Supporting Information

Using CoS cathode materials with 3D hierarchical porosity and an ionic liquid (IL) as electrolyte additive for high capacity rechargeable magnesium batteries

Mingguang Pan,[ab] Jianxin Zou,[*ab] Richard Laine,[*c] Darvaish Khan,[ab] Rui Guo,[d] Xiaoqin Zeng,[ab] and Wenjiang Ding[ab]

Table S1. Perfomance comparison of different TMC eletrode materials for RMBs.

Electrode material (Ref.)	Electrolyte	Voltage range (V vs. Mg^{2+}/Mg)	Current density (mA g^{-1})	Capacity (mAh g^{-1}) (Cycle number)
CoS_{HE} (this work)	**0.25M POC-0.2IL**	**0.0-1.8**	**20**	**~134 (1)-~360(68-85)**
CoS (*J. Power Sources*, 2015, **294**, 643)	0.25 M $Mg(AlCl_2EtBu)_2$/THF	0.1-2.0	50	~120 (1)-106 (60)
Mo_6S_8 (*Nature*, 2000, **407**, 724)	0.25 M $Mg(AlCl_2EtBu)_2$/THF	0.3-1.8	0.3 mA cm^{-2}	~60 (1-200)-~55(580)
graphene-like MoS_2 (*Adv. Mater.*, 2011, **23**, 640)	$Mg(AlCl_3Bu)_2$/THF	0.5-3.0	20	170 (1)-161 (50)
VS_4 (*Adv. Mater.*, 2018, **30**, 1802563)	0.4 M APC in THF	0.2-2.2	100	251(1)-150(180)
WSe_2 nanowires (*ACS Nano*, 2013, **7**, 8051)	0.25 M $Mg(AlCl_2EtBu)_2$/THF	0.3-3.0	50	~220 (1)-203 (160)
layered TiS_2 (60 ˚C) (*ACS Energy Lett.*, 2016, **1**, 297)	0.4 M APC in tetraglyme	0.4-1.8	47.8	250 (1)-~110 (40)
interlayer-expended TiS_2 (*Nat. Commun.*, 2017, **8**, 339)	0.25 M APC in THF	0.0-2.0	240	~155 (1)-~114(500)
Ti_2S_4 (60 ˚C) (*Energy Environ. Sci.*, 2016, **9**, 2273)	0.4 M APC in tetraglyme	0.5-1.8	24	190 (1)-140(40)
CuS (50 ˚C) (*Nano Energy*, 2018, **47**, 210)	0.25M APC/THF	0.2-1.9	50	~105 (1)-119 (30)
CuS nanosheet assemblies (*Appl. Mater. Interfaces*, 2019, **11**, 7046)	0.25 M $Mg(AlCl_2EtBu)_2$/THF	0.0-2.0	20	400 (1)-336 (15)

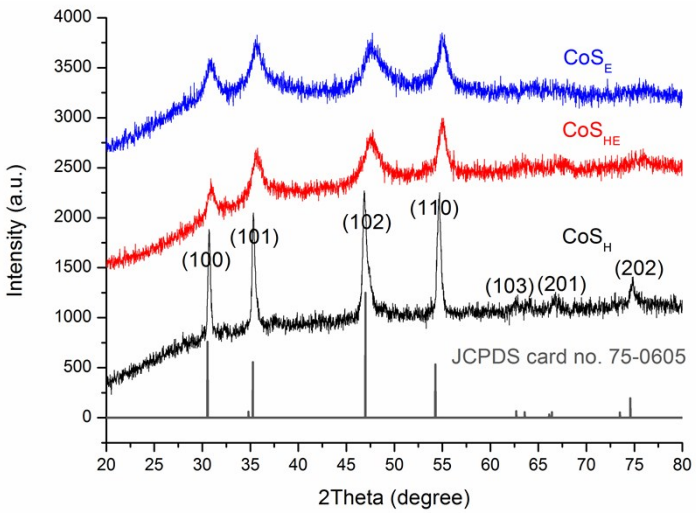

Figure S1. XRD patterns of CoS_H, CoS_{HE} and CoS_E.

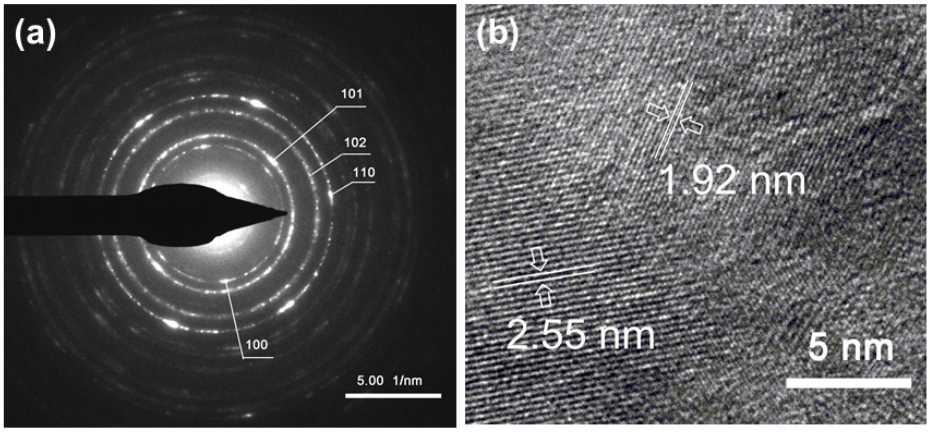

Figure S2. SAED (a) and HRTEM (b) images of CoS_{HE}.

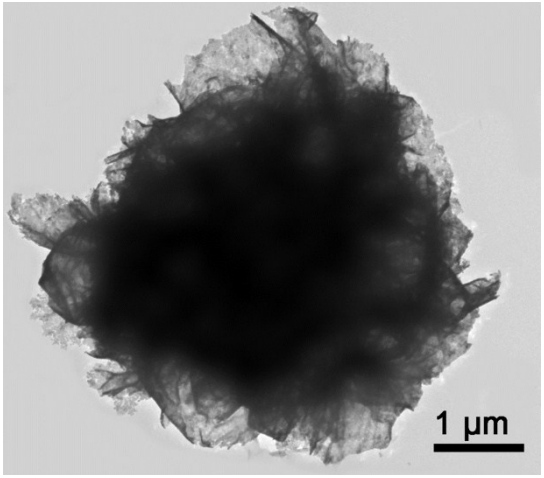

Figure S3. TEM image of CoS_{HE} sphere (~4.5 μm diameter).

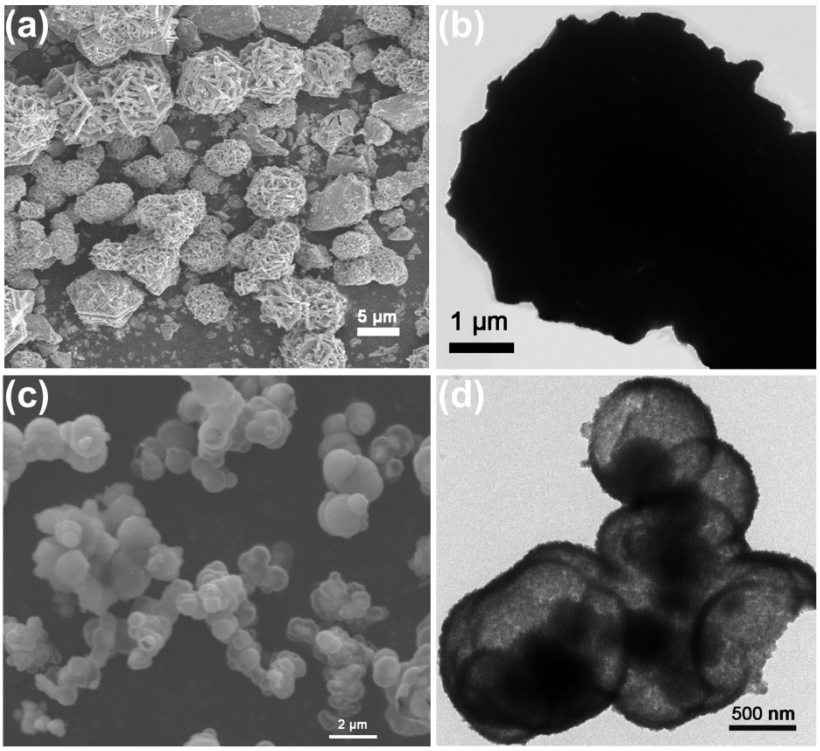

Figure S4. SEM (a,c) and TEM(b,d) images of CoS_H (a,b) and CoS_E (c,d).

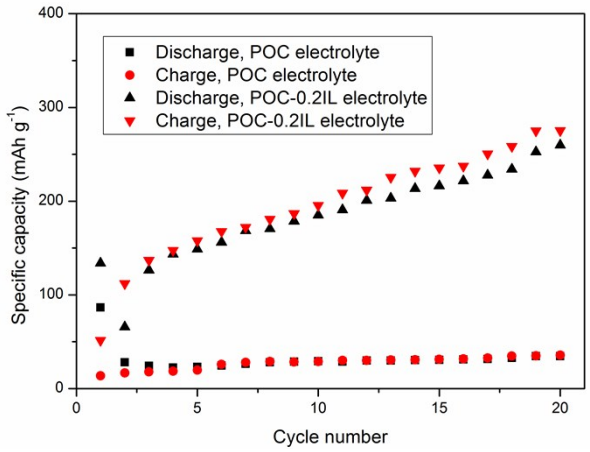

Figure S5. The cycling process (20 cycles) of CoS_{HE} cathode in POC electrolyte (■, Discharge; ●, Charge) and POC-0.2IL electrolyte (▲, Discharge; ▼, Charge).

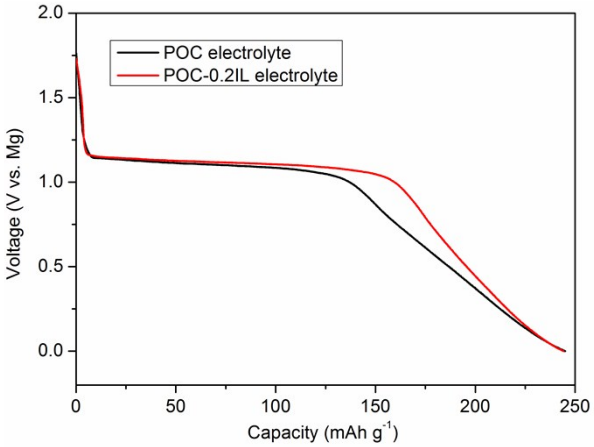

Figure S6. The discharge curves of CoS_{HE} cathode in POC-0.2IL electrolyte (red) and POC electrolyte (black) when discharging nearly identical capacity (around 245 mAh g^{-1}) at a current density of 20 mA g^{-1}.

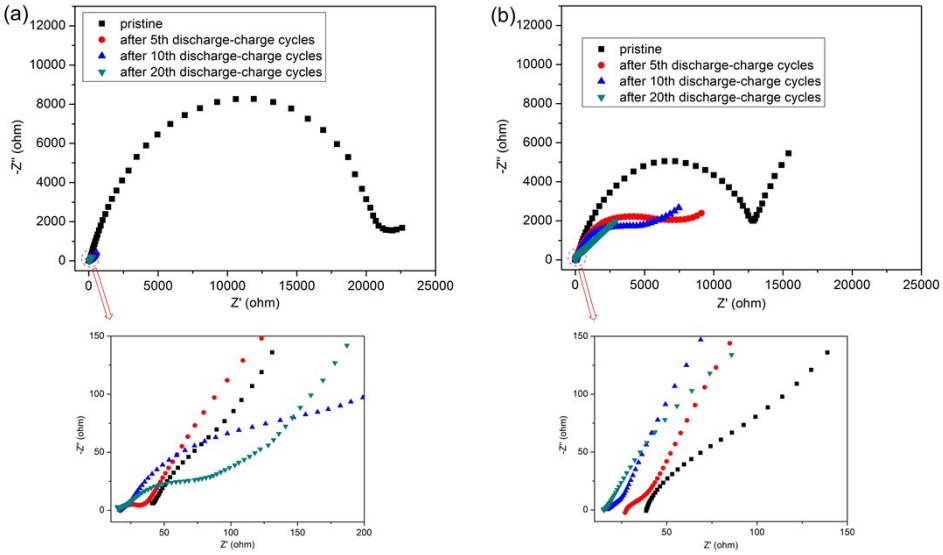

Figure S7. The EIS curve changes of CoS_{HE} cathode in (a) POC-0.2IL electrolyte and (b) POC electrolyte during the initial 20 cycles. The bottom is the enlarged parts for the dotted circle in (a) and (b), respectively.

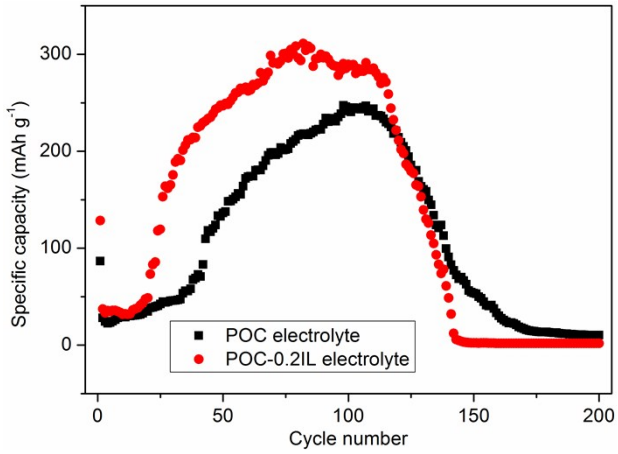

Figure S8. The discharge performances of CoS_{HE} cathode in POC-0.2IL electrolyte (red) and POC electrolyte (black).

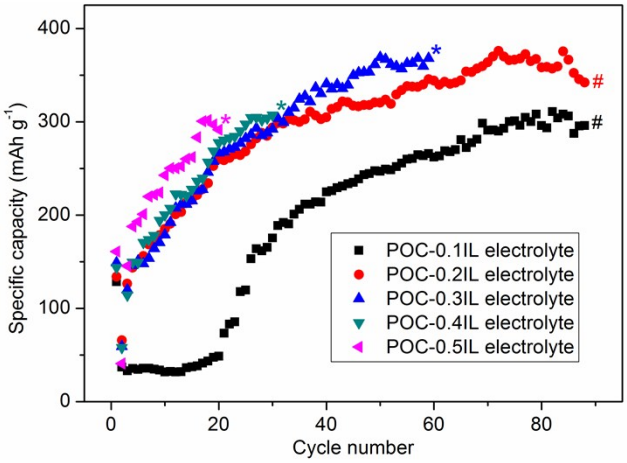

Figure S9. The discharge performances of CoS_{HE} cathode in POC-0.1IL (■), POC-0.2IL (●), POC-0.3IL (▲), POC-0.4IL (▼) and POC-0.5IL (◄) electrolytes, respectively. The marks, # and * in the figure denote that the batteries are still working and going to be dead, respectively.

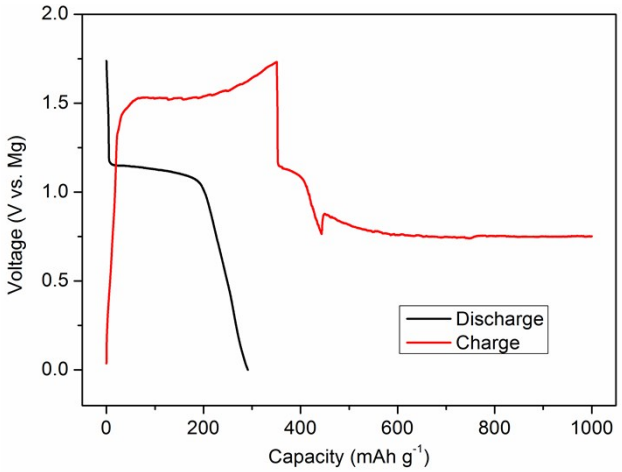

Figure S10. The discharge-charge curves of CoS_{HE} with POC-0.5IL electrolyte in the last cycle (20th).

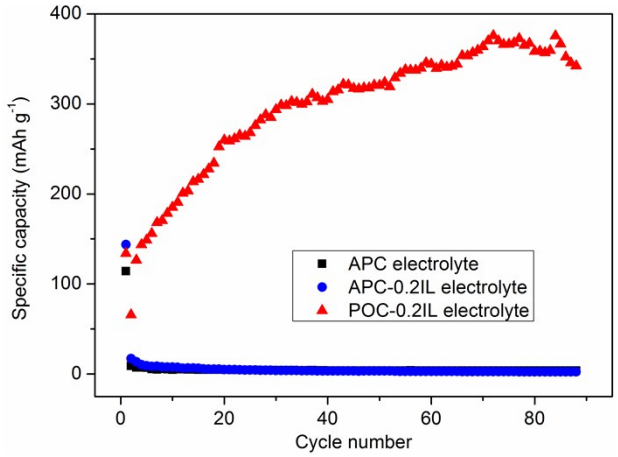

Figure S11. The discharge performances of CoS_{HE} cathode in APC (■), APC-0.2IL (●) and POC-0.5IL (▲) electrolytes, respectively.

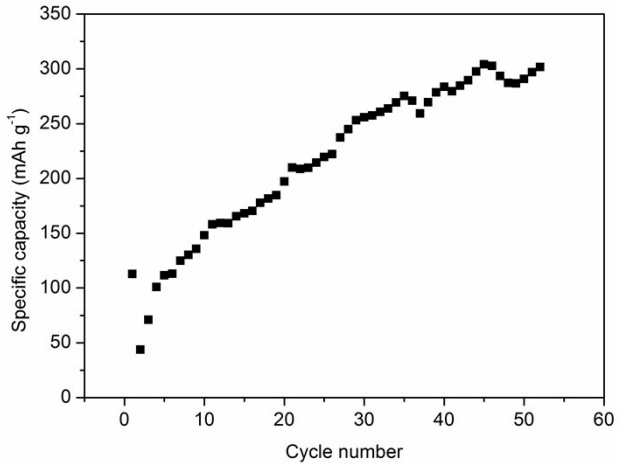

Figure S12. The discharge performances of CoS_{HE} cathode in POC-0.2IL electrolyte when the assembly of battery was exposure to the air.

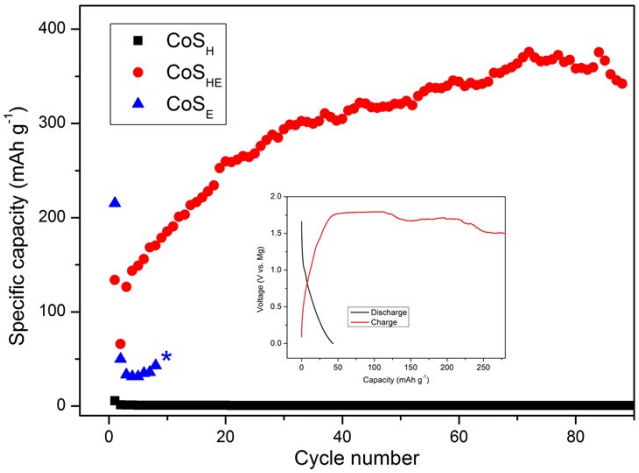

Figure S13. The discharge performances of CoS_H, CoS_{HE} and CoS_E cathodes in POC-0.2IL electrolyte. The mark (*) denotes the battery is going to be dead. Insert: the discharge-charge curves of CoS_E cathode in the last cycle (8th).

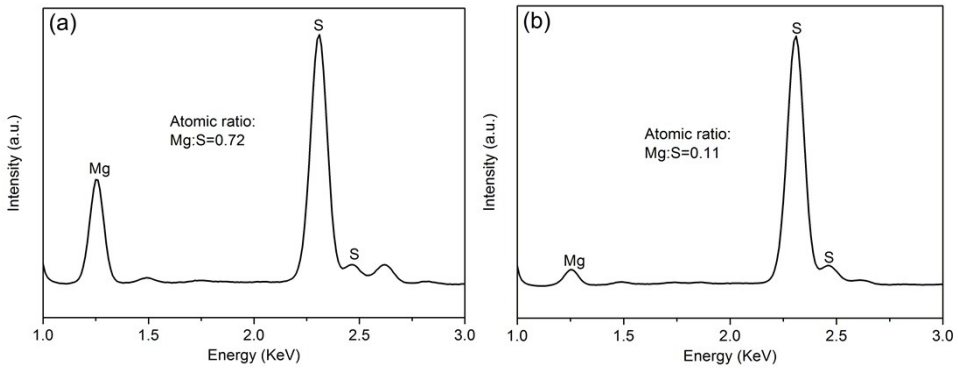

Figure S14. Partial EDX spectra of CoS_{HE} cathode (a) after 70th discharge and (b) after 70th charge, respectively. The content of Co is not real in the EDX spectra because of its magnetic property so that the data are not given. And we assume that the content of S after 70th discharge should be the same with that after 70th charge.

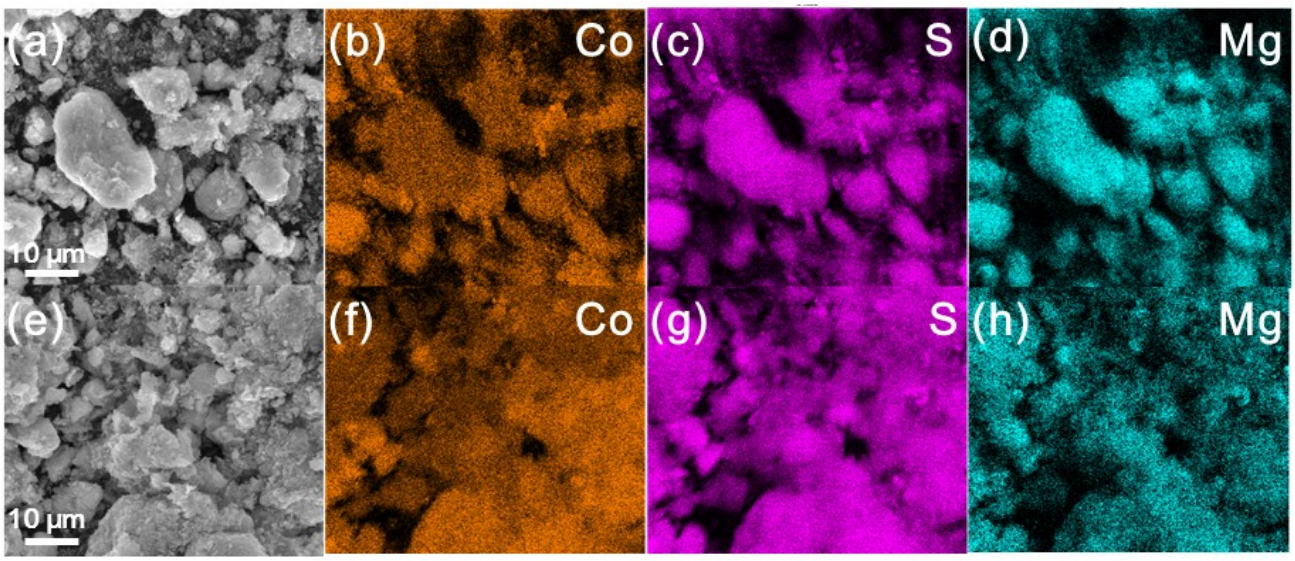

Figure S15. (a) SEM image of CoS_{HE} cathode after 70th discharge, and cooresponding (b) Co, (c) S, (d) Mg elemental mapping images; (e) SEM image of CoS_{HE} cathode after 70th charge, and cooresponding (f) Co, (g) S, (h) Mg elemental mapping images.

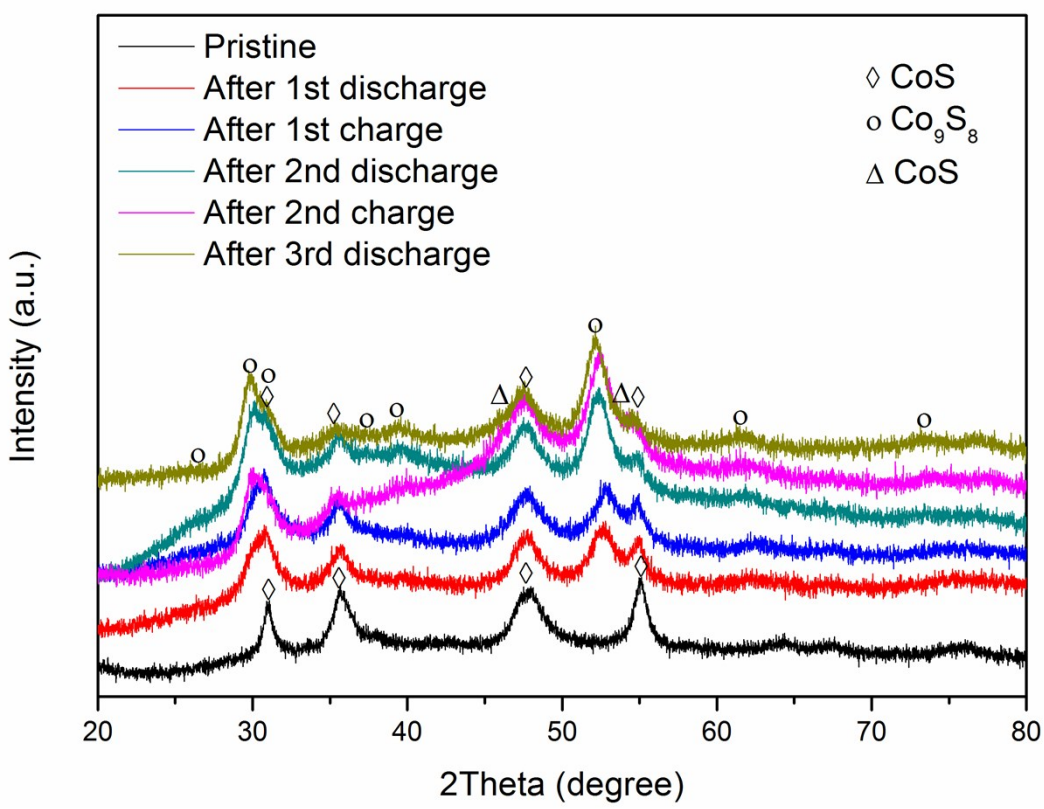

Figure S16. Ex situ XRD patterns of CoS_{HE} at the pristine state and after initial discharge-charge cycles.