Electronic Supplementary Information (ESI) for Catalysis Science & Technology

# A stable, efficient 3D Cobalt-graphene composite catalyst for

# the hydrolysis of ammonia borane

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- 1. TGA and FT-IR characterization of PEI-GO<sub>2D amide</sub>;
- 2. Morphology observation of PEI-GO<sub>2D amide</sub> and PEI-GO<sub>2D amide</sub> /Co;
- 3. Chemical Composition of PEI-GO<sub>2D amide</sub>/Co materials;
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- 5. Hydrogen evolution of AB catalyzed by PEI-GO<sub>2D amide</sub>/Co at different temperatures.

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#### 1. TGA and FT-IR characterization of PEI-GO<sub>2D amide</sub>



Figure S1. (a) TGA curve and (b) FT-IR sprectrum of PEI-GO<sub>2D amide</sub>.

Fig. S1a shows the TGA curve of PEI-GO<sub>2D amide</sub>. The mass loss of PEI molecules mainly occurs between 300-400 °C; therefore, PEI-GO<sub>2D amide</sub> contains about 30% PEI, lower than PEI-GO<sub>3D</sub> (45%). Fig. S1b gives the FT-IR spectrum of PEI-GO<sub>2D amide</sub>. The peak at 1658 cm<sup>-1</sup> indicates the existence of large amount of amide group. However, the peaks at 1562 (NH<sub>2</sub>) and 1450 cm<sup>-1</sup> (C-N) are weak, meaning that the content of amine groups is quite low.

## 2. Morphology observation of PEI-GO<sub>2D amide</sub> and PEI-GO<sub>2D amide</sub>/Co



Figure S2. (a) TEM and (b) FESEM images of PEI-GO<sub>2D amide</sub>. (c, d) HRTEM of PEI-GO<sub>2D amide</sub>/Co.

Fig. S2a is the TEM image of PEI-GO<sub>2D amide</sub>. No obvious corrugations are obtained, indicating a low content of PEI molecules linked on GO. The FESEM image in Fig. S2b shows that PEI-GO<sub>2D</sub>  $_{amide}$  has a typical 2D structure. HRTEM images of Fig. S2c and d shows the uniform and dense

deposition of Co NPs on PEI-GO<sub>2D amide</sub>, with the average size of ~4 nm.

3. Chemical Composition of PEI-GO<sub>2D amide</sub>/Co materials



Figure S3. XPS survey of (a) C<sub>1s</sub>, (b)N<sub>1s</sub> and (c) Co<sub>2p</sub> core-level spectra of PEI-GO<sub>2D amide</sub>/Co.

Fig. S3a presents the  $C_{1s}$  core-level spectrum. The peaks at 284.8 and 288.1 eV are ascribed to C-C and C(O)NH species, respectively. And the  $N_{1s}$  spectrum in Fig. S3b indicates that only amide groups exist in PEI-GO<sub>2D amide</sub>, amine groups are not significant. This is consistent with FT-IR results.

#### 4. TEM images of PEI-GO<sub>3D</sub>/Co and PEI-GO<sub>2D amide</sub>/Co after stability test



**Figure S4.** TEM image of (a) PEI-GO<sub>3D</sub>/Co and (b) PEI-GO<sub>2D amide</sub>/Co after five cycles of the catalytic hydrolysis of AB.

Fig. S4a shows the particle size of PEI-GO<sub>3D</sub>/Co after five cycles catalytic hydrolysis of AB. The particle size doesn't have obvious increment. Fig. S4b shows the particle size of PEI-GO<sub>2D amide</sub>/Co after five cycles. The average particle size increases from ~4 nm to ~5 nm. Therefore, PEI-GO<sub>3D</sub> has much better dispersing ability for Co NPs than PEI-GO<sub>2D amide</sub>.

# 5. Hydrogen evolution of AB catalyzed by PEI-GO<sub>2D amide</sub>/Co at different temperatures



**Figure S5.** (a) Hydrolysis of AB by PEI-GO<sub>2D amide</sub>/Co at different temperatures and (b) the corresponding Arrhenius plot of ln rate vs. (1/T)

Fig. S5a shows the AB hydrolysis behavior catalyzed by PEI-GO<sub>2D amide</sub>/Co at different temperatures. The linear part of these hydrogen evolution curves are used to calculate hydrogen generation rate (r). Then, ln(r) verses 1/T is re-plotted in Fig. S5b. The calculated activation energy of PEI-GO<sub>2D amide</sub>/Co is  $27.55 \pm 0.94$  kJ mol<sup>-1</sup>, which is slightly higher than that of PEI-GO<sub>3D</sub>/Co.