Supplementary Information for

Chemical Interaction and Imaging of Single Co_3O_4/Graphene Sheet Studied by Scanning Transmission X-ray Microscopy and X-ray Absorption Spectroscopy

Jian Wang*, Jigang Zhou*, Yongfeng Hu, and Tom Regier

Canadian Light Source Inc., University of Saskatchewan, Saskatoon, SK S7N 0X4, Canada

Contents (supplementary figures and table)

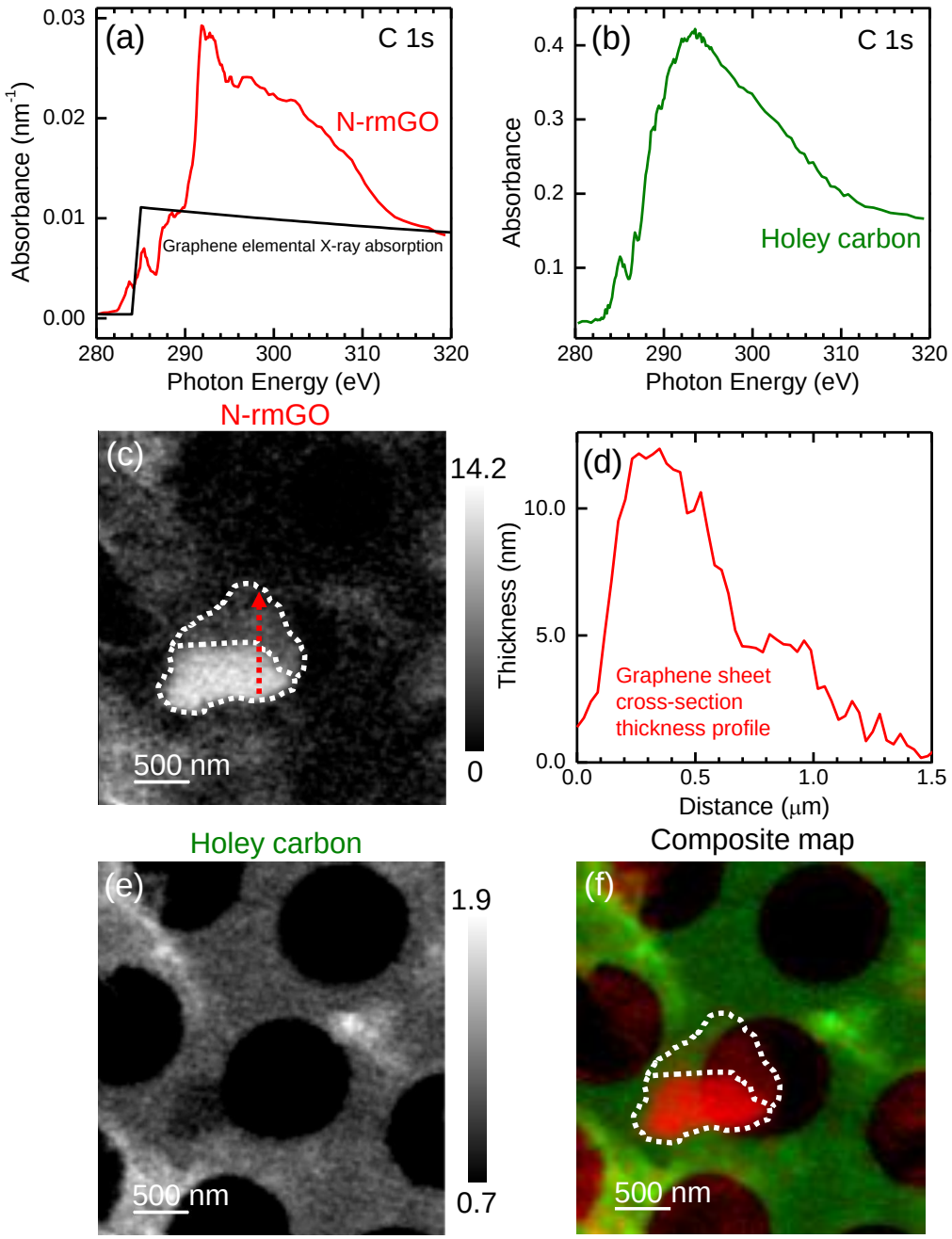

Figure S1 C K-edge STXM chemical imaging of individual Co_3O_4/N-rmGO sheets on a holey carbon film coated TEM grid, (a) linearly scaled reference spectrum of N-rmGO (spectrum scaled to the calculated graphene elemental X-ray absorption profile of 1 nm thick material, assuming graphene density is 2.16 g/cm^3, same as that of graphite); (b) reference spectrum of holey carbon obtained at a pure region on the sample; (c) thickness map of N-rmGO, the vertical grey scale represents the material thickness in nm, the enclosed white dotted lines indicate the regions of interest, i.e. a folded graphene sheet; (d) cross-section thickness profile along the red dotted arrow in (c); (e) map of holey carbon, the vertical grey scale represents the intensity ratio to the reference spectrum; (f) colour composite map, red color: N-rmGO, green color: holey carbon; the enclosed white dotted lines indicate the regions of interest.

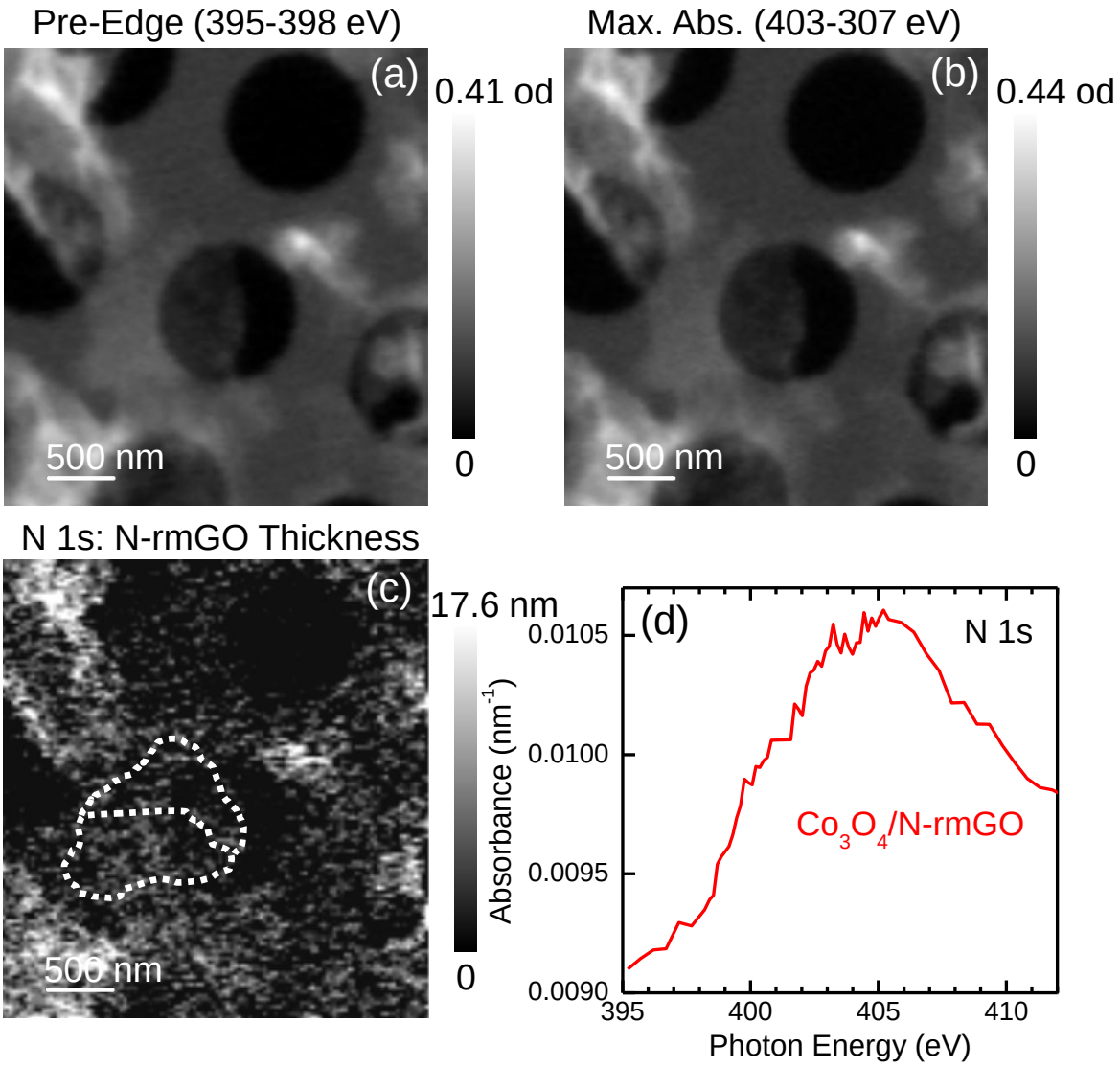

Figure S2 N K-edge STXM chemical imaging of individual Co_3O_4/N-rmGO sheets on a holey carbon film coated TEM grid, (a) N 1s pre-edge averaged optical density (od) image from 395 to 398 eV; (b) N 1s averaged od image at the maximum absorption from 403 to 407 eV; (c) N-rmGO thickness map derived from the N 1s, i.e. maximum absorption image (b) subtracts pre-edge image (a), then scaled by the edge-jump (difference of the pre-edge and the absorption maximum, i.e. 0.0015 od nm^{-1}) of (d) Co_3O_4/N-rmGO linear reference spectrum (nm^{-1}) at the N 1s. The enclosed white dotted lines indicate the regions of interest, i.e. a folded graphene sheet; (d) was obtained from the absolute absorption spectrum of Co_3O_4/N-rmGO, i.e. Fig. 2c, divided by the thickness of the sample region of interest (5.6 nm) from which the spectrum was obtained.

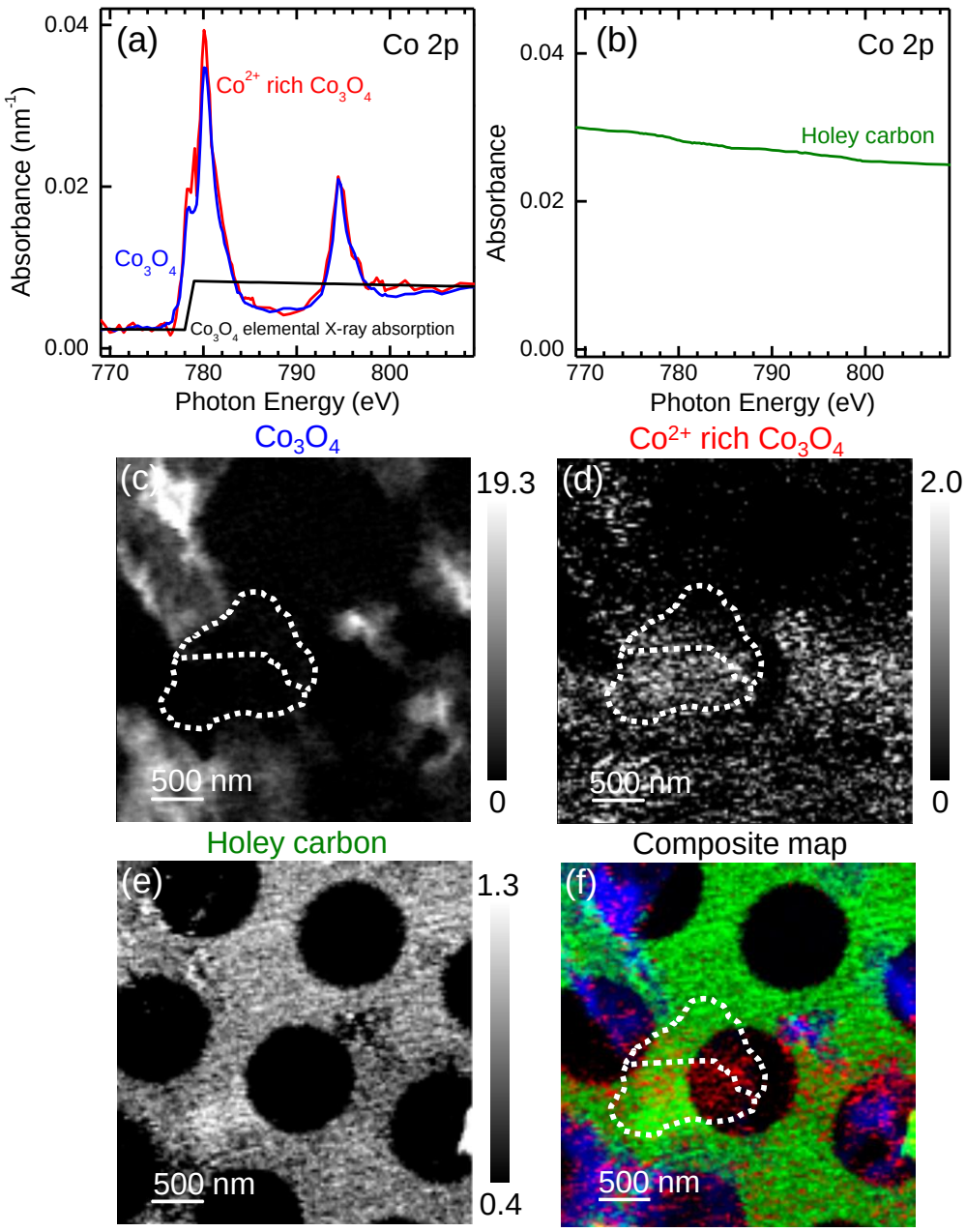

Figure S3 Co L-edge STXM chemical imaging of individual Co_3O_4/N-rmGO sheets on a holey carbon film coated TEM grid, (a) linearly scaled reference spectra of Co^{2+} rich Co_3O_4 and stoichiometric Co_3O_4 (spectra scaled to the calculated Co_3O_4 elemental X-ray absorption profile of 1 nm thick material, d = 6.11 g/cm^3); (b) reference spectrum (i.e. absorption baseline) of holey carbon at the Co L-edge; (c) and (d) thickness maps of stoichiometric Co_3O_4 and Co^{2+} rich Co_3O_4 respectively, the vertical grey scales represent the material thickness in nm; (e) map of holey carbon, the vertical grey scale represents the intensity ratio to the reference spectrum; (f) colour composite map, red color: Co^{2+} rich Co_3O_4, green color: holey carbon, blue color: stoichiometric Co_3O_4. The enclosed white dotted lines in (c), (d) and (f) indicate the regions of interest, i.e. a folded graphene sheet.

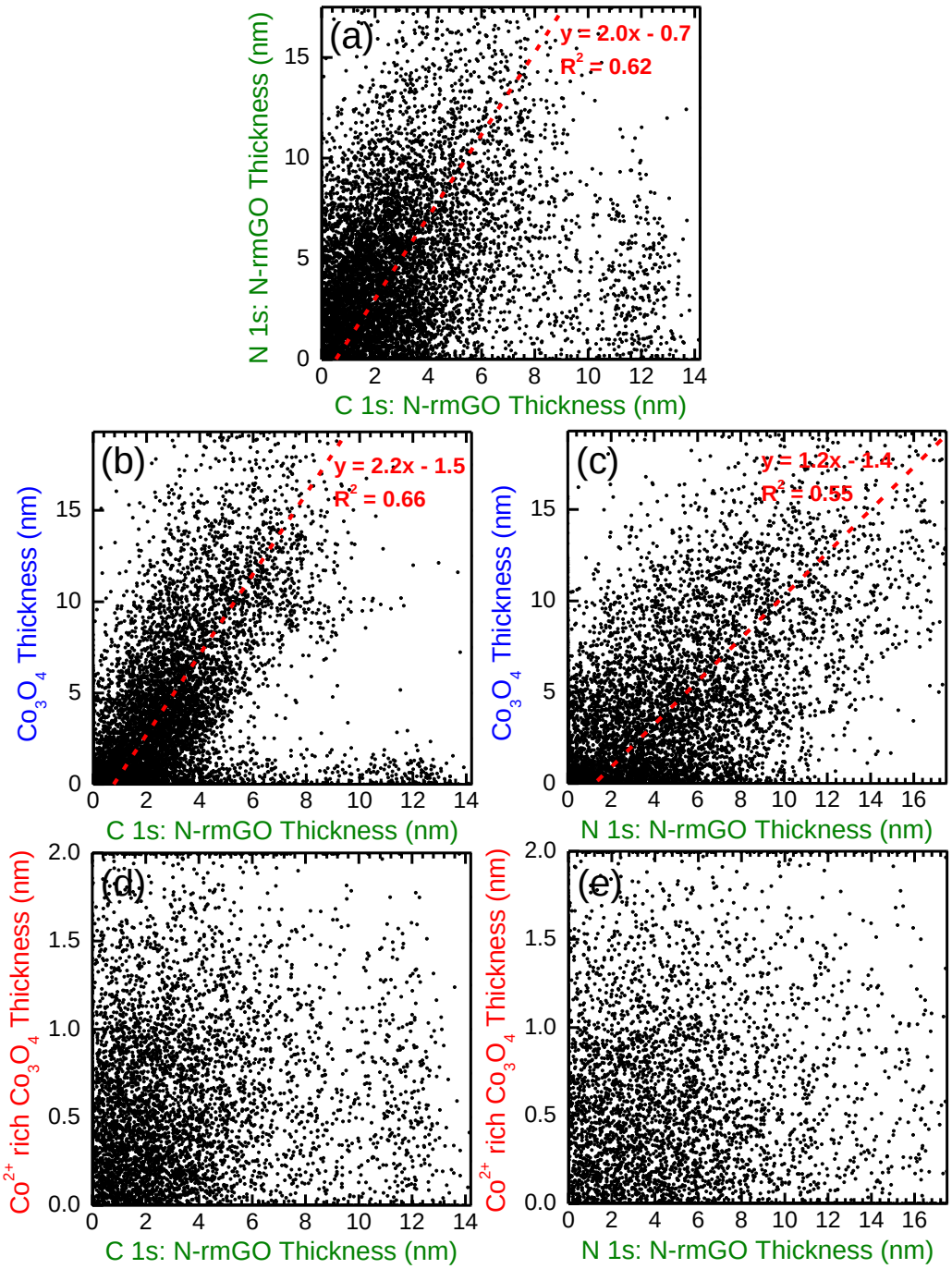

Figure S4 Correlation analysis for images from Fig. 3a to 3d of the main text, (a) correlation plot between N-rmGO thickness derived at the C 1s and N 1s edges, i.e. Fig. 3a versus Fig. 3b; (b) and (c) correlation plots between Co_3O_4 and the N-rmGO thickness derived at the C 1s and N 1s, respectively, i.e. Fig. 3c versus Fig. 3a and 3b respectively; (d) and (e) correlation plots between Co^{2+} rich Co_3O_4 and the N-rmGO thickness derived at the C 1s and N 1s, respectively, i.e. Fig. 3d versus Fig. 3a and 3b respectively. The red dashed lines in (a), (b) and (c) plots are fitted trendlines labelled with linear relationship equations and the correlation coefficients.

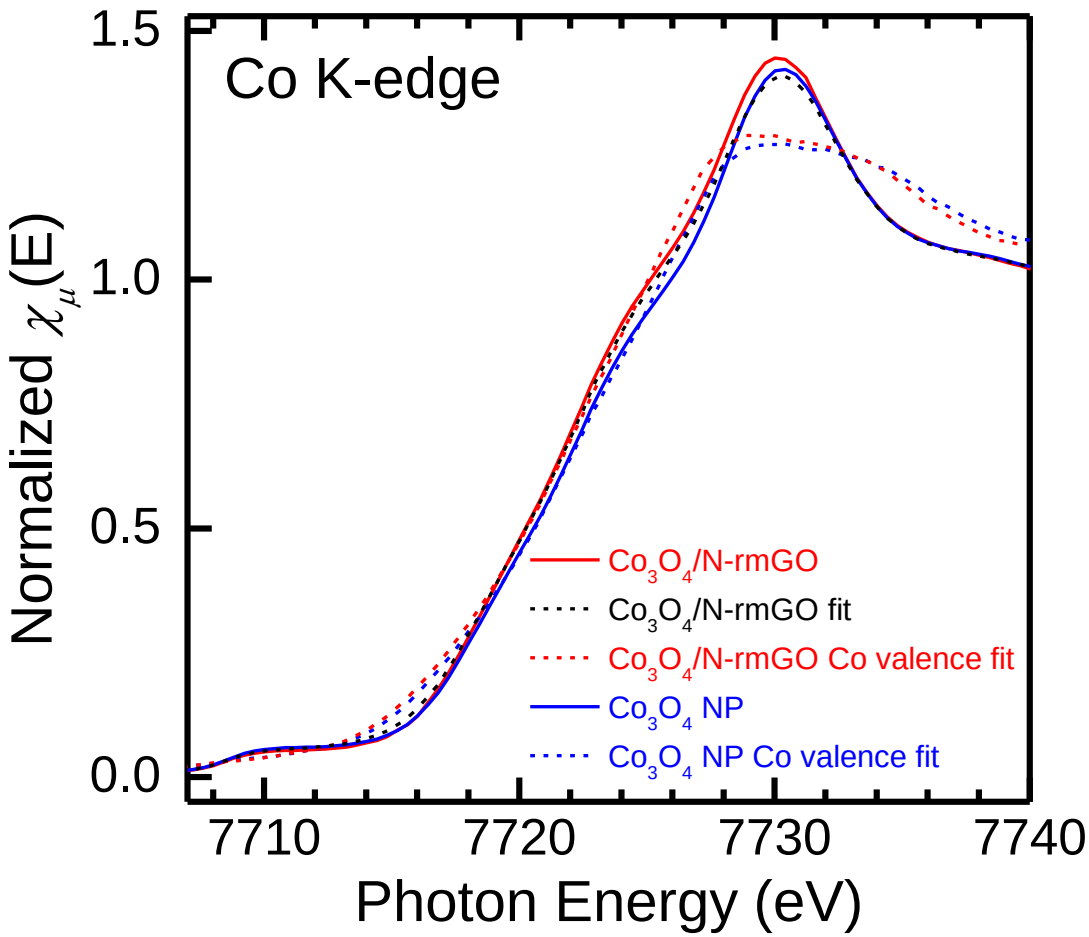

Figure S5 Co K-edge XANES spectra of Co_3O_4/N-rmGO and Co_3O_4 NP together with the linear combination fit of the spectra. Red line: Co_3O_4/N-rmGO XANES spectrum, black dashed line: Co_3O_4/N-rmGO spectrum fitted by the normalized reference spectra of CoO, Co_2O_3 and Co_3O_4 NP (for reference spectra see Figure 4 in the main text), red dashed line: Co_3O_4/N-rmGO spectrum Co valence fitted by the normalized reference spectra of CoO and Co_2O_3, blue line: Co_3O_4 NP XANES spectrum, blue dashed line: Co_3O_4 NP spectrum Co valence fitted by the normalized reference spectra of CoO and Co_2O_3. All fitting coefficients are listed in Table S1.

Table S1 Linear combination fitting coefficients and fitting quality (χ^2) of Co K-edge XANES spectra of Co_3O_4/N-rmGO and Co_3O_4 NP fitted by the normalized reference spectra of CoO, Co_2O_3 and Co_3O_4 NP.

	Normalized Reference Spectra			χ^2
	CoO	**Co_2O_3**	**Co_3O_4 NP**	
Co_3O_4/N-rmGO fit	0.131(0.000)	0.000(0.000)	0.869(0.000)	0.02931
Co_3O_4/N-rmGO Co valence fit	0.431(0.049)	0.569(0.049)		0.51597
Co_3O_4 NP Co valence fit	0.311(0.049)	0.689(0.049)		0.49889

N.B.: values in the brackets are the fitting errors.