ADVANCED FUNCTIONAL MATERIALS

Supporting Information

for *Adv. Funct. Mater.,* DOI: 10.1002/adfm.202210343

A Low-Cost and Recyclable Mg/$SOCl_2$ Primary Battery Via Synergistic Solvation and Kinetics Regulation

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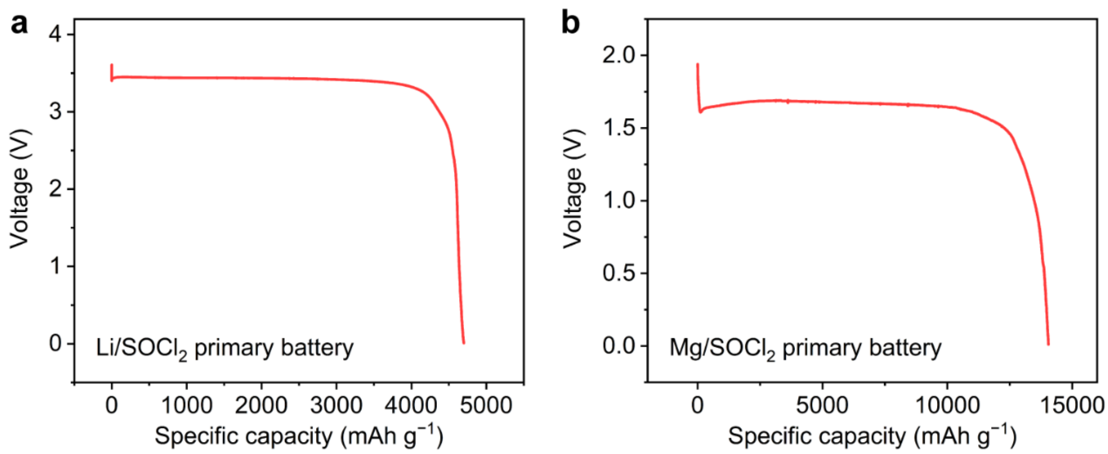

Figure S1. a, b) Galvanostatic discharge curves of Li and Mg/$SOCl_2$ primary batteries at the current density of 50 mA g^{-1}, respectively.

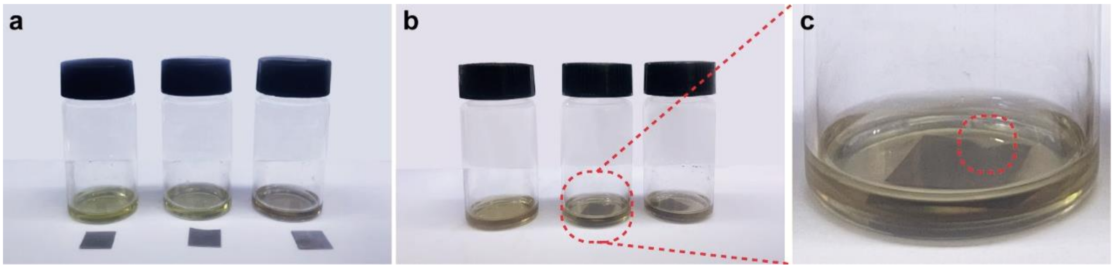

Figure S2. a, b) Mg anodes before and after immersing in a variety of electrolytes (left to right: HC-A-$SOCl_2$, A-$SOCl_2$, A-M-$SOCl_2$) overnight, respectively. **c)** Enlarged photograph from **b** indicates the Mg metal corrosion in A-$SOCl_2$ electrolyte.

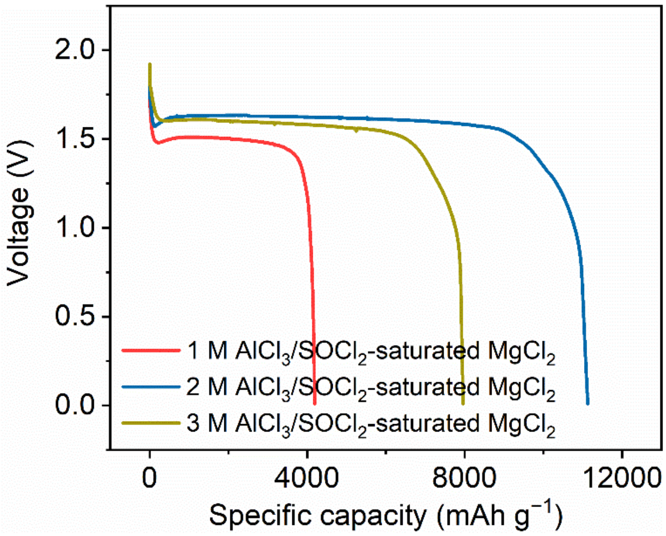

Figure S3. The discharging profiles of Mg/$SOCl_2$ batteries using different electrolytes comprised of increasing $AlCl_3$ concentrations (1, 2 and 3 M) with the addition of excess $MgCl_2$ (0.4, 0.8 and 1.2 M) for saturation. Current density, 100 mA g^{-1}.

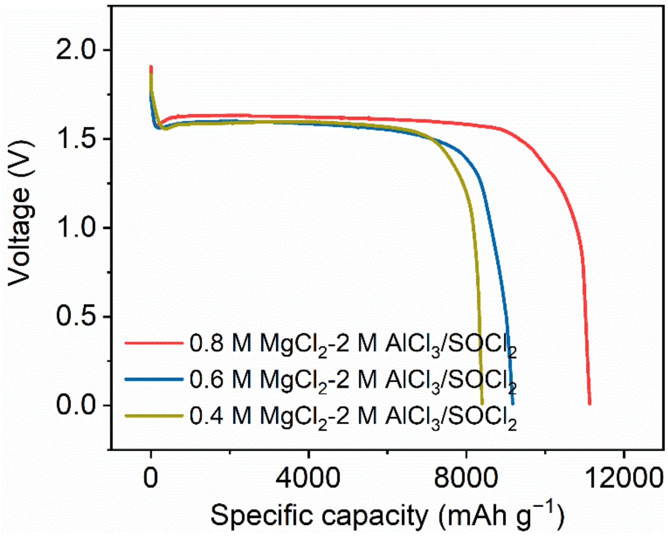

Figure S4. The discharging profiles of $Mg/SOCl_2$ primary batteries using different electrolytes comprised of increasing $MgCl_2$ concentration (0.4, 0.6 and 0.8 M) with the same $AlCl_3$ concentration of 2 M. Current density, 100 mA g^{-1}.

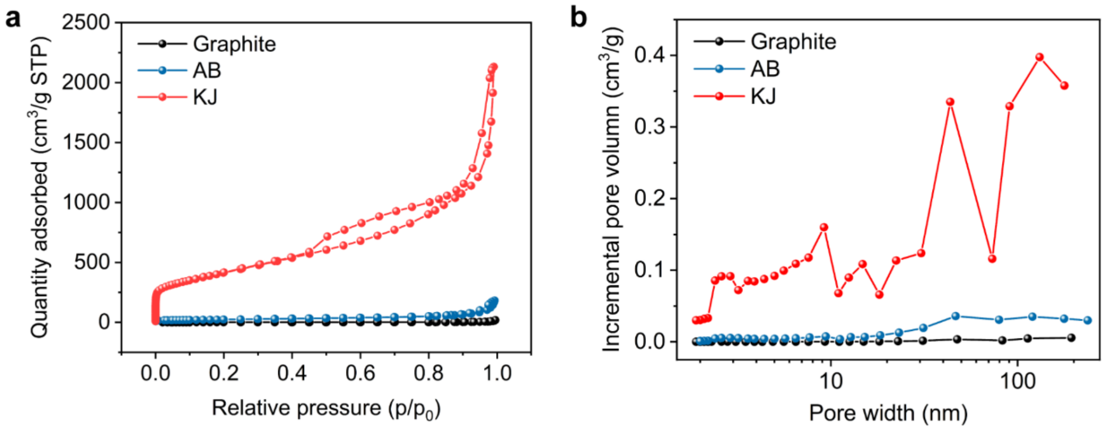

Figure S5. a) Nitrogen adsorption-desorption isotherms of graphite, AB and KJ using Brunauer-Emmett-Teller (BET) method. **b)** Pore size distribution derived from the adsorption branch by the Barrett-Joyner-Halenda (BJH) method.

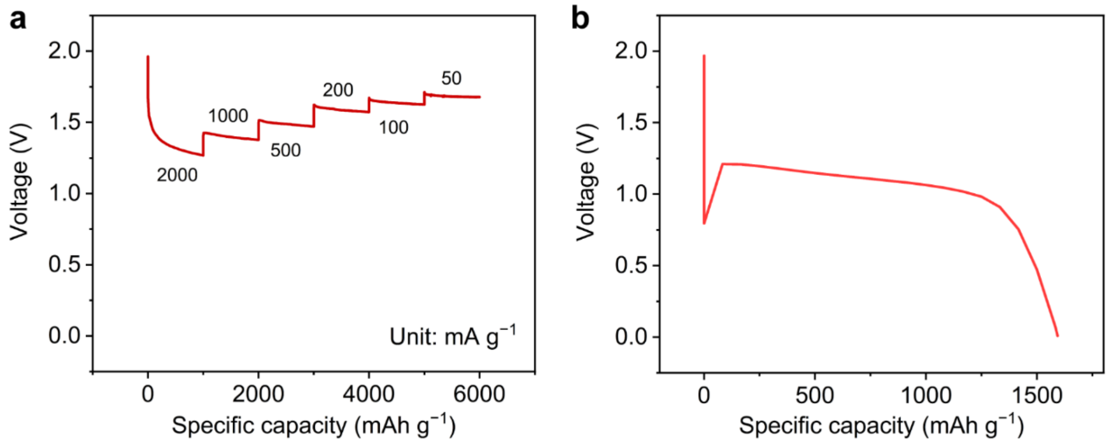

Figure S6. a) The discharging profile of a $Mg/SOCl_2$ primary battery with different discharge current densities from 2,000 to 50 mA g^{-1}. **b)** Galvanostatic discharge curve of a $Mg/SOCl_2$ primary battery at the current density of 10,000 mA g^{-1} at 50 °C.

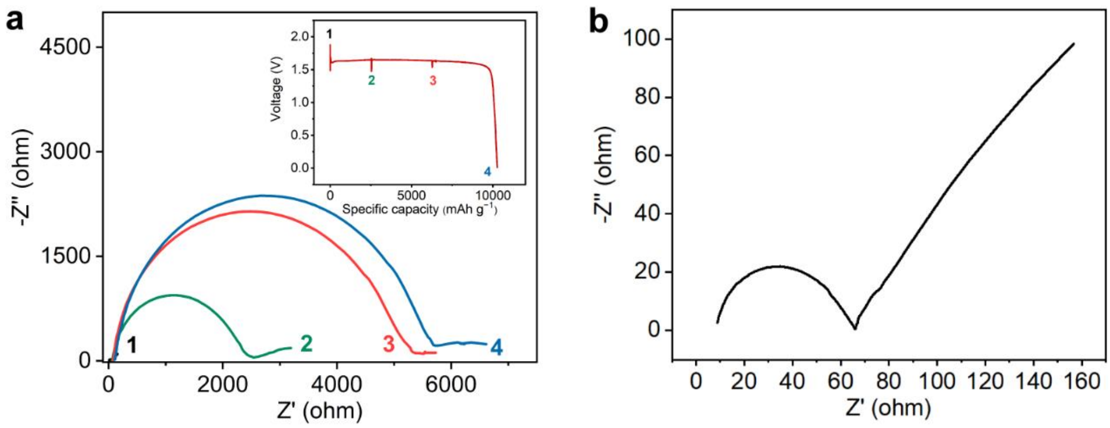

Figure S7. a) EIS measurement of a Mg/$SOCl_2$ primary battery at a variety of discharge states as indicated in the discharging curve of the inset. **b)** EIS measurement of a Mg/$SOCl_2$ primary battery before discharge.

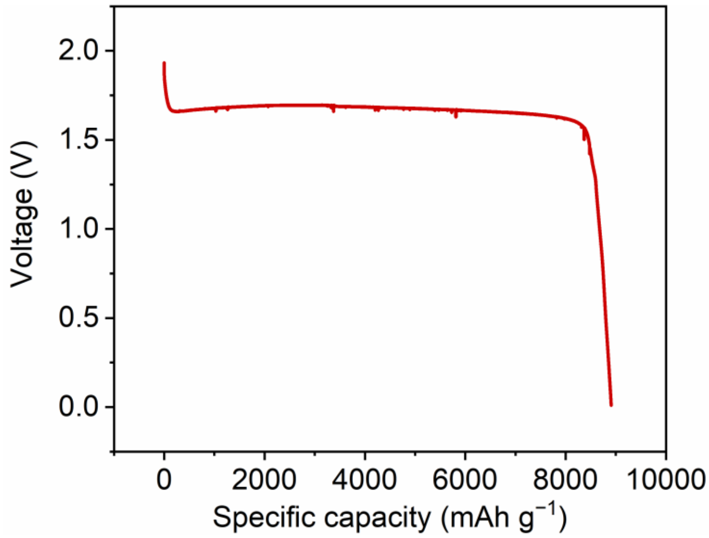

Figure S8. Galvanostatic discharge curve of the $Mg/SOCl_2$ primary battery with a cathode mass loading of 2–3 mg cm^{-2} at the current density of 50 mA g^{-1}.

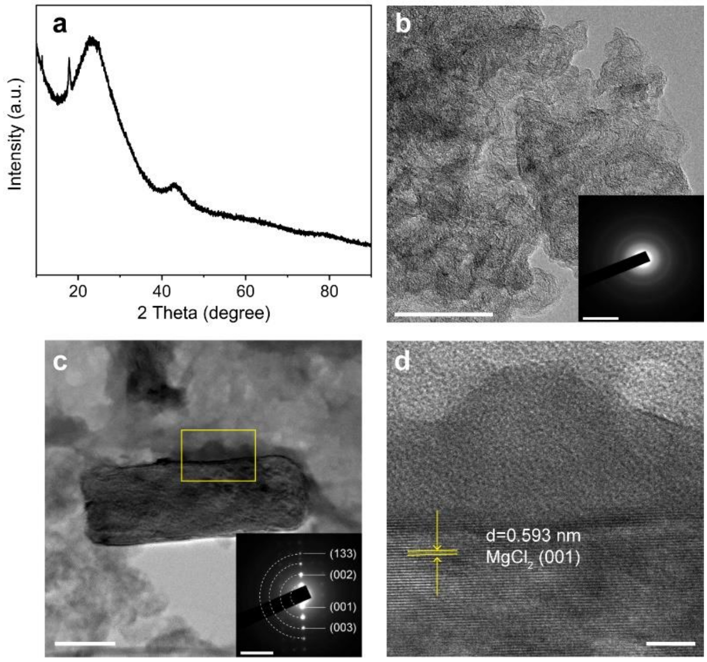

Figure S9. a, b) XRD pattern and TEM image of the KJ, respectively. Inset in **b**, SAED pattern. Scale bar in **b**, 50 nm. Inset, 1/5 nm. **c, d)** TEM and HRTEM images of the KJ after discharge. Scale in **c**, 100 nm. Inset, 1/5 nm. Scale bar in **d**, 10 nm.

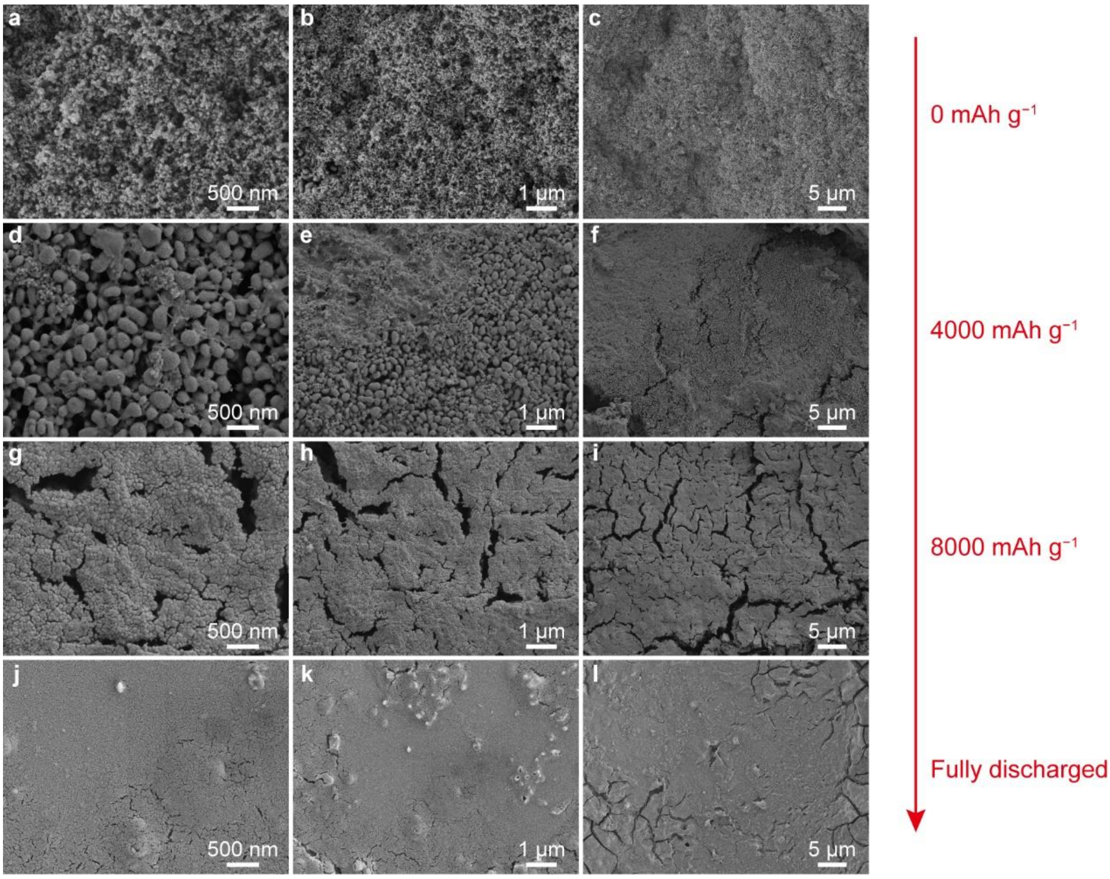

Figure S10. SEM images of the discharge products on KJ cathode at different discharge depths. The discharge current density is 100 mA g^{-1}.

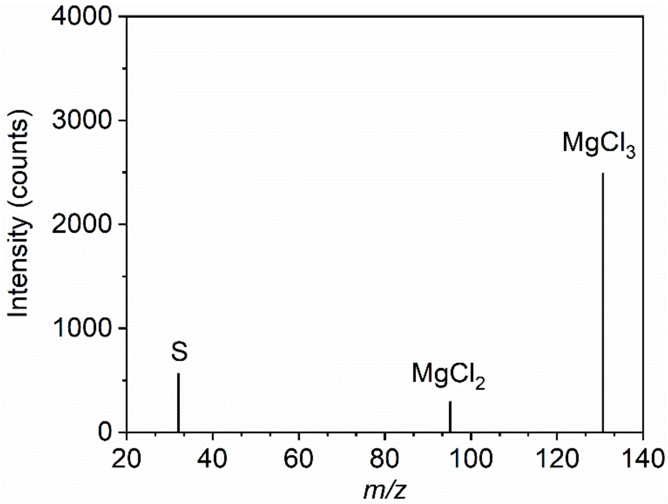

Figure S11. TOF-SIMS spectra probing the $MgCl_3$, $MgCl_2$ and S secondary ion fragments from a fully discharged KJ cathode.

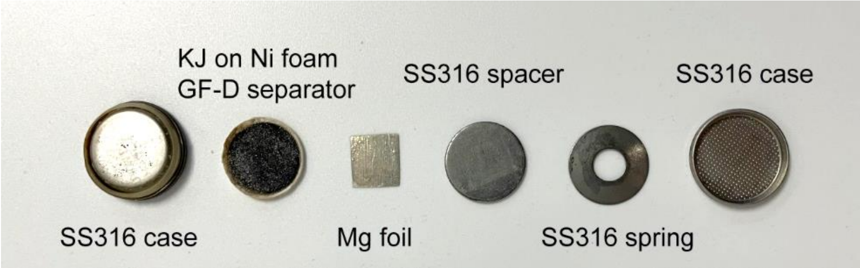

Figure S12. Photograph of the disassembled components from a fully discharged Mg/$SOCl_2$ primary battery.

Table S1. The detailed cost evaluation of the $Mg/SOCl_2$ and $Li/SOCl_2$ chemistries.

	KJ ($ kg^{-1})	Mg ($ kg^{-1})	$SOCl_2$ ($ kg^{-1})	$AlCl_3$ ($ kg^{-1})	$MgCl_2$ ($ kg^{-1})	P_{cell} ($ kWh^{-1})
$Mg/SOCl_2$	68.8	8.9	0.688	4.96	0.104	4
	AB ($ kg^{-1})	Li ($ kg^{-1})	$SOCl_2$ ($ kg^{-1})	$AlCl_3$ ($ kg^{-1})	LiCl ($ kg^{-1})	P_{cell} ($ kWh^{-1})
$Li/SOCl_2$	4.64	169.6	0.688	4.96	40.96	28

Note: The prices of these components were collected on Aladdin, Alibaba. Average bulk metal prices were obtained from the Asian Metal Online Database (www.asianmetal.cn). The prices of separators and current collectors were not included in the calculation. The abbreviations of P_{cell} refers to per individual price of battery.

Table S2. The detailed processability evaluation of the metal anodes of $Mg/SOCl_2$ and $Li/SOCl_2$ primary batteries.

	Mg metal	Li metal
Moisture resistance	Good	Poor
Thermal fatigue resistance	Good	Poor
Malleability	Good	Poor
Scalability	Good	Medium
Overall rating	Good	Poor

Table S3. Comparison of the specific surface area, pore volume (micropores and mesopores) and discharge specific capacity of different carbon materials including graphite, acetylene black (AB) and ketjenblack (KJ).

	Graphite	AB	KJ
Specific surface area ($m^2\ g^{-1}$)	4.15	85.31	1497.28
Pore volume ($cm^3\ g^{-1}$)	0.02138	0.25370	2.96151
Micropore pore volume ($cm^3\ g^{-1}$)	0.00002	0.00467	0.02199
Mesopore pore volume ($cm^3\ g^{-1}$)	0.02136	0.24903	2.93952
Average pore diameter (nm)	21.01	13.24	8.80
Discharge specific capacity (mAh g^{-1} carbon)	4252	6737	11131

Table S4. Comparison on the main parameters of different Mg-based primary batteries.

	Specific capacity (mAh g^{-1})	Average voltage (V)	Ref
Mg/Hg_2Cl_2	113	1.55	1
Mg/Pb_2Cl_2	192	0.95	1
Mg/CuCl	270	1.30	1
Mg/AgCl	183	1.55	1
Mg/AgCl	130	1.20	2
Mg/AgCl	179	1.42	3
Mg/MnO_2	768	1.44	4
Mg/MnO_2	1539	1.31	5
$Mg/SOCl_2$[a]	11131	1.62	This work
$Mg/SOCl_2$[b]	14046	1.67	This work

[a)] (Discharge current density, 100 mA g^{-1}); [b)] (Discharge current density, 50 mA g^{-1}).

Table S5. The elemental composition of the AZ91D Mg-Al alloy.

	Mg	Al	Zn	Mn	Si	Cu	Ni	Fe
AZ91D	Rest	8.5–9.5	0.45–0.9	0.17–0.4	<0.05	<0.025	<0.001	<0.004

Note: The AZ91D is a commercial Mg-Al alloy with better mechanical strengths and corrosion resistances compared with pure Mg metal.

References

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