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Electronic Supplementary Information

## Preparation of metal oxide/polyaniline/N-MWCNT hybrid composite electrodes for electrocatalytic synthesis of ammonia at atmospheric pressure

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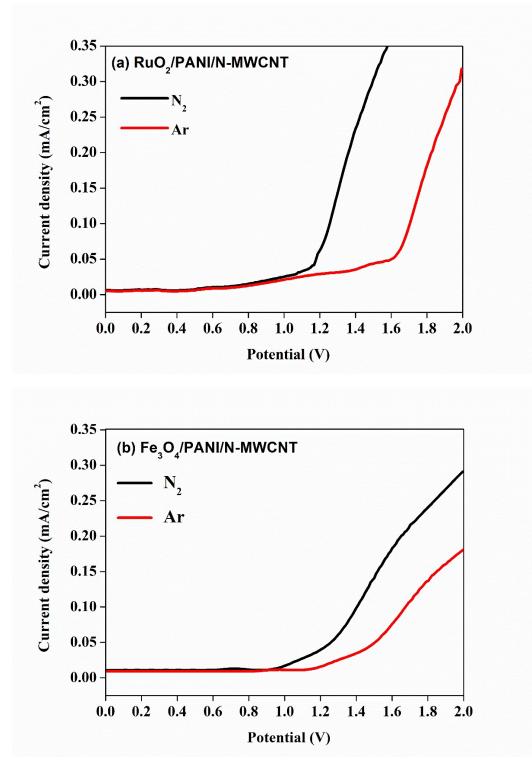


Fig. S1. LSV curves for (a)  $RuO_2/PANI/N-MWCNT$  and (b)  $Fe_3O_4/PANI/N-MWCNT$  electrocatalysts in the presence of argon and nitrogen.

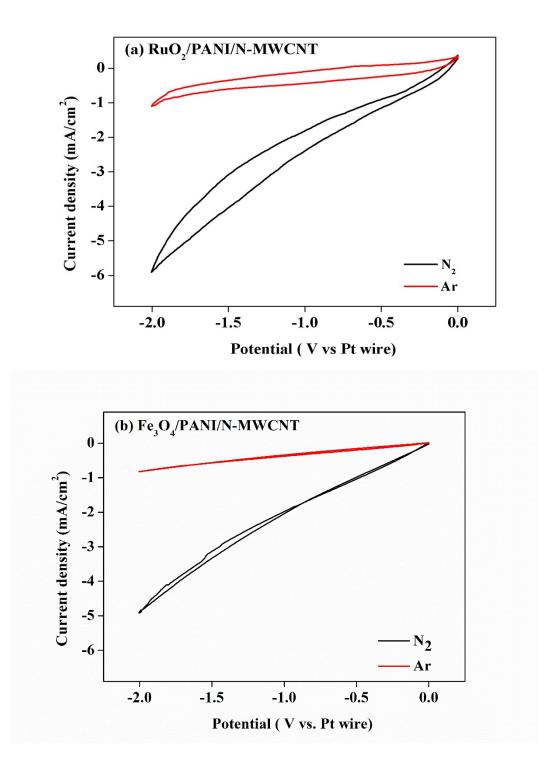
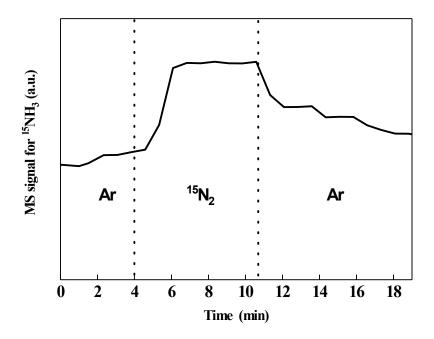


Fig. S2. CV curves for (a)  $RuO_2/PANI/N-MWCNT$  and (b)  $Fe_3O_4/PANI/N-MWCNT$  electrocatalysts in the presence of argon and nitrogen.



**Fig. S3.** Mass spectrometer signal of m/z=18 for  ${}^{15}NH_3$  from the electrolytic cell with RuO<sub>2</sub>/PANI/N-MWCNT during potentiostatic electrolysis with supply of Ar or  ${}^{15}N_2$  at a cell voltage of 1.2 V and 25°C.

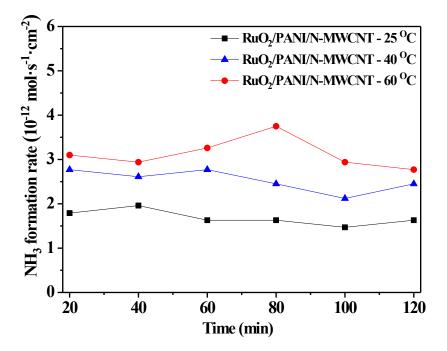


Fig. S4. Results of continuous  $N_2$  reduction over 2 h using an electrolysis cell with RuO<sub>2</sub>/PANI/N-MWCNT in the temperature range of 25–60°C at an applied potential of 1.2 V.

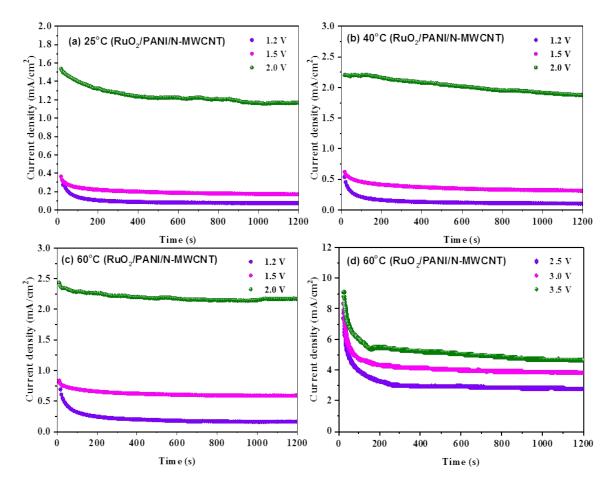


Fig. S5. Trend of current density versus time for the  $RuO_2/PANI/N-MWCNT$  electrocatalyst at different temperatures.

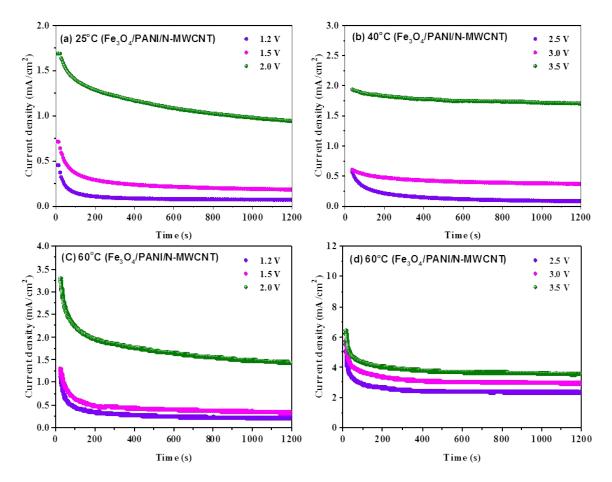


Fig. S6. Trend of current density versus time for the  $Fe_3O_4/PANI/N-MWCNT$  electrocatalyst at different temperatures.

Table S1. Overall faradaic efficiencies  $(H_2+NH_3)$  from the RuO<sub>2</sub>/PANI/N-MWCNT electrocatalyst under an applied potential of 1.2 - 2.0 V at  $25^{\circ}$ C

Applied potential (V)	FE H <sub>2</sub> (%)	FE NH <sub>3</sub> (%)	Overall FE
1.2	98.11	0.49	98.6
1.5	97.27	0.34	97.61
2.0	94.53	0.07	94.60

\*Definition of faradaic efficiency of hydrogen production:

$$FE_{H_2}(\%) = \frac{R_{H2}(mol \cdot cm^{-2} \cdot s^{-1}) \times t(s) \times S(cm^{-2}) \times F}{2 \times I(A) \times t(s)} \times 100\%$$

where  $R_{H2}$  is the hydrogen produced per electrode area (S) and time (t), I (A) is the average current during the reaction, and F is the Faraday constant.