h). Top: dispersions immediately after sonication. Bottom: dispersions 3 weeks after sonication. The yellow color of the *o*-xylene sample is due to the solvent itself.

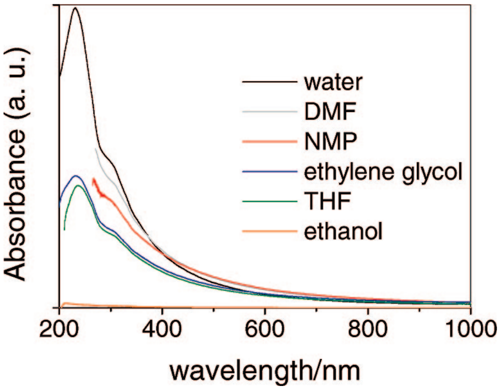

Figure 5. UV−vis absorption spectra of as-prepared graphite oxide dispersed in different solvents by means of bath ultrasonication (1 h). The spectra were recorded for stabilized dispersions (i.e., 3 weeks after preparation).

smaller than those of the other three solvents. In any case, the concentration of dispersed graphite oxide in all of these solvents is estimated to be in the range of a few tenths of 1 mg mL^{-1}, and it can be increased through further sonication. Such concentration values should be sufficient for most practical uses of this material.[3,8,10,11]

Both visual inspection and UV−vis absorption spectroscopy indicated that as-prepared graphite oxide dispersions with long-term stability could be prepared in ethylene glycol, DMF, NMP, and THF, in addition to water. To investigate the degree of exfoliation of the graphite oxide material in these solvents, AFM imaging of the dispersions deposited onto mica substrates was carried out. Representative results are shown in Figure 6. For samples prepared from graphite oxide dispersions in water, the AFM images revealed the presence of irregularly shaped sheets of uniform thickness and lateral dimensions ranging from a few hundred nanometers to a few micrometers (Figure 6a). As illustrated in Figure 6b for the sheet marked by the green line in panel a, the sheets were typically between 1.0 and 1.4 nm thick. This observation is in agreement with previous AFM studies of graphite oxide dispersions in water, which assigned the 1.0−1.4-nm-thick objects to single-layer graphene oxide sheets.[4,9,11,12,14] Such thickness is significantly larger than that of single-layer pristine graphene (∼0.34 nm) and is generally attributed to the presence of oxygen-containing functional groups attached on both sides of the graphene sheet and to the atomic scale roughness arising from structural defects (sp^3 bonding) generated on the originally atomically flat graphene sheet.[14] Thus, individual graphene oxide sheets are expected to be thicker (∼1.0−1.4 nm) than individual pristine graphene sheets (∼0.34 nm).

As could be expected, the AFM investigations indicated that an almost complete exfoliation of graphite oxide into individual graphene oxide sheets was achieved in water (Figure 6a,b). More significantly, a similar degree of exfoliation was also attained for the four organic solvents in which stable graphite oxide dispersions could be prepared (DMF, THF, NMP, and ethylene glycol). This is exemplified in Figure 6c,d for a sample prepared from a dispersion in DMF. Again in this case, sheets of uniform thickness (∼1.0−1.4 nm, Figure 6e) and with the same range in lateral dimensions as that reported for the aqueous dispersions were observed, implying that the exfoliation of as-prepared graphite oxide down to the single layer level was accomplished in DMF. For THF, single-layer graphene oxide sheets were usually encountered (Figure 6f,g), although bilayer sheets were also relatively common (Figure 6h,i). Thus, a high degree of exfoliation was also achieved in THF. Similar conclusions were obtained when dispersions in ethylene glycol and NMP were investigated (e.g., Figure 6j,k for ethylene glycol). For both water and the four successful organic solvents (DMF, THF, NMP, and ethylene glycol), AFM inspection of a large number of sheets revealed that about 80% of the sheets were indeed single-layer graphene oxide whereas the remaining ∼20% corresponded to graphite oxide platelets made up of only two or three graphene oxide layers. Therefore, we can conclude that almost full exfoliation of as-prepared graphite oxide into individual graphene oxide sheets was generally achieved for all of the solvents that afforded stable dispersions of the material. This is important because most of the attractive properties of graphene and graphene-based sheets are mainly associated with their existence as individually separated entities.[4,10]

At present, the mechanisms that allow the stable dispersion of graphene oxide in these organic solvents are not clear. A necessary, but not sufficient, condition seems to be that the solvent molecules be considerably polar. This is reasonable because the graphene oxide sheets are thought to be heavily decorated with polar oxygen-containing functionalities (hydroxyl, carbonyl, carboxyl; see XPS and FTIR spectroscopy results), which should

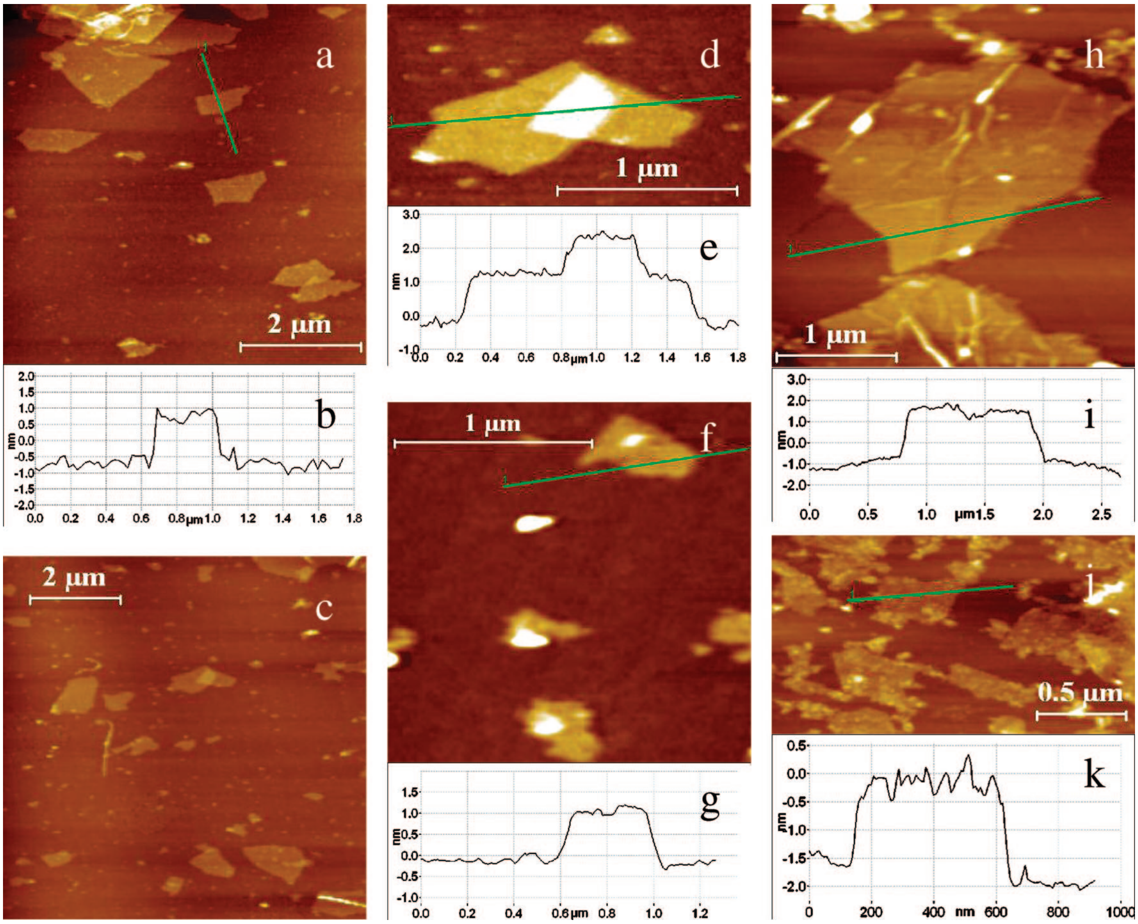

Figure 6. AFM images and line profiles of graphene oxide sheets deposited from dispersions in different solvents onto mica substrates. (a) Sheets deposited from a dispersion in water. (b) Line profile for the sheet marked by the green line in panel a. (c, d) Sheets deposited from a dispersion in DMF. (e) Line profile taken along the green line for the two overlapping sheets in panel d. (f) Sheets deposited from a dispersion in THF. (g) Line profile for the sheet marked by the green line in panel f. (h) Image of an ~2-μm-wide sheet deposited from a dispersion in THF, displaying several wrinkles. Wrinkles were frequently observed for the larger sheets deposited from dispersions in any solvent. (i) Line profile taken along the green line for the sheet in panel h. (j) Sheets deposited from a dispersion in ethylene glycol. Some remnants of the solvent are still present on the mica substrate. (k) Line profile for the sheet marked by the green line in panel j.

promote a good graphene oxide sheet−solvent interaction. We note that such a mechanism has been suggested to contribute to the good dispersability of nitric acid-treated, and therefore oxygen group-containing, carbon nanotubes in DMF and NMP.[32] Both water and the four successful organic solvents exhibit significant electrical dipole moment values: 1.82 D (water), 3.24 D (DMF), 4.09 D (NMP), 1.75 D (THF), and 2.31 D (ethylene glycol).[33] On the contrary, solvents with small dipole moment (*n*-hexane, 0.085 D; *o*-xylene, 0.45 D)[33] clearly failed to disperse the as-prepared graphite oxide material. However, there is a range of solvents with high dipole moments (particularly DMSO, 4.09 D) that also failed to provide graphite oxide dispersions with long-term stability, which suggests that other factors besides solvent polarity are important for determining good dispersability. Previous knowledge of the dispersion behavior of carbon nanotubes in organic solvents cannot provide much more insight into this question because the solvent characteristics required to disperse carbon nanotubes efficiently have not yet been well established.[32,34,35] Clarifying this point will necessitate further in-depth investigations.

4. Conclusions

We have identified several organic solvents (DMF, NMP, THF, and ethylene glycol) in which as-prepared graphite oxide can form dispersions with long-term stability. Furthermore, the graphite oxide material in these solvents is exfoliated mostly into individual, single-layer graphene oxide sheets with lateral dimensions between a few hundred nanometers and a few micrometers. Such dispersions are thus comparable to graphene oxide dispersions in water, which are currently used for the preparation of many graphene-based materials and therefore should facilitate the further manipulation and processing of these materials.

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