Supporting Information

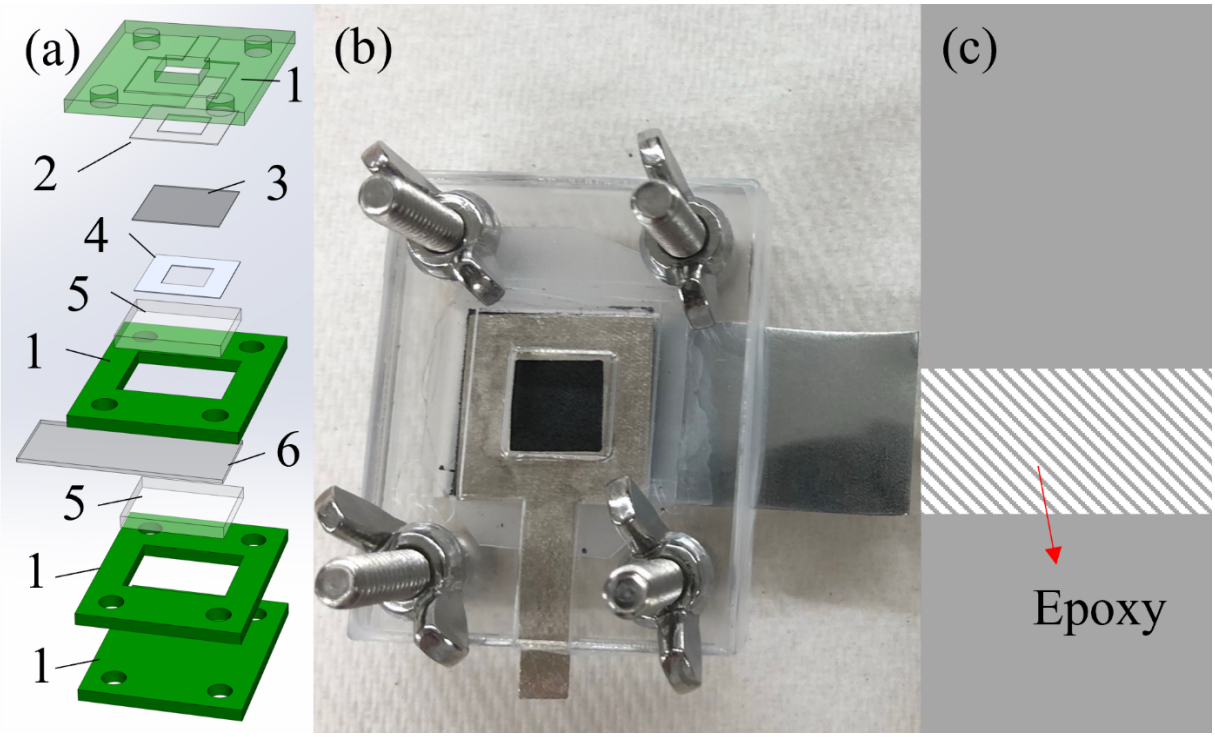

Figure S1. (a) Schematic of the sandwich-type cell in a disassembled view. (b) Optical image of the sandwich-type cell in an assembled view. (c) Illustration of Zn foil coated with epoxy. Note that the numbers correspond to the following: 1) Acrylic sheets, 2) Ni current collector, 3) GDL or carbon cloth coated with electrocatalyst, 4) polypropylene film, 5) GPE, and 6) Zn foil.

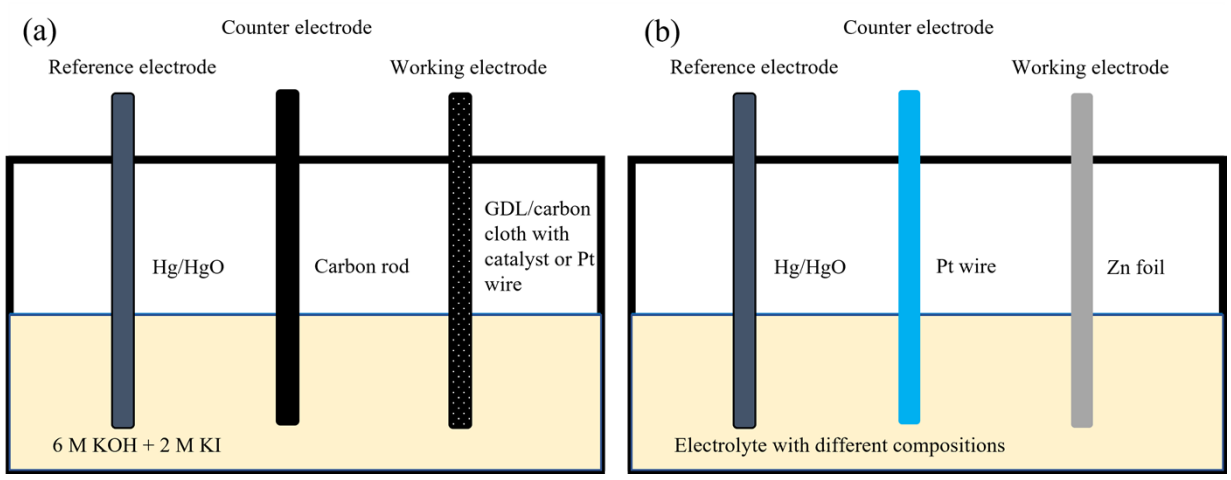

Figure S2. (a) Three-electrode setup for CV tests at the air electrode. (b) Three-electrode setup for CV tests at the Zn electrode.

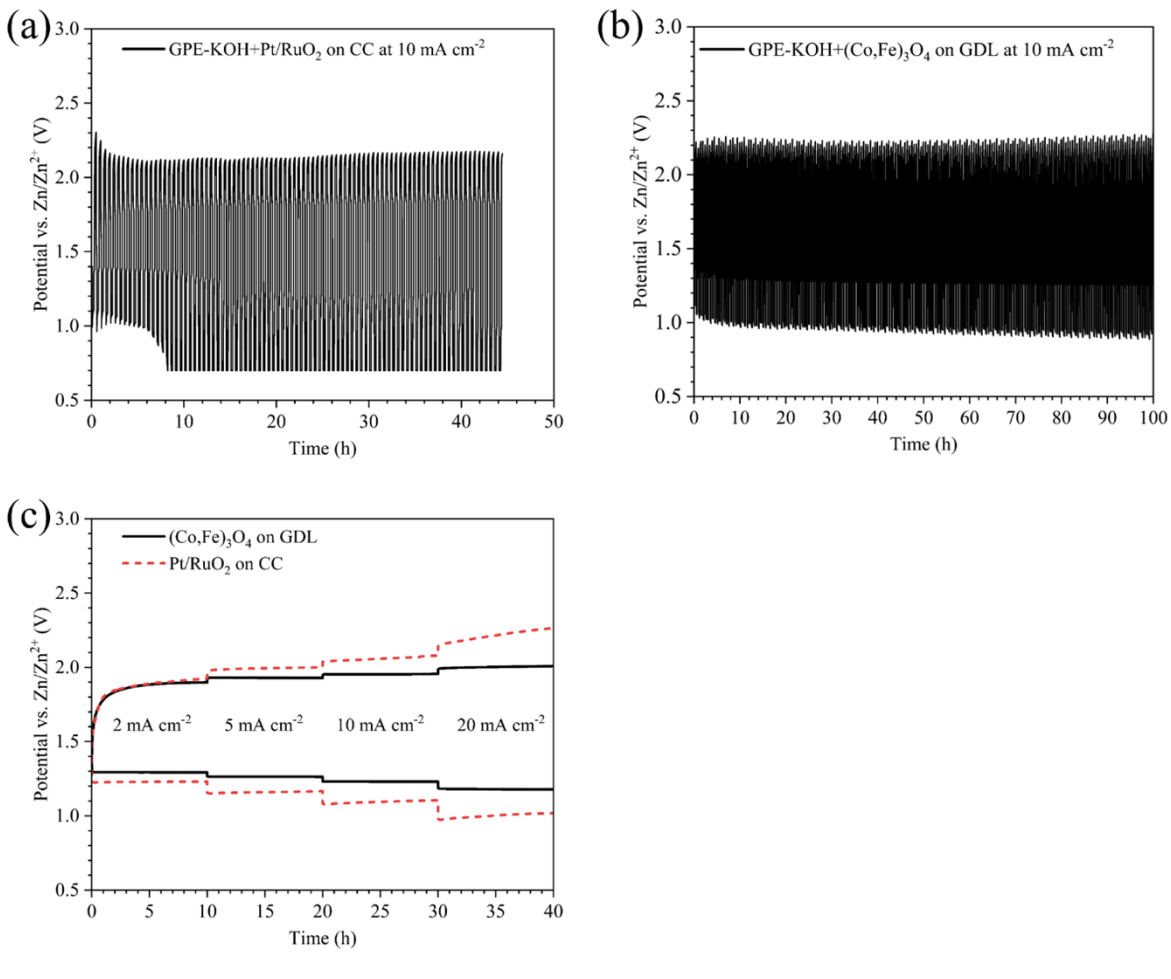

Figure S3. (a) Cyclability test at 21℃ and 10 mA cm^{-2} for ZAB using GPE-KOH and Pt/RuO_2 coated CC as the air electrode. (b) Cyclability test at 21℃ and 10 mA cm^{-2} for ZAB using GPE-KOH and $(Co,Fe)_3O_4$ coated GDL. (c) Rate test results for ZABs using GPE-KOH with $(Co,Fe)_3O_4$ on GDL and Pt/RuO_2 on CC as the air electrode at 21℃.

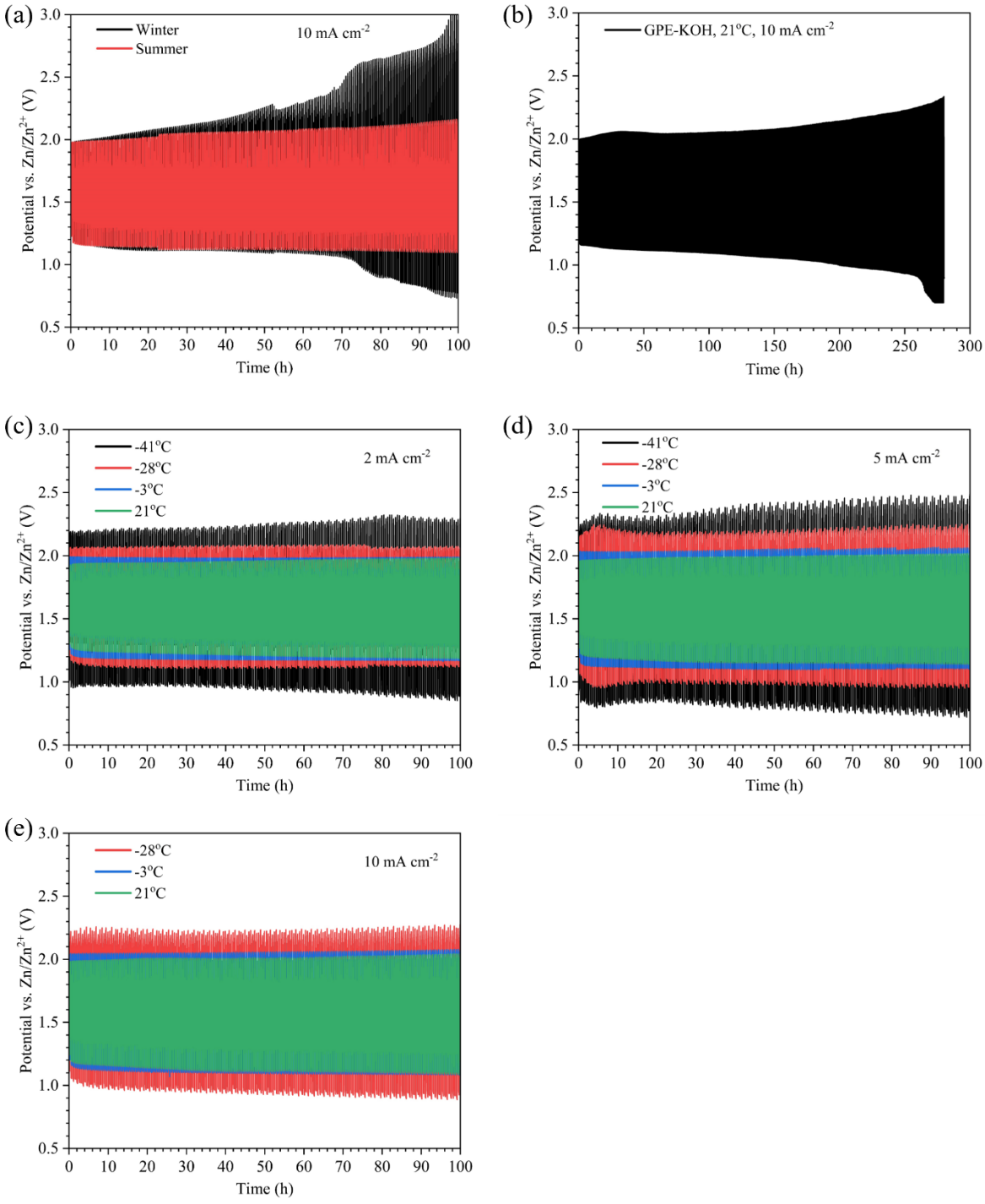

Figure S4. (a) Cyclability test at 21℃ and 10 mA cm^{-2} for ZAB using GPE-KOH in different seasons. (b) Full view of the lifetime test for ZAB using GPE-KOH. (c) Cyclability tests at different temperatures for ZABs using GPE-KOH and $(Co,Fe)_3O_4$ coated GDL at (c) 2 mA cm^{-2}, (d) 5 mA cm^{-2}, (e) 10 mA cm^{-2}.

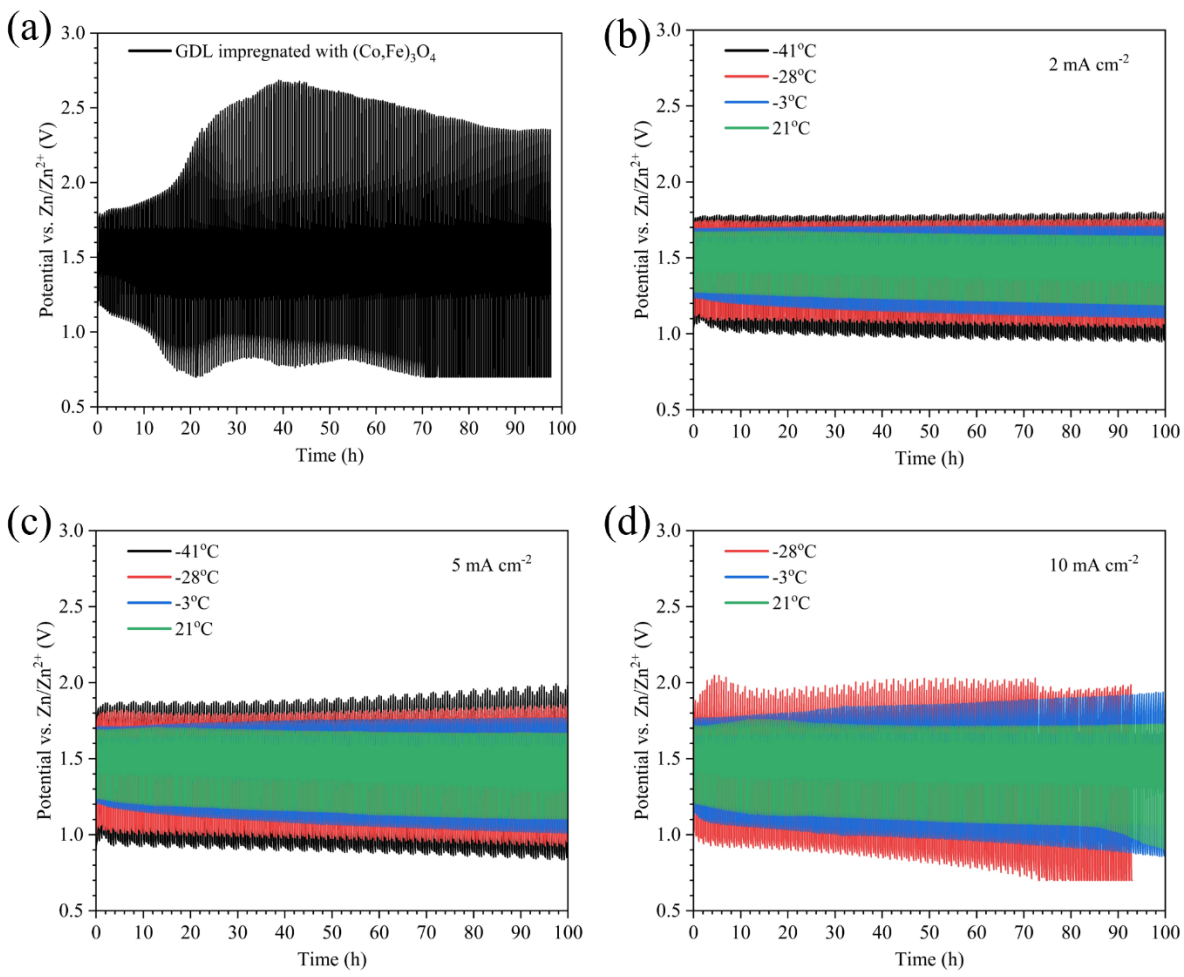

Figure S5. (a) Cyclability test at 10 mA cm^{-2} using aqueous 6 M KOH+2 M KI+0.4 M ZnO as the electrolyte and GDL with $(Co,Fe)_3O_4$ decorated N-CNTs as the air electrode. Cyclability tests at different temperatures for ZABs using GPE-KOH-KI and Pt/RuO_2 coated CC at (b) 2 mA cm^{-2}, (c) 5 mA cm^{-2}, and (d) 10 mA cm^{-2}.

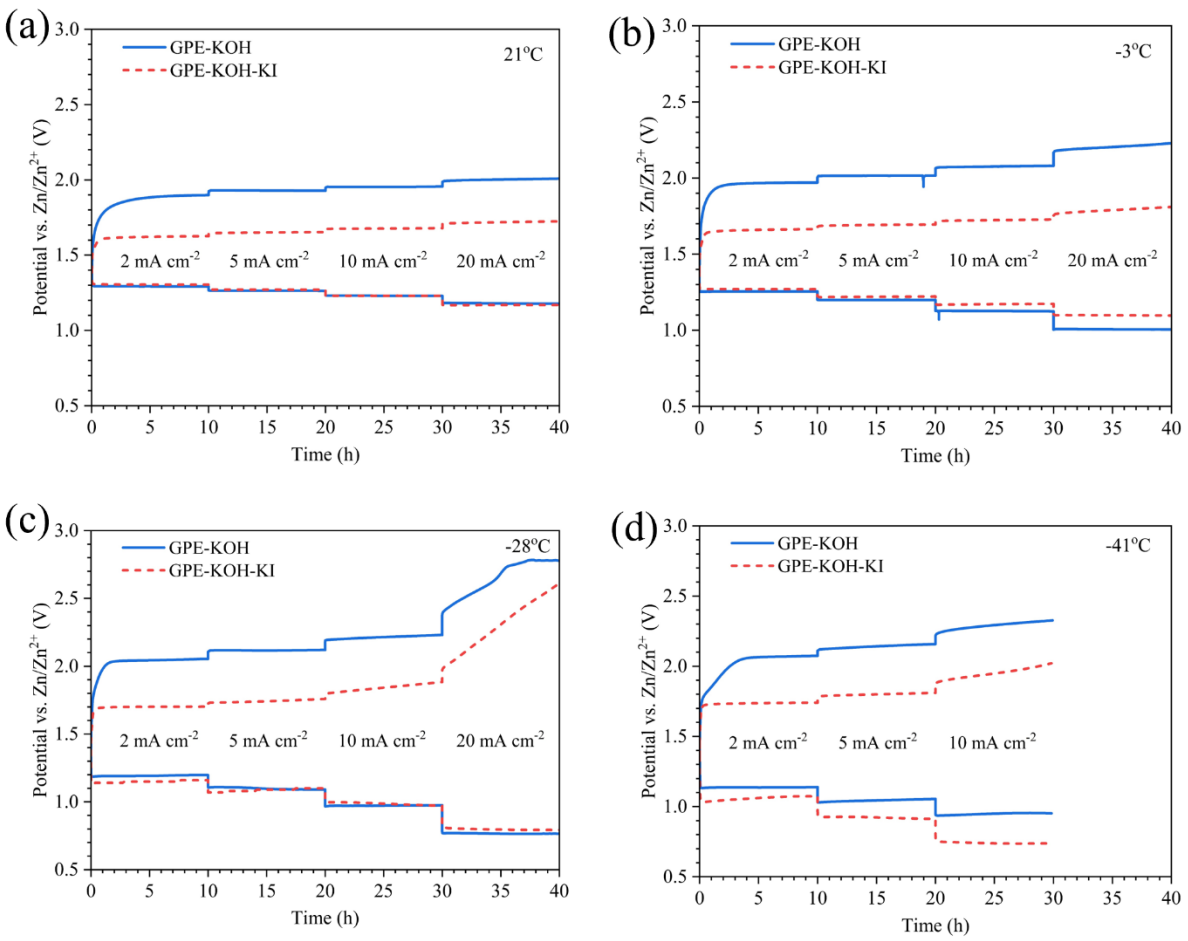

Figure S6. Comparison of rate test results for ZABs using $(Co,Fe)_3O_4$ coated GDL with GPE-KOH and Pt/RuO_2 coated CC with GPE-KOH-KI at (a) $21°C$, (b) $-3°C$, (c) $-28°C$, and (d) $-41°C$.

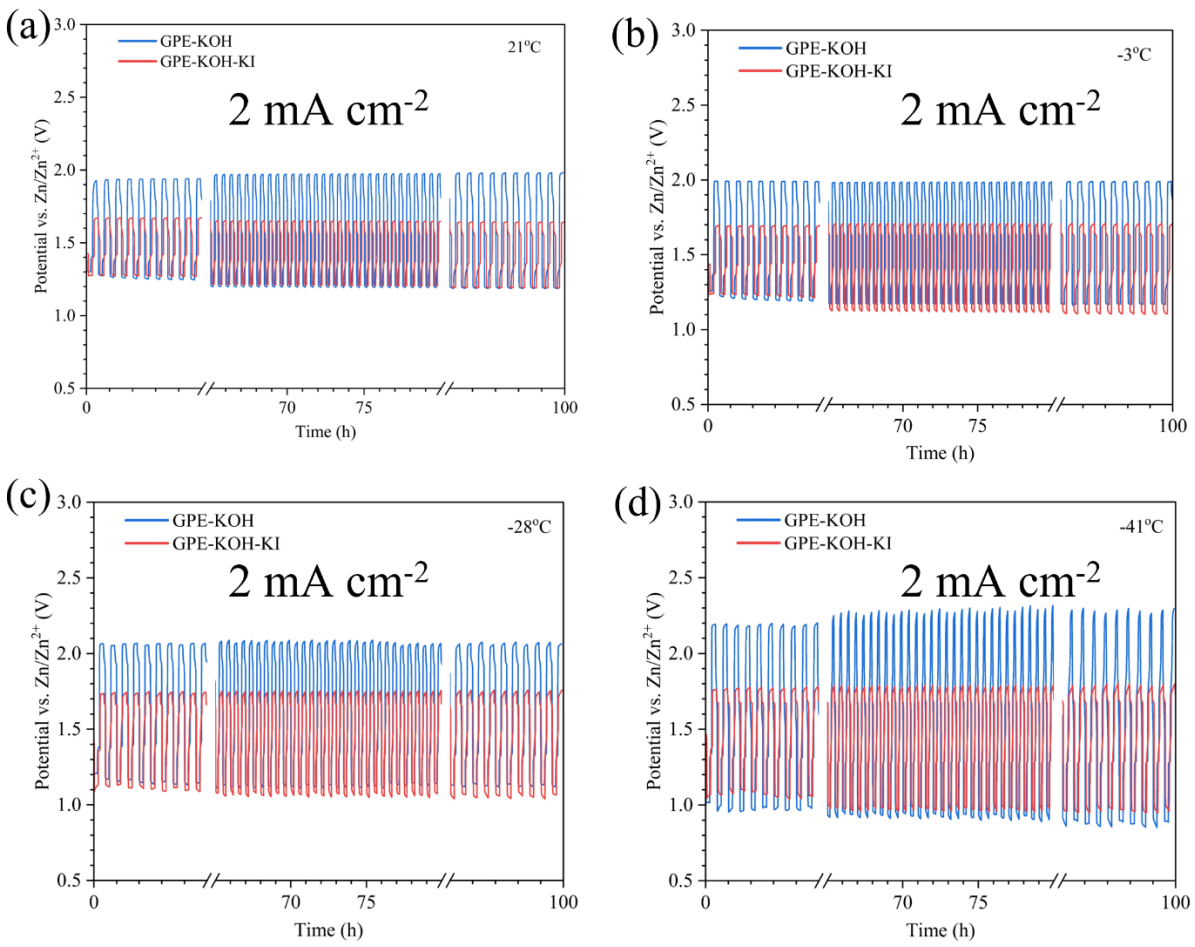

Figure S7. Cyclability test results for ZABs using $(Co,Fe)_3O_4$ coated GDL with GPE-KOH and Pt/RuO_2 coated CC with GPE-KOH-KI at 2 mA cm^{-2} at (a) $21℃$, (b) $-3℃$, (c) $-28℃$, and (d) $-41℃$.

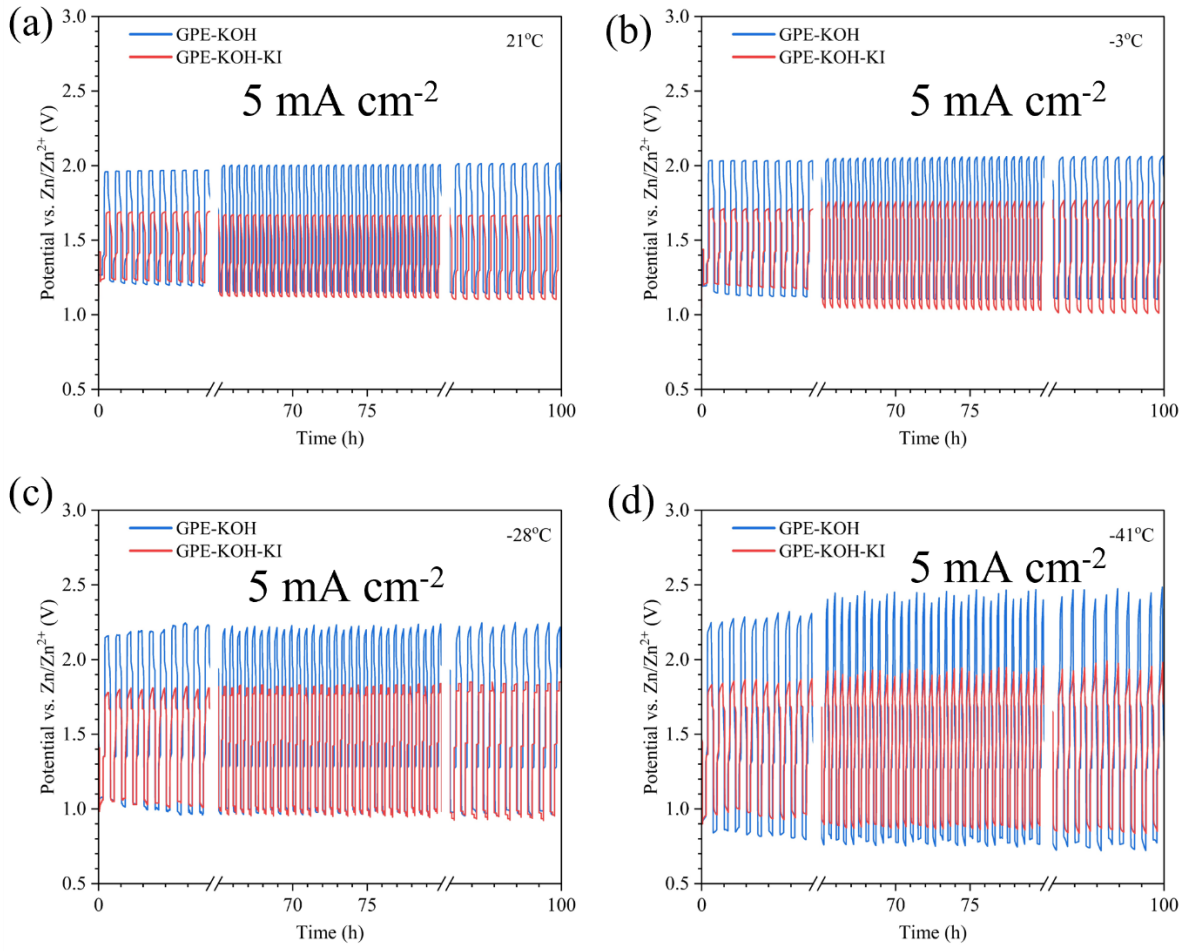

Figure S8. Comparison of cyclability test results for ZABs using $(Co,Fe)_3O_4$ coated GDL with GPE-KOH and Pt/RuO_2 coated CC with GPE-KOH-KI at 5 mA cm^{-2} at (a) 21°C, (b) −3°C, (c) −28°C, and (d) −41°C.

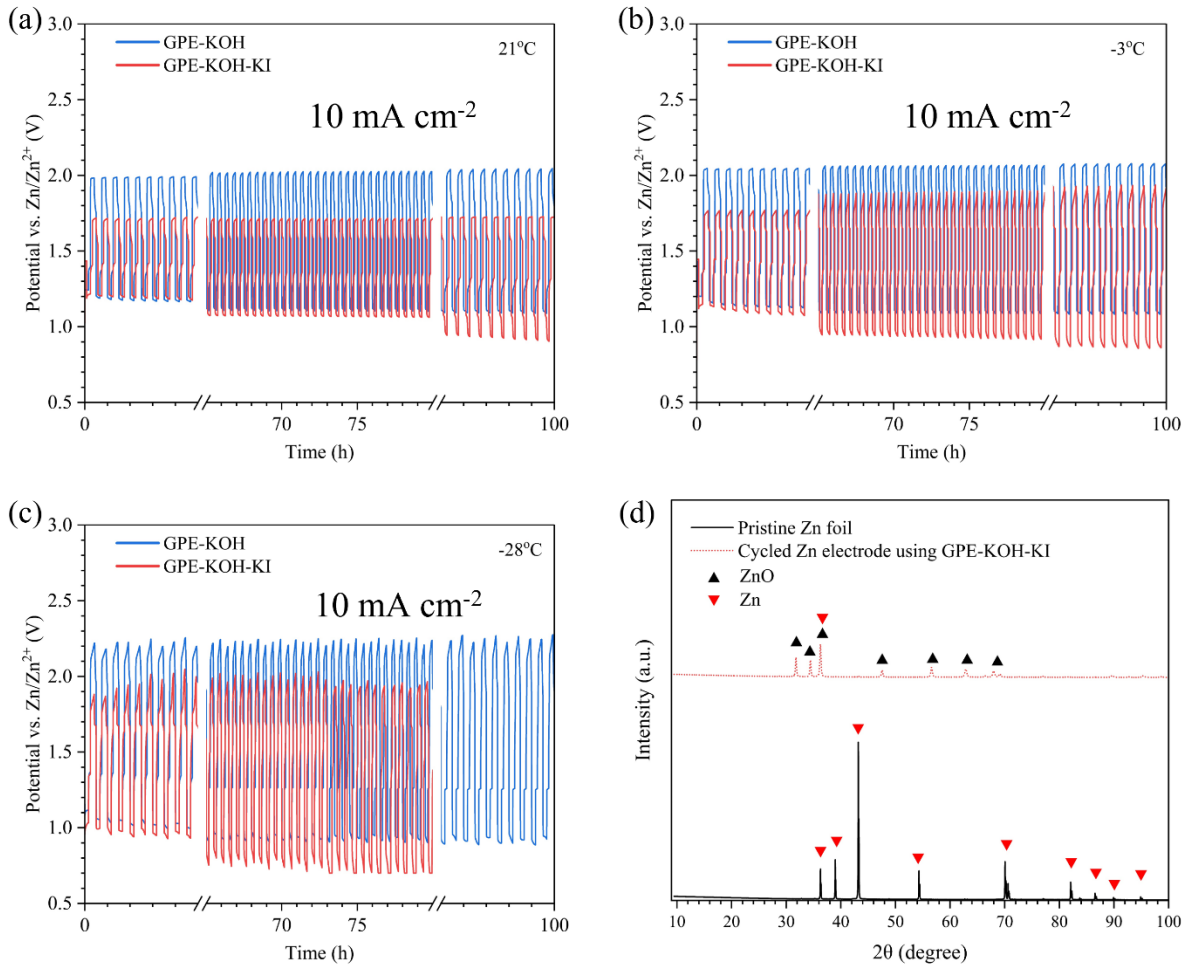

Figure S9. Cyclability test results for ZABs using $(Co,Fe)_3O_4$ coated GDL with GPE-KOH and Pt/RuO_2 coated CC with GPE-KOH-KI at 10 mA cm^{-2} at (a) 21℃, (b) −3℃, and (c) −28℃. (d) XRD patterns for pristine Zn foil and the Zn electrode after cycling using GPE-KOH-KI at 21℃ and 10 mA cm^{-2} for 100 h.

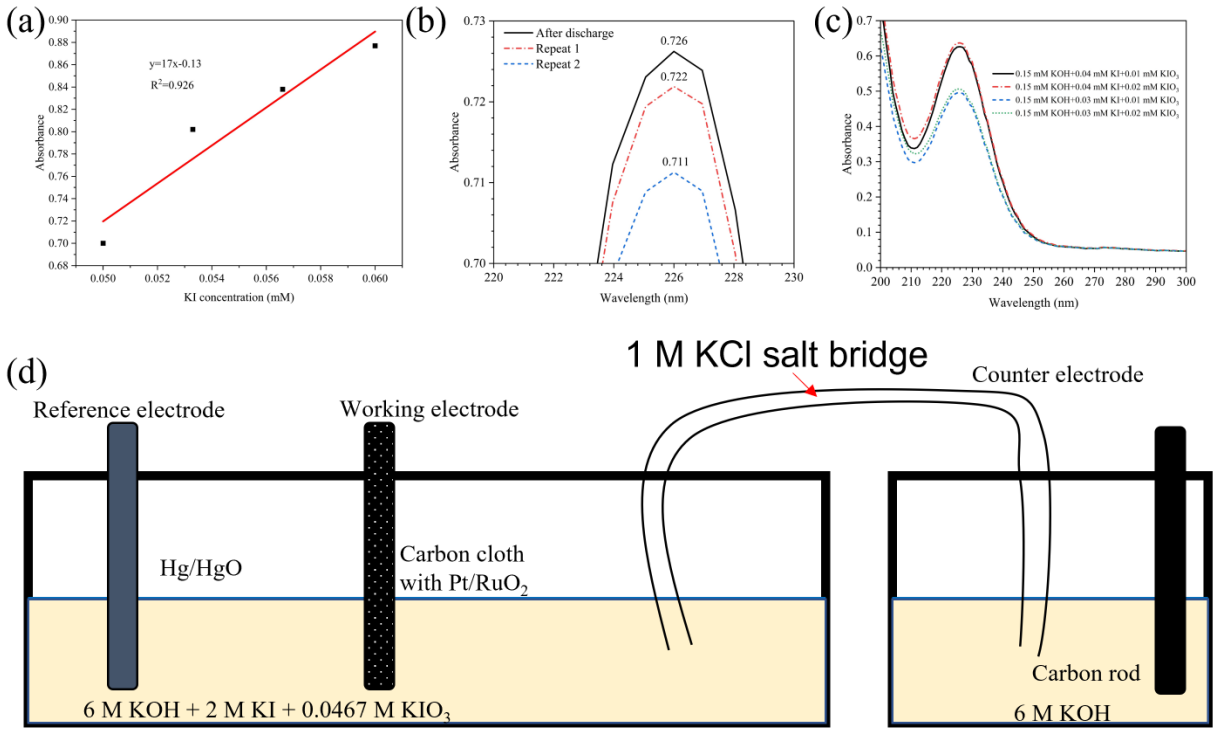

Figure S10. (a) Absorbance vs. KI concentration. This plot was created using the data in Figure 7d. UV-vis spectra for (b) sample A after discharge and with repeated diluted samples to confirm reproducibility, and (c) solutions with different concentrations of KI and KIO_3. (d) The three-electrode setup for the half-cell discharge process.

Table S1. Summary of rate test battery efficiencies and power densities for ZABs using GPE-KOH under different test conditions

	Efficiency at different current densities (%)				
Temperature (℃)	2 mA cm^{-2}	5 mA cm^{-2}	10 mA cm^{-2}	20 mA cm^{-2}	Peak power density (mW cm^{-2})
21	68	67	63	59	127
-3	64	60	54	45	38
-28	58	51	43	27	17
-41	55	49	41	Failed	11

Table S2. Summary of cyclability test battery efficiencies for ZABs using GPE-KOH under different test conditions

	2 mA cm^{-2}		5 mA cm^{-2}		10 mA cm^{-2}	
Temperature (℃)	Initial Efficiency (%)	Final Efficiency (%)	Initial Efficiency (%)	Final Efficiency (%)	Initial Efficiency (%)	Final Efficiency (%)
21	68	60	64	57	63	53
-3	63	59	59	53	59	52
-28	58	54	50	43	50	41
-41	46	39	42	32	Failed	Failed

Table S3. Summary of rate test battery efficiencies and power densities for ZABs using GPE-KOH-KI under different test conditions

	Efficiency at different current densities (%)				
Temperature (℃)	2 mA cm^{-2}	5 mA cm^{-2}	10 mA cm^{-2}	20 mA cm^{-2}	Peak power density (mW cm^{-2})
21	80	77	73	67	98
-3	76	72	68	61	27
-28	68	63	54	31	22
-41	61	50	37	Failed	11

Table S4. Summary of rate test battery efficiency improvement for ZABs using GPE-KOH-KI under different testing conditions when compared with ZABs using GPE-KOH

	Efficiency improvement at various current densities (%)			
Temperature (℃)	2 mA cm^{-2}	5 mA cm^{-2}	10 mA cm^{-2}	20 mA cm^{-2}
21	18	15	16	14
-3	19	20	26	36
-28	17	24	26	15
-41	11	2	-10	Both failed

Table S5. Summary of cyclability test battery efficiencies for ZABs using GPE-KOH-KI under different test conditions

	2 mA cm^{-2}		5 mA cm^{-2}		10 mA cm^{-2}	
Temperature (℃)	Initial Efficiency (%)	Final Efficiency (%)	Initial Efficiency (%)	Final Efficiency (%)	Initial Efficiency (%)	Final Efficiency (%)
21	77	73	74	66	71	52
-3	73	65	71	57	65	45
-28	65	61	60	52	55	Failed
-41	61	53	52	43	Failed	Failed

Table S6. Summary of cyclability test initial and final battery efficiency improvements for ZABs using GPE-KOH-KI under different test conditions when compared with ZABs using GPE-KOH

	2 mA cm^{-2}		5 mA cm^{-2}		10 mA cm^{-2}	
Temperature (℃)	Initial Efficiency (%)	Final Efficiency (%)	Initial Efficiency (%)	Final Efficiency (%)	Initial Efficiency (%)	Final Efficiency (%)
21	13	22	16	16	13	-2
-3	16	10	20	8	10	-13
-28	12	13	20	21	10	None
-41	33	36	24	34	N/A	N/A

Table S7. Comparison of ZABs using GPEs reported in this work and in the literature

Electrolyte	Current Density (mA cm^{-2})	Temp. (℃)	Catalyst	Maximum Power Density (mW cm^{-2})	Initial Battery Efficiency (%)	Cycling Time (h)	Ref.
GPE-KOH	2	21	$(Co,Fe)_3O_4$	127	68	100	This work
		-28		17	58	100	
		-41		11	46	100	
	10	21		127	61	260	
GPE-KOH-KI	2	21	Pt/RuO_2	98	77	100	
		-28		22	65		
		-41		11	61		
	5	21		98	77		
PAM-PAA	1	25	Pt/RuO_2	11.8	66	10	1
		-20		8.2	55	10	
Cellulose-PAA	2	25	MnO_2, Co_3O_4	40.25	69	11	2
PAM	5	25	MnO_2, GO	105	63	23.3	3
PVA	3	25	Co_3O_4	62.6	63	48	4
PAA	2	25	FeCo based catalyst	160	65	105	5
	5	25			~60	80	
	2	-20		80.5	57	5	
PAMPS-K/MC	1	25	Co_3O_4	73.9	64	24	6
		0		N/A	~60		
		-20		54.2	~55		
PAM-KI	2	20	Pt/RuO_2	43	71	75	7
		-40		10	59	40	
PANa-starch	1	25	Pt/RuO_2	67.5	68	28.7	8
		-20		30.7	~53	44.1	
PAA	2	25	FeCo based catalyst	128.8	65.9	92	9
		-30		63.6	60.4	92	
PANa	2	25	Pt/RuO_2	88	65.5	160	10
PANa-cellulose	5	25	Fe based	108.6	60	110	11
PVA-GG	2	25	Pt/IrO_2	50	~60	10	12
PVA	1	25	Co_3O_4	N/A	~38	70	13
PVA	1	25	Co_3O_4	16	~39	40	14
PVA-PAA-GO-KI	2	25	$Pt/C+Co_3O_4$	78.6	73	200	15
Aqueous 6 M KOH+3 M KI+0.2 M Zn(Ac)2	5	25	Pt/C	148.8	76.5	80	16
PEVA	2	25	Co_3O_4	N/A	~60	230	17
PAM-SA-KI	1	25	PtC/RuO_2	132	80	110	18

Abbreviations:
PAMPS-K/MC: poly(2-acrylamido-2-methylpropanesulfonic acid potassium salt) with methyl cellulose
PANa-starch: sodium polyacrylate-starch
GG: guar hydroxypropyltrimonium chloride
GO: graphene oxide
SA: sodium alginate
PEVA: poly(ethylene vinyl acetate)

Table S8. Summary of rate test battery efficiencies and power densities for ZABs using GPE-KOH and GPE-KOH-KI under different conditions

GPE-KOH					
Temperature (℃)	2 mA cm^{-2}	5 mA cm^{-2}	10 mA cm^{-2}	20 mA cm^{-2}	Peak power density (mW cm^{-2})
21	68	67	63	59	127
-3	64	60	54	45	38
-28	58	51	43	27	17
-41	55	49	41	Failed	11
GPE-KOH-KI					
Temperature (℃)	2 mA cm^{-2}	5 mA cm^{-2}	10 mA cm^{-2}	20 mA cm^{-2}	Peak power density (mW cm^{-2})
21	80	77	73	67	98
-3	76	72	68	61	27
-28	68	63	54	31	22
-41	61	50	37	Failed	11

Table S9. Summary of cyclability performance of ZABs using GPE-KOH and GPE-KOH-KI under different conditions

GPE-KOH						
	2 mA cm^{-2}		5 mA cm^{-2}		10 mA cm^{-2}	
Temperature (℃)	Initial Efficiency (%)	Final Efficiency (%)	Initial Efficiency (%)	Final Efficiency (%)	Initial Efficiency (%)	Final Efficiency (%)
21	68	60	64	57	63	53
-3	63	59	59	53	59	52
-28	58	54	50	43	50	41
-41	46	39	42	32	Failed	Failed
GPE-KOH-KI						
	2 mA cm^{-2}		5 mA cm^{-2}		10 mA cm^{-2}	
Temperature (℃)	Initial Efficiency (%)	Final Efficiency (%)	Initial Efficiency (%)	Final Efficiency (%)	Initial Efficiency (%)	Final Efficiency (%)
21	77	73	74	66	71	52
-3	73	65	71	57	65	45
-28	65	61	60	52	55	Failed
-41	61	53	52	43	Failed	Failed

Table S10. Ionic mobility of selected ions in water

Ion	Mobility (10^{-8} m^2 s^{-1} V^{-1})	Reference
K^+	7.62	19
OH^-	20.64	19
I^-	6.23±0.04	20

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