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Graphene oxides as sorbents for purification and separation of liquids: a physicochemical study

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The features of sorption properties of oxidized graphite materials with respect to polar and non-polar liquids were examined. The sorption properties of a highly oxidized material (graphene oxide synthesized by the improved Hummers' method) and weakly oxidized thermally expanded graphite were compared directly. Differences in a number of physicochemical parameters were confirmed by X-ray diffraction analysis, infrared spectroscopy, X-ray photoelectron spectroscopy, and differential scanning calorimetry. Graphene oxide synthesized by the Hummers' method with ratio C/O ≈ 2.5 sorbs only polar liquids. Thermally expanded graphite, which was synthesized via heat processing of electrochemically oxidized graphite, with C/O ≈ 9.5 sorbs both polar and non-polar liquids. Differential scanning calorimetry was used to demonstrate that the parameters of sorption of polar liquids are comparable within the error limits for materials that differ significantly in their degree of oxidation. It was found that water sorption by materials based on thermally expanded graphite does not lead to their swelling. The capacity to maintain a constant interplanar spacing in interaction with aqueous media may make thermally expanded graphite a promising material for the fabrication of membranes for removal of impurities from water.

Keywords: graphene oxide, thermally expanded graphite, sorption, swelling.

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Introduction

Graphene oxide (GO) is a non-stoichiometric product of graphite oxidation in a highly acidic environment. Oxygen-containing functional groups (carboxyl, epoxy, carbonyl, and hydroxyl ones) emerge in the graphite structure in the process of oxidation. Individual oxidized graphene sheets may only exist on a substrate as a monolayer or in suspension. Such individual sheets are referred to as „graphene oxide" in literature. When GO materials dry, layers „stick together," forming particles consisting of tens and hundreds of sheets. In the present study, both individual and „bound" sheets of oxidized graphites are denoted as GO.

Unlike the original graphite, GO is hydrophilic and swells in polar liquids with an increase in interplanar spacing [1,2]. Deeply oxidized GOs ($\approx$ C/O $2-4$) do not sorb non-polar liquids, such as octane or benzene, and do not swell in them [1,3]. There are two main approaches to GO synthesis: the Brodie method (BGO) [4] and the Hummers method (HGO) [5]. In the former method, the original graphite is oxidized with potassium chlorate and fuming nitric acid. In order to achieve complete oxidation of the original material, the oxidation procedure is repeated several times. The Brodie method is potentially explosive. Different variations of the Hummers method are used more often at present. The idea of this method is that graphite is oxidized in a sulfuric acid environment or a mixture of sulfuric and orthophosphoric acids with potassium permanganate [6–8]. The synthesis rate is higher, and a uniformly oxidized product is obtained already at the first stage.

The main physicochemical parameters for characterization of GO materials are the interplanar spacing (X-ray diffraction analysis method), the C/O ratio (X-ray photoelectron spectroscopy method), and the ratio of sp^3 and sp^2 carbon atoms (Raman spectroscopy). It was also demonstrated in [1,3,8–12] that the sorption values for certain liquids (differential scanning calorimetry (DSC) and isopiestic experiment) may serve as reproducible characteristics of GO materials. The DSC method is used to estimate the sorption of liquid by GO at the melting temperature of the sorbed substance in direct contact with GO. In an isopiestic experiment, GO contacts the sorbed substance via the gas phase at room temperature. The sorption properties of GO materials are being studied actively at present, since they are related directly to the permeability of GO membranes that are used for separation and purification of liquids. Although a large number of studies focused on these issues have already been published, the mechanism of sorption of liquids by GO has not been characterized.

The sorption of polar substances by GO is normally accompanied by swelling of the sorbent. The patterns of variation of sorption/swelling in transition from HGO to BGO, upon a change in temperature and pressure, and in transition from adsorbate to adsorbate in a series of normal alcohols have been reported in literature [9–11].

The C/O ratio is one of the parameters that may be adjusted in order to control the sorption properties of GO. Reduced GO (rGO) has been shown to sorb water in a similar manner to HGO from which it was synthesized [12]. More detailed comparative data on the sorption properties of GO materials with different degrees of oxidation have not been published.

In the present study, we propose to compare HGO and thermally expanded graphite (TEG) to identify the features of sorption properties of graphite materials. TEG-based materials are used widely to purify water from oil contamination. TEG may be regarded as a highly underoxidized graphite oxide with a low concentration of oxygen-containing groups. It is normally synthesized in several stages. At the first stage, graphite is intercalated with strong Brønsted acids (HNO_3, H_2SO_4, and $HClO_4$). Intercalation proceeds as graphite interacts with an intercalating acid in the presence of an oxidizing agent [13–15]. Anions and neutral acid molecules penetrate into the interplanar space of graphite as the graphite matrix gets oxidized. Electrochemical oxidation of the graphite matrix, wherein graphite is oxidized in an intercalate solution that serves as an electrolyte, is also possible [16]. This method allows one to obtain overoxidized forms of graphite. The next stage of TEG synthesis involves rinsing the intercalated compounds with water, which results in the formation of oxygen functional groups on the graphite surface. The obtained oxidized graphite is then subjected to high-temperature processing, which induces a significant expansion of the graphite matrix and results in the formation of TEG [17,18].

Thermal expansion is caused by the destruction of oxygen groups in oxidized graphite with the formation of gaseous CO, CO_2, and H_2O, which generate dispersive pressure in the graphite matrix and induce a significant increase in its volume. Thermal processing is normally carried out at temperatures of 600 °C–1000 °C, since it is at these temperatures that the interplanar spacing in the graphite matrix increases significantly. However, oxidized graphite obtained by electrochemical anodic overoxidation of graphite is characterized by a high degree of expansion at temperatures above 580 K [16]. The graphite matrix of such TEG contains oxygen groups that did not decompose during thermal processing.

The aim of the present study was to compare the sorption properties of GO materials with different degrees of oxidation. GO synthesized by the Hummers method and TEG with just a small amount of oxygen-containing groups on its surfaces were compared. Sorption values for typical polar and non-polar liquids were determined experimentally. The sorption rates and swelling process parameters were compared.

1. Experimental

1.1. Materials

HGO was synthesized by the improved Hummers method. Manganese impurities were removed by rinsing the material with hydrochloric acid (30 %) and ethanol (96 %). The cryochemical method was used for HGO drying.

At the first stage of TEG synthesis, electrochemical overoxidation of graphite was performed in accordance with the procedure detailed in [20]. Electrochemical oxidation of graphite in a 60 % nitric acid solution was carried out in a Teflon cell. Graphite served as the anode, which was positioned on a platinum current lead; a platinum wire acted as the cathode; and a nitric acid solution was the electrolyte. Graphite was oxidized in the galvanostatic mode with $Q = 500\,\mathrm{mA{\cdot}h{\cdot}g^{-1}}$. The obtained graphite nitrate was rinsed in water at a ratio of $m(\mathrm{graphite}):m(H_2O) \sim 1:7$ on a glass porous filter to form oxidized graphite. The resulting oxidized graphite was then dried in a drying oven at 335 K for 6 h. Thermally expanded graphite was produced via thermal processing of oxidized graphite in a quartz cup for 120 s in a muffle furnace at a temperature of 300 °C.

1.2. Instruments

X-ray diffraction studies were carried out using a Rigaku D/MAX 2500 diffractometer (Japan) (Cu$K\alpha$ radiation). The diffraction patterns were recorded in 0.02° steps within the range of angles $2\theta = 2 - 80^\circ$ with an exposure of 2 s per point. Prior to experiments, the samples were dried in a desiccator with phosphorus (V) oxide. To record diffraction patterns of swollen samples in liquid, they were first kept in contact with the studied liquid for seven days. The X-ray patterns were recorded at room temperature.

A temperature dependence of XRD spectra within the temperature range of 250−350 K was obtained additionally for TEG samples in direct contact with liquid water. These samples were heterogeneous mixtures of TEG and a significant excess of water. A Synergy-DW HyPix-Arc 1501 (Rigaku Oxford Diffraction) instrument with Mo$K\alpha$ ($\lambda = 0.7090$ Å) radiation was used for measurements. It was fitted with a Cobra Plus cryosystem (Oxford Cryosystems). It took 15 min to establish an equilibrium temperature before each measurement.

Prior to the measurement of X-ray photoelectron spectra (Kratos Axis Ultra DLD), the samples were degassed at a pressure of 10^{-9} Torr for two hours. An aluminum anode (Al$K\alpha$) was the radiation source. Survey spectra were recorded at a transmission energy of 160 eV. Range: 5−1320 eV. High-resolution spectra were recorded at a transmission energy of 40 eV. The spectra were calibrated against the binding energy of carbon C1s − 284.5 eV.

Differential scanning calorimetry was used to study the sorption characteristics of TEG and GO materials. The obtained sorption values correspond to the melting temperature of the sorbed substances. The method allows one to determine the mass of liquid sorbed in micropores ($d < 2\,\mathrm{nm}$) [8]. The mass ratio of liquid and the graphite material in samples was varied from 3:1 to 1:1. Prior to experiments, the graphite material samples were first dried in a desiccator with phosphorus (V) oxide, and a given amount of polar or non-polar liquid was then added to them. A Mettler DSC−30 low-temperature calorimeter (Switzerland) was used in DSC experiments. The temperature scale and heat flow of the device were calibrated against the temperature and enthalpy of melting of pure indium.

The following liquids were tested: water (deionized), octane (Sigma-Aldrich), 1-octanol (Sigma-Aldrich), and acetonitrile (Sigma-Aldrich).

EPR spectra were measured with a Bruker EMX plus spectrometer fitted with a highly sensitive Bruker ER 4119 HS resonator.

The spectra were recorded at a microwave power of 1 mW and a modulation amplitude of 1 G. It was found that these parameters do not distort the shape of the spectrum line of nitroxyl radicals sorbed on the inner surface of TEG. TEG samples containing intercalated water were prepared as follows: a solution of the nitroxyl radical ((2,2,6,6-tetramethylpiperidin-1-yl)oxyl, TEMPO) in octane was added to vacuum-treated TEG powder (vacuum degassing at $5 \cdot 10^{-4}$ Torr, for 12 h). The amount of nitroxyl radicals corresponded approximately to $(2-3) \cdot 10^{16}$ particles per 1 mg of material. The sample was then dried in air and subjected to vacuum treatment to remove octane. TEG containing a spin probe was then introduced into a glass capillary with an internal diameter of 1 mm. A certain volume of liquid water (equal to or below the level of equilibrium sorption at room temperature) was added to the sample in the capillary with a mass of $\sim 2\,\mathrm{mg}$. The capillary was sealed and centrifuged for 10 min at a speed of 2500 rot/min to distribute water uniformly throughout the material.

2. Results and discussion

XRD is the primary analysis method for characterization of dry and swollen graphite materials. The interplanar spacing (reflection (001)) in the original graphite was 3.4 Å (Fig. 1). This value is a reference for comparison with other graphite materials. Following oxidation, the interplanar spacing increases in dry oxidized materials due to the formation of oxygen-containing functional groups. The magnitude of interlayer distance extension is an indicator of the degree of oxidation of the material. Specifically, HGO has an interplanar spacing of 7.5 Å. TEG has the same interplanar spacing as the original graphite, since there are much fewer oxygen-containing groups in the interplanar space of TEG than in HGO. X-ray photoelectron spectroscopy measurements were performed to assess quantitatively the oxidation state (C/O ratio) and the content of functional groups.

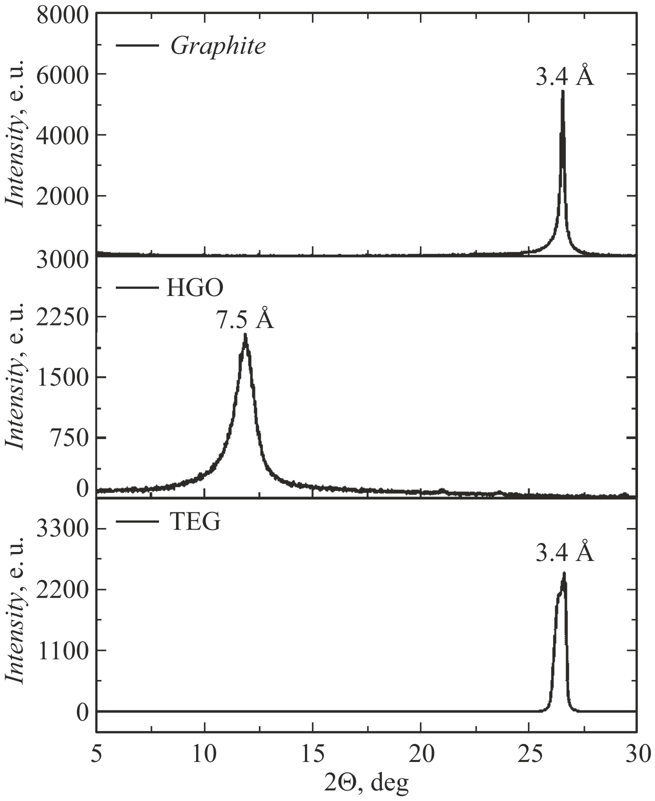

Figure 1. XRD data.

Table 1. Concentrations of elements on the surface of the studied samples (at.%) calculated from survey XPS spectra

Sample	C/O	O	C	N	Si	S	Cl
TEG	9.5	9.4	89.7	0.4	0.4	–	0.1
HGO	2.5	28.4	70.3	0.6	–	0.5	0.2

Table 1 presents the data on the elemental composition of HGO and TEG material samples. Minor impurity inclusions of elements contained in the oxidizing mixtures are seen. The C/O ratios for HGO (2.5) and TEG (9.5) differ by a factor of almost 4. This ratio was determined both from the survey XPS spectrum and from the spectrum after deconvolution. Although TEG has a significantly lower degree of oxidation, the qualitative composition of functional groups is almost the same as in HGO (Table 2).

Table 3 lists the values of equilibrium sorption of liquids by HGO and TEG. Despite significant differences in the degree of oxidation and interlayer spacing, the sorption of water, acetonitrile, and 1-octanol by TEG is comparable with the values obtained for HGO. Non-polar octane is not sorbed by HGO due to high oxidation of the material.

Table 2. Binding energies and fractions of components in high-resolution XPS spectra and the corresponding bond types for HGO and TEG samples

<table>
<tr><td colspan="6">HGO</td></tr>
<tr><td rowspan="2">O 1s</td><td rowspan="2">26.19</td><td>532.7</td><td>18.48</td><td colspan="2">C = O, O-C-O</td></tr>
<tr><td>533.2</td><td>7.71</td><td colspan="2">O-(C = O), O-H</td></tr>
<tr><td rowspan="3">C 1s</td><td rowspan="3">72.55</td><td>285.0</td><td>36.20</td><td colspan="2">C-C (sp^3)</td></tr>
<tr><td>287.1</td><td>26.64</td><td colspan="2">C = O</td></tr>
<tr><td>288.2</td><td>9.71</td><td colspan="2">O-C = O</td></tr>
<tr><td rowspan="2">N 1s</td><td rowspan="2">0.62</td><td>399.3</td><td>0.36</td><td colspan="2">O = C-NH$_2$</td></tr>
<tr><td>402.1</td><td>0.26</td><td colspan="2">NH$_4^+$, C-N-O, nitrate</td></tr>
<tr><td rowspan="2">S 2p</td><td rowspan="2">0.51</td><td>168.8</td><td>0.34</td><td>$2p_{3/2}$</td><td rowspan="2">Sulfate</td></tr>
<tr><td>170.0</td><td>0.17</td><td>$2p_{1/2}$</td></tr>
<tr><td>Cl 2p</td><td>0.13</td><td>200.2</td><td>0.09</td><td>$2p_{3/2}$</td><td>C-Cl</td></tr>
<tr><td colspan="6">TEG</td></tr>
<tr><td>Line</td><td>Element concentration, at.%</td><td>Bond energy, eV</td><td>Fraction, at.%</td><td colspan="2">Bond type</td></tr>
<tr><td rowspan="3">O 1s</td><td rowspan="3">9.02</td><td>531.3</td><td>1.24</td><td colspan="2">C-O</td></tr>
<tr><td>532.3</td><td>6.23</td><td colspan="2">Si-O</td></tr>
<tr><td>533.6</td><td>1.55</td><td colspan="2">O-H</td></tr>
<tr><td rowspan="4">C 1s</td><td rowspan="4">86.47</td><td>284.5</td><td>78.20</td><td colspan="2">C-C (sp^2)</td></tr>
<tr><td>286.2</td><td>3.13</td><td colspan="2">C = O, O-C-O</td></tr>
<tr><td>288.4</td><td>1.00</td><td colspan="2">$\pi - \pi^*$</td></tr>
<tr><td>291.1</td><td>4.14</td><td colspan="2">$\pi - \pi^*$</td></tr>
<tr><td rowspan="2">N 1s</td><td rowspan="2">0.39</td><td>400.0</td><td>0.29</td><td colspan="2">C-NH$_3^+$</td></tr>
<tr><td>405.4</td><td>0.10</td><td colspan="2">Nitrate</td></tr>
<tr><td rowspan="2">Si 2p</td><td rowspan="2">0.41</td><td>102.2</td><td>0.27</td><td colspan="2">$2p_{3/2}$</td></tr>
<tr><td>102.9</td><td>0.14</td><td colspan="2">$2p_{1/2}$</td></tr>
</table>

DSC allows one to determine only the values of equilibrium sorption into micropores of the material being studied. The method is insensitive to liquid contained in macropores ($>$ 30 nm). It provides an opportunity to determine separately the mass of liquid in mesopores ($1 < d < 30$ nm), but characteristic peaks corresponding to this type of sorption were not detected in the DSC curves in the present study. Thus, DSC sorption measurements are representative of the mass of adsorbed liquid in slit micropores (i.e., in the interplanar space of graphite oxides).

It has been demonstrated in literature that liquid molecules are sorbed into the interlayer space of HGO and BGO [1]. The material swells as a result, and the interlayer spacing increases from approximately 7−8 Å to 12−20 Å. The swelling of TEG in the process of sorption of liquids was studied using the X-ray diffraction method for samples soaked in the studied liquid at room temperature (Fig. 2). TEG swells only in non-polar octane and weakly polar 1-octanol. In both cases, the intercalation of octane and 1-octanol into the interlayer space faces kinetic difficulties. The original graphite peak was observed even after the material was left immersed in liquid for a week. Note that highly oxidized materials (HGO and BGO) reached sorption equilibrium in 15−20 min. The interplanar spacing in the „TEG+octane“ and „TEG+1-octanol“ systems increases by the same amount (from 3.4 to 4.5−4.6 Å). The reflection corresponding to the interplanar spacing in swollen structures is also broadened significantly, which is indicative of disordering of the graphite structure as a result of its interaction with molecules of the intercalated liquid. It was demonstrated in [1,7,9] that the interlayer distance in HGO increases by 4.5 Å on average for such solvents as water and acetonitrile. In the case of TEG, 4.6−4.7 Å is the absolute value of interplanar spacing determined after the material is brought into contact with octane and 1-octanol.

Table 3. DSC data

Sample	Water (g/g), $T = 273\,K$	Acetonitrile (g/g), $T = 229\,K$	Octanol-1 (g/g), $T = 257\,K$	Octane (g/g), $T = 216\,K$
TEG	0.5 ± 0.1	0.45 ± 0.07	0.75 ± 0.07	0.67 ± 0.07
HGO	0.63 ± 0.04	0.47 ± 0.08	0.71 ± 0.09	≈ 0

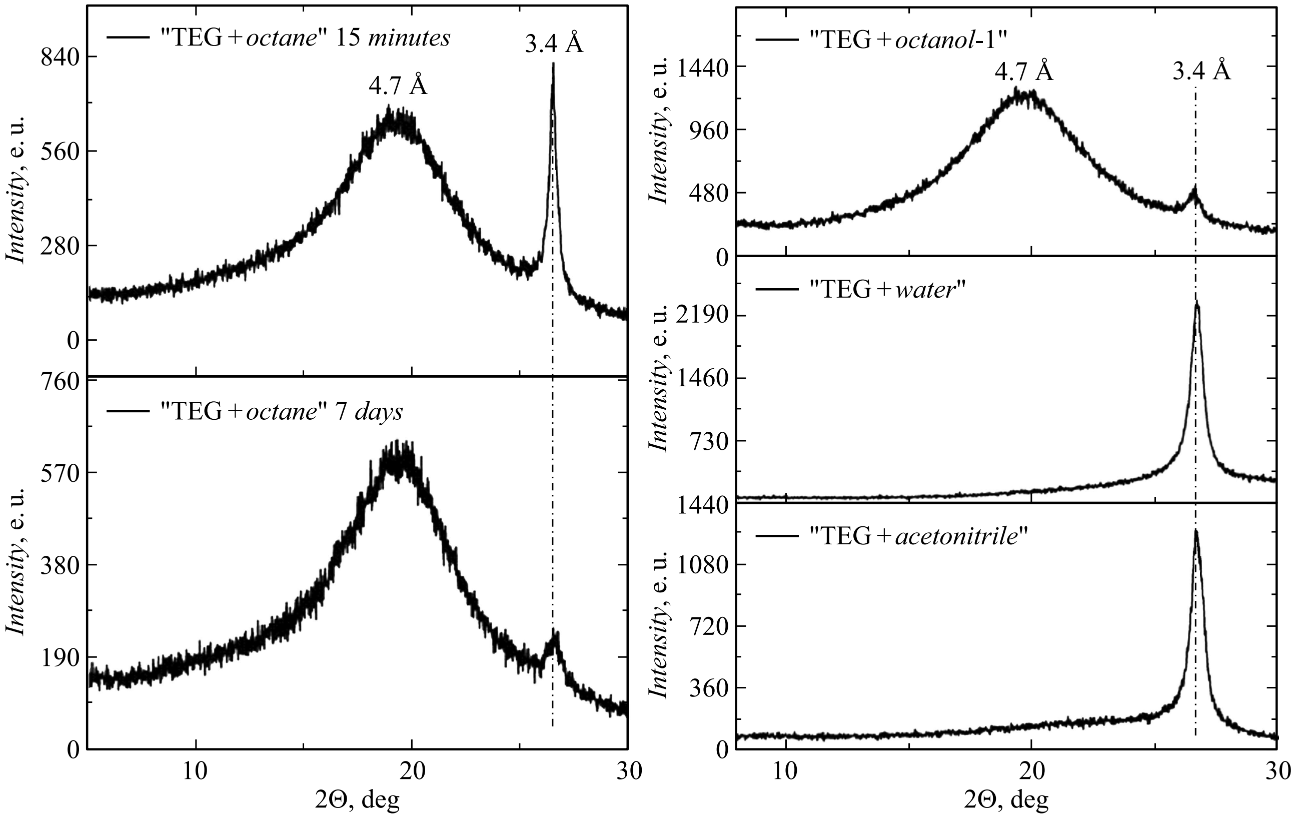

Figure 2. XRD data for „TEG + liquid“ systems.

The DSC and XRD data for swollen structures suggest that the intercalation of a certain volume of liquid requires the same distance for both strongly and weakly oxidized graphite materials. The presence of at least a small amount of oxygen-containing functional groups is a fundamental factor for water sorption. It has been demonstrated earlier that the original graphite does not sorb water [1,3,19].

When water and acetonitrile are in direct contact with TEG, the graphite structure does not swell. It may be noted that the level of sorption into micropores (interlayer distance) of TEG determined by DSC is comparable to the HGO sorption values for water and acetonitrile.

It has been demonstrated in literature that an increase in sorption by HGO at lower temperatures is accompanied by an increase in the interlayer distance. In order to determine the interlayer distances at low temperatures in the „TEG+water“ system, diffraction patterns were recorded within the temperature range of 293–250 K (Fig. 3). The (002) reflection corresponding to the interlayer distance in TEG is superimposed on reflections of the crystalline structure of ice in the diffraction patterns recorded below 273 K. It is evident that the TEG reflection does not shift toward lower 2θ values as the freezing temperature decreases. Swelling in the „TEG+water“ system is not observed even at below-room temperatures.

The obtained DSC and XRD data (at different temperatures) cannot provide a clear indication where water sorbed in the material is located. The EPR spectroscopy method (in the spin probe variation) was used to verify that water molecules may intercalate into the interplanar space of TEG without causing swelling of the material.

EPR spectra of nitroxyl radicals are highly sensitive to the rotational mobility of probes, which correlates with the microviscosity of the medium. Thus, we may gain an insight into the molecular mobility of the medium, its microviscosity, and the phase state by analyzing the

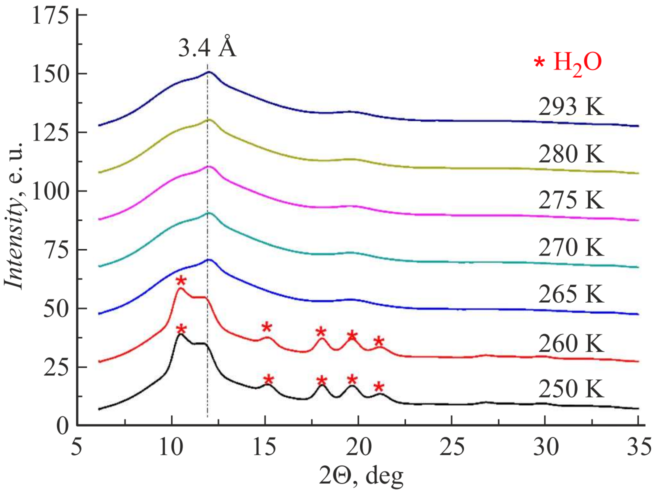

Figure 3. XRD data for the „TEG+H_2O“ system at low temperatures.

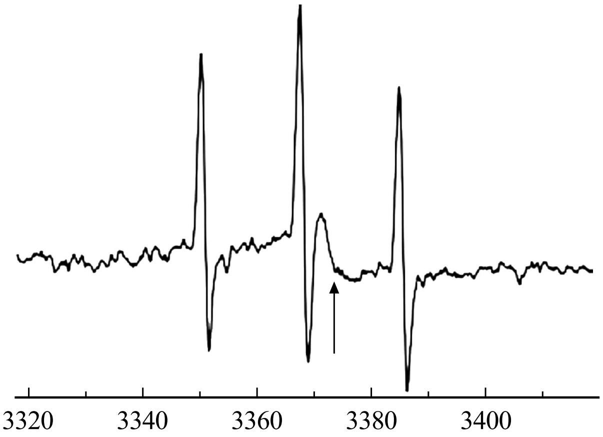

Figure 4. EPR spectrum of the „TEG+H_2O+TEMPO“ system.

line shape in the spectra of nitroxyl radicals. The spin probe technique was proven to be an efficient tool for determining the mobility of polar liquids intercalated into graphite oxides synthesized by various methods [8]. TEG is known to be a highly conductive material, and its intrinsic EPR signal is an asymmetric singlet with a characteristic Dyson line shape. This EPR signal shape is associated with the presence of conduction electrons in the material. Charge carriers are expected to affect the shape of the EPR spectrum line of spin probes, allowing one to determine the material regions into which water molecules are intercalated.

The water-saturated material features an EPR spectrum combining of two signals: the intrinsic TEG signal and the nitroxyl radical signal. Notably, the nitroxyl radical signal is a resolved triplet, which implies that water intercalated into TEG is characterized by high mobility. However, each component of the triplet signal is asymmetric, and the line shape of all components corresponds to the Dyson one. This suggests that nitroxyl radicals are localized in the mobile fraction located between slightly oxidized TEG layers spaced by 3.4 Å. An exchange of unpaired electrons of nitroxyl radicals and conduction electrons is possible in this case, which induces an asymmetric distortion of the line shape. A certain fraction of nitroxyl radicals may be localized in water intercalated into TEG macropores (larger than 30 nm in size). The mentioned exchange of unpaired and conduction electrons may be impeded in this case, and the EPR spectrum of spin probes will assume the form of a symmetrical triplet signal. Thus, the overall EPR spectrum may be represented as a sum of two signals (symmetrical and asymmetrical), which yields an asymmetrical resulting signal (Fig. 4).

3. Discussion and conclusion

A comparative study of sorption/swelling of the TEG material, which may be regarded as a weakly oxidized graphite oxide with C:O $\approx$ 9.5, was carried out. TEG is not exactly a reduced form of ordinary graphite oxide, since a slightly different method was used to produce it.

In the experimental study of TEG, its sorption properties were compared systematically with those of highly oxidized HGO. Adsorbents were selected in such a way as to examine the effect of polarity (water and octane) and the size of the sorbent molecule (water, acetonitrile and octane, octanol). It was demonstrated that a reduction in the number of oxygen-containing groups does not lead to the transformation of a hydrophilic material into a hydrophobic one. TEG sorbs 30–50 % more water than HGO. A similar result was obtained in [12], where water sorption by HGO increased in the process of its reduction. At the same time, TEG sorbs octane, while its sorption by HGO is virtually zero. It can be said that a reduction in the number of oxygen-containing groups does not lead to suppression of hydrophilicity of the material, but enhances its oleophilicity. TEG sorbs octane significantly better, but loses its capacity for selective sorption in the process. The material becomes amphiphilic. A reduction in the number of oxygen-containing groups translates into a significant reduction in the rate of sorption. This made isopiestic measurements, wherein the sorbent and the adsorbate contact only through the gas phase, impossible.

The swelling of TEG is significantly less pronounced than that of HGO. In the case of octane and octanol, the interplanar spacing increases by 1–1.5 Å. However, the resulting distance between GO planes is 4.5–4.6 Å, which corresponds to the interplanar spacing increase after the introduction of a single layer into HGO and BGO. It can be argued that in TEG, which has no oxygen-containing groups, the sorbed molecules may occupy the entire space between oxidized graphene planes (Fig. 5). In the case of water, which consists of molecules with a characteristic size below 3 Å, a single layer may be sorbed without swelling, which is what was observed in the experiment. DSC experiments revealed reproducible sorption of water into

Figure 5. Model characterizing the mechanism of sorption and intercalation by TEG.

the interplanar space; however, the interplanar spacing was found to remain unchanged even after prolonged contact with liquid water and a reduction in temperature. EPR experiments confirmed that sorbed water is located between the GO planes.

One of the possible areas of application of GO membranes is water desalination. The size effect (i.e., a GO material with small channels with diameter $d \leq 3-4$ Å) is proposed to be used for removal of cations and anions formed in the process of dissociation of electrolytes. This small diameter is not an impediment to the penetration/sorption of water into pores (the diameter of a water monolayer is 3 Å) and lets it pass through the membrane, but prevents the adsorption of hydrated ions in narrow channels. A problem arises in contact with liquid water. The sorption of water by HGO is normally accompanied by swelling; as the interplanar spacing increases, large particles get a chance to penetrate the material freely and will not be retained by the membrane [1]. GO modified with various spacer molecules, which have the capacity to maintain a certain fixed interplanar distance, may be used to prevent swelling. It has also been proposed to synthesize GO materials under controlled humidity and thus fix the interplanar spacing in them. TEG may be a solution to this problem, since sorption/swelling in this material is limited to a single monolayer, and an interplanar spacing of 3,4 Å does not increase upon contact with aqueous media. With this interplanar spacing, even the smallest cations (K^+ and Na^+) are unable to penetrate into channels and pass through the membrane.

Conclusion

The results of a comparative study of sorption properties of HGO and TEG graphite oxides with different oxygen concentrations suggest the following:

(1) a reduction in the number of oxygen-containing groups in a graphite oxide does not necessarily result in suppression of sorption of polar liquids;

(2) when the number of oxygen-containing groups decreases, the material starts to sorb typical non-polar liquids (octane) and becomes amphiphilic;

(3) the sorption rate decreases noticeably following a reduction in the number of oxygen-containing groups; and

(4) the sorption of small molecules (water, acetonitrile) by weakly oxidized TEG is not accompanied by an increase in interplanar spacing (swelling). This indicates that TEG may potentially be used to produce membranes retaining a fixed interplanar spacing in aqueous media.

Conflict of interest

The authors declare that they have no conflict of interest.

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