Supporting Information

Simultaneous removal of residual sulfate and heavy metals from spent electrolyte of lead-acid battery after precipitation and carbonation

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Thermodynamic modeling

The thermodynamic calculations were carried out with Matlab® R2010a software using thermodynamic parameters of all possible reactions. Due to the fact that calcium, sulfate and carbonate are all divalent ions with quite low ionic activity coefficient (γ), which are γ_{Ca2+} = 0.28 (I = 0.7, 25°C and 1 atm), γ_{SO42-} = 0.12 and γ_{CO32-} = 0.20 (I = 0.7, 25°C and 1 atm), respectively. The activity corrections are also taken into consideration in the thermodynamic modeling process. The thermodynamic parameters used in the modeling process were listed in Table S1.

Table S1. Chemicals, formulas, reactions and equilibrium constants used in the thermodynamic calculations.

Formulas	Possible Chemical Reactions		$Log_{10}(K)$[1]
H^+ / OH^-	$H_2O \leftrightarrow H^+ + OH^-$	(S1)	-13.99
$CaCO_3$	$Ca^{2+} + CO_3^{2-} \leftrightarrow CaCO_3$	(S2)	3.339
$CaHCO_3^+$	$Ca^{2+} + H^+ + CO_3^{2-} \leftrightarrow CaHCO_3^+$	(S3)	11.44
$CaOH^+$	$Ca^{2+} + H_2O \leftrightarrow H^+ + CaOH^+$	(S4)	-12.83
$CaCO_3(s)$	$Ca^{2+} + CO_3^{2-} \leftrightarrow CaCO_3(s)$	(S5)	8.3
$Ca(OH)_2(s)$	$Ca^{2+} + 2H_2O \leftrightarrow 2H^+ + Ca(OH)_2(s)$	(S6)	-22.62
$Cd(CO_3)_2^{2-}$	$Cd^{2+} + 2CO_3^{2-} \leftrightarrow Cd(CO_3)_2^{2-}$	(S7)	7.2
$CdHS^+$	$Cd^{2+} + HS^- \leftrightarrow CdHS^+$	(S8)	8.24
$Cd(HS)_2$	$Cd^{2+} + 2HS^- \leftrightarrow Cd(HS)_2$	(S9)	15.46
$Cd(HS)_3^-$	$Cd^{2+} + 3HS^- \leftrightarrow Cd(HS)_3^-$	(S10)	17.36
$Cd(HS)_4^{2-}$	$Cd^{2+} + 4HS^- \leftrightarrow Cd(HS)_4^{2-}$	(S11)	19.32
CO_2	$2H^+ + CO_3^{2-} \leftrightarrow H_2O + CO_2$	(S12)	16.68
$CO_2(g)$	$2H^+ + CO_3^{2-} \leftrightarrow H_2O + CO_2(g)$	(S13)	18.15
H_2CO_3	$2H^+ + CO_3^{2-} \leftrightarrow H_2CO_3$	(S14)	16.70
HCO_3^-	$H^+ + CO_3^{2-} \leftrightarrow HCO_3^-$	(S15)	10.33

S^{2-}	$HS^{-} \leftrightarrow H^{+} + S^{2-}$	(S16)	-12.88
H_2S	$HS^{-} + H+ \leftrightarrow H_2S$	(S17)	7.023
$H_2S(g)$	$HS^{-} + H+ \leftrightarrow H_2S(g)$	(S18)	8.018
$Pb(CO_3)_2^{2-}$	$Pb^{2+} + 2CO_3^{2-} \leftrightarrow Pb(CO_3)_2^{2-}$	(S19)	10.1
$PbCO_3$	$Pb^{2+} + CO_3^{2-} \leftrightarrow PbCO_3$	(S20)	6.6
$PbHCO_3^{+}$	$Pb^{2+} + H^{+} + CO_3^{2-} \leftrightarrow PbHCO_3^{+}$	(S21)	13.35
$Cd(OH)_2(s)$	$Cd^{2+} + 2\ H_2O \leftrightarrow 2H^{+} + Cd(OH)_2(s)$	(S22)	-13.65
$CdCO_3(s)$	$Cd^{2+} + CO_3^{2-} \leftrightarrow CdCO_3(s)$	(S23)	12.1
$CdS(s)$	$Cd^{2+} + HS^{-} \leftrightarrow H^{+} + CdS(s)$	(S24)	14.13
$Pb(OH)_2(s)$	$Pb^{2+} + 2H_2O \leftrightarrow Pb(OH)_2(s) + 2H^{+}$	(S25)	-13.6
$PbCO_3(s)$	$Pb^{2+} + CO_3^{2-} \leftrightarrow PbCO_3(s)$	(S26)	13.2
$PbS(s)$	$Pb^{2+} + HS^{-} \leftrightarrow H^{+} + PbS(s)$	(S27)	14.86
$Fe(CO_3)_2^{2-}$	$Fe^{2+} + H^{+} + CO_3^{2-} \leftrightarrow Fe(CO_3)_2^{2-}$	(S28)	20.53
$Fe(OH)_2(s)$	$Fe(OH)_2(s) \leftrightarrow Fe(OH)_2$	(S29)	-7.3
$Fe(OH)_3^{-}$	$Fe(OH)_2(s) + H_2O \leftrightarrow Fe(OH)_3^{-} + H^{+}$	(S30)	-19.9
$Fe(OH)_4^{2-}$	$Fe(OH)_2(s) + 2H_2O \leftrightarrow Fe(OH)_4^{2-} + 2H^{+}$	(S31)	-32.85
$FeHCO_3^{+}$	$3H^{+} + CO_3^{2-} + Fe(OH)_2(s) \leftrightarrow FeHCO_3^{+} + 2H_2O$	(S32)	24.93
Fe^{2+}	$2H+ + Fe(OH)_2(s) \leftrightarrow 2H_2O + Fe^{2+}$	(S33)	13.5
$FeCO_3(s)$	$2H^{+} + CO_3^{2-} + Fe(OH)_2(s) \leftrightarrow FeCO_3(s)$	(S34)	21.22
$FeS(s)$	$H^{+} + HS^{-} + Fe(OH)_2(s) \leftrightarrow FeS(s) + 2H_2O$	(S35)	16.95

[1.] *K* data obtained from HSC Chemistry® 6.0 database.

HUMAS Kit analysis

HUMAS kits were utilized to analysis the concentrations of SO_4^{2-} (HS-SO4-H, Cat. No. 08014) and S^{2-} (HS-S, Cat. No. 37010) in the aqueous solution. The sample solutions were first diluted and then injected into the tube of corresponding kit to measure the concentrations of SO_4^{2-} and S^{2-} in each step.

Raman spectroscopy analysis

The Raman spectroscopy analysis was utilized to analyze the symmetric stretching mode of carbonate ions in the aqueous solution. The sample was directly obtained from the solution after SRB treatment and precipitation process, and filled into a quartz cell to be analyzed on the universal platform sampling accessory of Thermo Scientific DXR SmartRaman Spectrometer, with 532nm laser and filter, the characteristic wavenumber of carbonate ions were selected at 1066cm^{-1}, and the range of the wavenumber region is 500-4000cm^{-1}.