

Supporting Information 1:Parameters

The simulation of interface characteristics and charge transfer dynamics for layered electrodes using cascade capacitance in supercapacitors by COMSOL software

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1.Parameters of activated carbon as negative electrode

Property	Variable	Value	Unit	Property group
Electrical conductivity	σ_{iso} ; $\sigma_{mii} = \sigma_{iso}$, $\sigma_{mij} = 0$	32.4[S/m]	S/m	Basic
Electrolyte conductivity	$\sigma_{mal,iso}$; $\sigma_{mal,ii} = \sigma_{mal,iso}$, $\sigma_{mal,ij} = 0$	80[S/m]	S/m	Electrolyte conductivity
Diffusion coefficient	D_{iso} ; $D_{ii} = D_{iso}$, $D_{ij} = 0$	$3.9e-14[m^2/s]$	m^2/s	Basic
Density	ρ	2260[kg/m ³]	kg/m ³	Basic
Equilibrium potential	Eeq	Eeq_int1(soc)	V	Equilibrium potential
Temperature derivative of equilibrium potential	dEeqdT	0[V/K]	V/K	Equilibrium potential
Reference concentration	cEeqref	def.csmax	mol/m ³	Equilibrium potential
Maximum electrode state of charge	socmax	elpot.Eeq_inv(E_min)	1	Operational electrode state of charge
Minimum electrode state of charge	socmin	elpot.Eeq_inv(E_max)	1	Operational electrode state of charge
Maximum operational potential	E_max	4.2[V]	V	Operational electrode state of charge
Minimum operational potential	E_min	0[V]	V	Operational electrode state of charge
Equilibrium concentration	csEq	def.csmax*elpot.Eeq_inv(V)	mol/m ³	Equilibrium concentration

2.Parameters of KOH solution as electrolyte

Property	Variable	Value	Unit	Property group
Electrolyte conductivity	$\sigma_{mal,iso}$; $\sigma_{mal,ii} = \sigma_{mal,iso}$, $\sigma_{mal,ij} = 0$	$(A*M+B*M^2+C*M*T_K+D*M/T_K+E*M^3+F*M^2*T_K^2)*1[S/cm]$	S/m	Electrolyte conductivity
Diffusion coefficient	D_{iso} ; $D_{ii} = D_{iso}$, $D_{ij} = 0$	$3.75e-9[m^2/s]$	m^2/s	Basic
Density	ρ	$(A_{rho}(T_{degC})^2+B_{rho}(T_{degC})^3+C_{rho}(T_{degC}))*1[kg/m^3]$	kg/m ³	Basic
Transport number	transpNum	0.22	1	Species properties
Activity dependence	fcl	2	1	Species properties

3.Parameters of graphene oxide as positive electrode

Property	Variable	Value	Unit	Property group
Electrical conductivity	σ_{iso} ; $\sigma_{mii} = \sigma_{iso}$, $\sigma_{mij} = 0$	1000[S/m]	S/m	Basic
Electrolyte conductivity	$\sigma_{mal,iso}$; $\sigma_{mal,ii} = \sigma_{mal,iso}$, $\sigma_{mal,ij} = 0$	100[S/m]	S/m	Electrolyte conductivity
Young's modulus	E	E_int(c/csmax)	Pa	Basic
Poisson's ratio	ν	$\nu_{int}(c/csmax)$	1	Basic
Diffusion coefficient	D_{iso} ; $D_{ii} = D_{iso}$, $D_{ij} = 0$	$1.4523e-13*exp(68025.7/8.314*(1/(T_{ref}/1[K])-1/(T/1[K]))) [m^2/s]$	m^2/s	Basic
Thermal conductivity	k_{iso} ; $k_{ii} = k_{iso}$, $k_{ij} = 0$	1[W/(m*K)]	W/(m*K)	Basic
Heat capacity at constant pressure	Cp	750[J/(kg*K)]	J/(kg*K)	Basic
Density	ρ	2300[kg/m ³]	kg/m ³	Basic
Equilibrium potential	Eeq	Eeq_int1(soc)+dEeqdT_int1(soc)*(T-298[K])	V	Equilibrium potential
Temperature derivative of equilibrium potential	dEeqdT	dEeqdT_int1(soc)	V/K	Equilibrium potential
Reference concentration	cEeqref	def.csmax	mol/m ³	Equilibrium potential
Maximum electrode state of charge	socmax	elpot.Eeq_inv(E_min)	1	Operational electrode state of charge
Minimum electrode state of charge	socmin	elpot.Eeq_inv(E_max)	1	Operational electrode state of charge
Maximum operational potential	E_max	4.2[V]	V	Operational electrode state of charge
Minimum operational potential	E_min	0[V]	V	Operational electrode state of charge
Volumetric strain	dvolt	dvoltDSOL(c/def.csmax)	1	Intercalation strain
Equilibrium concentration	csEq	def.csmax*elpot.Eeq_inv(V)	mol/m ³	Equilibrium concentration

4.Parameters of ZnMn₂O₄ as positive electrode

Property	Variable	Value	Unit	Property group
<input checked="" type="checkbox"/> Electrical conductivity	σ_{iso} ; $\sigma_{\text{mai}} = \sigma_{\text{iso}}$, $\sigma_{\text{maj}} = 0$	100[S/m]	S/m	Basic
<input checked="" type="checkbox"/> Electrolyte conductivity	$\sigma_{\text{mal}}_{\text{iso}}$; $\sigma_{\text{mal}}_{\text{ii}} = \sigma_{\text{mal}}_{\text{iso}}$, $\sigma_{\text{mal}}_{\text{ij}} = 0$	100[S/m]	S/m	Electrolyte conductivity
Young's modulus	E	$E_{\text{int}}(c/c_{\text{smax}})$	Pa	Basic
Poisson's ratio	ν	$\nu_{\text{u_int}}(c/c_{\text{smax}})$	1	Basic
Diffusion coefficient	D_{iso} ; $D_{\text{ii}} = D_{\text{iso}}$, $D_{\text{ij}} = 0$	$1.4523 \times 10^{-13} \exp(68025.7/8.314 \cdot (1/(T_{\text{ref}}/1[\text{K}]) - 1/(T/1[\text{K}]))) [\text{m}^2/\text{s}]$	m^2/s	Basic
Thermal conductivity	k_{iso} ; $k_{\text{ii}} = k_{\text{iso}}$, $k_{\text{ij}} = 0$	$1 [\text{W}/(\text{m} \cdot \text{K})]$	$\text{W}/(\text{m} \cdot \text{K})$	Basic
Heat capacity at constant pressure	C_p	$750 [\text{J}/(\text{kg} \cdot \text{K})]$	$\text{J}/(\text{kg} \cdot \text{K})$	Basic
Density	ρ	$2300 [\text{kg}/\text{m}^3]$	kg/m^3	Basic
Equilibrium potential	Eeq	$E_{\text{eq_int}}(s_{\text{oc}}) + dE_{\text{eqdT_int}}(s_{\text{oc}}) \cdot (T - 298 [\text{K}])$	V	Equilibrium potential
Temperature derivative of equilibrium potential	dEeqdT	$dE_{\text{eqdT_int}}(s_{\text{oc}})$	V/K	Equilibrium potential
Reference concentration	cEeqref	def.csmax	mol/m^3	Equilibrium potential
Maximum electrode state of charge	socmax	elpot.Eeq_inv(E_min)	1	Operational electrode state of charge
Minimum electrode state of charge	socmin	elpot.Eeq_inv(E_max)	1	Operational electrode state of charge
Maximum operational potential	E_max	4.2[V]	V	Operational electrode state of charge
Minimum operational potential	E_min	0[V]	V	Operational electrode state of charge
Volumetric strain	dvol	$d\text{VOLdSOL}(c/\text{def.csmax})$	1	Intercalation strain
Equilibrium concentration	csEq	$\text{def.csmax} \cdot \text{elpot.Eeq_inv}(V)$	mol/m^3	Equilibrium concentration

5.Parameters of cyclic voltammetry test

Name	Expression	Value	Description
V_min	0[V]	0 V	
V_max	1.6[V]	1.6 V	
v	0.05[V/s]	0.05 V/s	Scanning rate
tH	$(V_{\text{max}} - V_{\text{min}})/v$	32 s	half-period

6. Electrolyte parameters preset by COMSOL software

Name	Expression	Unit	Description	Selection	Details
cd.sigmalrr	ionc.sigmal11	S/m	Electrolyte conductivity, rr-component	Domain 2	Meta
cd.sigmalphir	ionc.sigmal21	S/m	Electrolyte conductivity, phir-component	Domain 2	Meta
cd.sigmalzr	ionc.sigmal31	S/m	Electrolyte conductivity, zr-component	Domain 2	Meta
cd.sigmalrphi	ionc.sigmal12	S/m	Electrolyte conductivity, rphi-component	Domain 2	Meta
cd.sigmalphiphi	ionc.sigmal22	S/m	Electrolyte conductivity, phiphi-compon...	Domain 2	Meta
cd.sigmalzphi	ionc.sigmal32	S/m	Electrolyte conductivity, zphi-component	Domain 2	Meta
cd.sigmalrz	ionc.sigmal13	S/m	Electrolyte conductivity, rz-component	Domain 2	Meta
cd.sigmalphiz	ionc.sigmal23	S/m	Electrolyte conductivity, phiz-component	Domain 2	Meta
cd.sigmalzz	ionc.sigmal33	S/m	Electrolyte conductivity, zz-component	Domain 2	Meta
cd.Qh	-cd.ilr*phir-cd.ilz*philz	W/m ³	Total power dissipation density	Domain 2	+ operation
cd.lIMag	sqrt(realdot(cd.ilr,cd.ilr)+realdot(cd.ilphi,cd.ilphi)+realdot(cd.ilz,cd.ilz))	A/m ²	Electrolyte current density magnitude	Domain 2	
cd.Qli	0	A/m ³	Current source	Domain 2	+ operation
cd.tEr	-philTr	V/m	Tangential electric field, r-component	Boundaries 4–7	
cd.tEphi	0	V/m	Tangential electric field, phi-component	Boundaries 4–7	
cd.tEz	-philTz	V/m	Tangential electric field, z-component	Boundaries 4–7	
cd.Er	-phir	V/m	Electric field, r-component	Domain 2	
cd.Ephi	0	V/m	Electric field, phi-component	Domain 2	
cd.Ez	-philz	V/m	Electric field, z-component	Domain 2	
cd.ilr	-cd.sigmalrr*phir-cd.sigmalrz*philz	A/m ²	Electrolyte current density, r-component	Domain 2	+ operation
cd.ilphi	-cd.sigmalphir*phir-cd.sigmalphiz*philz	A/m ²	Electrolyte current density, phi-compone...	Domain 2	+ operation
cd.ilz	-cd.sigmalzr*phir-cd.sigmalzz*philz	A/m ²	Electrolyte current density, z-component	Domain 2	+ operation
cd.ilr	cd.ilr	A/m ²	Electrolyte current density vector, r-com...	Domain 2	
cd.ilphi	cd.ilphi	A/m ²	Electrolyte current density vector, phi-co...	Domain 2	
cd.ilz	cd.ilz	A/m ²	Electrolyte current density vector, z-com...	Domain 2	
cd.phil	phil	V	Electrolyte potential	Domain 2	
cd.sigmaleffrr	cd.sigmalrr	S/m	Electrolyte conductivity, rr-component	Domain 2	
cd.sigmaleffphir	cd.sigmalphir	S/m	Electrolyte conductivity, phir-component	Domain 2	
cd.sigmaleffzr	cd.sigmalzr	S/m	Electrolyte conductivity, zr-component	Domain 2	

7. Electrode parameters preset by COMSOL software

Name	Expression	Unit	Description
cd.ivtot	cd.ivdl	A/m ³	Electrode reaction source
cd.av_dl_pce1_pdl1	1000000[1/m]	1/m	Double-layer area
cd.idl	0	A/m ²	Double-layer current density
cd.ivdl	cd.idl*cd.av_dl_pce1_pdl1	A/m ³	Double-layer current source
cd.itot	cd.idl	A/m ²	Total interface current density

8.Second current distribution parameters preset by COMSOL

software

Name	Expression	Unit	Description
domflux.philr	$2 \cdot cd.llr \cdot \pi \cdot r \cdot cd.d$	A/m	Domain flux, r-component
domflux.philz	$2 \cdot cd.llz \cdot \pi \cdot r \cdot cd.d$	A/m	Domain flux, z-component
domflux.phisr	$2 \cdot cd.lsr \cdot \pi \cdot r \cdot cd.d$	A/m	Domain flux, r-component
domflux.phisz	$2 \cdot cd.lsz \cdot \pi \cdot r \cdot cd.d$	A/m	Domain flux, z-component
cd.d	1	1	Out-of-plane geometry extension
cd.bndflux_phil	$\text{if}(r > 0.001/\sqrt{\text{mean}(\text{emetric2})}), -0.5 \cdot dflux_spatial(\text{phil})/(\pi \cdot r), \text{NaN}$	A/m ²	Boundary flux
cd.bndflux_phil	$\text{if}(r > 0.001/\sqrt{\text{mean}(\text{emetric2})}), 0.25 \cdot (uflux_spatial(\text{phil}) - dflux_spatial(\text{phil}))/(\pi \cdot r), \text{NaN}$	A/m ²	Boundary flux
cd.nll	$cd.bndflux_phil/cd.d$	A/m ²	Normal electrolyte current density
cd.nR	nR	1	Normal vector, R-component
cd.nPHI	0	1	Normal vector, PHI-component
cd.nZ	nZ	1	Normal vector, Z-component
cd.nR	dnR	1	Normal vector, R-component
cd.nPHI	0	1	Normal vector, PHI-component
cd.nZ	dnZ	1	Normal vector, Z-component
cd.bndflux_phis	$\text{if}(r > 0.001/\sqrt{\text{mean}(\text{emetric2})}), -0.5 \cdot dflux_spatial(\text{phis})/(\pi \cdot r), \text{NaN}$	A/m ²	Boundary flux
cd.bndflux_phis	$\text{if}(r > 0.001/\sqrt{\text{mean}(\text{emetric2})}), -0.5 \cdot uflux_spatial(\text{phis})/(\pi \cdot r), \text{NaN}$	A/m ²	Boundary flux
cd.bndflux_phis	$\text{if}(r > 0.001/\sqrt{\text{mean}(\text{emetric2})}), 0.25 \cdot (uflux_spatial(\text{phis}) - dflux_spatial(\text{phis}))/(\pi \cdot r), \text{NaN}$	A/m ²	Boundary flux
cd.nls	$cd.bndflux_phis/cd.d$	A/m ²	Normal electrode current density
cd.nil	0	A/m ²	Inward electrolyte current density
cd.nis	0	A/m ²	Inward electrode current density
cd.Qsi	0	A/m ³	Current source
cd.mulstopcond	1	1	Multiplicative stop condition
cd.stopcond	cd.mulstopcond	1	Solver stop condition
cd.Ect	NaN	V	Electrode potential
cd.nr	dnr		Normal vector, r-component
cd.nphi	0		Normal vector, phi-component
cd.nz	dnz		Normal vector, z-component
cd.nr	dnr		Normal vector, r-component
cd.nphi	0		Normal vector, phi-component
cd.nz	dnz		Normal vector, z-component
cd.nr	dnr		Normal vector, r-component