

## Supporting Information

Figure S1. TEM and HRTEM imaging of ultrathin single crystal lepidocrocite nanosheets. a, TEM image of Fe-G 5%. b, HRTEM image of Fe-G 5%. c, TEM image of Fe-G 10%. d, HRTEM image of Fe-G 10%. e, TEM image of Fe-G 15%. f, HRTEM image of Fe-G 5%. g, TEM image of Fe-G 20%. h, HRTEM image of Fe-G 20%.



Figure S2. EDS spectrum of ultrathin single crystal lepidocrocite nanosheets.

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(b)

Figure S3. a) AFM image of nanosheets; b) Height analysis in AFM. Height for graphene + Fe nanosheets is 2.8 nm, graphene alone is 0.6 nm, thus the Fe nanosheet is 2.1 nm.



Figure S4. NEXAFS spectrum of reduced graphene oxide in Fe-G 20% nanocomplex.



Figure S5. Data fitting with the first-rate reaction kinetics for phenol degradation with Fe-G nanostructures of different Fe contents (shown in Fig. 5e).

The equation for the first-order reaction kinetics is

 $C/C_0 = a + b \times e^{-kt}$  (1) where "C" and "C<sub>0</sub>" are the concentration and original concentration of phenol, "t" is the reaction time and "k" stands for the first-order reaction rate, "a" and "b" are the fitting constants.

This equation can be transformed to  $t = a + k \times \ln(C/C_0)$  (2)

By using the equation (2), the phenol degradation data can be linearly fitted for the first 30 min ( $R^2$  is Correlation coefficient).

The following table is the parameters for fitting and the <i>k</i> values.			
	а	$k(\min^{-1})$	$R^2$
Fe-G 5%	-0.035	0.016	0.940
Fe-G 10%	0.057	0.025	0.954
Fe-G 15%	0.392	0.041	0.752
Fe-G 20%	0.423	0.075	0.885

The *k* values are also reported in Table 1.



Figure S6. XRD patterns of Fe-G 20% before and after phenol treatment.



Figure S7. FTIR spectra of Fe-G 20% before and after phenol treatment.

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Figure S8. SEM image of the nanosheet sample for XRD characterization.