Supporting Information

Insights into the electrochemical processes of rechargeable Magnesium-Sulfur batteries with new cathode design

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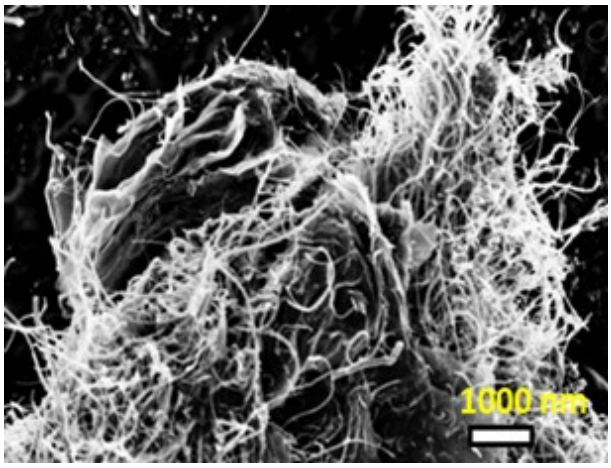


Fig. S1: SEM images of nitrogen doped (G-MWCNT) nanocomposite (NC).

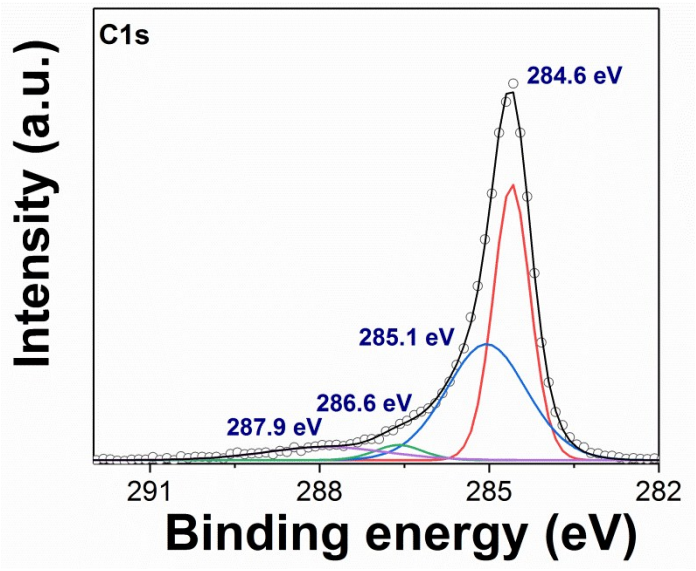


Fig. S2: C1s - XPS spectra of sulfur loaded NC.

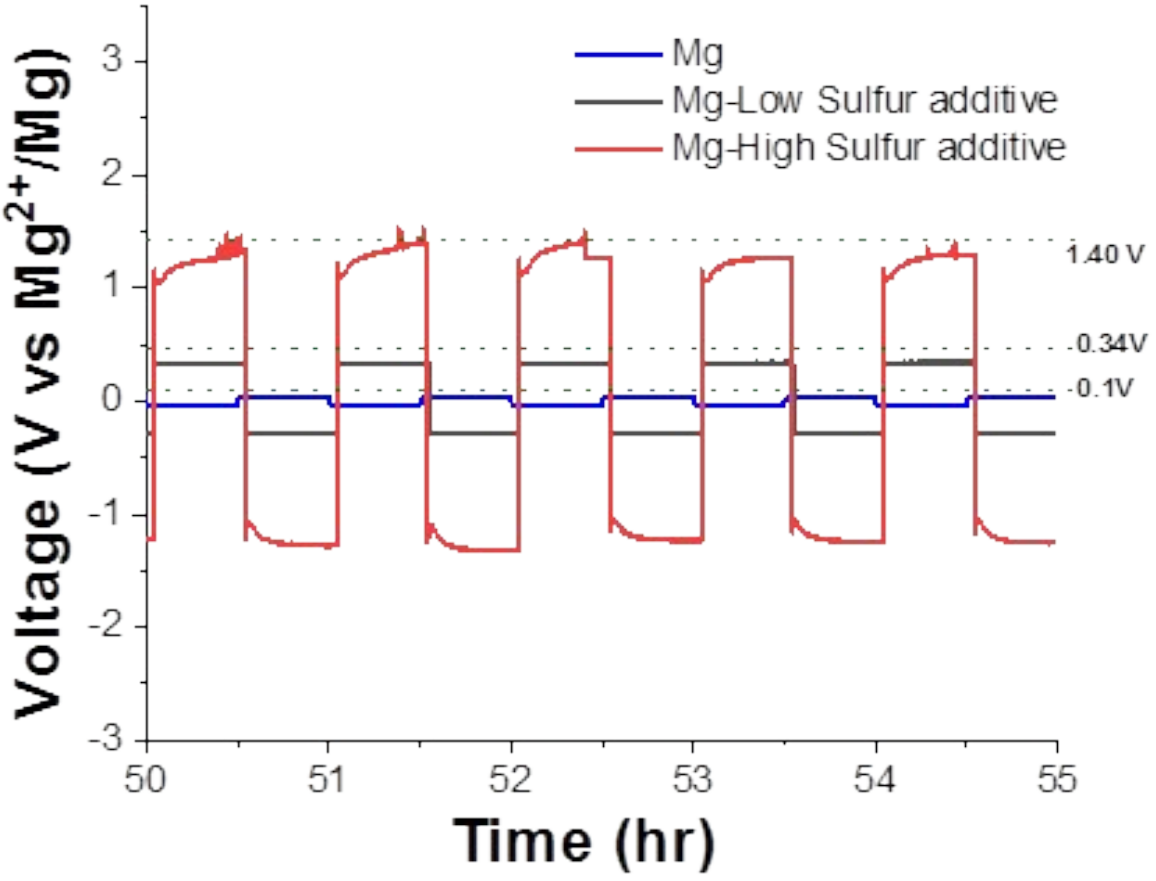


Fig. S3: Stripping/plating in an Mg/Mg symmetric cell with pure Mg electrolyte and electrolytes containing low and high amounts of sulfur additive, respectively. The graph demonstrates the growth of overvoltage with sulfur additive concentration.

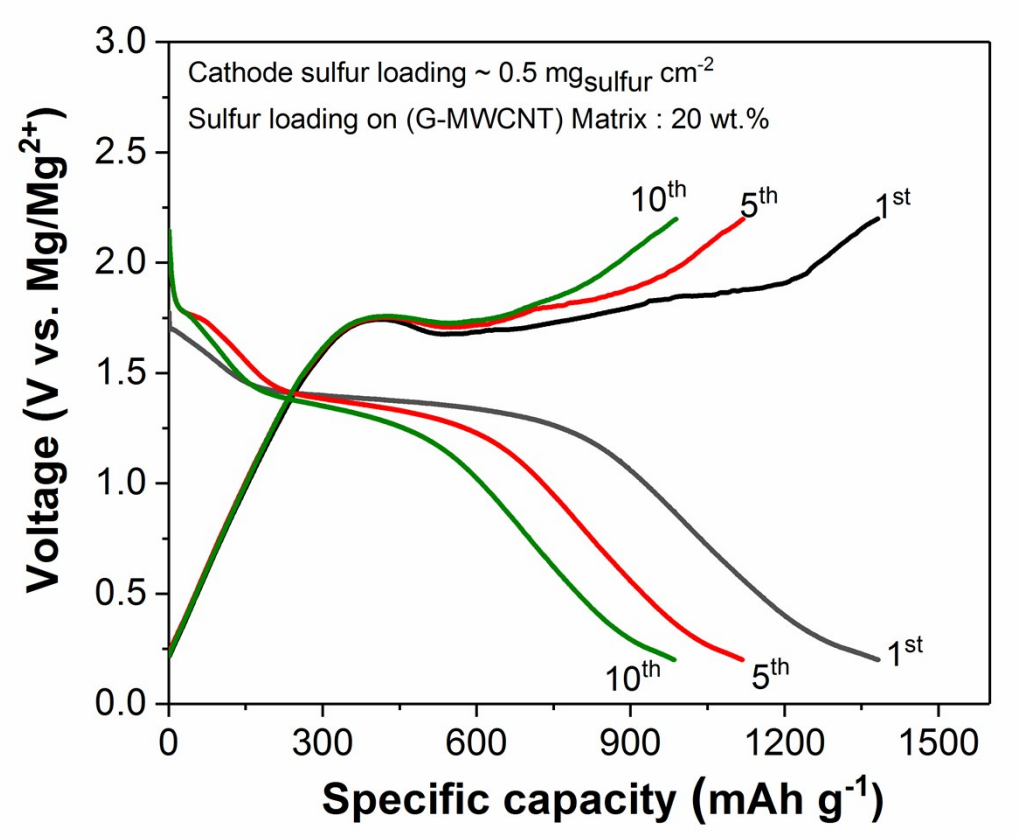


Fig. S4: Charge/discharge voltage profiles for the S/NC cathode with low sulfur loading ~0.5 mg_{sulfur} cm^{-2}. The sulfur loading on (G-MWCNT) matrix: 20 wt.%.

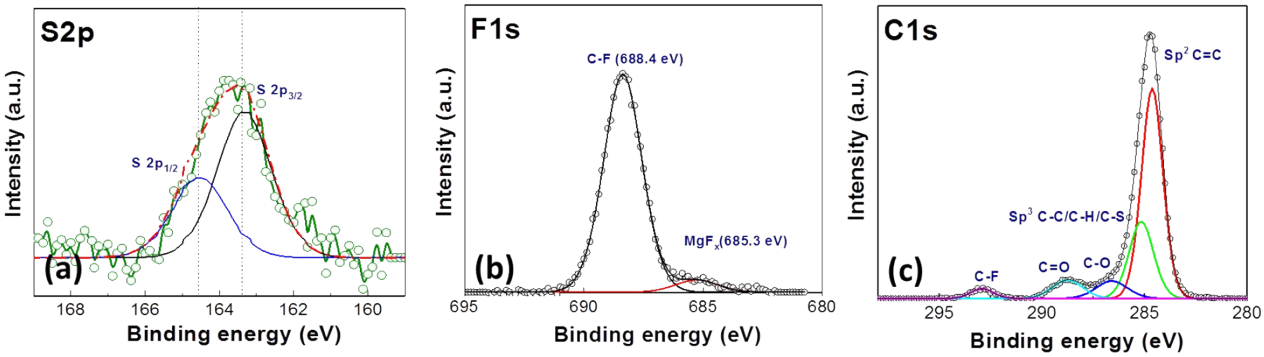


Fig. S5: (a) S2p, (b) F1s, and (c) C1s XP, detail spectra of the cycled Mg anode.

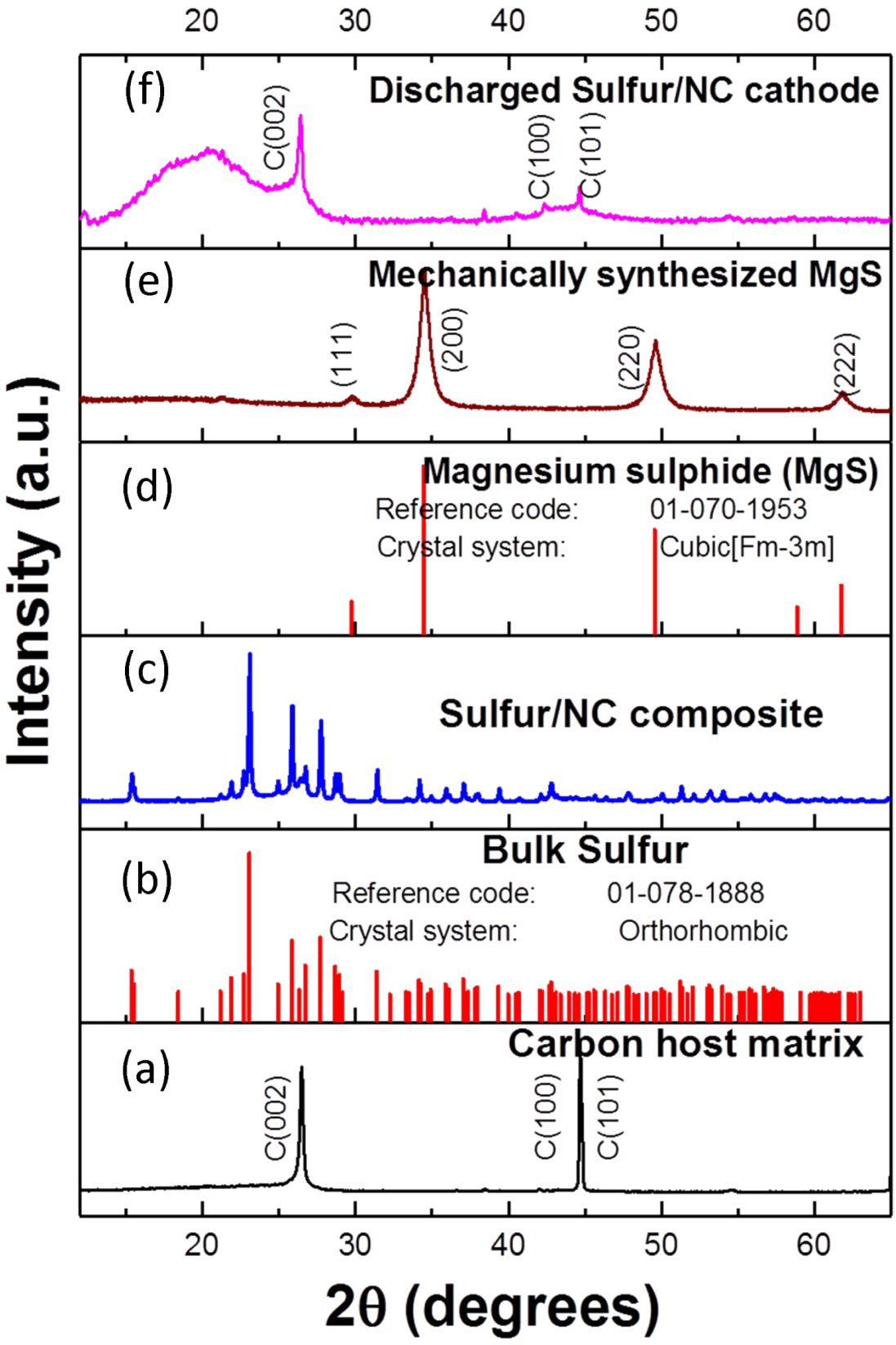


Fig. S6: XRD spectra of pristine sulfur cathode, discharged sulfur cathode, and mechanically synthesized MgS.

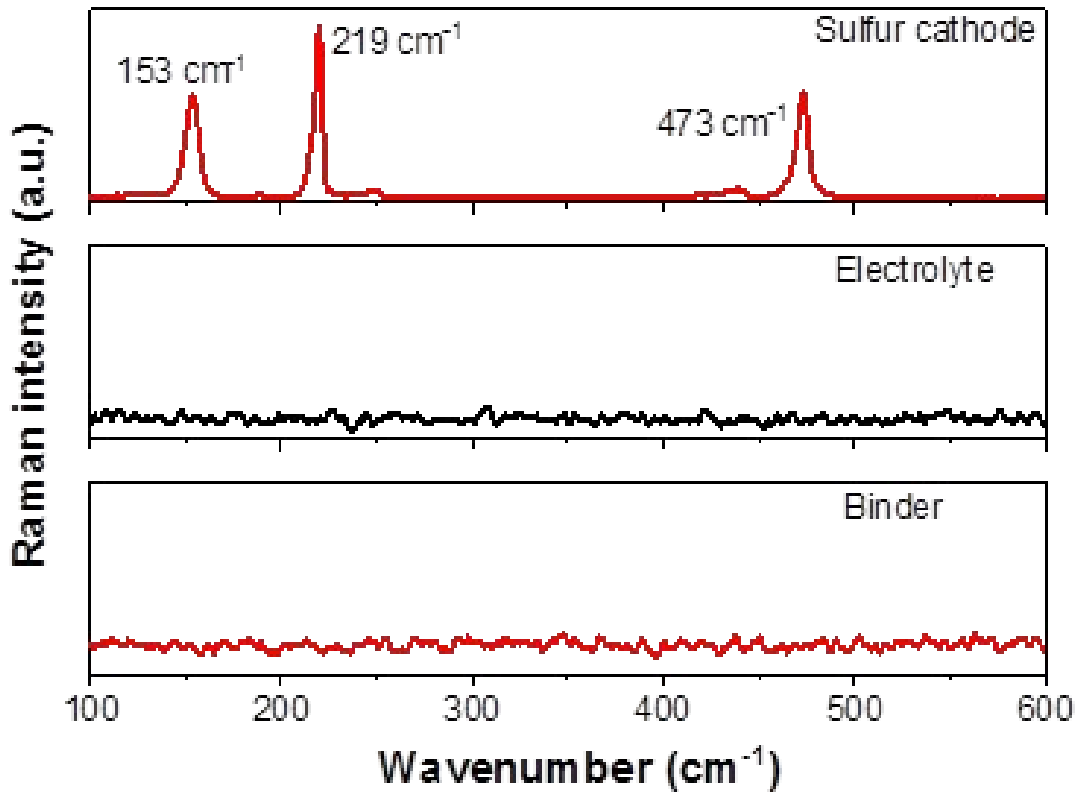
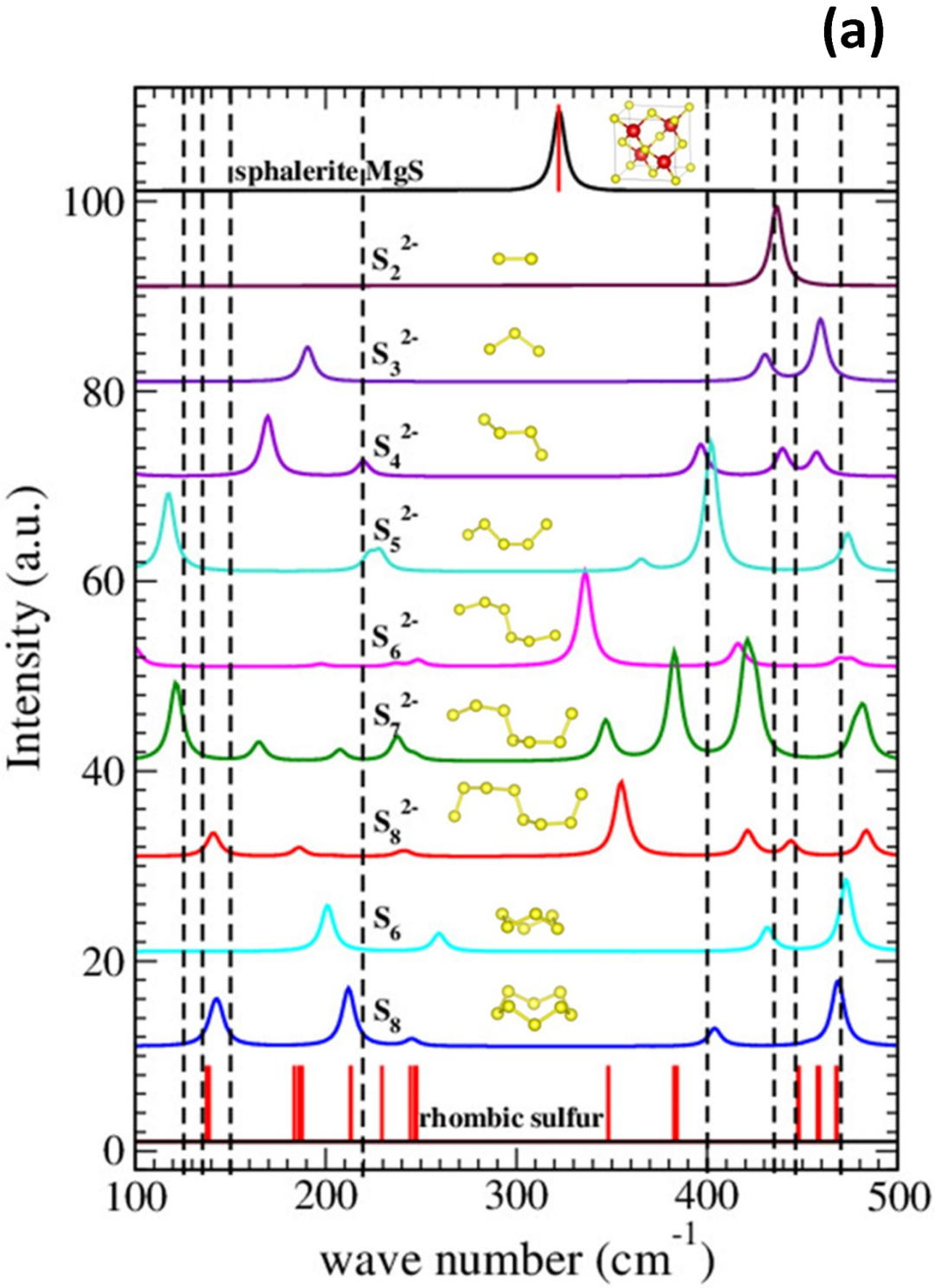


Fig. S7: Raman spectra of sulfur cathode, pure electrolyte and binder.


(a)
sphalerite MgS
S_2^{2-}
S_3^{2-}
S_4^{2-}
S_5^{2-}
S_6^{2-}
S_7^{2-}
S_8^{2-}
S_6
S_8
rhombic sulfur
Intensity (a.u.)
wave number (cm^{-1})
0
20
40
60
80
100
100
200
300
400
500

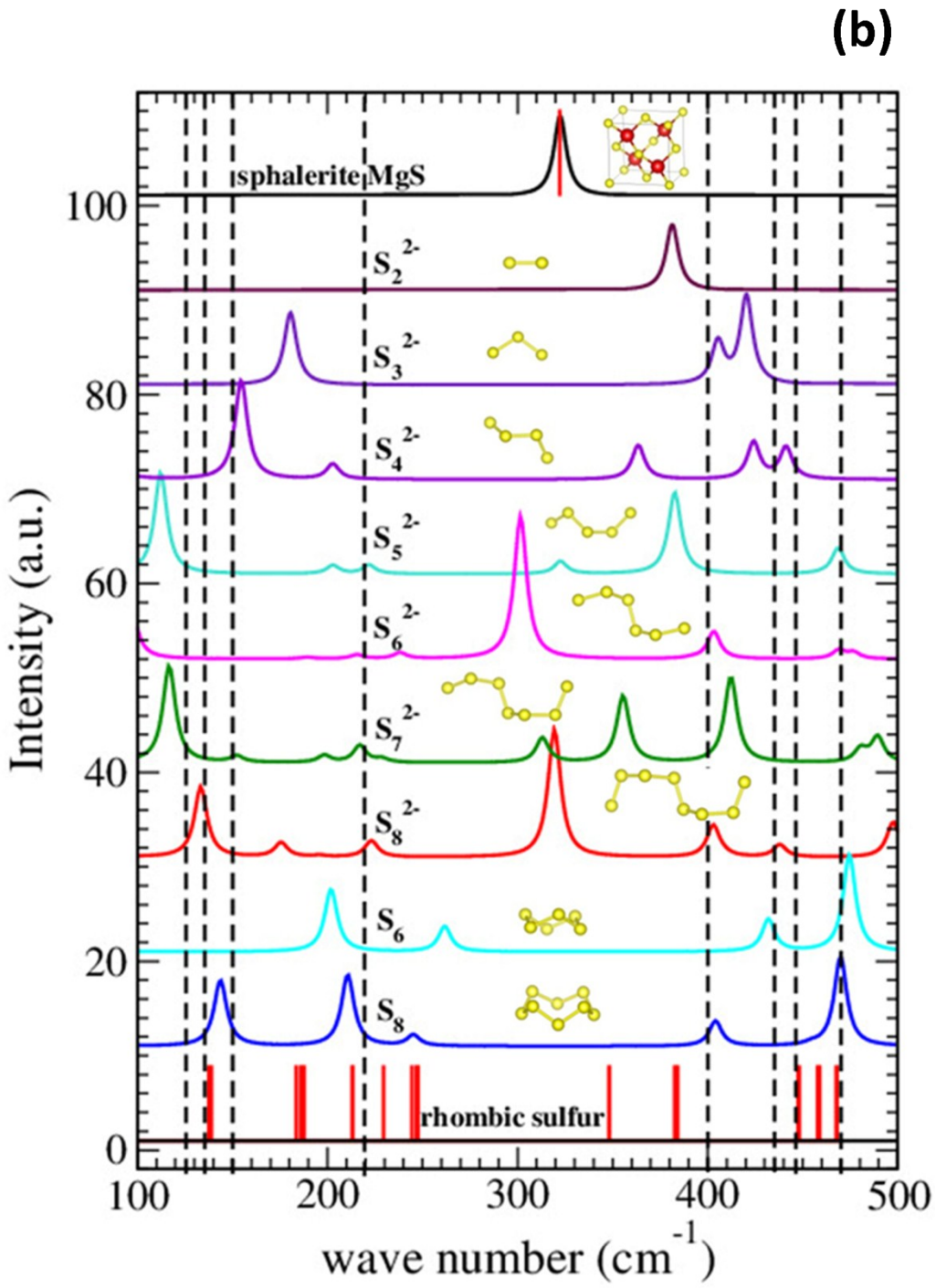
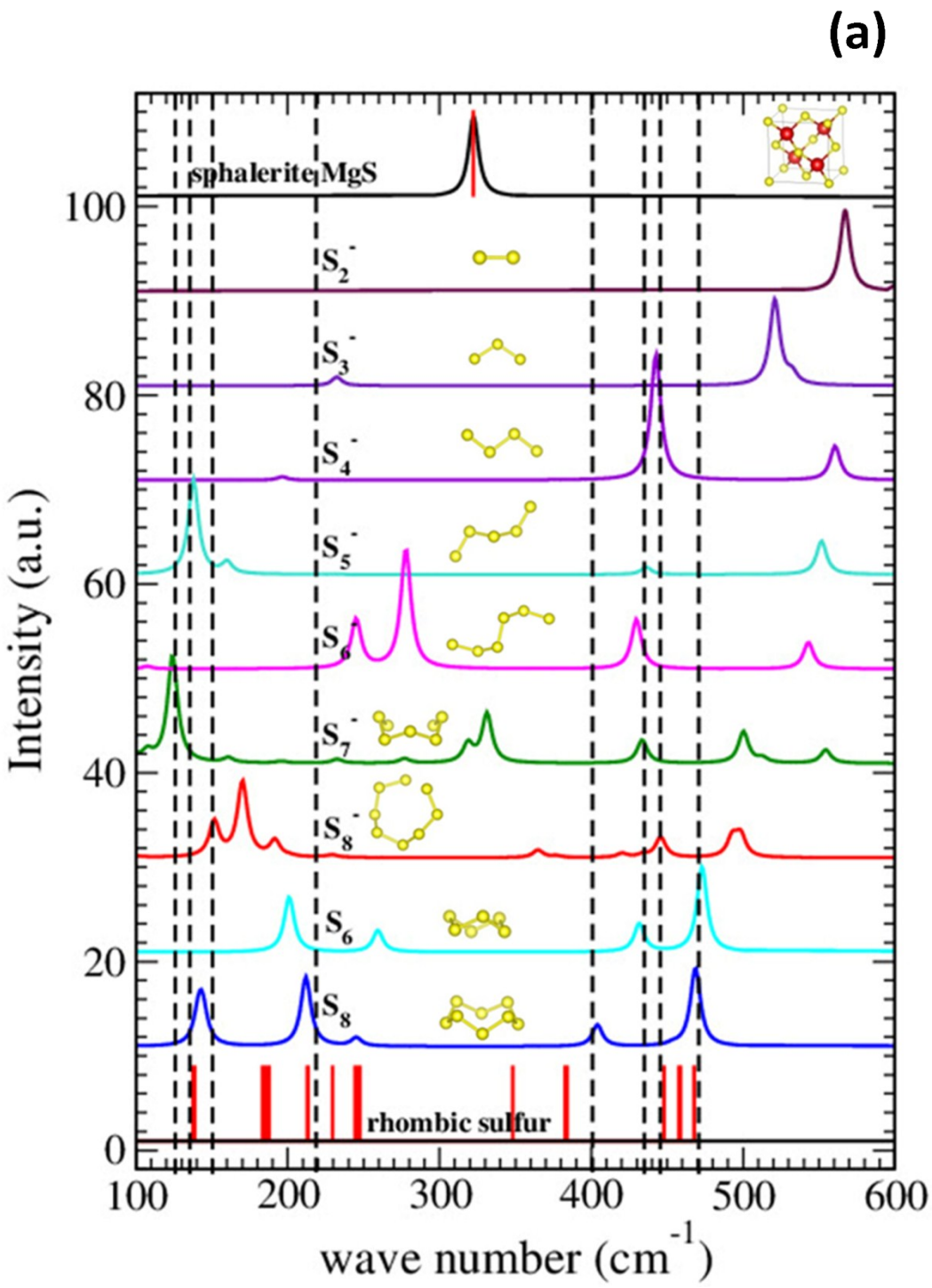


Fig. S8: Raman spectra of different polysulfide di-anions, elemental sulfur and sphalerite MgS as obtained from DFT. The depicted spectra correspond to the conformers that were found to be energetically most favorable. In (a) the polysulfide calculations consider the impact of the electrolyte while in (b) vacuum is assumed. Dashed lines correspond to experimentally observed frequencies.


(a)
sphalerite MgS
S_2^-
S_3^-
S_4^-
S_5^-
S_6^-
S_7^-
S_8^-
S_6
S_8
rhombic sulfur
Intensity (a.u.)
wave number (cm^{-1})
0
20
40
60
80
100
100
200
300
400
500
600

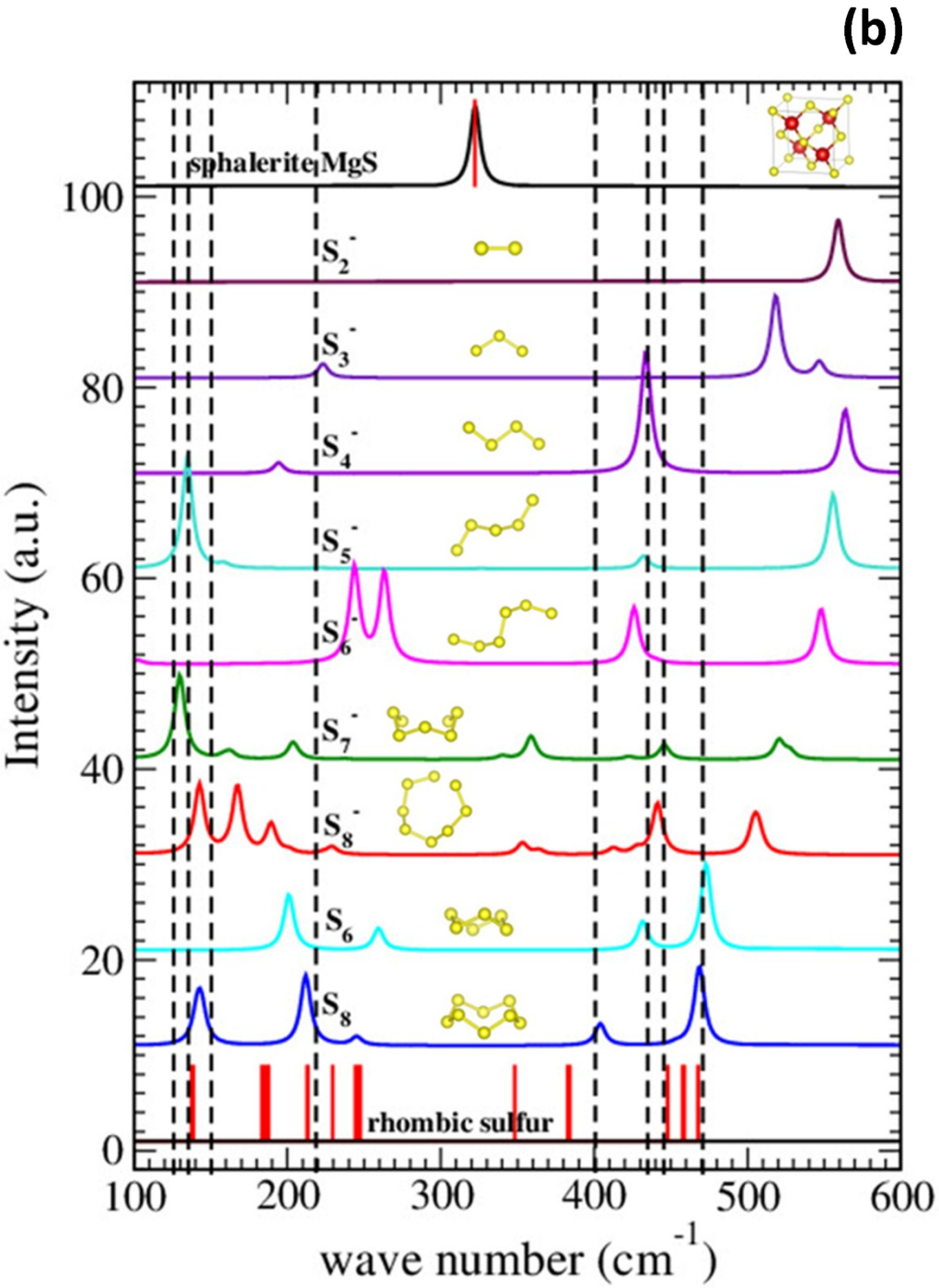


Fig. S9: Raman spectra of different polysulfide mono-anions, elemental sulfur and sphalerite MgS as obtained from DFT. The depicted spectra correspond to the conformers that were found to be energetically most favorable. In (a) the polysulfide calculations consider the impact of the electrolyte while in (b) vacuum is assumed. Dashed lines correspond to experimentally observed frequencies.

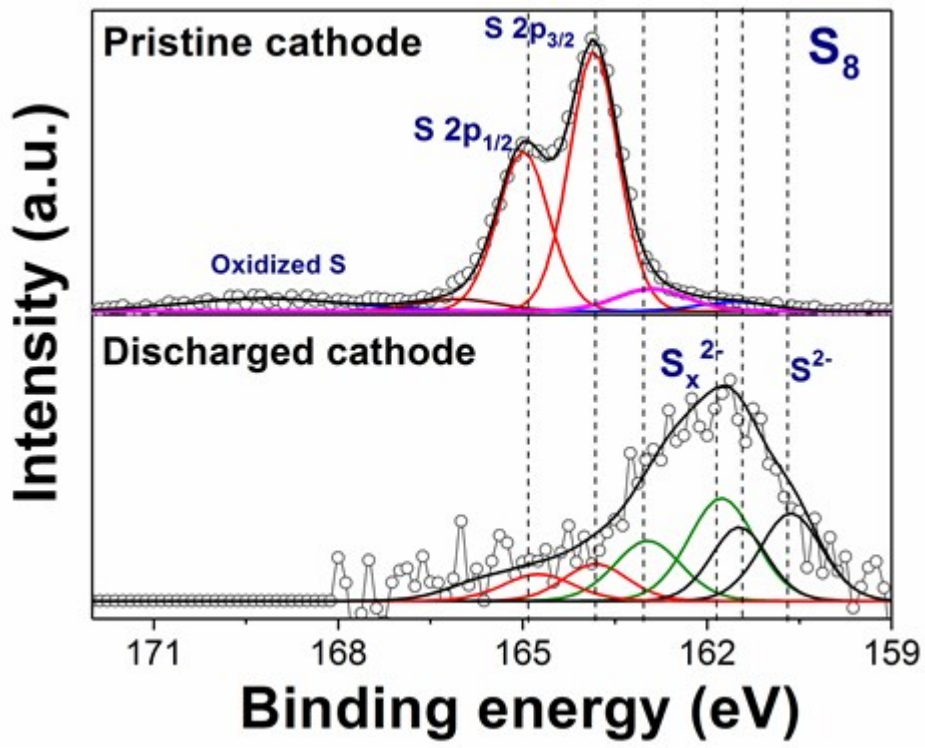


Fig. S10: S2p XP spectra for the pristine sulfur cathode and the discharged cathode at 0.5V.

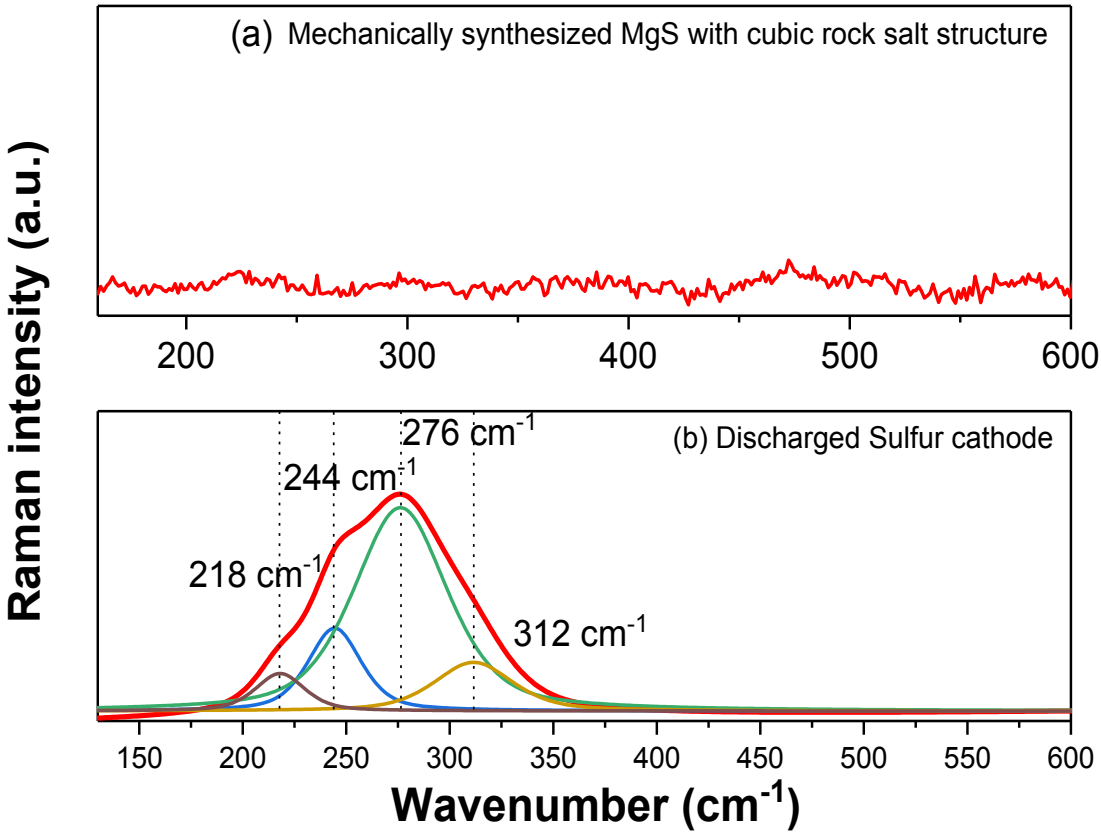


Fig. S11: Raman spectra for (a) mechanically synthesized MgS with cubic rock salt structure and (b) discharged sulfur cathode.

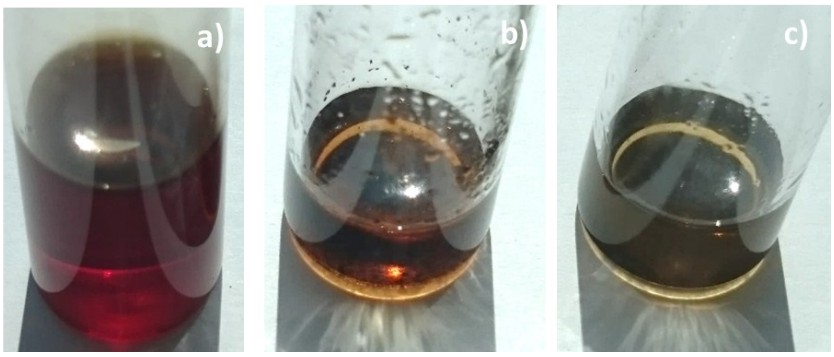


Fig. S12: (a) Mg polysulfide solution as prepared, (b) after treating with pristine G-MWCNT, and (c) after treating with nitrogen functionalized (G-MWCNT) for two days.

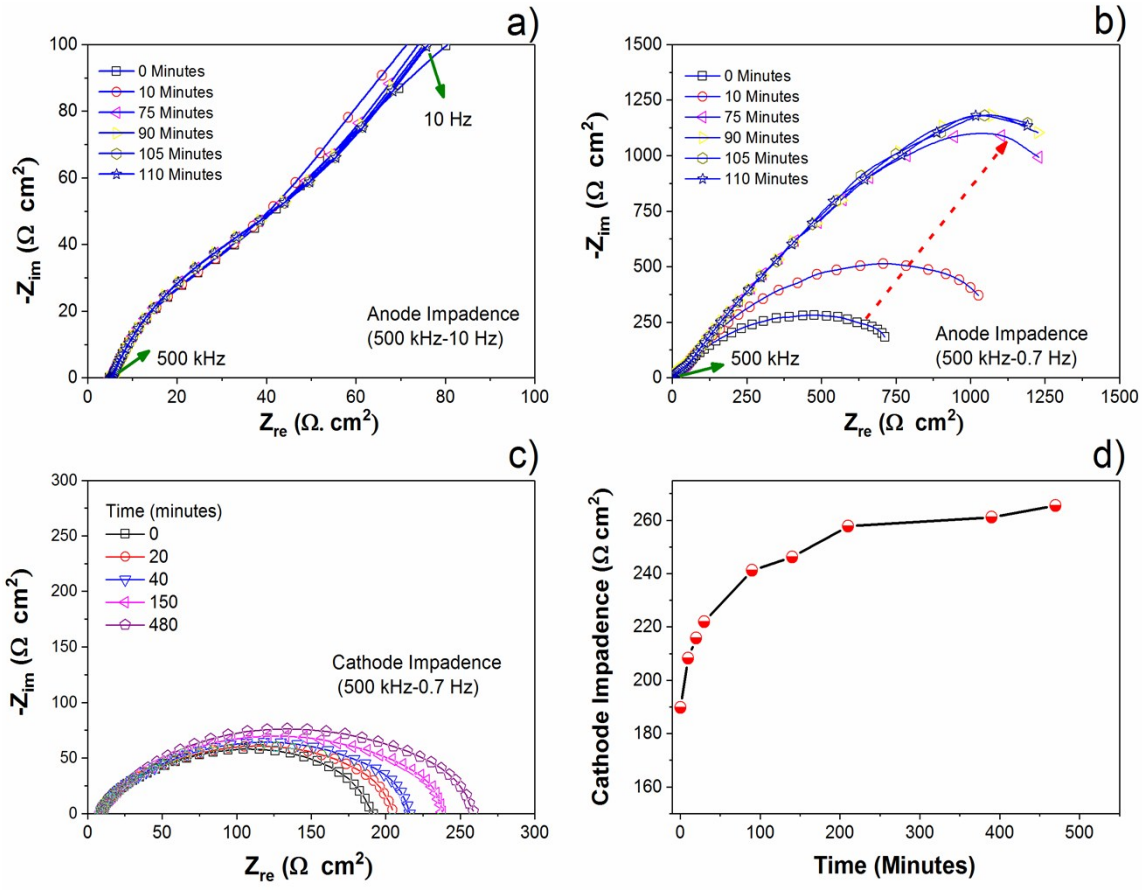


Fig. S13: Impedance spectra of the Mg anode of an Mg/S cell at OCV under different resting periods in the frequency range (a) 500 kHz -10 Hz, and (b) 500 kHz – 0.7Hz.

(c, d) Impedance spectra and the variation of impedance of the sulfur cathode of an Mg/S cell at OCV under different resting periods.

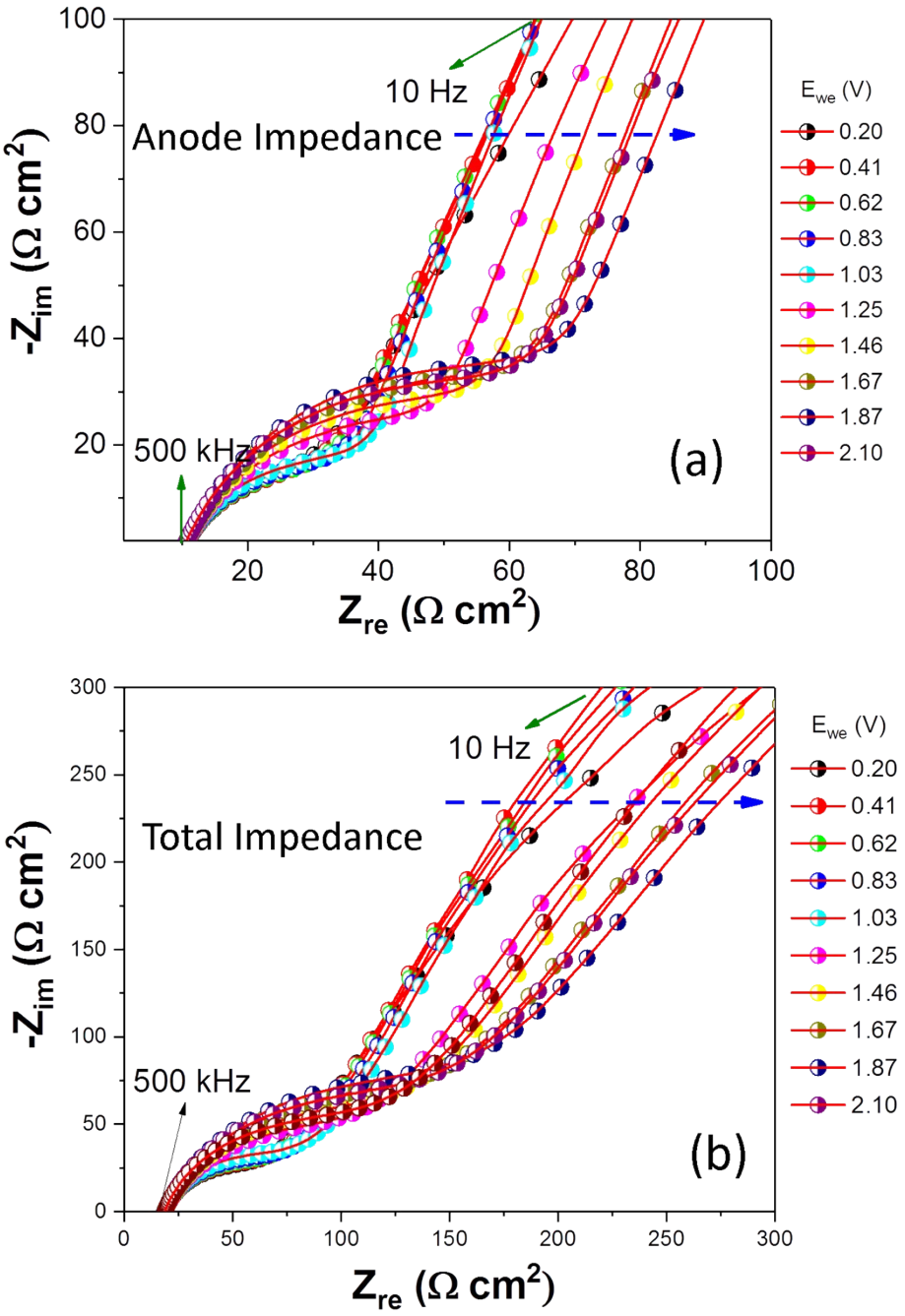


Fig. S14: Changes in the impedance spectra of the (a) Mg anode and (b) total Mg/S cell impedance under different cathode potentials over the frequency range 500 kHz -10 Hz. The spectra were collected during the charge of Mg/S cell.

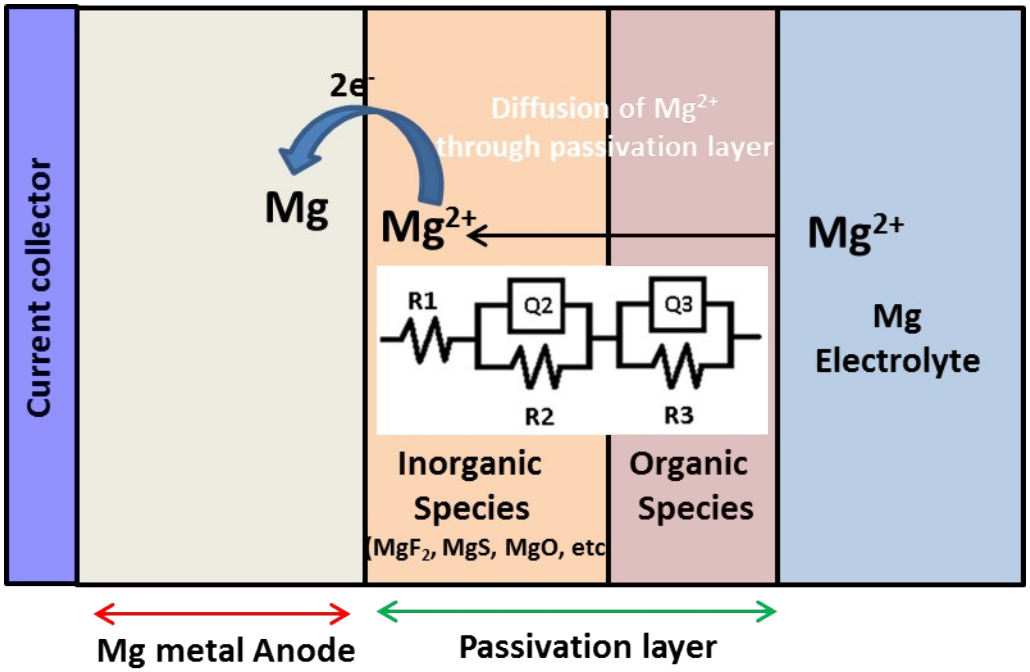


Fig. S15: The equivalent circuit for the Mg anode side of an Mg/S cell.

Voltage (V)	2.1	1.87	1.67	1.46	1.25	1.03	0.83	0.62	0.41	0.2
R1(Ω cm^{-2})	9.7	9.7	9.7	9.7	9.8	9.6	9.5	9.5	9.5	9.5
Q2-Yo	4.75E-05	5.30E-05	6.15E-05	7.24E-05	7.07E-05	8.25E-05	0.000102	0.000106	0.000121	0.000115
Q2-n	0.82	0.81	0.79	0.78	0.78	0.76	0.74	0.74	0.73	0.73
R2 (Ω cm^{-2})	55.1	59.6	57.5	53.3	48.0	45.6	46.0	38.6	33.6	27.5
Q3-Yo	0.000152	0.000149	0.000148	0.000153	0.000166	0.000167	0.000155	0.00017	0.000179	0.000196
Q3-n	0.90	0.91	0.91	0.90	0.87	0.87	0.89	0.87	0.86	0.84
R3 (Ω cm^{-2})	2714.9	2823.7	2932.5	3155.2	3697.5	2165.8	1609.7	1475.3	1084.3	681.0

Table S1: The equivalent circuit fitting values for the Mg anode impedances for the cathode discharge potentials from 2.1V to 0.2V