

## **Supporting Information**

### **Ultrathin and Vacancy-rich CoAl-Layered Double Hydroxide/Graphite Oxide Catalysts: Promotional Effect of Cobalt Vacancies and Oxygen Vacancies in Alcohol Oxidation**

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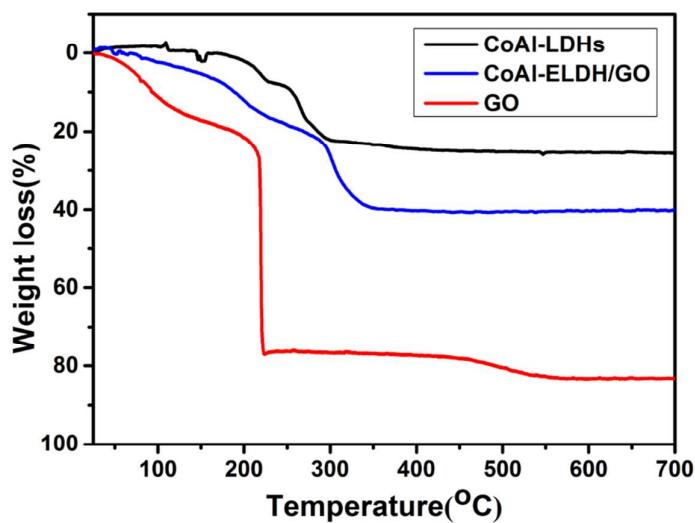
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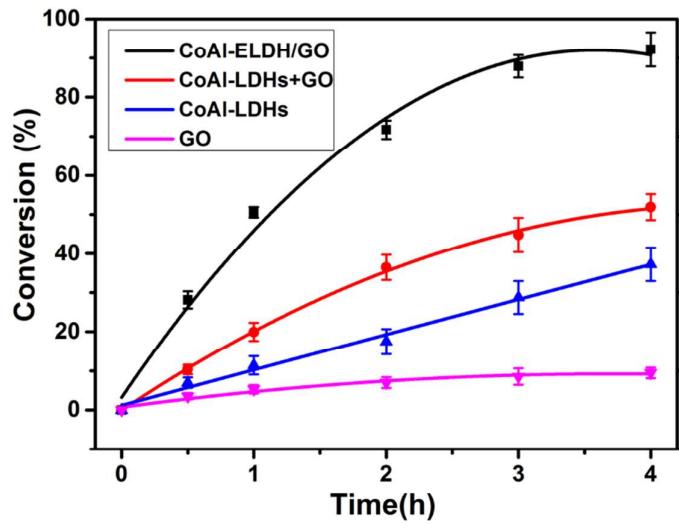
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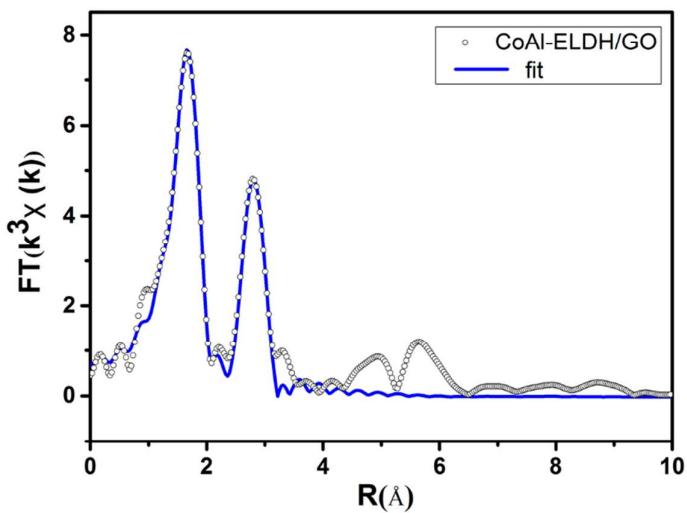
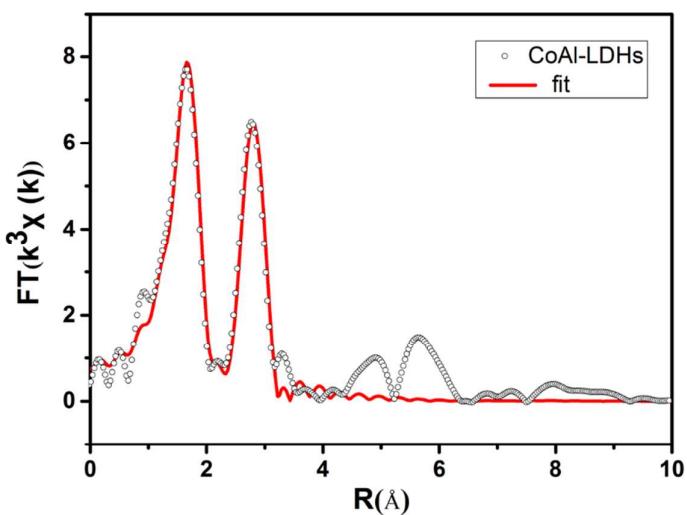
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**Figure S1** Thermogravimetric profiles of the catalysts.



**Figure S2** Evolution of the conversion with reaction time over different catalysts.



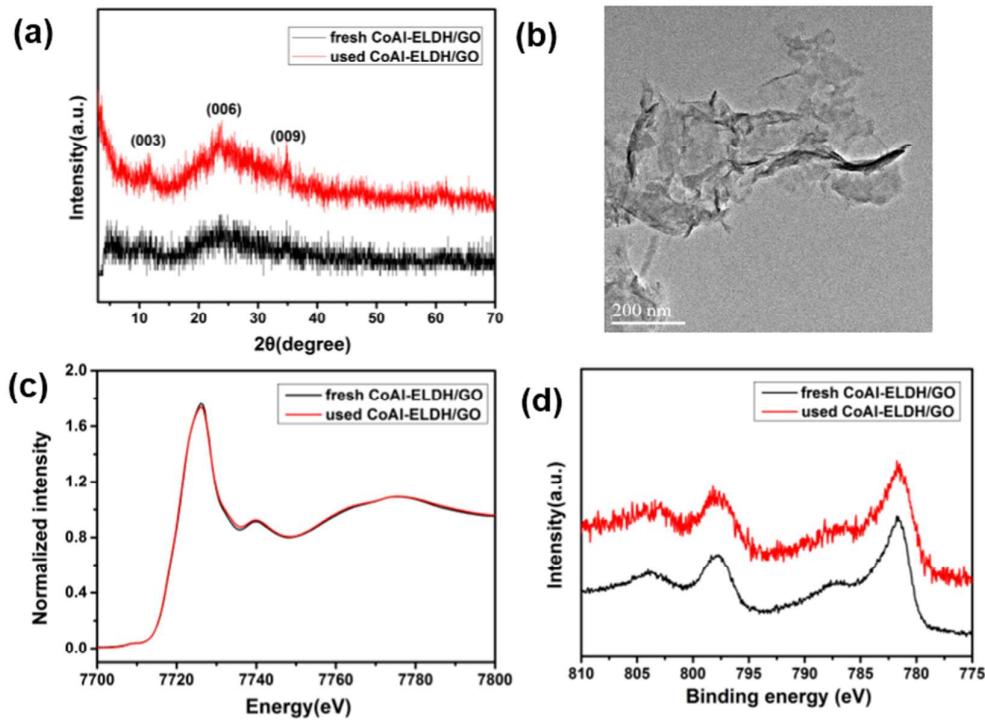
**Figure S3** EXAFS curve-fitting results for the bulk CoAl-LDHs and CoAl-ELDH/GO.

**Table S1** Local structure parameters around Co estimated by EXAFS analysis

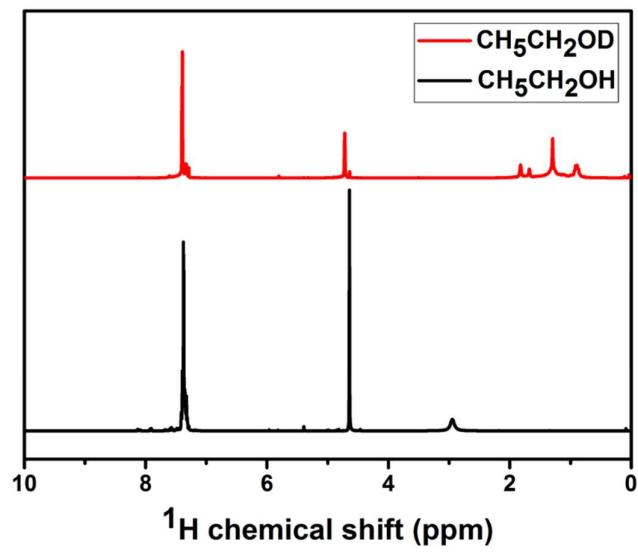
Sample	Shell	N	R(Å)	$\sigma^2(10^{-3}\text{\AA}^2)$	$\Delta E_0(\text{eV})$
<b>bulk CoAl-LDHs</b>	Co-O <sub>OH</sub>	6	2.09	8.0	0.82
	Co···Co	3.8	3.12	8.6	2.24
<b>CoAl-ELDH/GO</b>	Co-O <sub>OH</sub>	5.5	2.09	7.3	0.69
	Co-···Co	2.8	3.12	8.8	2.58

**Table S2** Carbonate mode assignment from CO<sub>2</sub> adsorption and desorption (all frequencies in cm<sup>-1</sup>)

Sample	bicarbonate			bidentate carbonate		unidentate carbonate	
	> 400cm <sup>-1</sup>			250–400 cm <sup>-1</sup>		150–250 cm <sup>-1</sup>	
	v <sub>asym</sub> OCO	v <sub>sym</sub> OCO	δCOH	v <sub>asym</sub> OCO	v <sub>sym</sub> OCO	v <sub>asym</sub> OCO	v <sub>sym</sub> OCO
<b>GO</b>	1696	1445	-	-	-	1591	1392
<b>bulk CoAl-LDHs</b>	1782	1359	1200	1588	1292	-	-
<b>CoAl-ELDH/GO</b>	1771	1405	1215	1651	1291	1537	1365



**Figure S4** (a) XRD patterns of fresh and used CoAl-ELDH/GO catalyst. (b) HRTEM image of used CoAl-ELDH/GO catalyst. (c) The normalized intensity of Co K-edge XANES spectra for fresh and used CoAl-ELDH/GO catalysts. (d) XPS spectra of Co 2p region for fresh and used CoAl-ELDH/GO catalysts.



**Figure S5** <sup>1</sup>H NMR spectra of  $\text{C}_6\text{H}_5\text{CH}_2\text{OH}$  and  $\text{C}_6\text{H}_5\text{CH}_2\text{OD}$ .