Supporting Information

Multilayered intercalation of 1-octanol into Brodie graphite oxide.

Alexey Klechikov,¹ Jinhua Sun¹, Igor A. Baburin,³ Gotthard Seifert,³ Anastasiia T. Rebrikova,² Natalya V. Avramenko,² Mikhail V. Korobov,² Alexandr V. Talyzin¹

¹ Umeå University, Department of Physics, S-90187 Umeå, Sweden

² Department of Chemistry, Moscow State University, Leninskie Gory 1-3, Moscow 119991, Russia

³ Technische Universität Dresden, Theoretische Chemie, Bergstraße 66b, 01062 Dresden, Germany

1. XRD data for B-GO in liquid 1-octanol for broader range of temperatures.



Figure 1S. Temperature dependence of d(001) summarized by results of two experiments: one with cooling below room temperature and second with heating from room temperature up to 1373K. Linear decrease of d(001) is observed for temperature range 273-353K and stabilization around 20.3Å at higher temperatures. The heating part

was not completely reversible. The initial value of d(001) at 298K was 23.49Å, after heating-cooling cycle d(001)=21.66Å was found and after 15 hours at ambient temperature it increased up to 22.31Å. Most likely slight irreversibility is connected to effects of heating, either reduction of chemical modification. The inter-layer distance which corresponds to d(001) at 363-383K which is close to 3-layered solvate found by vacuum drying of B-GO/octanol sample (Figure 5 in the main text). In principle, the temperature dependence shown in Figure 1S can be considered as combination of two steps: one is very broad and smeared over the temperature interval ~273-250K and second is in narrower temperature interval of 260-270K.

2. XRD data for precursor BGO and ambient temperature BGO-octanol.



Figure 2S. XRD patterns recorded from solvent free precursor BGO powder and BGO-octanol at ambient temperature. The figure illustrates solvent induced ordering of BGO. Octanol intercalated BGO shows much smaller FWHM of 0.09 degrees (compared to 0.43 degrees for BGO) and higher order reflections from 00ℓ set.

3. Tentative phase diagram of the system B-GO – 1 octanol.

In ref. 1 it was proposed to consider intercalation of GO with polar organic solvents as formation of equilibrium phases, namely, solid solutions and solid solvates of the corresponding solvent in GO. The model was found useful in description of temperature/pressure dependences of sorption of small polar molecules into BGO and HGO.

The sketch of the BGO-1-octanol T-x phase diagram is presented in Fig. 1S. The diagram is based on the DSC heating traces (see Fig.2 of the main text) and shows the incongruent melting temperature and the compositions of the L- and A- solvates. Also shown is the incongruent melting temperature of the intermediate "M-solvate". The M-solvate is placed tentatively in this diagram to explain peak observed on DSC (**Figure 2** in the main text) scan recorded with nOct/nB-GO = 0.14 and (c) nOct/nB-GO = 0.12, compositions

intermediate between A-solvate and L-solvates. The DSC peak due to melting of Msolvate phase is found at lower temperatures and weaker compared to L-solvate melting. The composition of this solvate was not determined. All the three solvates are depicted as "point compounds" for simplicity. In reality they have broad homogeneity ranges. This may explain the substantial width of the DSC peaks and the gradual change of the interplane distance with temperature (see Fig.3b of the main text). The area from the left-hand side of the A-solvate in Fig. 1S is a solid solution of 1-octanol in BGO. The usefulness of the model in this part of the phase diagram is not confirmed by experimental data.



Figure 3S. B-GO- 1-octanol phase diagram. Displayed are equilibrium phases and compositions of the A- and L-solvates. The compositions of the solvates are expressed in molar fractions of 1-octanol, X_{oct} .