Supplementary Material

for

Enhancing Molten Salt Oxidation Sustainability: Thermodynamic Insights for Spent Salt Reuse and Carbonate Cycle Replenishment

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Supporting Figures

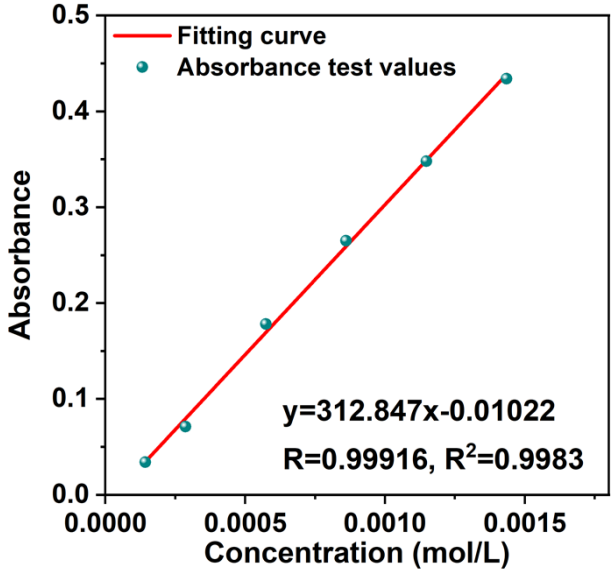

Fig. S1. The relation equation between standard solution concentrations and absorbance.

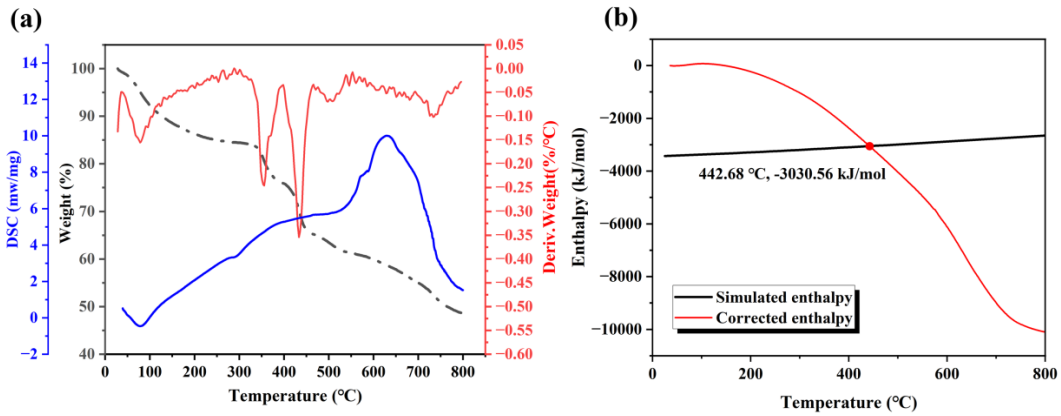

Fig. S2. (a) TG-DSC and (b) reaction enthalpy of CERs oxidized without carbonate process at 10 ℃/min rate.

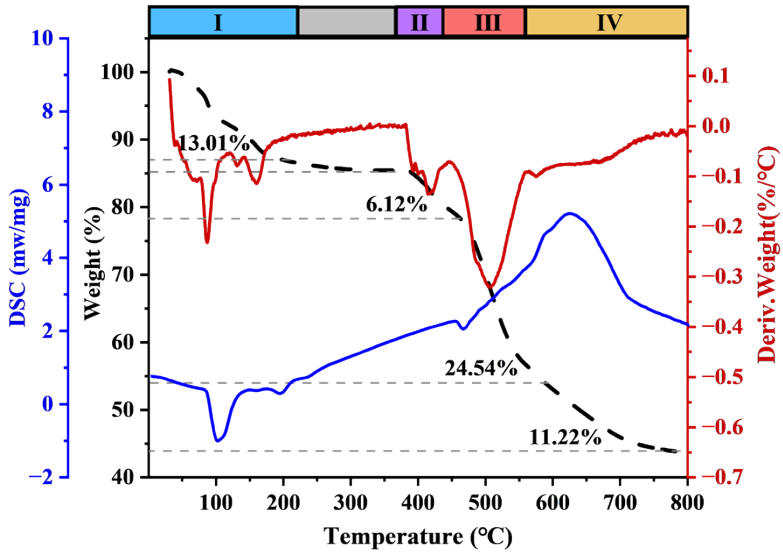

Fig. S3. TG-DSC curves and oxidation stage division of MSO process.

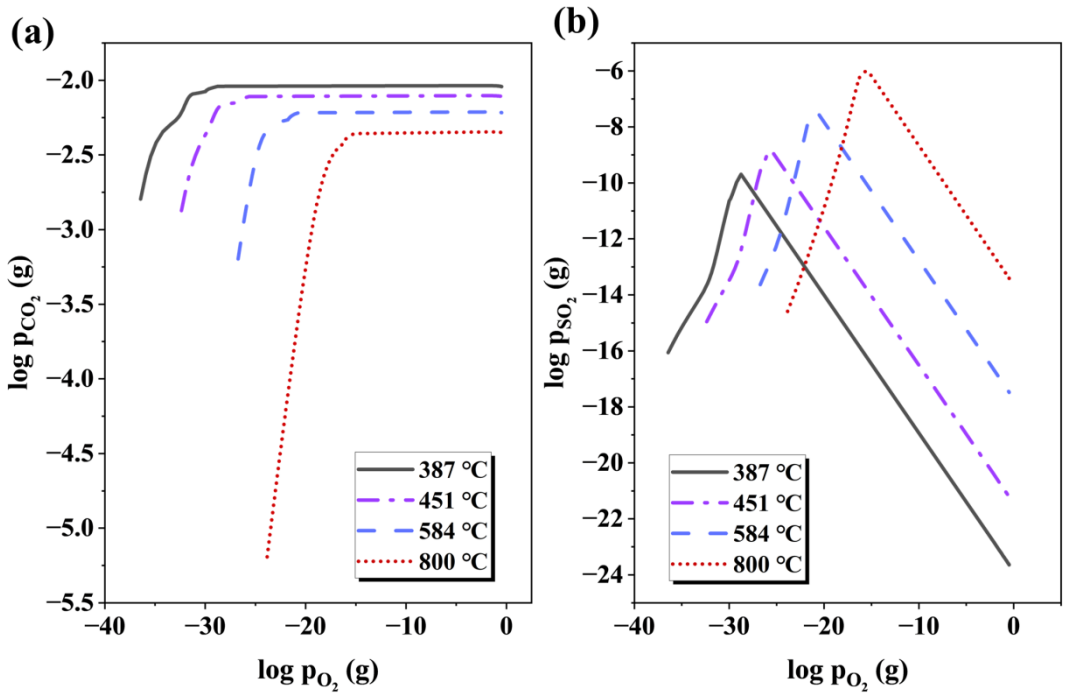

Fig. S4. Variation of CO_2 (a) and SO_2 (b) partial pressures in equilibrium calculations of MSO at different oxygen partial pressures and temperatures.

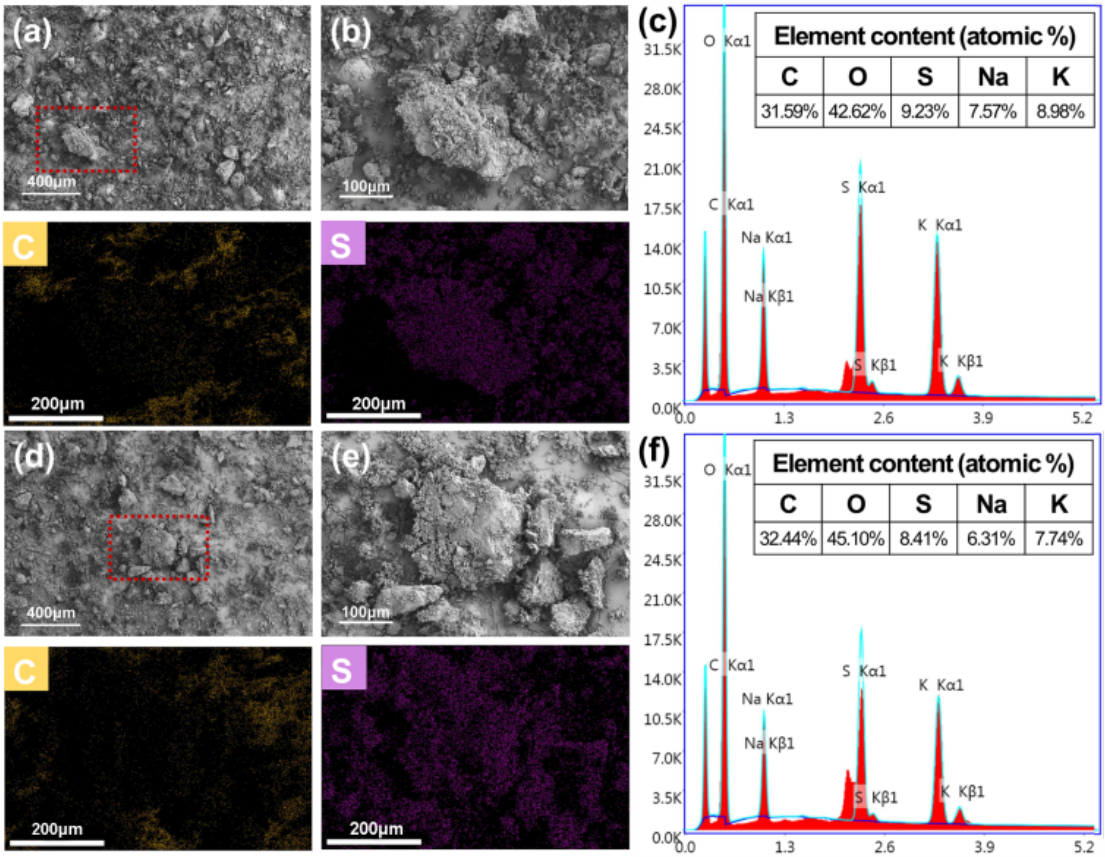

Fig. S5. SEM-mapping patterns and elemental analysis results of spent salt collected from MSO-DC (a-c) and MSO-RC (d-f).

Supporting Tables

Table S1 The input settings for the equilibrium calculations.

Species Formula	Feed Temperature℃	Activity Coefficient	Initial Amount (kmol)	51 Step Add Amount (kmol)
Initial Input Phase:				
$C_{17}H_{29}O_{14}S_2$	25	1	0.1924	0.0000
Na_2CO_3	25	1	0.3000	0.0000
Li_2CO_3	25	1	0.4400	0.0000
K_2CO_3	25	1	0.2600	0.0000
Gas Products:				
O_2 (g)	0	1	0.0000	0.1443
CO (g)	0	1	0.0000	0.0000
CO_2 (g)	0	1	0.0000	0.0000
H_2 (g)	0	1	0.0000	0.0000
SO_2 (g)	0	1	0.0000	0.0000
H_2S (g)	0	1	0.0000	0.0000
H_2O (g)	0	1	0.0000	0.0000
CH_4 (g)	0	1	0.0000	0.0000
C_2H_4 (g)	0	1	0.0000	0.0000
C_2H_6 (g)	0	1	0.0000	0.0000
Inorganic Salt Products:				
Na_2SO_4	0	1	0.0000	0.0000
K_2SO_4	0	1	0.0000	0.0000
Li_2SO_4	0	1	0.0000	0.0000
$K_3Na(SO_4)_2$	0	1	0.0000	0.0000
Na_2SO_3	0	1	0.0000	0.0000
K_2SO_3	0	1	0.0000	0.0000
Li_2SO_3	0	1	0.0000	0.0000

Table S2 Equilibrium amounts of components of the molten salt oxidation process at different temperatures when reaching 1.08 ThOD (3.8961 kmol) oxygen content.

Species Formula	Equilibrium amounts at different temperatures (kmol)				
	25 ℃	**387 ℃**	**451 ℃**	**584 ℃**	**800 ℃**
Initial Reaction Phase:					
$C_{17}H_{29}O_{14}S_2$	0.0000	0.0000	0.0000	0.0000	0.0000
Na_2CO_3	0.1752	0.1741	0.1731	0.1690	0.1635
Li_2CO_3	0.4400	0.4394	0.4388	0.4375	0.4360
K_2CO_3	0.0000	0.0016	0.0033	0.0088	0.0197
Gas Products:					
O_2 (g)	0.0519	0.0519	0.0519	0.0519	0.0519
CO (g)	0.0000	0.0000	0.0000	0.0000	0.0000
CO_2 (g)	3.6556	3.6556	3.6556	3.6556	3.6556
H_2 (g)	0.0000	0.0000	0.0000	0.0000	0.0000
SO_2 (g)	0.0000	0.0000	0.0000	0.0000	0.0000
H_2S (g)	0.0000	0.0000	0.0000	0.0000	0.0000
COS(g)	0.0000	0.0000	0.0000	0.0000	0.0000
H_2O (g)	2.7898	2.7898	2.7898	2.7898	2.7898
CH_4 (g)	0.0000	0.0000	0.0000	0.0000	0.0000
C_2H_4 (g)	0.0000	0.0000	0.0000	0.0000	0.0000
C_2H_6 (g)	0.0000	0.0000	0.0000	0.0000	0.0000
Inorganic Salt Products:					
Na_2SO_4	0.1248	0.1259	0.1269	0.1310	0.1365
K_2SO_4	0.2600	0.2584	0.2567	0.2512	0.2403
Li_2SO_4	0.0000	0.0006	0.0012	0.0025	0.0040
$K_3Na(SO_4)_2$	0.0000	0.0000	0.0000	0.0000	0.0000
Na_2SO_3	0.0000	0.0000	0.0000	0.0000	0.0000
K_2SO_3	0.0000	0.0000	0.0000	0.0000	0.0000
Li_2SO_3	0.0000	0.0000	0.0000	0.0000	0.0000

Table S3 Key parameters of unit for pyrolysis and MSO-RC processes.

Impact Category	Units	Pyrolysis Values	MSO-RC Values
Global warming	kg CO_2 eq	765	402
Stratospheric ozone depletion	kg CFC11 eq	0.000474	0.000246
Ionizing radiation	kBq Co-60 eq	1.26×10^{11}	2.80×10^{3}
Ozone formation, Human	kg NO_x eq	2.65	1.38
Fine particulate matter formation	kg PM2.5 eq	2.32	1.11
Ozone formation, Terrestrial	kg NO_x eq	2.66	1.38
Terrestrial acidification	kg SO_2 eq	6.39	2.99
Freshwater eutrophication	kg P eq	0.127	0.0687
Marine eutrophication	kg N eq	0.00805	0.00439
Terrestrial ecotoxicity	kg 1,4-DCB	362	206
Freshwater ecotoxicity	kg 1,4-DCB	6.12	3.51
Marine ecotoxicity	kg 1,4-DCB	8.58	4.89
Human carcinogenic toxicity	kg 1,4-DCB	23.4	12.5
Human non-carcinogenic toxicity	kg 1,4-DCB	142	79.1
Land use	m^2 a crop eq	9.14	4.98
Mineral resource scarcity	kg Cu eq	0.16	0.184
Fossil resource scarcity	kg oil eq	138	72.5
Water consumption	m^3	4.27	2.28