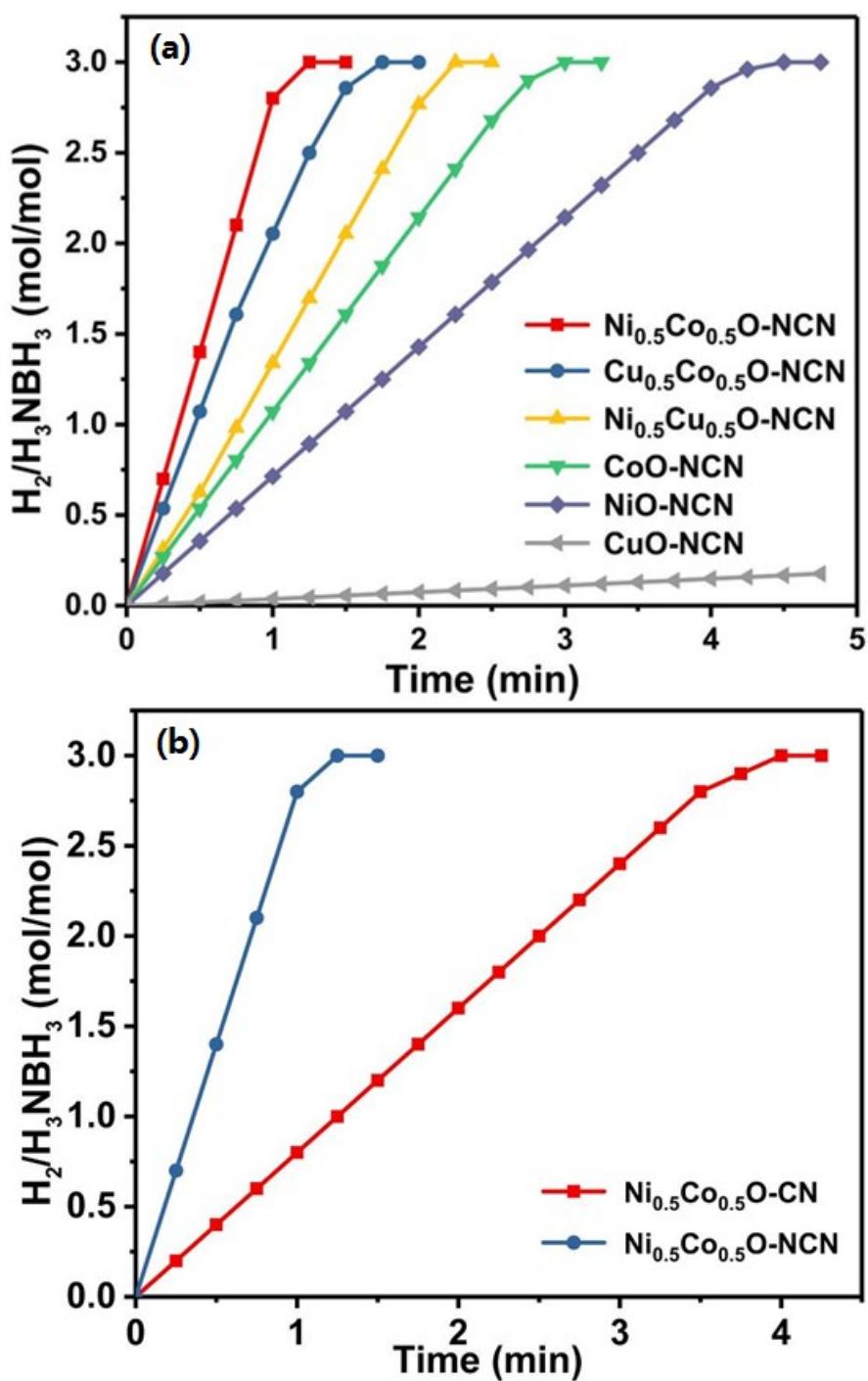


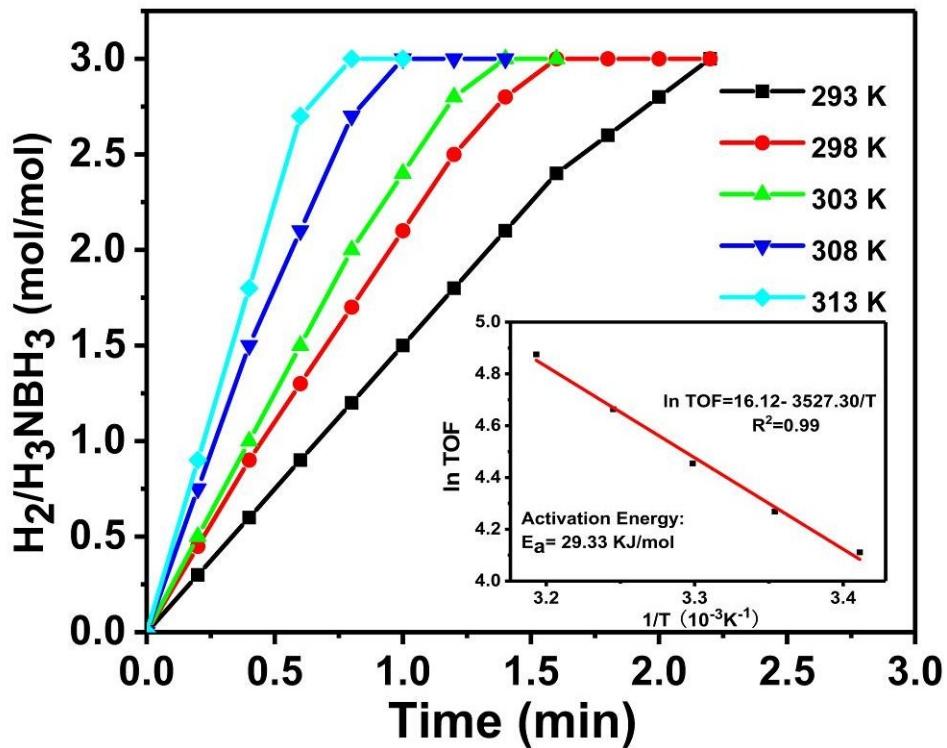
## Supporting information

# Carbon Nitride Supported Ni<sub>0.5</sub>Co<sub>0.5</sub>O Nanoparticles with Strong Interfacial Interaction to Enhance The Hydrolysis of Ammonia Borane

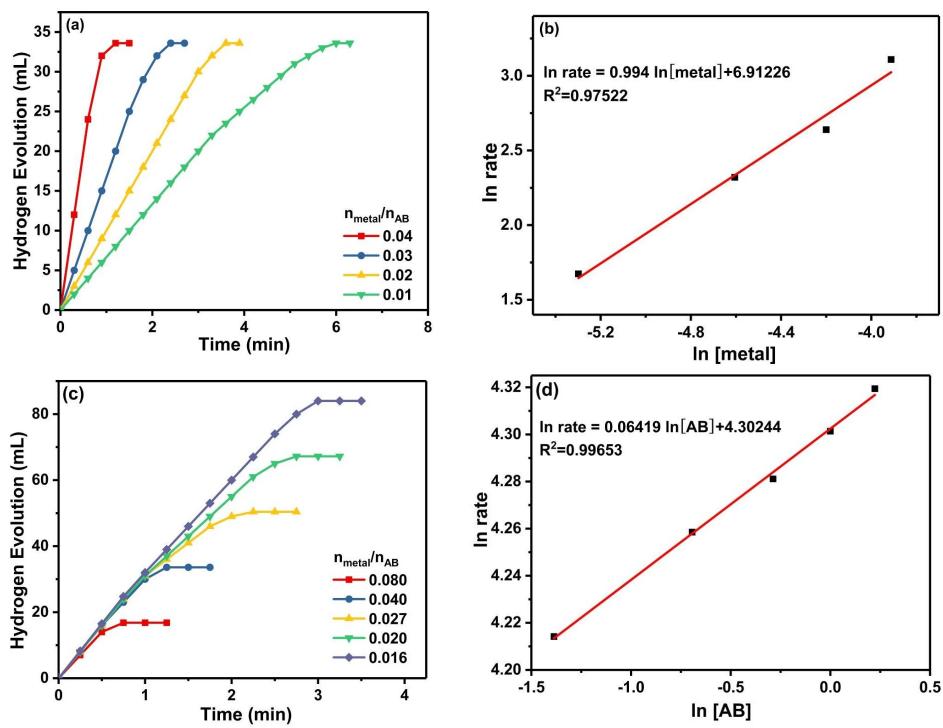
*Yunpeng Shang<sup>a</sup>, Kun Feng<sup>a</sup>, Yu Wang<sup>b\*</sup>, Xuhui Sun<sup>a</sup>, and Jun Zhong<sup>a\*</sup>*



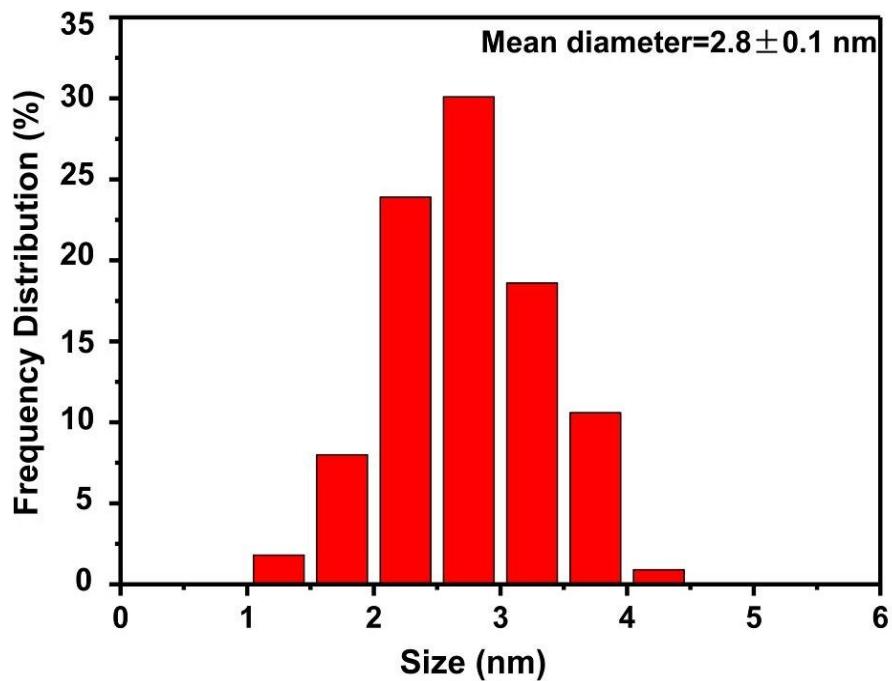
**Figure S1:** (a) Hydrogen evolution curves of the hydrolysis of AB aqueous solution catalyzed by  $\text{Ni}_{0.5}\text{Co}_{0.5}\text{O-NCN}$ ,  $\text{Cu}_{0.5}\text{Co}_{0.5}\text{O-NCN}$ ,  $\text{Ni}_{0.5}\text{Cu}_{0.5}\text{O-NCN}$ ,  $\text{CoO-NCN}$ ,  $\text{NiO-NCN}$  and  $\text{CuO-NCN}$ . (b) Comparison of the hydrogen evolution curves catalyzed by  $\text{Ni}_{0.5}\text{Co}_{0.5}\text{O-CN}$  and  $\text{Ni}_{0.5}\text{Co}_{0.5}\text{O-NCN}$ .



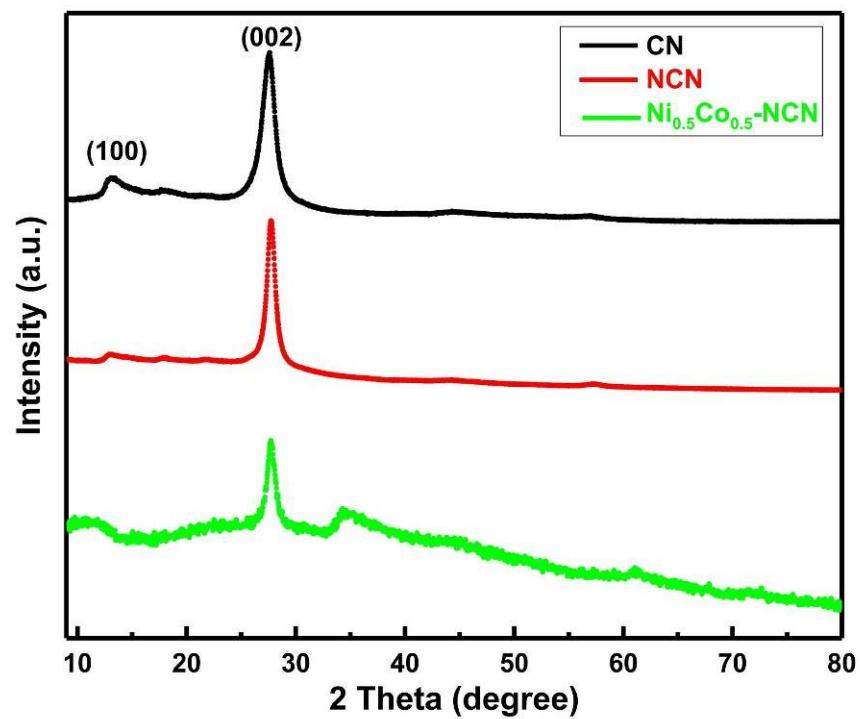
**Figure S2:** Hydrogen-generating rate as a function of temperature in the hydrolysis of AB catalyzed by  $\text{Ni}_{0.5}\text{Co}_{0.5}\text{O-NCN}$ . Since at a high temperature the reaction will be finished quickly, we have used less  $\text{Ni}_{0.5}\text{Co}_{0.5}\text{O-NCN}$  (2.5 mg) in this reaction. Inset: Arrhenius plot of  $\ln(\text{TOF})$  versus  $1/T$ . The activation energy is 43.18 kJ/mol.



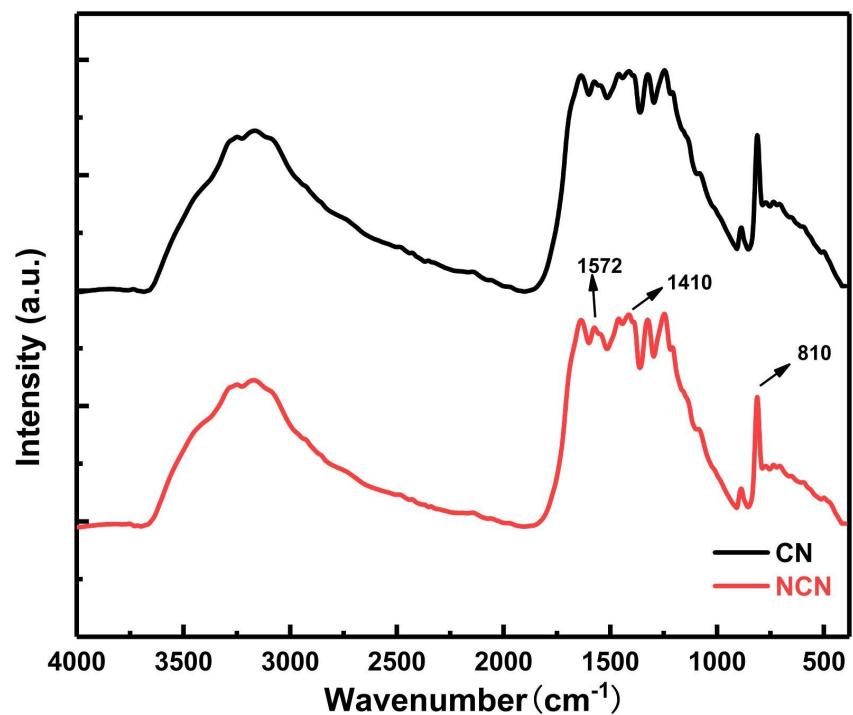
**Figure S3:** (a) Stoichiometric hydrogen evolution in aqueous solution at a fixed amount of AB with various  $\text{Ni}_{0.5}\text{Co}_{0.5}\text{O-NCN}/\text{AB}$  molar ratios at 298 K; (c) Relationship between hydrogen-generating rate and AB concentration at a fixed amount of  $\text{Ni}_{0.5}\text{Co}_{0.5}\text{O-NCN}$  in aqueous solution at 298 K; (b) and (d): Logarithmic plots of rate versus  $[\text{Ni}_{0.5}\text{Co}_{0.5}\text{O-NCN}]$  and  $[\text{AB}]$ , respectively.



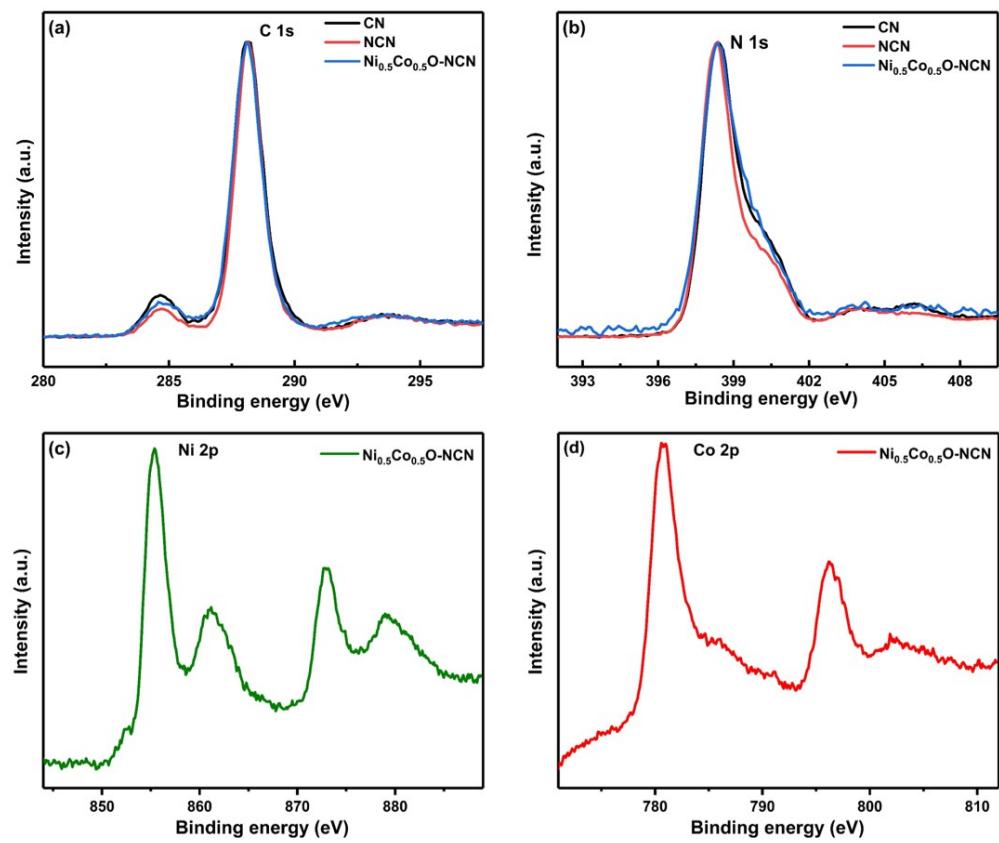
**Figure S4:** The particle size distribution of  $\text{Ni}_{0.5}\text{Co}_{0.5}\text{O-NCN}$  with an average size of 2.8 nm.



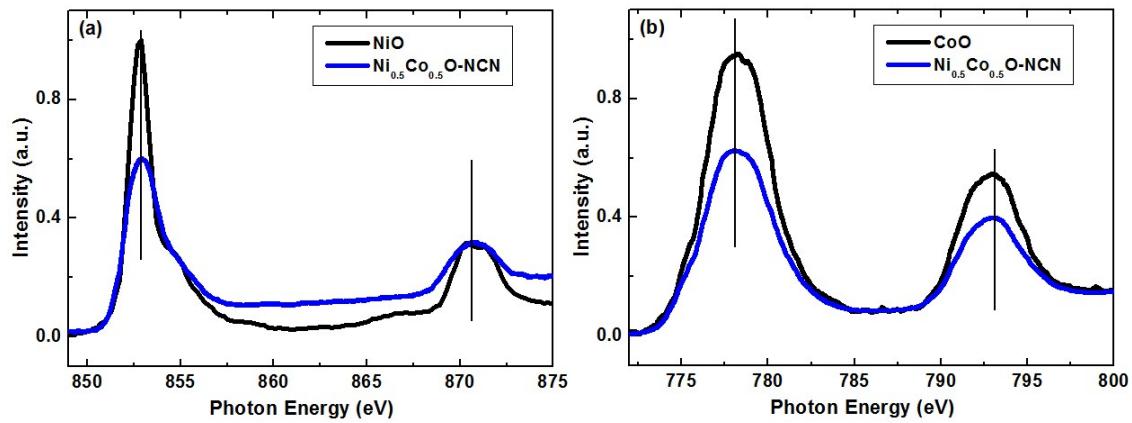
**Figure S5:** XRD spectra of CN, NCN and  $\text{Ni}_{0.5}\text{Co}_{0.5}\text{-NCN}$ .



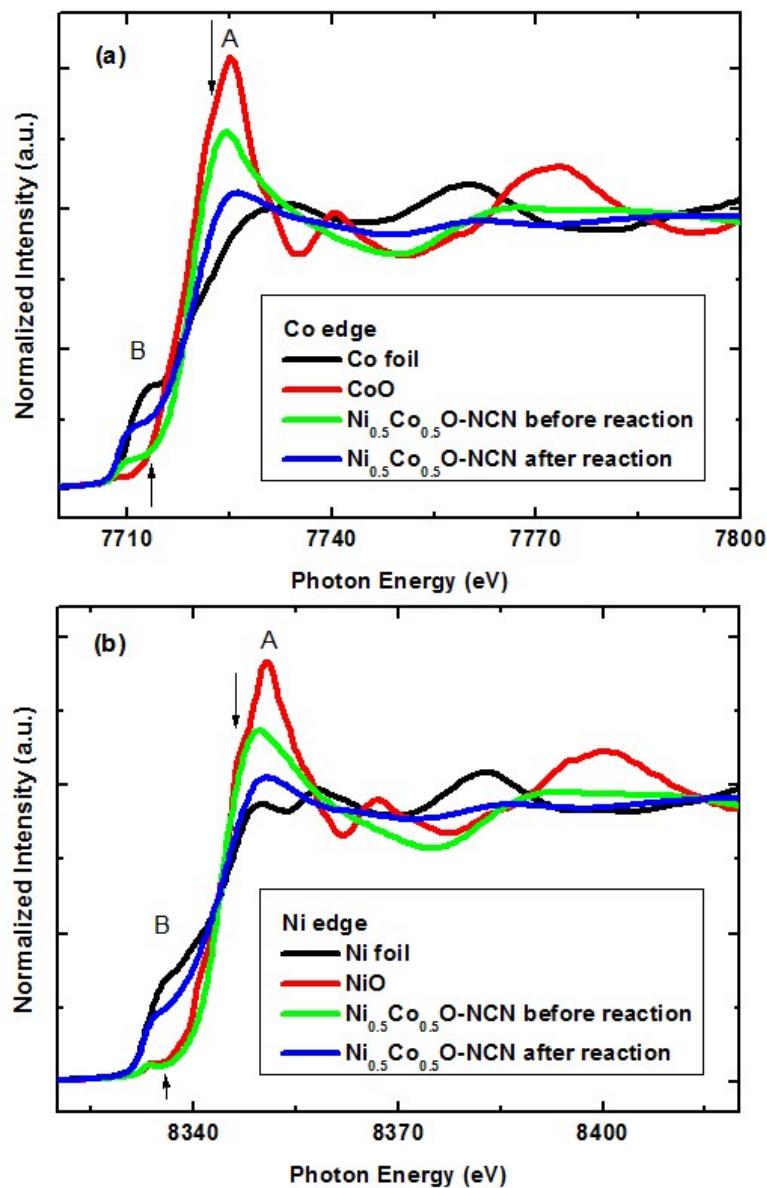
**Figure S6:** FTIR spectra of CN and NCN.



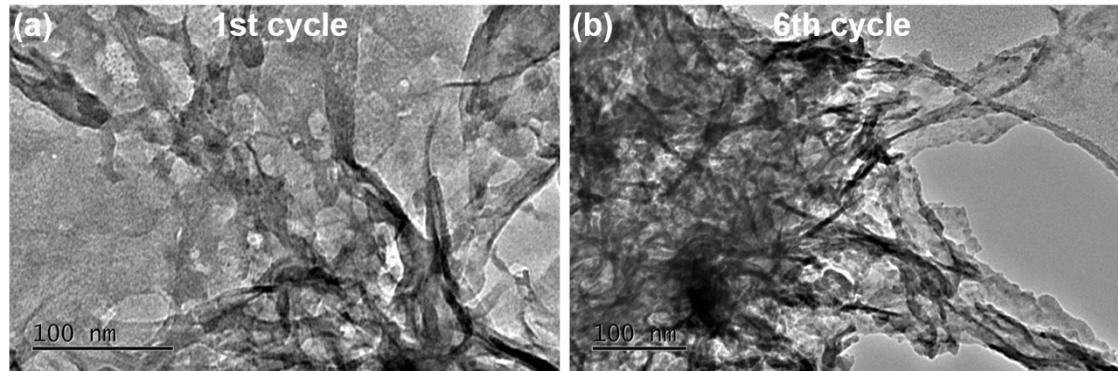
**Figure S7:** XPS spectra of CN, NCN and  $\text{Ni}_{0.5}\text{Co}_{0.5}\text{O}$ -NCN at C 1s (a), N 1s (b), Ni 2p (c) and Co 2p (d) edges, respectively.



**Figure S8:** XAS spectra of  $\text{Ni}_{0.5}\text{Co}_{0.5}\text{O-NCN}$  and the reference samples at Ni *L*-edge (a) and Co *L*-edge (b), respectively.



**Figure S9:** XAS spectra of  $\text{Ni}_{0.5}\text{Co}_{0.5}\text{O-NCN}$  before and after the hydrolysis reaction at Co *K*-edge (a) and Ni *K*-edge (b), respectively.



**Figure S10:** TEM images of the  $\text{Ni}_{0.5}\text{Co}_{0.5}\text{O}$ -NCN samples after the first cycle (a) and the 6<sup>th</sup> cycle (b).

Samples	Ni-loading/wt%	Co-loading/wt%	TOF (H <sub>2</sub> ) mol/(Cat-M)mol·min
<b>Ni<sub>0.8</sub>Co<sub>0.2</sub>O-NCN</b>	<b>17.1</b>	<b>4.3</b>	<b>42.9</b>
<b>Ni<sub>0.6</sub>Co<sub>0.4</sub>O-NCN</b>	<b>12.2</b>	<b>8.4</b>	<b>50.3</b>
<b>Ni<sub>0.5</sub>Co<sub>0.5</sub>O-NCN</b>	<b>11.6</b>	<b>10.5</b>	<b>76.1</b>
<b>Ni<sub>0.4</sub>Co<sub>0.6</sub>O-NCN</b>	<b>8.7</b>	<b>12.8</b>	<b>46.6</b>
<b>Ni<sub>0.2</sub>Co<sub>0.8</sub>O-NCN</b>	<b>3.9</b>	<b>16.2</b>	<b>35.2</b>
<b>NiO-NCN</b>	<b>19.1</b>	-	<b>5.5</b>
<b>CoO-NCN</b>	-	<b>15.8</b>	<b>13.3</b>
<b>NCN</b>	-	-	<b>0</b>

**Table S1.** Ni and Co contents and the TOF values of various Ni<sub>x</sub>Co<sub>1-x</sub>O-NCN samples.

Catalyst	TOF (H <sub>2</sub> ) mol/(Cat-M)mol·min	Solution	T (°C)	Ref.
<b>Ni<sub>0.5</sub>Co<sub>0.5</sub>O-NCN</b>	<b>76.1</b>	<b>Water</b>	<b>25</b>	<b>This work</b>
<b>Cu<sub>0.72</sub>Co<sub>0.18</sub>Mo<sub>0.1</sub></b>	<b>119.0</b>	<b>NaOH</b>	<b>25</b>	<b>1</b>
<b>Co/MIL-101(Cr)-NH<sub>2</sub></b>	<b>117.7(light)</b>	<b>Water</b>	<b>25</b>	<b>2</b>
<b>Ni<sub>0.3</sub>Co<sub>1.3</sub>P/GO</b>	<b>109.4</b>	<b>NaOH</b>	<b>25</b>	<b>3</b>
<b>Co-C<sub>3</sub>N<sub>4</sub>-580</b>	<b>93.8(light)</b>	<b>Water</b>	<b>25</b>	<b>4</b>
<b>Ni/ZIF-8</b>	<b>85.7</b>	<b>NaOH</b>	<b>25</b>	<b>5</b>
<b>Cu<sub>0.5</sub>Co<sub>0.5</sub>O-rGO</b>	<b>81.7</b>	<b>Water</b>	<b>25</b>	<b>6</b>
<b>CuCo/g-C<sub>3</sub>N<sub>4</sub></b>	<b>75.1(light)</b>	<b>Water</b>	<b>25</b>	<b>7</b>
<b>CoP</b>	<b>72.2</b>	<b>NaOH</b>	<b>25</b>	<b>8</b>
<b>Cu<sub>0.8</sub>Co<sub>0.2</sub>O-GO</b>	<b>70.0</b>	<b>Water</b>	<b>25</b>	<b>9</b>
<b>Ni<sub>0.9</sub>Mo<sub>0.1</sub>/graphene</b>	<b>66.7</b>	<b>Water</b>	<b>25</b>	<b>10</b>
<b>CuO-NiO</b>	<b>60.0</b>	<b>Water</b>	<b>25</b>	<b>11</b>
<b>Cu<sub>0.5</sub>Ni<sub>0.5</sub>/CMK-1</b>	<b>54.8</b>	<b>Water</b>	<b>25</b>	<b>12</b>
<b>CuCo/MIL-101-1-U</b>	<b>51.7</b>	<b>Water</b>	<b>25</b>	<b>13</b>
<b>Co NPs (in-situ)</b>	<b>49.8</b>	<b>Water</b>	<b>25</b>	<b>14</b>
<b>Ni NPs@3D-(N)GFs</b>	<b>41.7</b>	<b>Water</b>	<b>25</b>	<b>15</b>
<b>Ni<sub>2</sub>P</b>	<b>40.4</b>	<b>Water</b>	<b>25</b>	<b>16</b>
<b>Cu NPs@SCF</b>	<b>40.0</b>	<b>Water</b>	<b>25</b>	<b>17</b>
<b>PEI-GO/Co</b>	<b>39.9</b>	<b>Water</b>	<b>25</b>	<b>18</b>
<b>Ni@MCS-30</b>	<b>30.7</b>	<b>Water</b>	<b>25</b>	<b>19</b>
<b>Cu<sub>0.49</sub>Co<sub>0.51</sub>/C</b>	<b>28.7</b>	<b>Water</b>	<b>25</b>	<b>20</b>
<b>Ni NPs/CNT</b>	<b>23.5</b>	<b>Water</b>	<b>25</b>	<b>21</b>
<b>Cu<sub>0.1</sub>@Co<sub>0.45</sub>Ni<sub>0.45</sub>/graphene</b>	<b>15.46</b>	<b>Water</b>	<b>25</b>	<b>22</b>
<b>Ni NPs/C</b>	<b>8.8</b>	<b>Water</b>	<b>25</b>	<b>23</b>

<b>Cu/rGO</b>	<b>3.6</b>	<b>Water</b>	<b>25</b>	<b>24</b>
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**Table S2:** Activities of various non-noble metal based catalysts for the hydrolysis of AB.

References listed in Table S2:

1. Q. L. Yao, K. Yang, X. L. Hong, X. S. Chen and Z. H. Lu, *Catal. Sci. Technol.*, 2018, **8**, 870-877.
2. J. Song, X. Gu, J. Cheng, N. Fan, H. Zhang and H. Su, *Appl. Catal. B: Environ.*, 2018, **225**, 424-432.
3. C. C. Hou, Q. Li, C. J. Wang, C. Y. Peng, Q. Q. Chen, H. F. Ye, W. F. Fu, C. M. Che, N. López and Y. Chen, *Energy Environ. Sci.*, 2017, **10**, 1770-1776.
4. H. Zhang, X. J. Gu, J. Song, N. Fan and H. Q. Su, *ACS Appl. Mater. Inter.*, 2017, **9**, 32767-32774.
5. C. Wang, J. Tuninetti, Z. Wang, C. Zhang, R. Ciganda, L. Salmon, S. Moya, J. Ruiz and D. Astruc, *J. Am. Chem. Soc.*, 2017, **139**, 11610-11615.
6. H. Zheng, K. Feng, Y. Shang, Z. Kang, X. Sun and J. Zhong, *Inorg. Chem. Front.*, 2018, **5**, 1180-1187.
7. H. Zhang, X. Gu, P. Liu, J. Song, J. Cheng and H. Su, *J. Mater. Chem. A*, 2017, **5**, 2288-2296.
8. Z. C. Fu, Y. Xu, S. L. Chan, W. W. Wang, F. Li, F. Liang, Y. Chen, Z. S. Lin, W. F. Fu and C. M. Che, *Chem. Commun.*, 2017, **53**, 705-708.
9. K. Feng, J. Zhong, B. H. Zhao, H. Zhang, L. Xu, X. H. Sun and S. T. Lee, *Angew. Chem. Int. Ed.*, 2016, **55**, 11950-11954.
10. Q. L. Yao, Z. H. Lu, W. Huang, X. S. Chen and J. Zhu, *J. Mater. Chem. A*, 2016, **4**, 8579-8583.
11. H. Yen and F. Kleitz, *J. Mater. Chem. A*, 2013, **1**, 14790-14796.
12. H. Yen, Y. Seo, S. Kaliaguine and F. Kleitz, *ACS Catal.*, 2015, **5**, 5505-5511.
13. P. Liu, X. Gu, K. Kang, H. Zhang, J. Cheng and H. Su, *ACS Appl. Mater. Interfaces*, 2017, **9**, 10759–10767.
14. J. M. Yan, X. B. Zhang, H. Shioyama and Q. Xu, *J. Power Sources*, 2010, **195**, 1091-1094.
15. M. Mahyari and A. Shaabani, *J. Mater. Chem. A*, 2014, **2**, 16652-16659.
16. C. Y. Peng, L. Kang, S. Cao, Y. Chen, Z. S. Lin and W. F. Fu, *Angew. Chem. Int. Ed.*, 2015, **127**, 15951-15955.
17. M. Kaya, M. Zahmakiran, S. Özkar and M. Volkan, *ACS Appl. Mater. Interfaces*, 2012, **4**, 3866-3873.
18. J. T. Hu, Z. X. Chen, M. X. Li, X. H. Zhou and H. B. Lu, *ACS Appl. Mater. Interfaces*, 2014, **6**, 13191-13200.
19. P. Z. Li, A. Aijaz and Q. Xu, *Angew. Chem. Int. Ed.*, 2012, **51**, 6753-6756.

20. A. Bulut, M. Yurderi, İ. E. Ertas, M. Celebi, M. Kaya and M. Zahmakiran, *Appl. Catal. B: Environ.*, 2016, **180**, 121-129.
21. G. Q. Zhao, J. Zhong, J. Wang, T. K. Sham, X. H. Sun and S. T. Lee, *Nanoscale*, 2015, **7**, 9715-9722.
22. X. Y. Meng, L. Yang, N. Cao, C. Du, K. Hu, J. Su, W. Luo and G. Z. Cheng, *ChemPlusChem*, 2014, **79**, 325-332.
23. O. Metin, V. Mazumder, S. Özkar and S. H. Sun, *J. Am. Chem. Soc.*, 2010, **132**, 1468-1469.
24. Y. W. Yang, Z. H. Lu, Y. J. Hu, Z. J. Zhang, W. M. Shi, X. S. Chen and T. T. Wang, *RSC Adv.*, 2014, **4**, 13749-13752.

Cycles	TOF (H <sub>2</sub> ) mol/(Cat-M)mol·min	Catalytic Efficiency
1 <sup>st</sup>	76.1	100%
2 <sup>nd</sup>	73.9	97.1%
3 <sup>rd</sup>	71.5	94.0%
4 <sup>th</sup>	69.0	90.7%
5 <sup>th</sup>	66.3	87.1 %
6 <sup>th</sup>	63.3	83.2%

**Table S3.** TOF values and the catalytic efficiencies of Ni<sub>0.5</sub>Co<sub>0.5</sub>O-NCN in different cycles during the stability test.