Supplemental Information†

A bimetallic thermally-regenerative ammonia-based battery for high power density and efficiently harvesting low-grade thermal energy

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Calculations using the Nernst Equations:

The Nernst equations for calculating the electrode potentials of Cu/Zn-TRAB during the discharge:

Anode: $E = E^0 - \frac{RT}{2F} \ln \frac{[\alpha(NH_3)]^4}{\alpha\left(Zn(NH_3)_4^{2+}\right)}$ (S1)

Cathode: $E = E^0 - \frac{RT}{2F} \ln \frac{1}{\alpha\left(Cu^{2+}\right)}$ (S2)

where E^0 is the standard potential of electrode reaction, R the gas constant (J K^{-1} mol^{-1}), T the solution temperature (K), F the Faraday constant (96485 C mol^{-1}), $\alpha(NH_3)$ the activity of ammonia, $\alpha\left(Cu^{2+}\right)$ the activity of copper ion and $\alpha\left(Zn(NH_3)_4^{2+}\right)$ the activity of zinc ammonia complex.

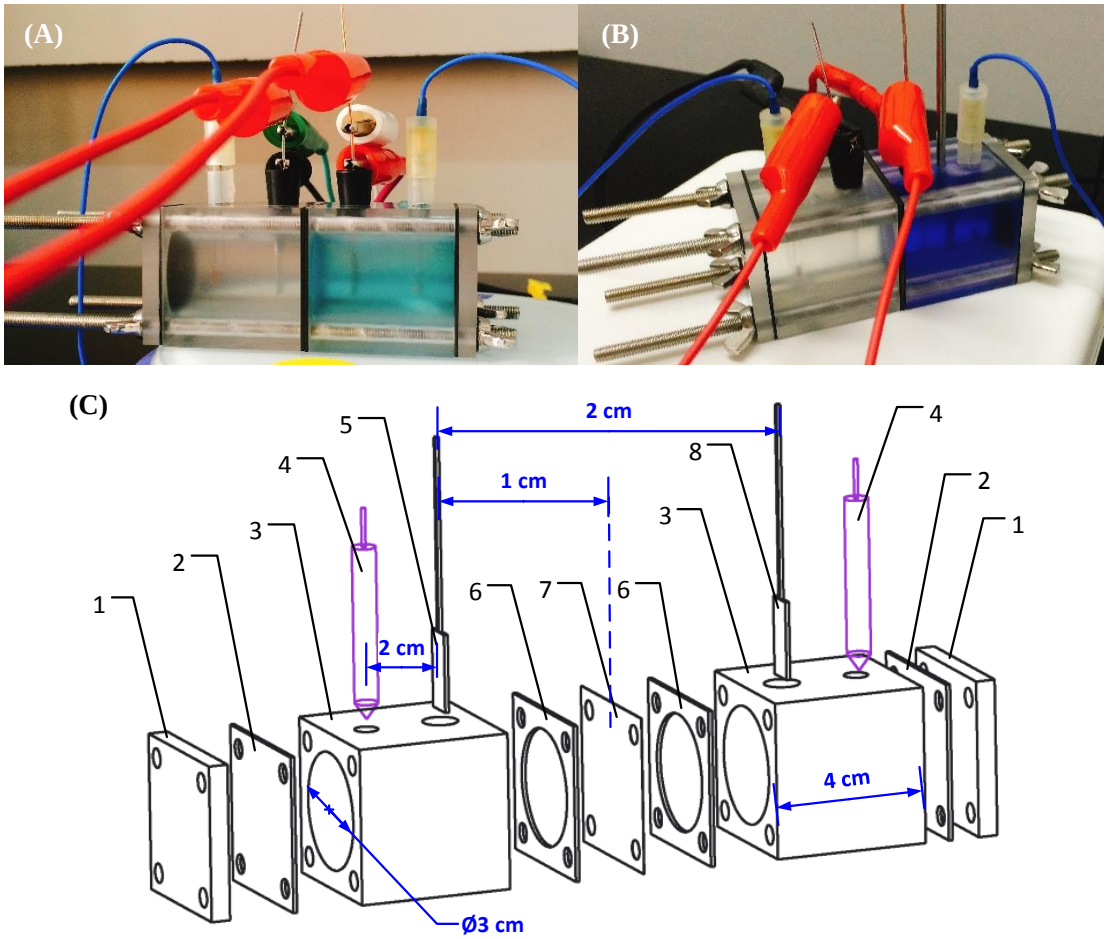


Figure S1. Test set-up for a Cu/Zn-TRAB. Photographs of a laboratory set-up during (A) discharge and (B) charge. (C) Exploded view drawing of the test Cu/Zn-TRAB (1-polycarbonate endplate, 2-fluoroelastomer gasket, 3-polycarbonate chamber, 4-Ag/AgCl reference electrode, 5-Zinc electrode, 6-another fluoroelastomer gasket, 7-anion exchange membrane, 8-copper electrode).

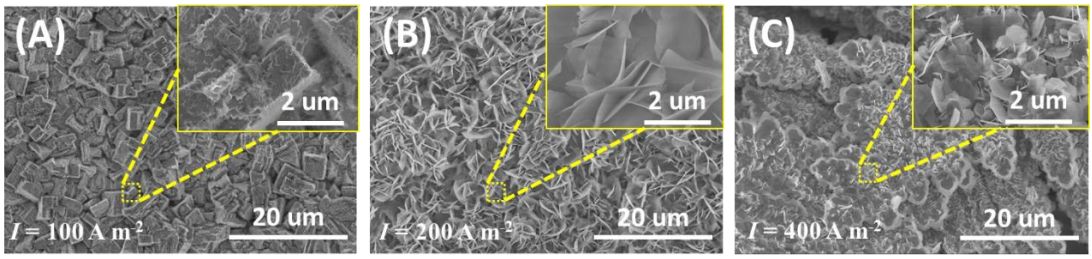


Figure S2. The SEM images of the zinc electrode after 30 minutes of the constant-current charge tests: (A) 100 A m^{-2} (B) 200 A m^{-2} (C) 400 A m^{-2}.

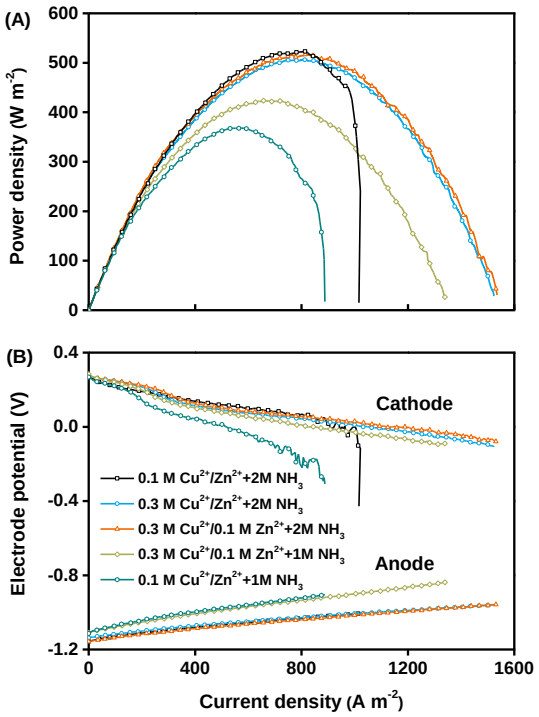


Figure S3. (A) Power density and (B) electrode potentials using different concentrations of Cu(II) and Zn(II) in the discharge process, with 1 M $(NH_4)_2SO_4$ as the supporting electrolyte and 1 M or 2 M NH_3 in the anolyte.

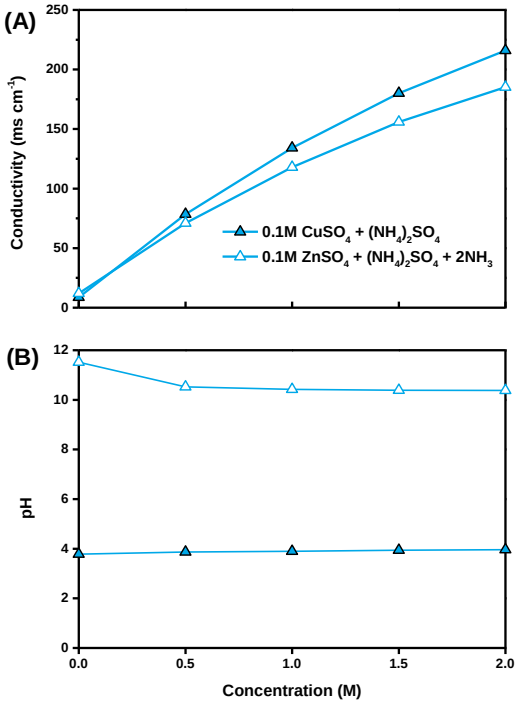


Figure S4. (A) Conductivity and (B) pH of 0.1 M $Cu(SO_4)_2$ or 0.1 M $Zn(SO_4)_2$ solutions with various concentrations of $(NH_4)_2SO_4$, with addition of 2 M NH_3 in the Zn(II) solutions.

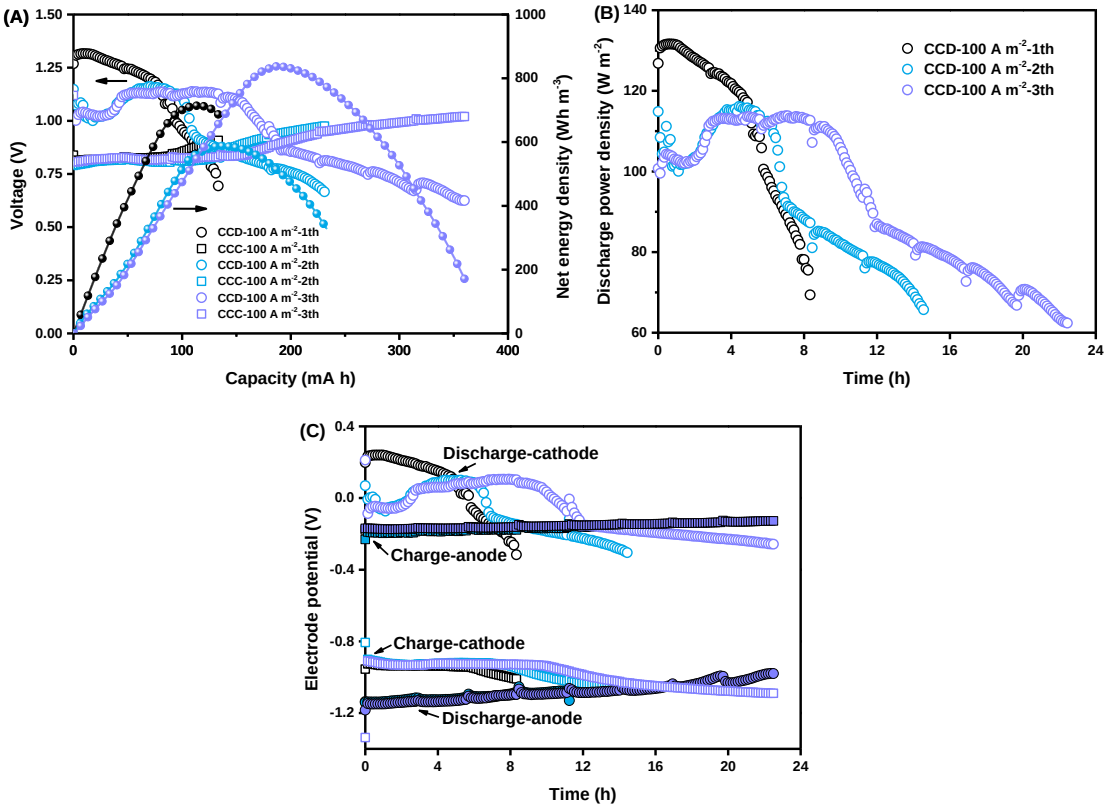


Figure S5. Whole batch cycle performance (A) voltage and net energy density (B) discharge power density (C) electrode potentials of three successive regeneration cycles at a current density of 100 A m^{-2}. The initial electrolyte contained 0.1 M Cu(II) and 1 M $(NH_4)_2SO_4$ in the catholyte and 0.1 M Zn(II), 1 M $(NH_4)_2SO_4$ and 2 M NH_3 in the anolyte. After discharging and charging, the anolyte effluent was regenerated and then operated in next process. "CCD" and "CCC" represent "constant current discharge" and "constant current charge" respectively.

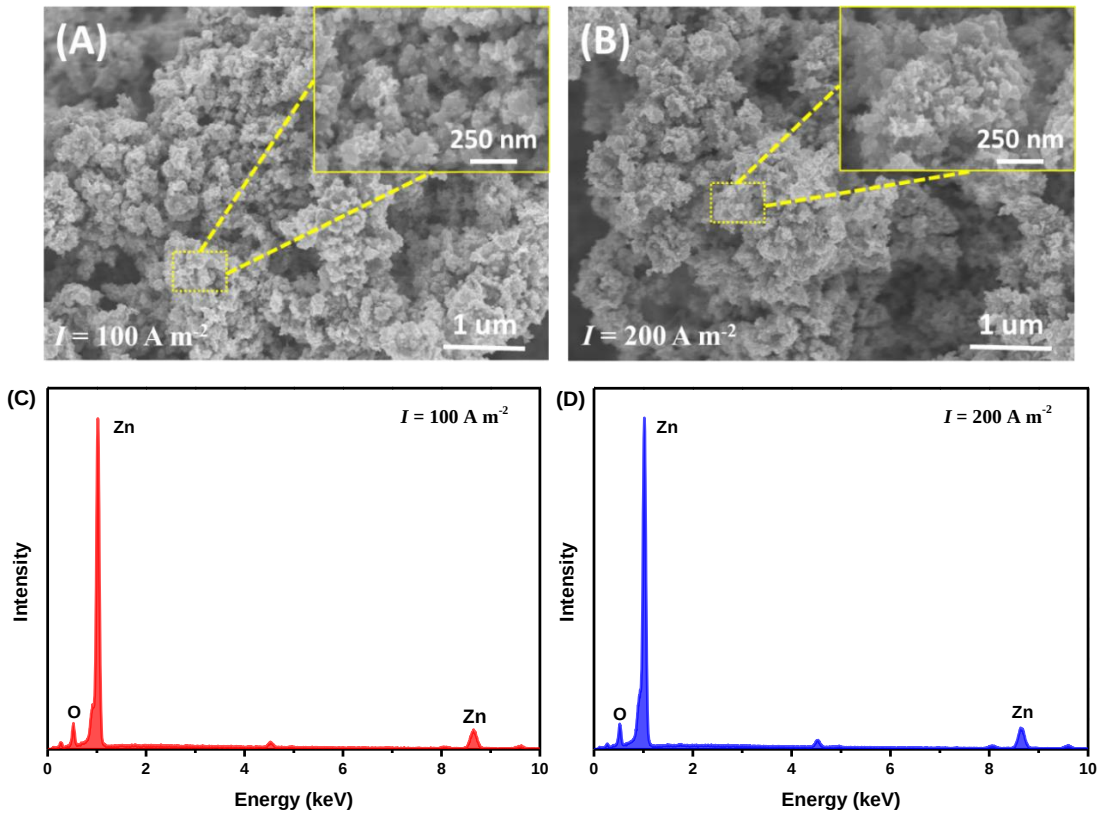


Figure S6. The SEM images of the zinc electrode and corresponding EDS spectrums after the constant-current discharge tests: (A) and (C) 100 A m^{-2}; (B) and (D) 200 A m^{-2}.

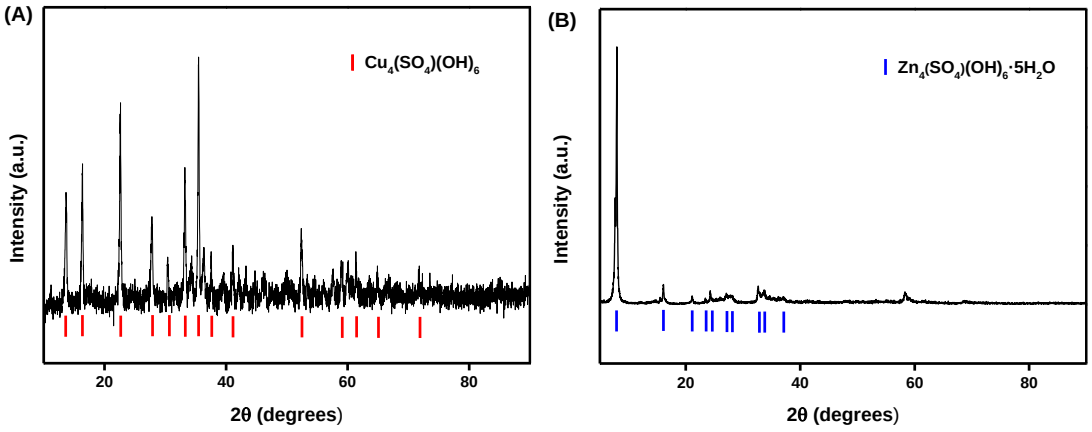


Figure S7. XRD patterns of the precipitates that produced during the thermal regeneration of anolyte effluent after (A) discharge and (B) charge processes.

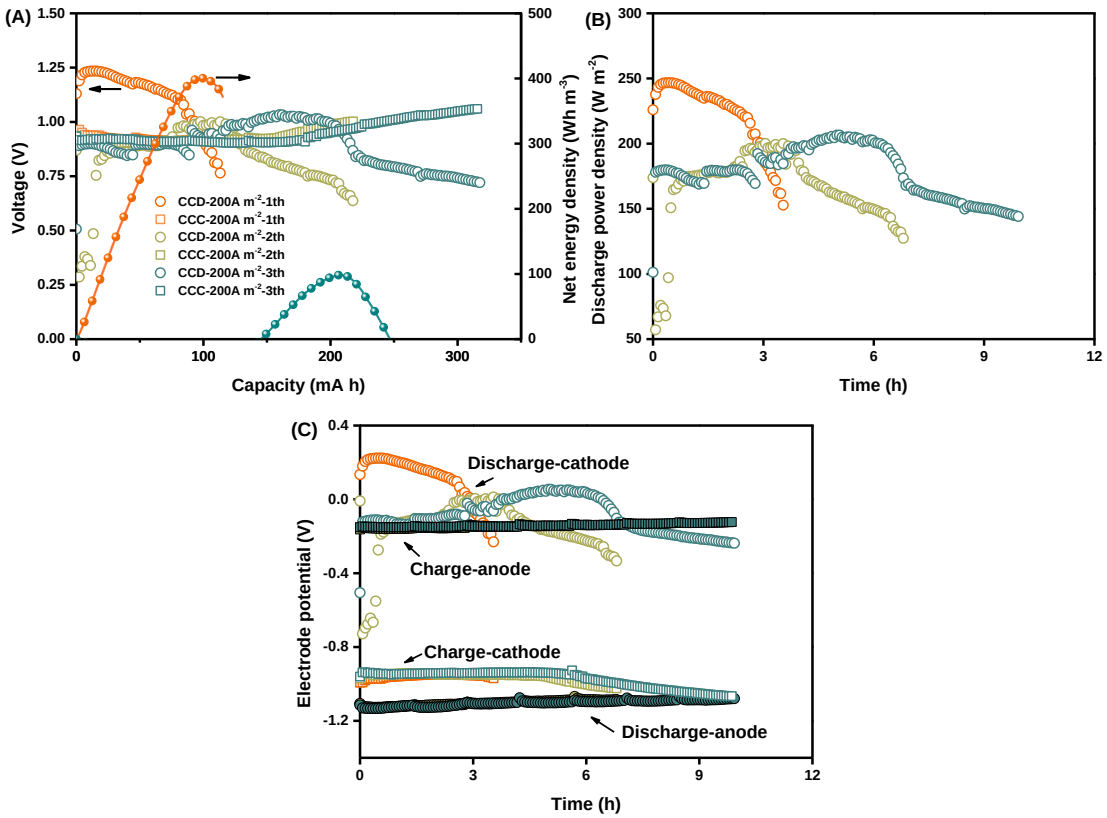
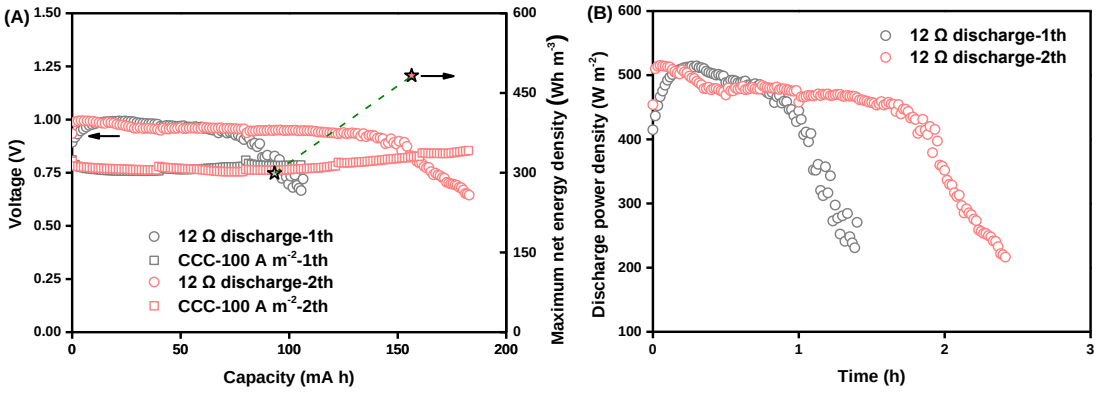


Figure S8. Whole batch cycle performance (A) voltage and net energy density (B) discharge power density (C) electrode potentials of three successive regeneration cycles at a current density of 200 A m^{-2}. The initial electrolyte contained 0.1 M Cu(II) and 1 M $(NH_4)_2SO_4$ in the catholyte and 0.1 M Zn(II), 1 M $(NH_4)_2SO_4$ and 2 M NH_3 in the anolyte. After discharging and charging, the anolyte effluent was regenerated and then operated in next process. "CCD" and "CCC" represent "constant current discharge" and "constant current charge" respectively.



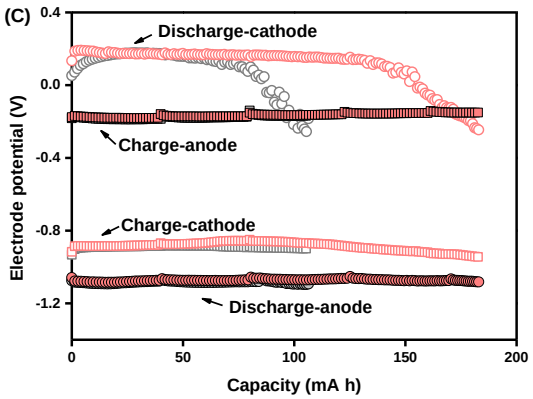


Figure S9. Whole batch cycle performance (A) voltage and maximum net energy density (B) discharge power density (C) electrode potentials of two successive regeneration cycles at a constant discharging load of 12 Ω (external resistance) and a constant charging current density of 100 A m^{-2}. The initial electrolyte contained 0.1 M Cu(II) and 1 M $(NH_4)_2SO_4$ in the catholyte and 0.1 M Zn(II), 1 M $(NH_4)_2SO_4$ and 2 M NH_3 in the anolyte. After discharging and charging, the anolyte effluent was regenerated and then operated in next process. Acid was used for fully dissolving the precipitates that were produced in the two regeneration processes of one cycle. "12 Ω discharge" and "CCC" represent "constant discharge load of 12 Ω" and "constant current charge" respectively.

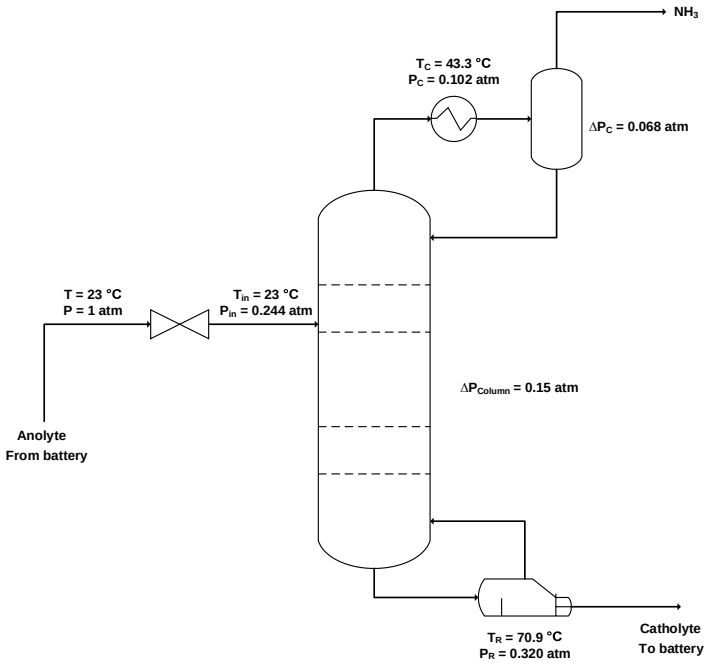


Figure S10. Schematic of the distillation model for ammonia separation from the anolyte based on Aspen HYSYS.