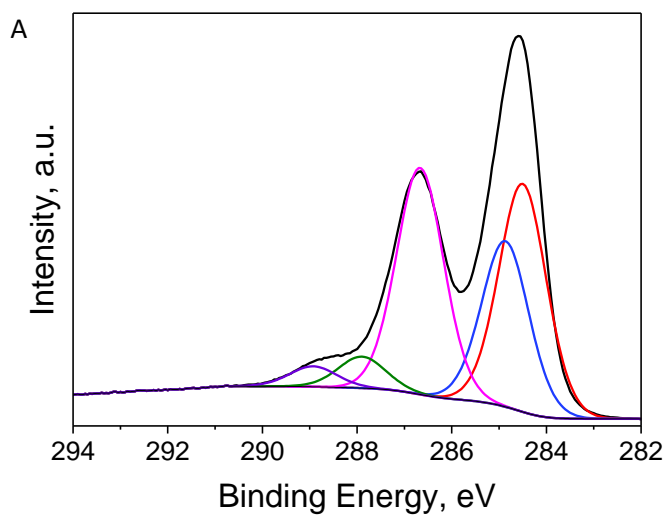


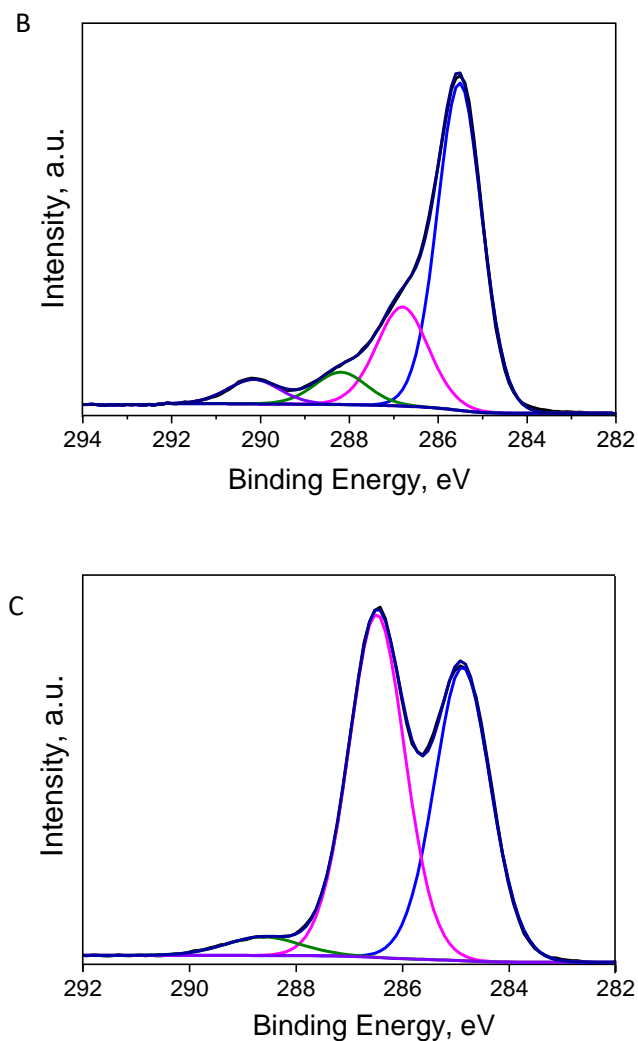
# Reduced graphene oxide-melamine and –polyurethane composites for the removal of organic and oil wastes

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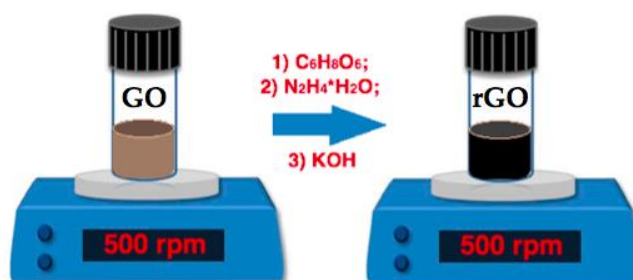
**Table S1.** Element concentrations on the surface of initial substances obtained by XPS spectra, at. %.

	C	O	N	Na	S	Cl	Si
Graphene oxide	63.3	34.4	-	0.8	1.0	0.2	-
Melamine	65.4	19.2	7.6	5.6	2.2	-	-
Polyurethane foam	69.0	24.3	-	3.2	-	-	3.5





**Figure S1.** C1s XPS spectrum of a(A) Graphene oxide, (B) Melamine and (C) Polyurethane sample

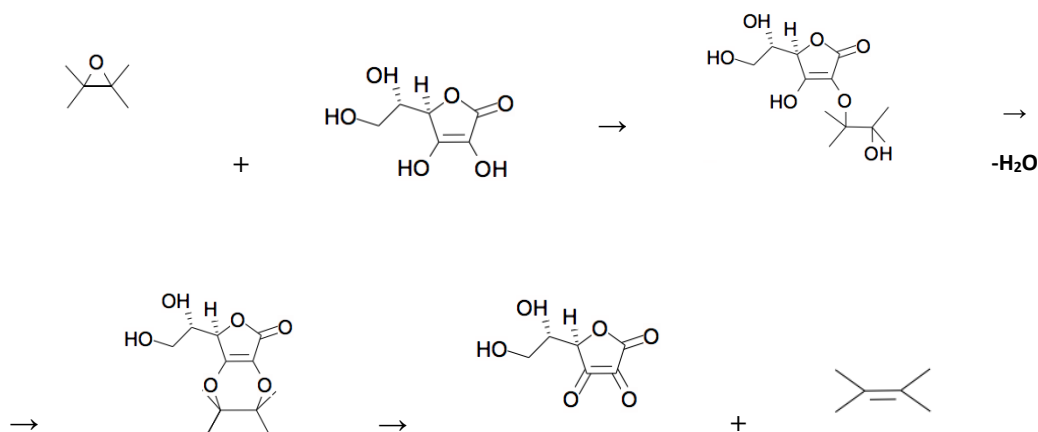


**Figure S2.** General scheme reduction of graphene oxide by using: 1) ascorbic acid; 2) hydrazine hydrate; 3) potassium hydroxide

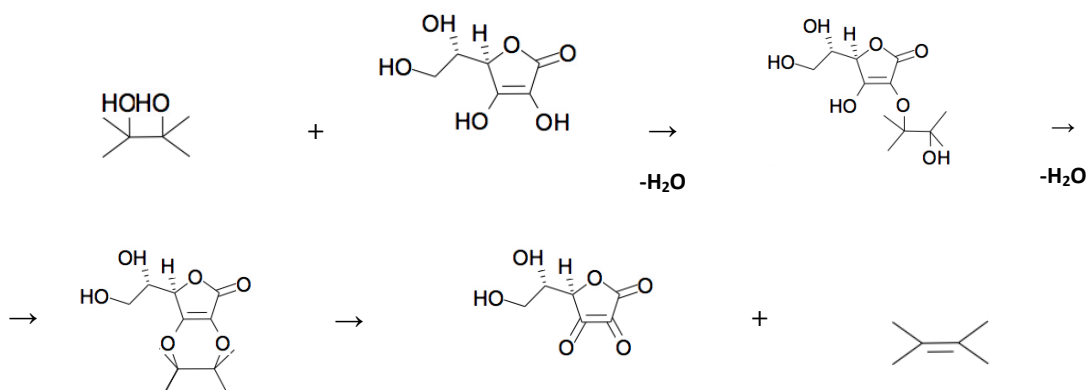
#### *Graphene oxide reduction mechanisms*

The GO reduction process with ascorbic acid as a reduction agent is based on  $\text{S}_{\text{N}}2$  mechanism of two-stage nucleophilic substitution [43]. The proposed ways of reduction of hydroxyl and epoxy functional groups of graphene oxide are presented below:

(a) restoration of the epoxy group:

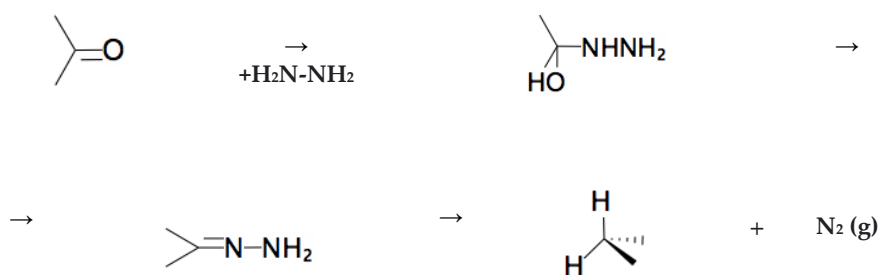


b) reduction of two OH-groups:



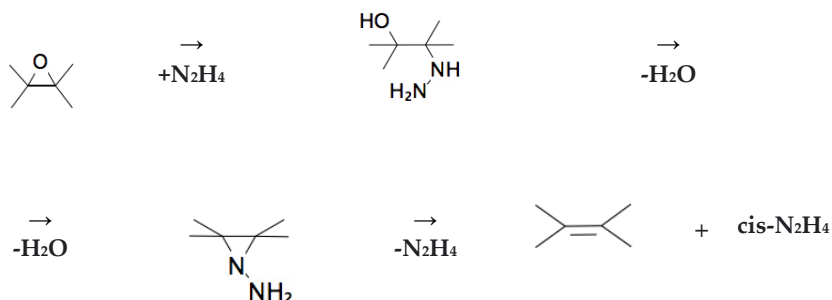
In the GO reduction process using hydrazine hydrate as a reducing agent is based on the Wolff-Kishner reduction mechanism (a), which starts with a nucleophilic attack of hydrazine on the carbon atom of the carbonyl functional group. The resulting products from this reduction are alkane and  $N_2$  gas.

(a) reduction of the carbonyl functional group:

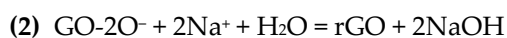
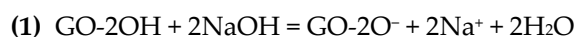


By using hydrazine hydrate, reduction can also be performed on the epoxide ring. This reduction mechanism is based on the research previously reported by Gao et al. [64]. The reduction of epoxide ring (de-epoxidation) starts with the hydrazine attack on the carbon atom of the backside of the epoxide rings forming the hydrazino alcohol. This is followed by hydrogen transfer from the hydrazino group to an alcohol group, which results in the formation of C-C and cis-diazene (cis- $N_2H_2$ ).

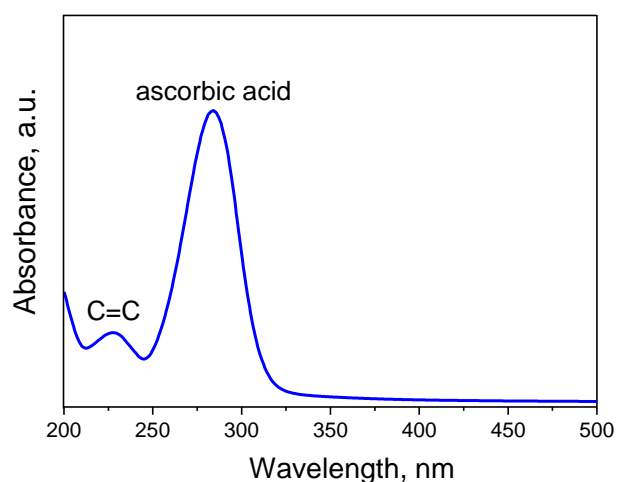
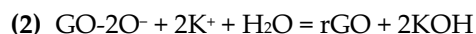
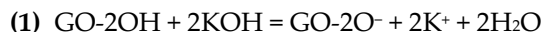
(b) reduction of the epoxy group:



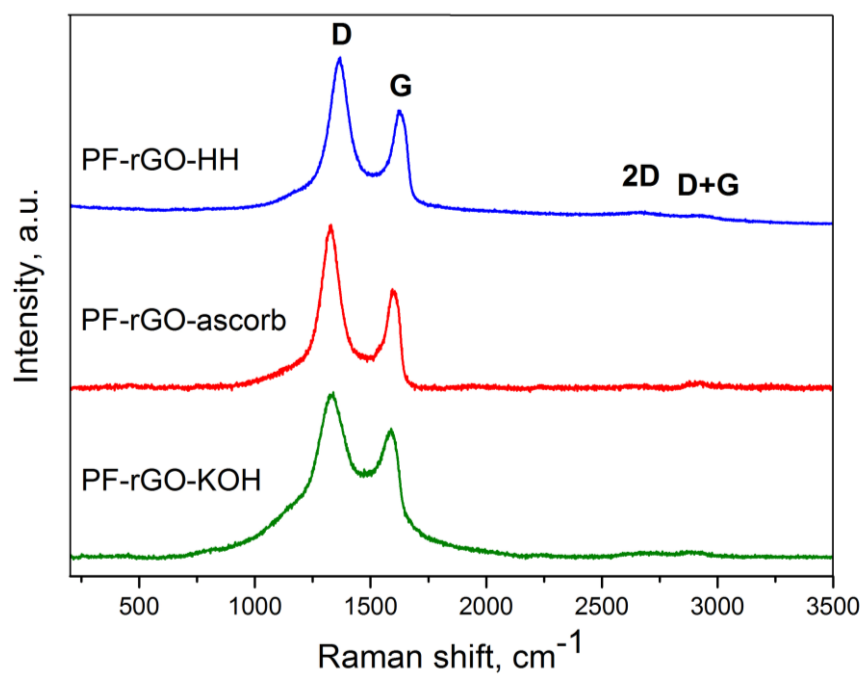
Potassium hydroxide was also used as a reagent for the reduction of graphene oxide. A simulation report showed that, in a NaOH solution, the OH<sup>-</sup> anion can interact with a hydroxyl group in GO to generate negatively charged GO. Due to this electron transfer, it is easy for the epoxy ring to be opened with a small energy barrier. These oxygen atoms on negatively charged GO can be removed with the assistance of Na<sup>+</sup> and a water molecule. In this process, NaOH can be considered as a catalyst [65]. The reduction reactions:



A similar mechanism is also evident for the case when potassium hydroxide is used as a reducing agent:



**Figure S3.** Absorption spectra of rGO-ascorb



**Figure S4.** Raman spectra of polyurethane-rGO composites

**Table S2.** Data obtained from Raman Spectra calculations

Sample	Position, cm <sup>-1</sup>				I <sub>D</sub> /I <sub>G</sub>	L <sub>a</sub> , nm
	D-mode	G-mode	2D-mode	D+G-mode		
PF-rGO-KOH	1335	1591	2671	2887	1.22	31.6
PF-rGO-ascorb	1330	1595	2649	2894	1.42	27.1
PF-rGO-HH	1331	1584	2633	2889	1.45	26.6

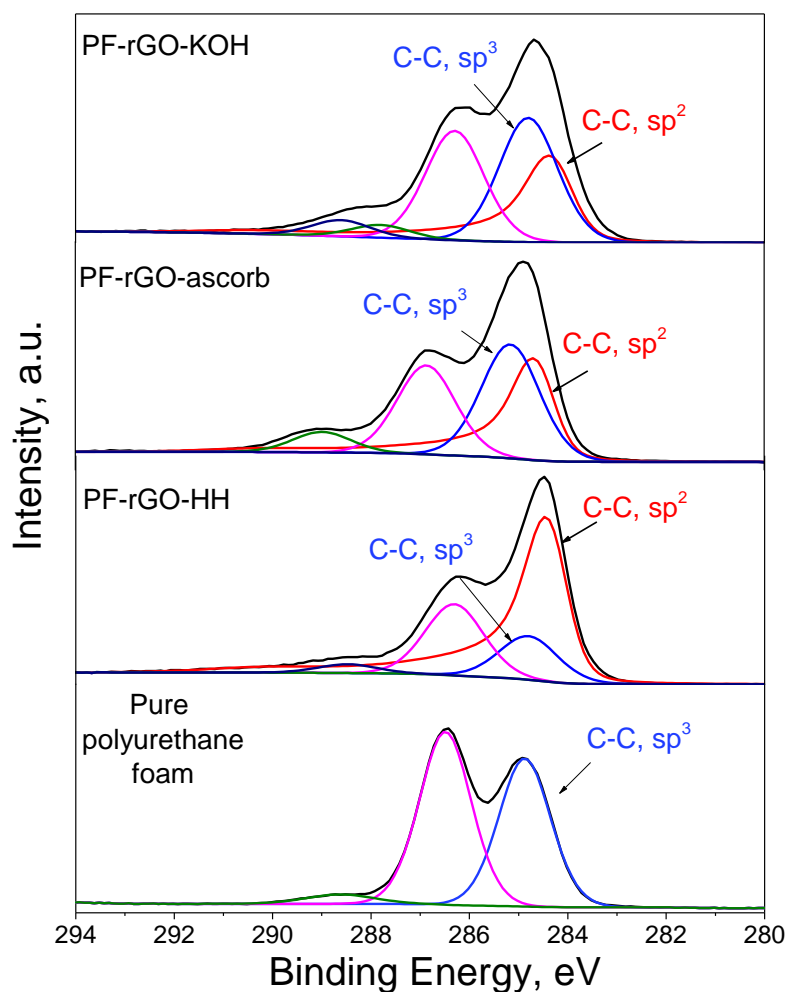


Figure S5. C1s XPS spectra of polyurethane-rGO composites

**Table S3.** Contents of different carbon species on the surface of polyurethane and polyurethane-based composites determined by XPS

Sample	Fraction, at. %			
	C-C (sp <sup>2</sup> )	C-C (sp <sup>3</sup> ), C-N	C-O	O=C-O, O=C-N, carbonate
Pure polyurethane foam	–	32.3	37.8	2.7
PF-rGO-HH	48.0	11.7	19.1	2.4
PF-rGO-ascorb	23.7	24.3	19.0	4.4
PF-rGO-KOH	22.2	26.2	23.2	6.2

## References

64. Gao, X.; Jang, J.; Nagase, S. Hydrazine and thermal reduction of graphene oxide: Reaction mechanisms, product structures, and reaction design. *J. Phys. Chem. C* 2010, 114, 832–842, doi:10.1021/jp909284g.
65. Chen, C.; Kong, W.; Duan, H.M.; Zhang, J. Theoretical simulation of reduction mechanism of graphene oxide in sodium hydroxide solution. *Phys. Chem. Chem. Phys.* 2014, 16, 12858–12864, doi:10.1039/c4cp01031k.