Supporting Information

Molecularly Engineered Cellulose: the Next-Generation Sustainable Polymer Electrolyte Materials

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Table S1. Performance comparison between inorganic electrolytes, conventional polymer electrolytes, and cellulose-mediated electrolyte materials.

Property	**Inorganic electrolyte materials**	**Polymer electrolyte materials**	**Cellulose-mediated electrolyte materials**
Ionic Conductivity	Moderate to high (~10^{-4} to 10^{-3} S cm^{-1})	Low to moderate (~10^{-8} to 10^{-4} S cm^{-1})	Moderate to high (~10^{-4} to 10^{-2} S cm^{-1})
Electrochemical Stability	High	Moderate	Moderate
Thermal Stability	Very high	Moderate	Moderate
Processability	High	Low to moderate	Moderate to high
Mechanical Stability	Hard, brittle, and poor flexibility	Moderate	Moderate to high
Interfacial contact	Poor interfacial contact with electrodes	Good interfacial contact	Good interfacial contact

Table S2. Comparison of properties between cellulose-mediated PEM and other electrolyte materials.

Device	Battery type	Ionic conductivity ($mS\ cm^{-1}$)	Tensile strength	Ion transfer number	Capacitance retention rate	Ref.
$LiFeO_4$//CNF//Li	Li-metal battery	1.50	29.2	0.78	94% after 200 cycles	1
$LiFeO_4$//PP//Li		1.22	-	0.39	84% after 200 cycles	2
$Na_2Fe_2(SO_4)_3$//CNC-CNF//Na	Na-ion battery	2.32	-	0.86	76.6% after 50 cycles	3
$Na_3V_2(PO_4)_3$//HSE//Na		2.25	-	-	-	4
Graphite//BC/K	K-ion/metal battery	0.72	2.22	0.69	96% after 200 cycles	5
Graphite//GF/K		0.70	0.85	0.34	88% after 200 cycles	5
rGO//PAM/LiCl/WSCA//rGO	Supercapacitor	179.0	0.34	0.28	91% after 1000 cycles	6
AC//PNA/SA/PEG//AC		-	-	-	81.9% after 1000 cycles	6

Table S3. The properties of cellulose-based PEMs prepared using molecular engineering strategies.

PEMs	Cellulose materials	Molecular engineering strategy.	Functional components	Electrolyte uptake (%)	Tensile strength	Ionic conductivity (mS/cm)	Oxidative/thermal stability	Scalability	Ref.
CEQC	Cellulose	Etherification/Addition reaction	$-(CH_3)_3N^+$/-COO^-	-	-	31	-	-	7
Zn-CCNF@XG	Cellulose	Oxidation reaction	$-COO^-$	-	60 MPa	11.7	235 ℃	10 cm × 10 cm	8
COOH-f-Cell	Cellulose	Oxidation reaction	$-COO^-$	83.3	220 kPa	10.6	-	-	9
SBC	Cellulose	Oxidation + Sulfonation reaction	$-SO_3^-$	-	-	1.6	-	5 cm × 5 cm	10
PCS	Cellulose	Phosphorization reaction	$-PO_3^{2-}$			0.76	-	2000 g	11
$CF-SO_3$	Cellulose	Sulfonation reaction	$-SO_3^-$	685	18.7 MPa	52.1	-	-	12
CP	Cellulose	Esterification reaction	$-COO^-$	-	12 MPa	1.09	274 ℃	20 cm × 20 cm	13
C-CLA	Cellulose	Esterification reaction	$-COO^-$	-	1.4 MPa	1.1	250 ℃	-	14
CLA-x-LATP	Cellulose acetate	Condensation reaction	$-COO^-$, $-CF_3$, -CN			1.25	No phase transition temperature	-	15
Li-Cu-CNF	Cellulose nanofiber	Ion corsslinking	Cu^{2+}		30 MPa	1.5	-	1 m × 20 cm	1
Fe^{3+}-PAA/CNF	Cellulose nanofiber	Ion corsslinking	Fe^{3+}	-	45 kPa	32	-	-	16
PLCZ	Cellulose nanofiber	Ion corsslinking	Sodium polyacrylate	250	-	40	-	10 cm × 10 cm	17
CNF/CMC	Nanocellulose-carboxymethylcellulose	Molecule bonding	$-COO^-$	194	70 MPa	26	-	-	18

Table S4. Summary of the properties of cellulose-mediated PEMs.

PEMs	Cellulose materials	Preparation method	Porosity (%)	Electrolyte uptake (%)	Tensile strength (MPa)	Ionic conductivity (mS/cm)	Ion transference number	Flame retardance	Cycling performance	Ref.
PEO	-	Casting	-	-	-	0.02	0.18	-	50 cycles at 2C	19
PP/Cotton	Cotton pulp	Vacuum filtration	65	180	1.6	1.76	-	-	92% after 100 cycles at 1C	20
TOCN	Pulp	Vacuum filtration	62	200	-	1.06	-	-	94.5% after 100 cycles at 1C	21
ECM	Pulp	Vacuum filtration	53	210	65	0.35	-	-	125 mAh/g after 100 cycles at 0.5C	22
CNFs/PSA	Pulp	Vacuum filtration	61.77	294	43	0.68	-	-	78.8% after 100 cycles at 1C	23
TOBC	Bacterial cellulose	Vacuum filtration	91.1	339	97	13.49	-	-	94% after 100 cycles at 0.2C	24
BC/Li-FR	Bacterial cellulose	Casting	52	265	19.8	4.69	0.75	Yes	94.7% after 95 cycles at 0.2C	25
BC/Al_2O_3	Bacterial cellulose	Hot-pressing	74.7	625	140	4.91	-	-	89% after 50 cycles at 0.5C	26
ZIF-8@BC/ANFs	Bacterial cellulose	Vacuum filtration	73.2	340.5	70.7	1.6	-	-	91.7% after 100 cycles at 0.5C	27
BC/PPS	Bacterial cellulose	Vacuum filtration	62.7	216.2	-	1.55	-	-	91.3% after 100 cycles at 0.2C	28
BA@ATP	Bacterial cellulose	Vacuum filtration	58	470	3.71	1.73	-	Yes	94.6% after 300 cycles at 1C	29
BPM-QSPE	Bacterial cellulose	Vacuum filtration	-	-	-	0.41	0.68	-	70.1% after 420 cycles at 1C	30

Al_2O_3@BC	Bacterial cellulose	Casting	-	-	-	0.46	0.52		84% after 120 cycles at 0.2C	31
PEO/PAM-BC	Bacterial cellulose	Casting	-	-	15.8	418	0.65	-	85.4% after 500 cycles at 0.1C	32
LDR	Bacterial cellulose	Casting	-	-	2.0	135	0.46	-	86.7% after 250 cycles at 1C	33
PVDF-CA/$Al(OH)_3$	Cellulose acetate	Casting	68.6	403.9	-	2.85	-	Yes	151.97 mAh/g after 100 cycles at 1C	34
PAN/CA/AlOOH	Cellulose acetate	Electrospinning	82	608	-	1.69	-	-	99.3% after 100 cycles at 0.2C	35
CA/PVDF-HFP	Cellulose acetate	Phase inversion	85.3	310	1.44	1.89	0.89	-	136 mAh/g at 8C	36
CPM	Cellulose acetate	Casting	86	403	-	1.68	-	-	97.3% after 100 cycles at 1C	37
NCA	Cellulose acetate	Electrospinning	-	586	13.84	0.81	0.81	-	96.3% after 1600 cycles at 10C	38
PVDF/TPP/CA	Cellulose acetate	Electrospinning	90	301	8.5	4.4	-	-	86.9% after 100 cycles at 0.2C	39
CA/PU-MOFs	Cellulose acetate	Electrospinning	38.7	522.1	82.6	0.48	0.71	-	78.6% after 900 cycles at 1C	40
Cellulose/PI-COOH	Cellulose acetate	Electrospinning	78	638	34.2	0.51	-	Yes	97% after 150 cycles at 1C	41
CQSE	Cellulose triacetate	Casting	-	-	7.4	1.8	0.43	Yes	92% after 260 cycles at 2C	42
NCSP	Cellulose fibers	Vacuum filtration	61.5	-	45.3	1.08	-	-	91.3% after 400 cycles at 1C	43
TOCF	Cellulose fiber	Vacuum filtration	64.8	323	75	2.83	-	-	92.3% after 100 cycles at 0.2C	44

Cel/G-gradient	Cellulose fibers	Casting	70	600	6.25	4.5	0.57	-	108.5 mAh/g after 1000 cycles	45
SCEs	Cellulose fibers	Casting	-	-	19.8	1.21	0.67	-	97.5% after 200 cycles at 0.1C	46
ANF-CP	Cellulose fibers	Vacuum filtration	-	238	23	2.74	-	-	92.6% after 500 cycles at 3C	47
TOCN-COOH	Cellulose fibers	Vacuum filtration	75.3	240	-	2.7	-	-	122 mAh/g at 0.5C	48
CTNF	Cellulose fibers	Vacuum filtration	-	-	75	7.43	0.59	-	90.4% after 10000 cycles at 1C	49
CF@HAP	Cellulose fibers	Template-assisted growth	-	-	9.1	1.47	0.63	Yes	94% after 500 cycles at 2C	50
CNF-XNW	Cellulose fibers	Vacuum filtration		450	53	3.7	0.72	-	90% after 300 cycles at 1C	51
DCZ-gel	Cellulose fibers	Casting	-	-	2.08	38.6	0.73	-	99.4% after 2000 cycles at 1C	52
PLCZ	Cellulose fibers	Casting	-	243	-	263	0.89		192.9 mAh/g after 1000 cycles at 1C	17
CHMOP-Li	Cellulose fibers	Casting	-	-	6.33	1.02	0.75	-	91.5% after 200 cycles at 0.5C	53
BGPE	Carboxymethyl cellulose	Casting	1.01	-	24.2	1.26	0.13	-	70% after 750 cycles at 1C	54
SA/CMC/THL	Carboxymethyl cellulose	Casting	-	-	2.1	22.8	0.7	-	99.42% after 1200 cycles at 2C	55
CMC/GO	Carboxymethyl cellulose	Casting	-	-		20.5	0.58	-	86% after 2000 cycles at 0.5C	56
Ani-hydrogel	Carboxymethyl cellulose	Casting	-	-	0.65	27.35	0.42	-	57.9 mAh/g after 1000 cycles at 1C	57

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