Supporting Information

Scale up of fully eco-friendly zinc-O_2(ads) batteries from the lab scale to the prototype level

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Table S1. Summary of the equivalent circuit elements (shown in Fig 4 in the article) resulting from fitting the EIS data.

Cycle load (No.)	DC current* (mA)	R_s (Ω)	R_{ct} (Ω)	Q_{dl} Q_0 ($S \cdot s^n$)	Q_{dl} n (-)	Q_w Q_0 ($S \cdot s^n$)	Q_w n (-)
New cell	OCV	0.613	0.822	8.05e-4	0.759	0.794	0.193
	35	0.617	1.330	7.61e-4	0.769		
	-35	0.620	1.288	6.98e-4	0.770		
	55	0.621	1.211	6.96e-4	0.782		
	-55	0.625	1.184	8.83e-4	0.787		
	75	0.626	1.129	7.39e-4	0.781		
	-75	0.627	1.105	5.48e-4	0.793		
200	OCV	0.628	1.505	6.26e-4	0.786	0.884	0.419
	35	0.627	1.509	7.69e-4	0.770		
	-35	0.630	1.476	6.29e-4	0.781		
	55	0.628	1.419	7.70e-4	0.774		
	-55	0.632	1.383	5.75e-4	0.788		
	75	0.628	1.329	8.07e-4	0.773		
	-75	0.632	1.295	5.44e-4	0.792		
400	OCV	0.626	1.745	6.68e-4	0.781	1.203	0.443
	35	0.625	1.705	8.37e-4	0.766		
	-35	0.629	1.673	6.27e-4	0.783		
	55	0.626	1.597	8.72e-4	0.766		
	-55	0.629	1.564	5.80e-4	0.789		
	75	0.626	1.488	9.35e-4	0.764		
	-75	0.629	1.469	5.50e-4	0.793		
600	OCV	0.635	2.165	6.94e-4	0.780	1.410	0.502
	35	0.639	2.014	8.25e-4	0.770		
	-35	0.643	1.998	5.81e-4	0.790		
	55	0.639	1.847	9.20e-4	0.765		
	-55	0.643	1.839	5.43e-4	0.795		
	75	0.640	1.688	1.02e-3	0.760		
	-75	0.643	1.697	5.22e-4	0.797		

* Positive current means charge, negative current means discharge.

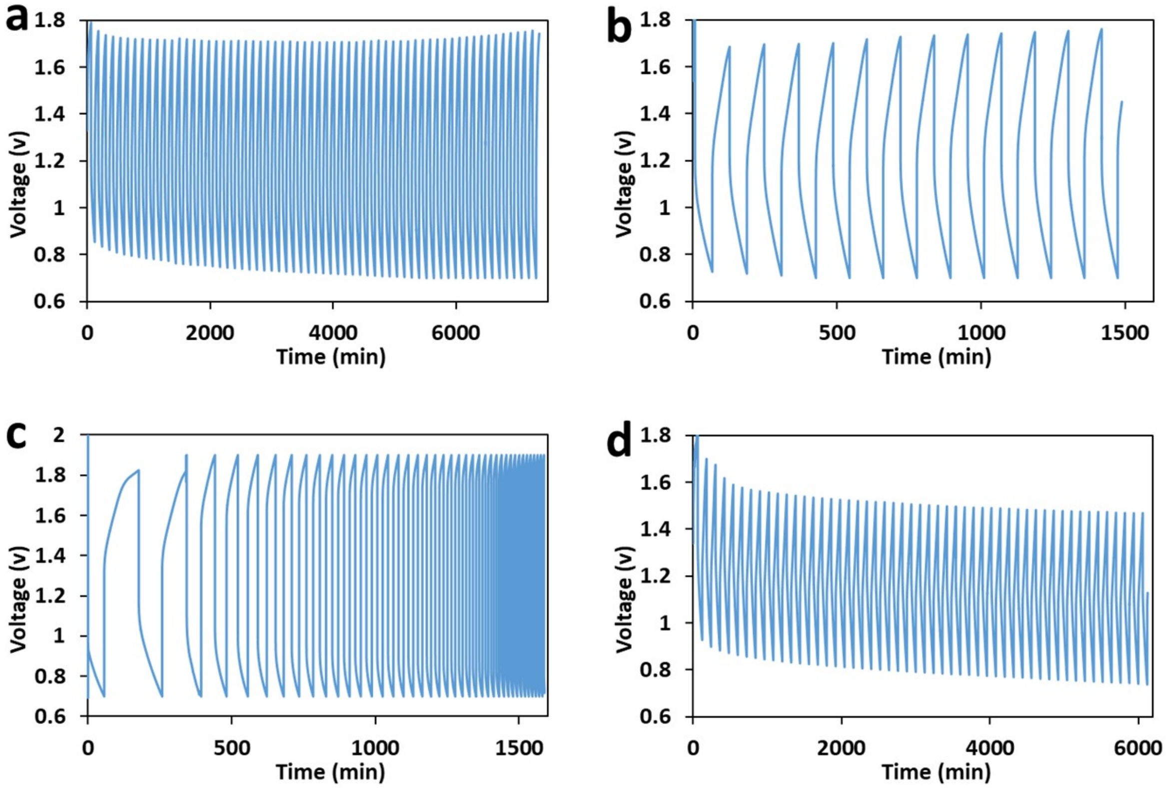


Figure S1 Improvement of cell geometry, galvanostatic charge/discharge battery cycling tests of various cell dimensions a) 50 × 50 × 0.65 mm (width, height, and thickness), 60 cm3, 6 mA/cm2; b) 25 × 50 × 0.65 mm, 30 cm3, 12 mA/cm2; c) 25 × 25 × 0.65 mm, 15 cm3, 8 mA/cm2; d) 25 × 50 × 0.65 mm, 16 cm3, 6 mA/cm2; for the zinc anode size, cathode volume, and current density, respectively. The length of the charge/discharge cycles and depth of discharge (DoD) were 120 min and 50 %, respectively.

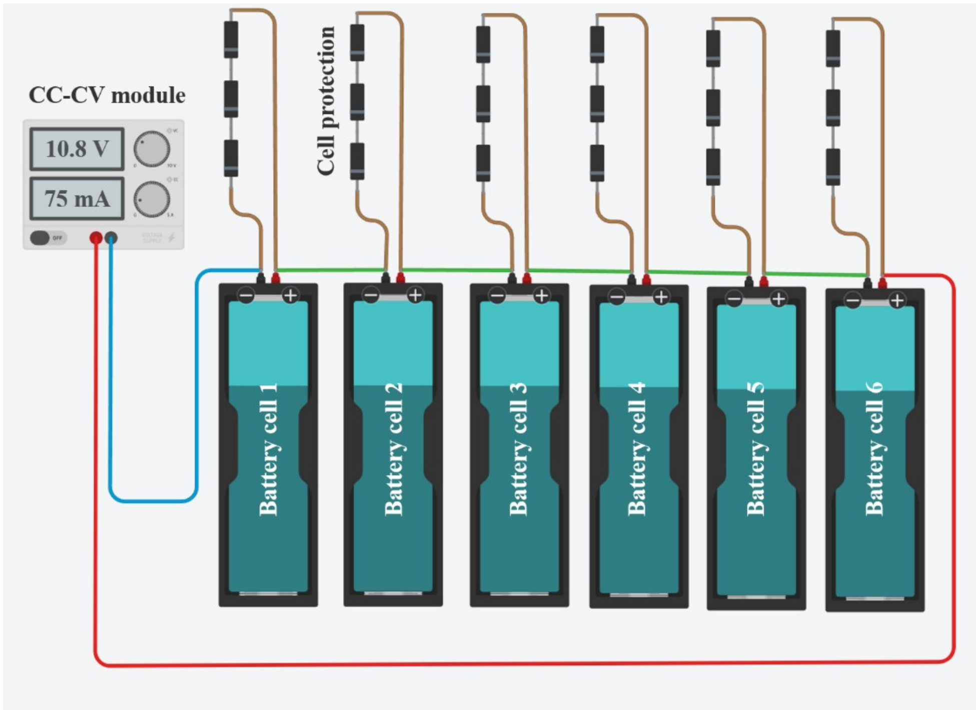


Figure S2. Simple homemade battery charge controller

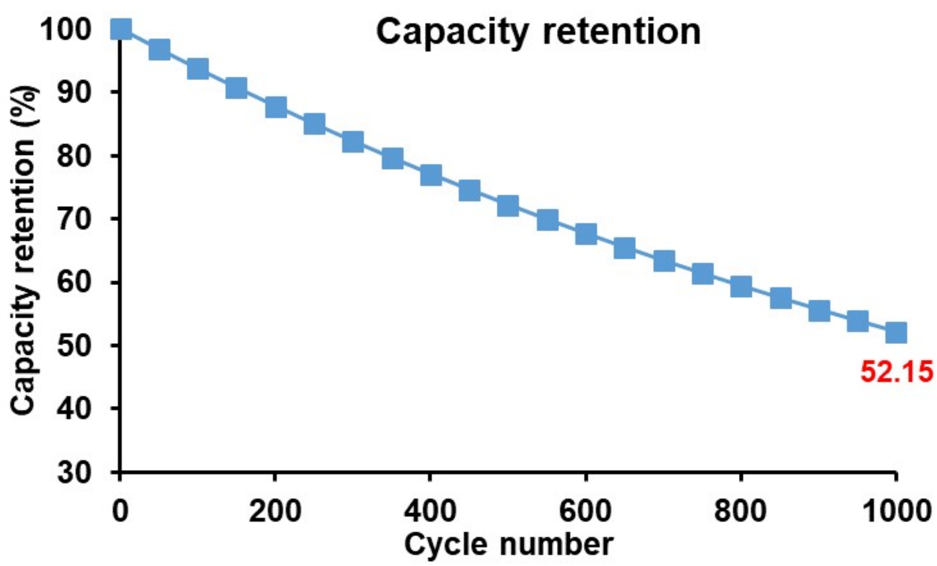


Figure S3. Plot of cell capacity retention as a function of cycle number recorded at 75 mA

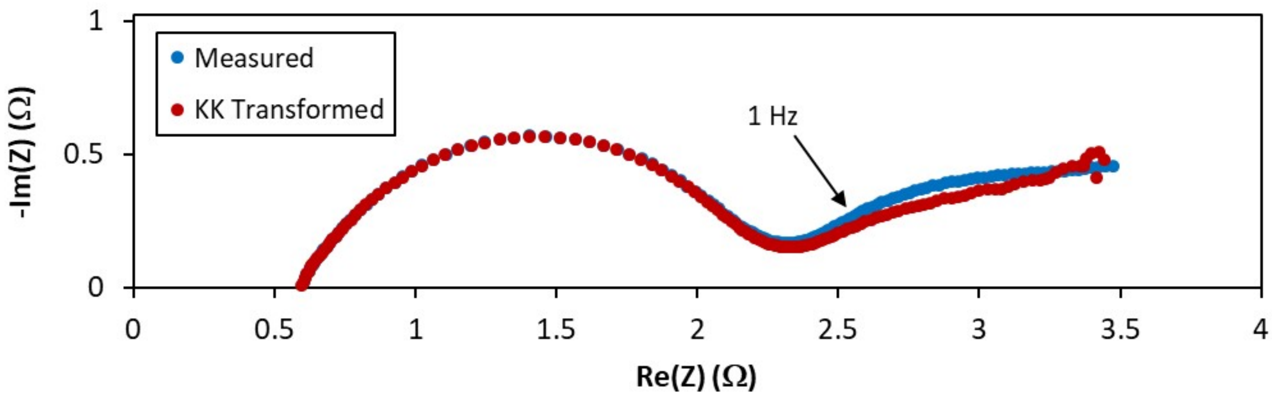


Figure S4. Combination of the measured data and its KK-transform. After 200 cycles, recorded at open-circuit voltage (OCV).

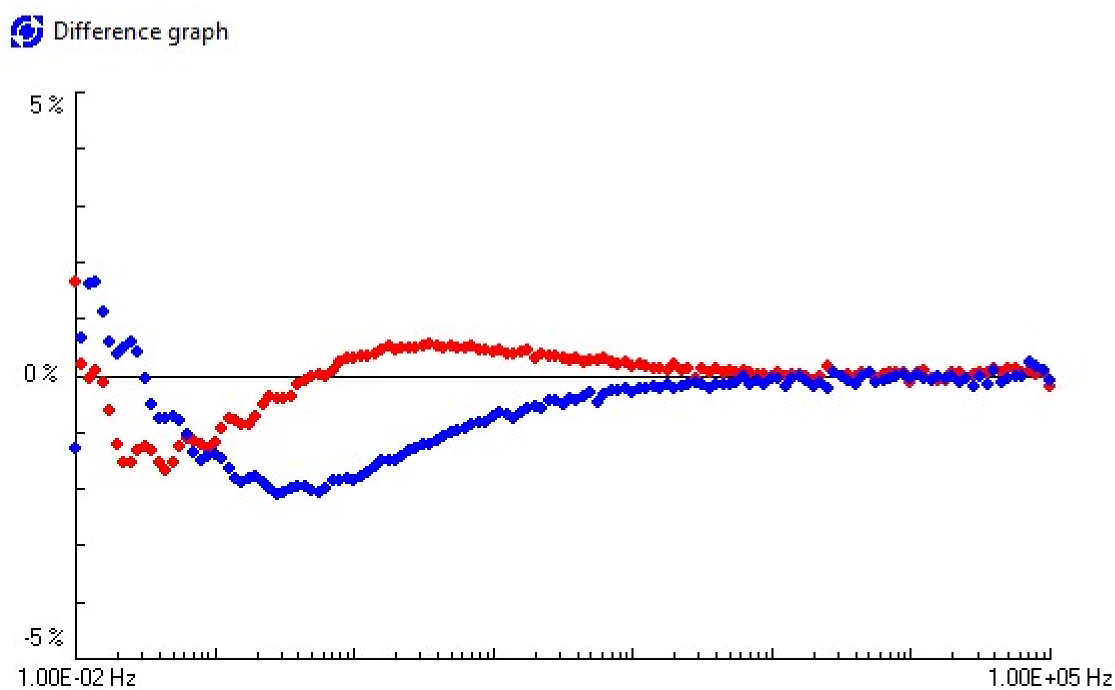
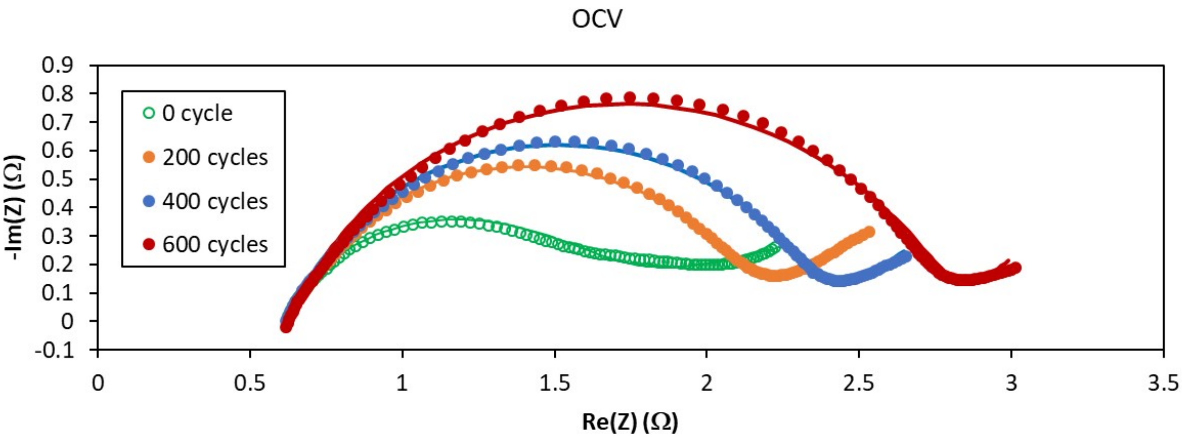


Figure S5. The real part (red) and imaginary part (blue) residuals of the KK-test. After 200 cycles, recorded at open-circuit voltage (OCV).



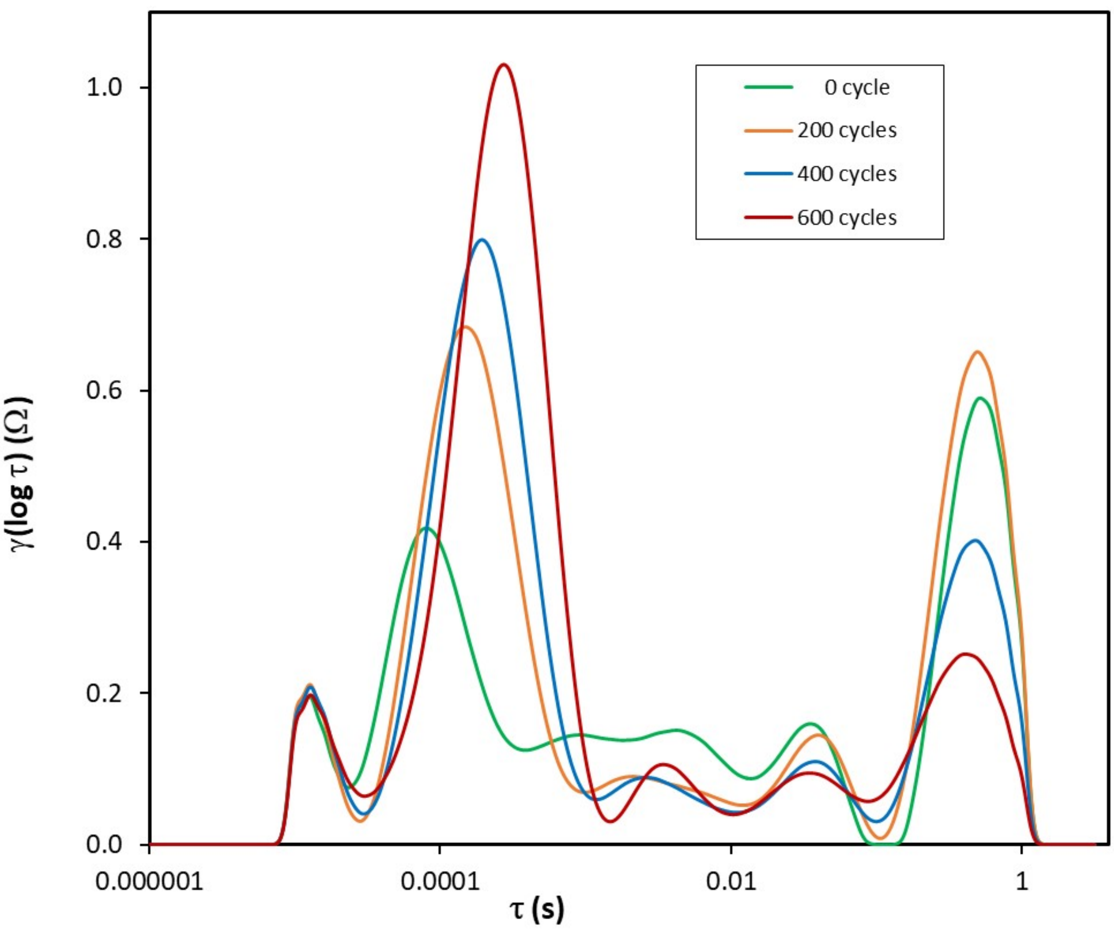
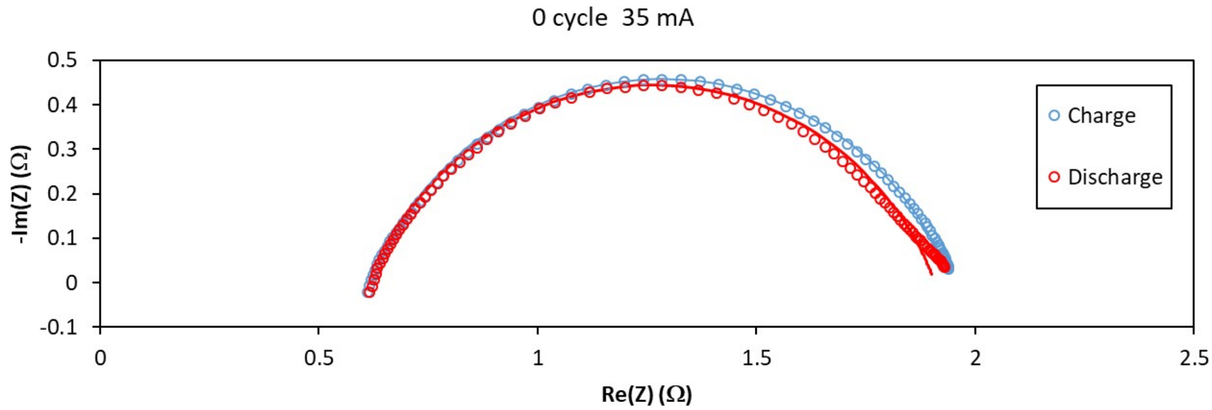


Figure S6. Nyquist-plots obtained by EIS at OCV and the corresponding DRT spectra of a single zinc–O_2(ads) battery cell. In the top chart, the solid lines represent the fitted impedance data using the ECM shown in the inset of Fig. 4c in the article.



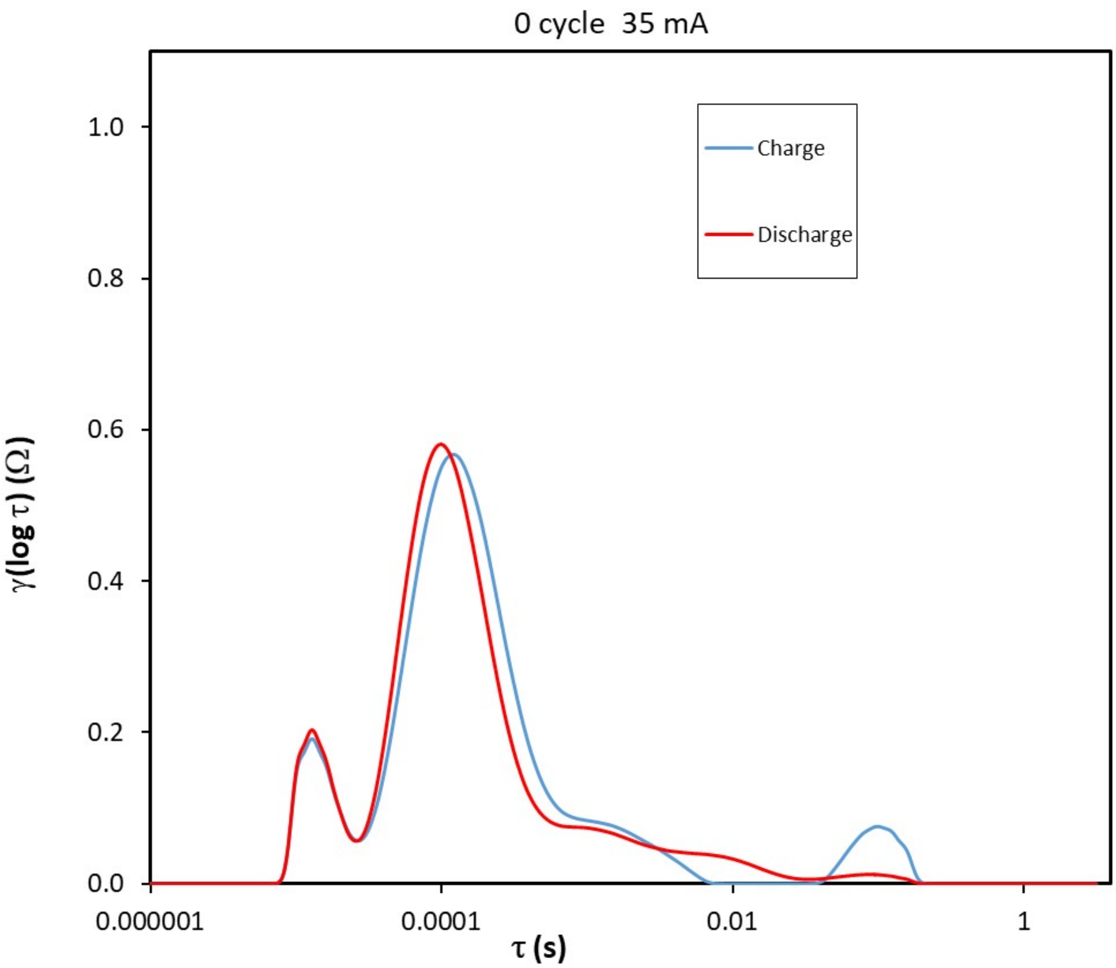


Figure S7a. Nyquist-plots obtained by EIS and the corresponding DRT spectra of a single new zinc–O_2(ads) battery cell recorded at a DC current of 35 mA during charging and discharging. The solid lines represent the fitted impedance data using the ECM shown in the inset of Fig. 4a in the article.

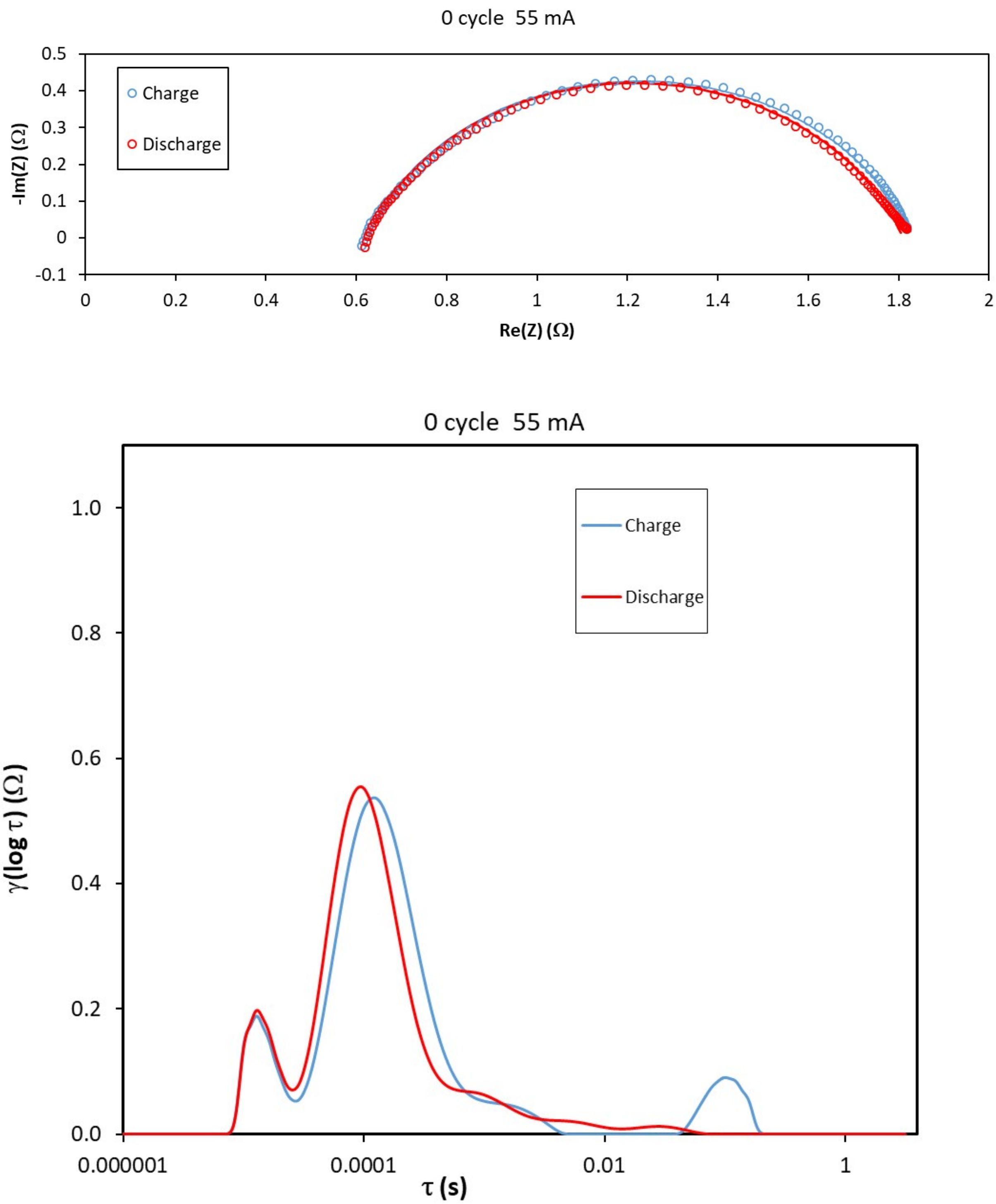
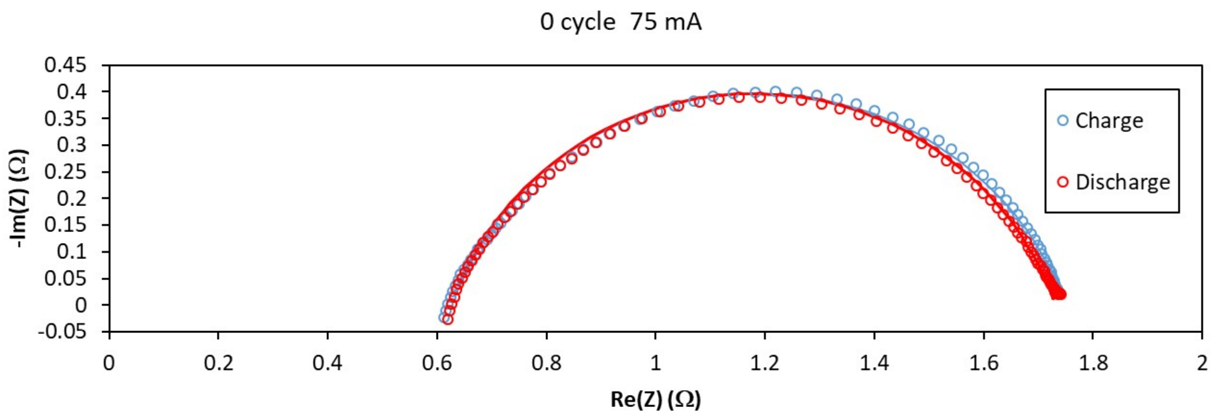


Figure S7b. Nyquist-plots obtained by EIS and the corresponding DRT spectra of a single new zinc–O_2(ads) battery cell recorded at a DC current of 55 mA during charging and discharging. The solid lines represent the fitted impedance data using the ECM shown in the inset of Fig. 4a in the article.



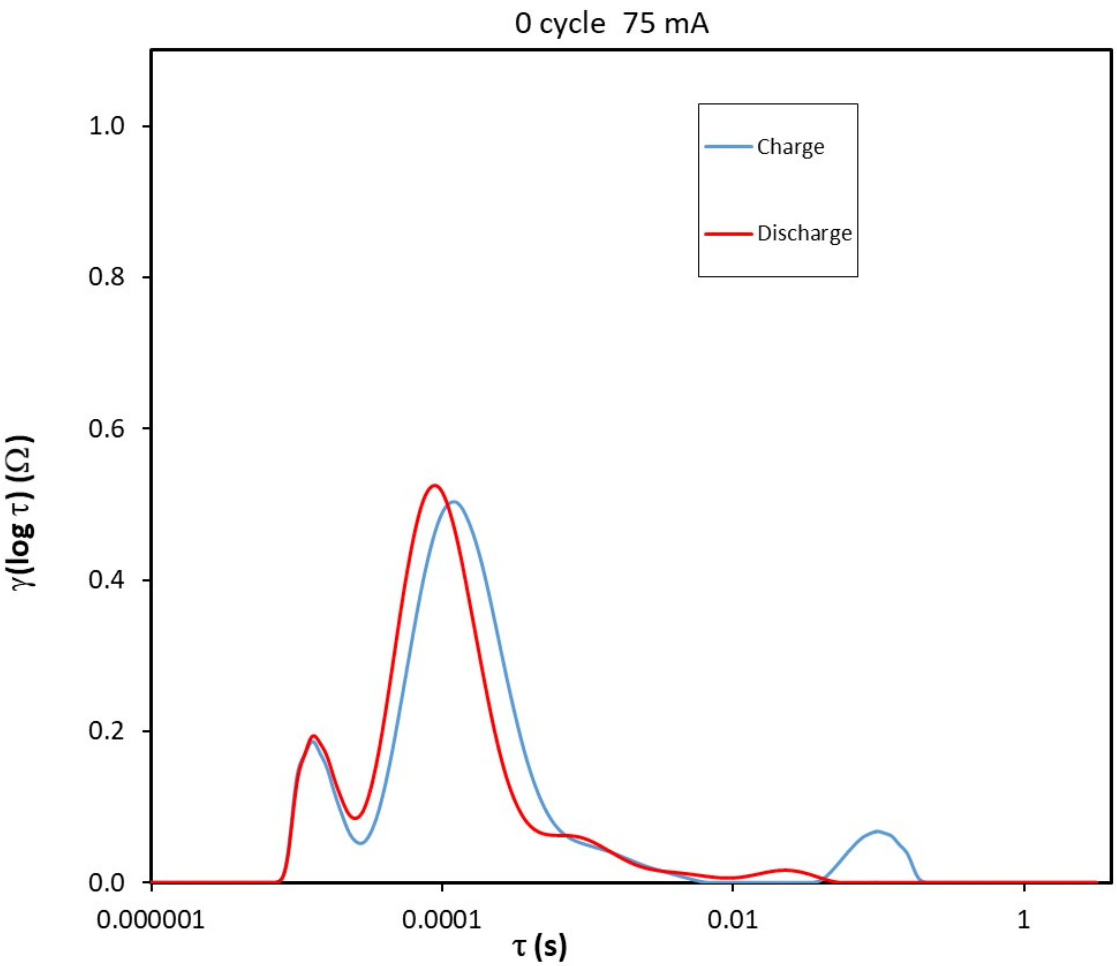
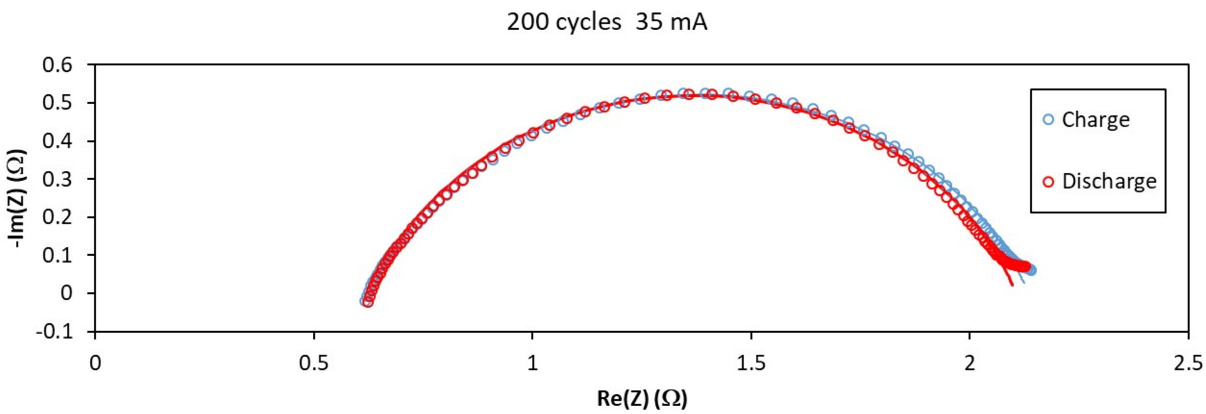


Figure S7c. Nyquist-plots obtained by EIS and the corresponding DRT spectra of a single new zinc–O_2(ads) battery cell recorded at a DC current of 75 mA during charging and discharging. The solid lines represent the fitted impedance data using the ECM shown in the inset of Fig. 4a in the article.



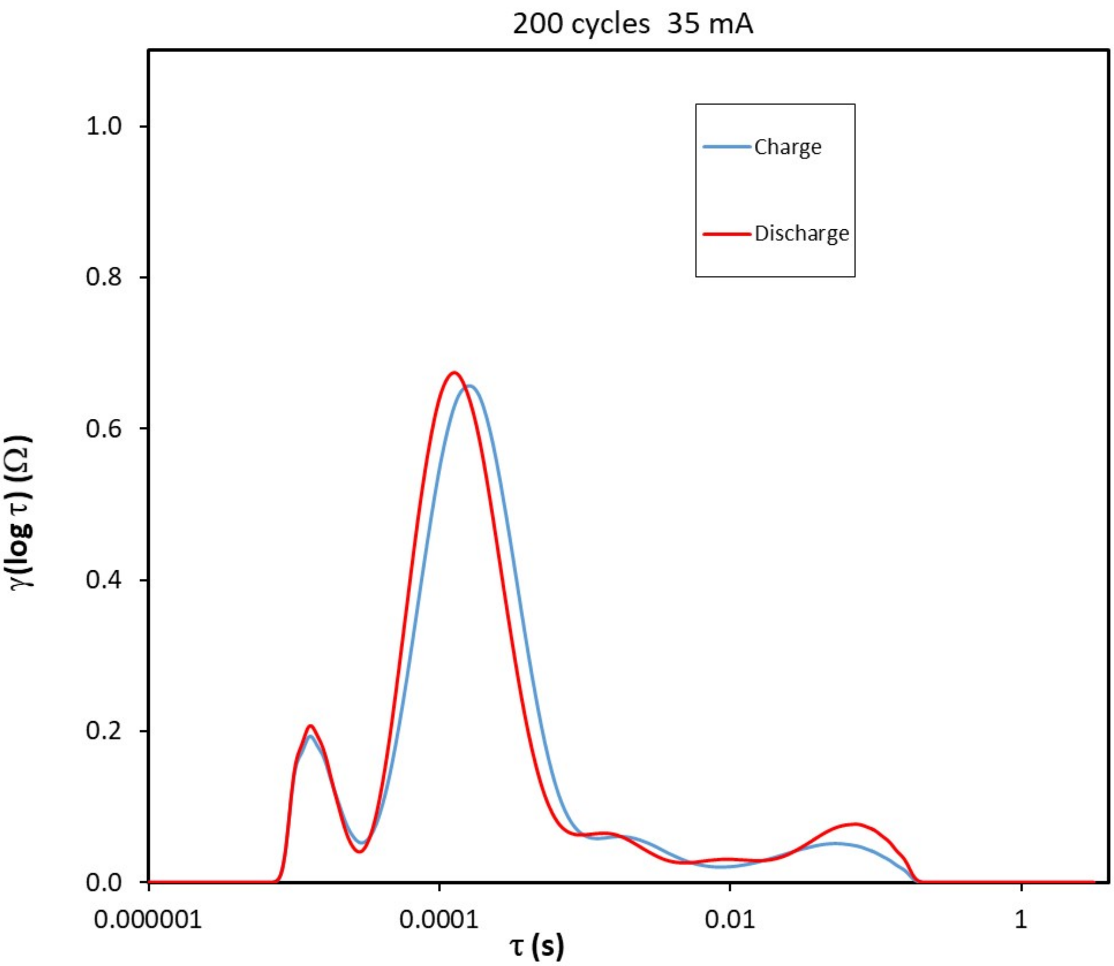
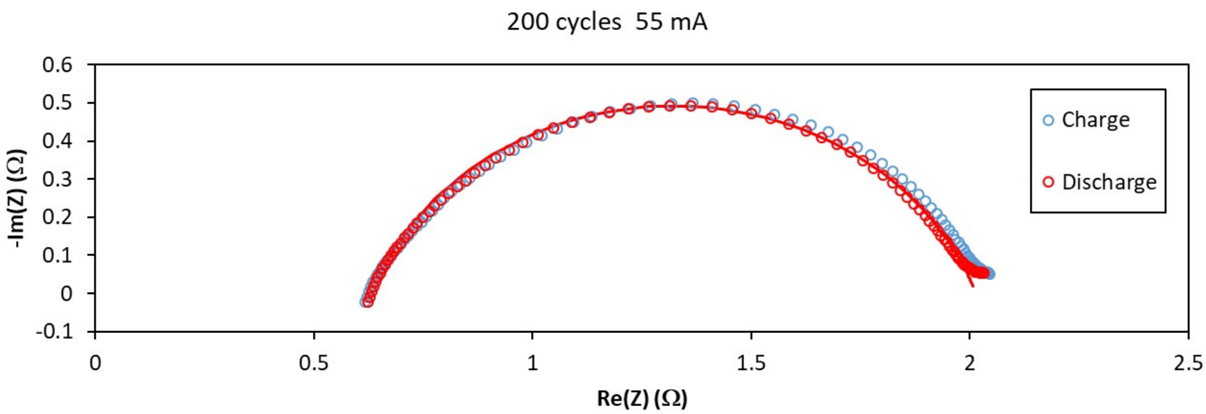


Figure S7d. Nyquist-plots obtained by EIS and the corresponding DRT spectra of a single zinc–O_2(ads) battery cell recorded after 200 cycles at a DC current of 35 mA during charging and discharging. The solid lines represent the fitted impedance data using the ECM shown in the inset of Fig. 4a in the article.



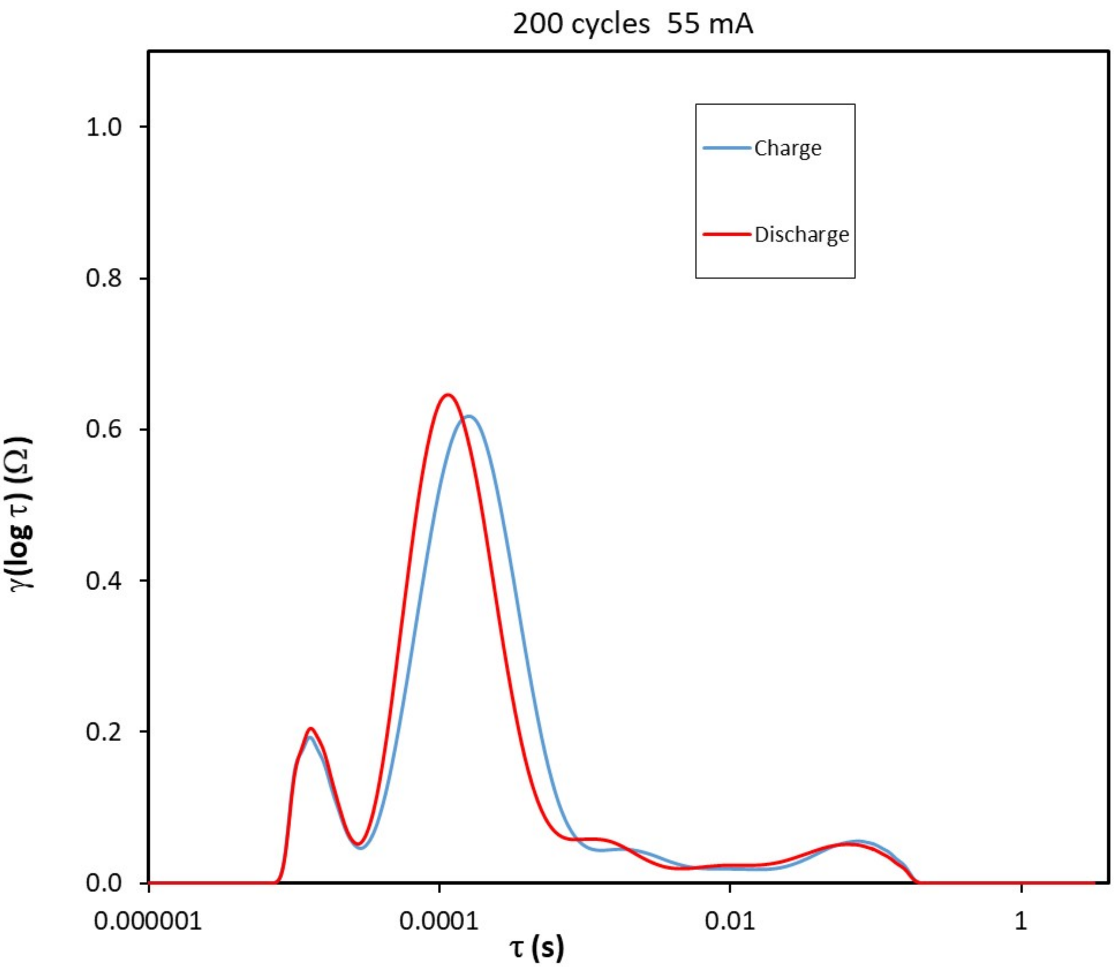
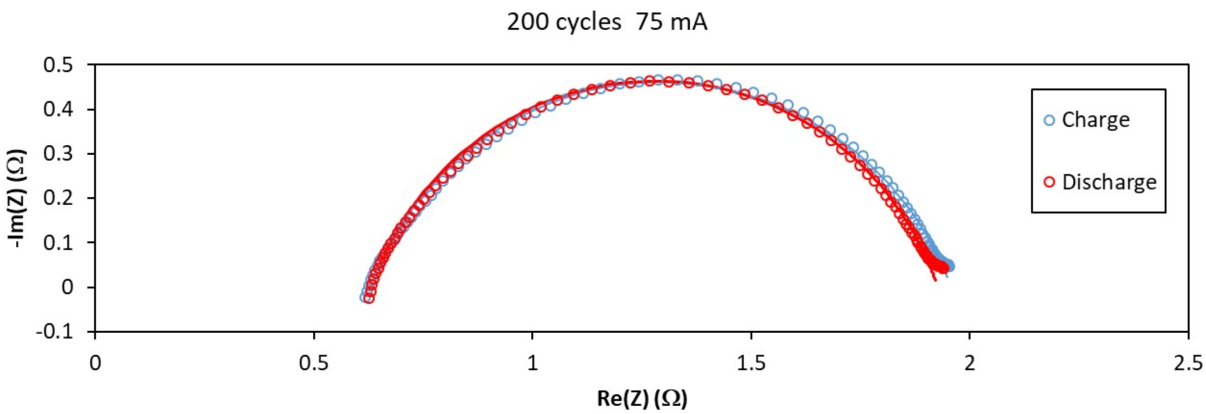


Figure S7e. Nyquist-plots obtained by EIS and the corresponding DRT spectra of a single zinc–O_2(ads) battery cell recorded after 200 cycles at a DC current of 55 mA during charging and discharging. The solid lines represent the fitted impedance data using the ECM shown in the inset of Fig. 4a in the article.



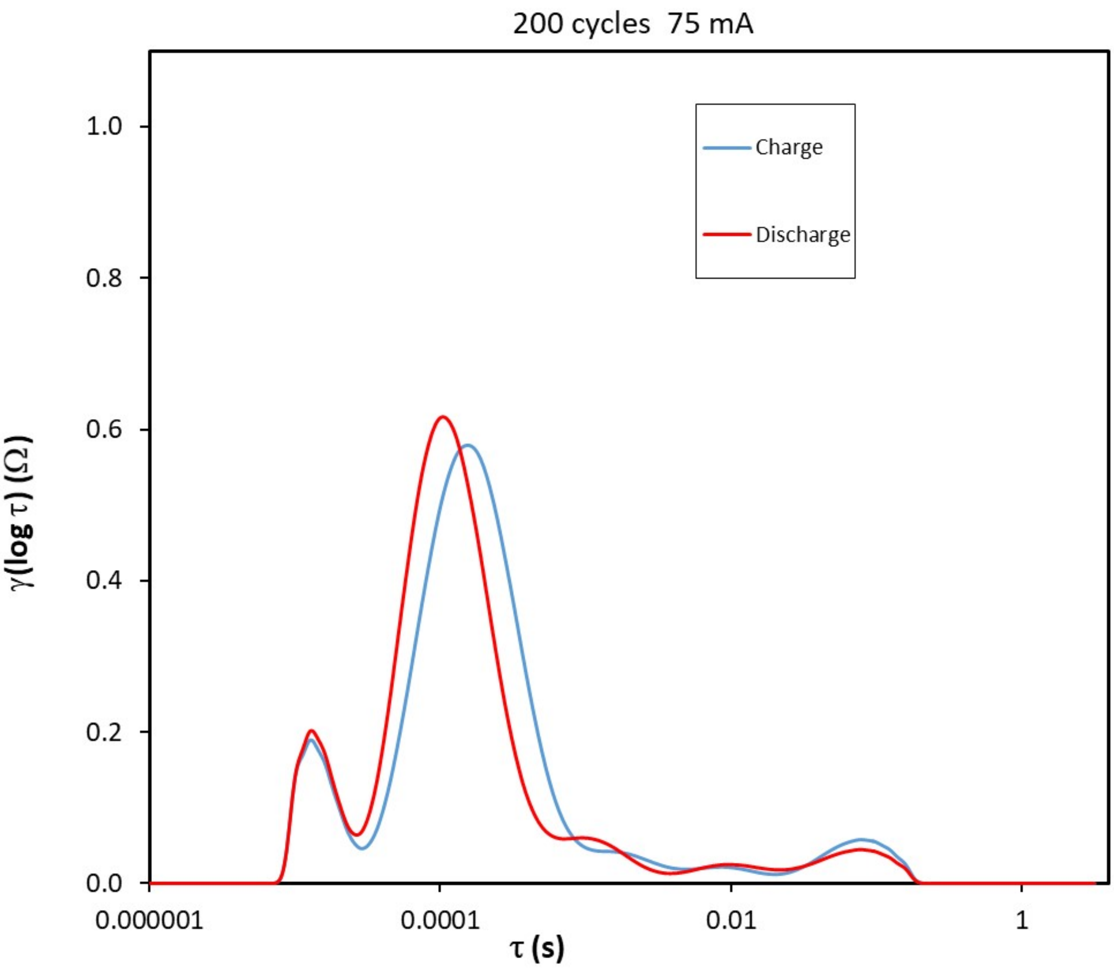
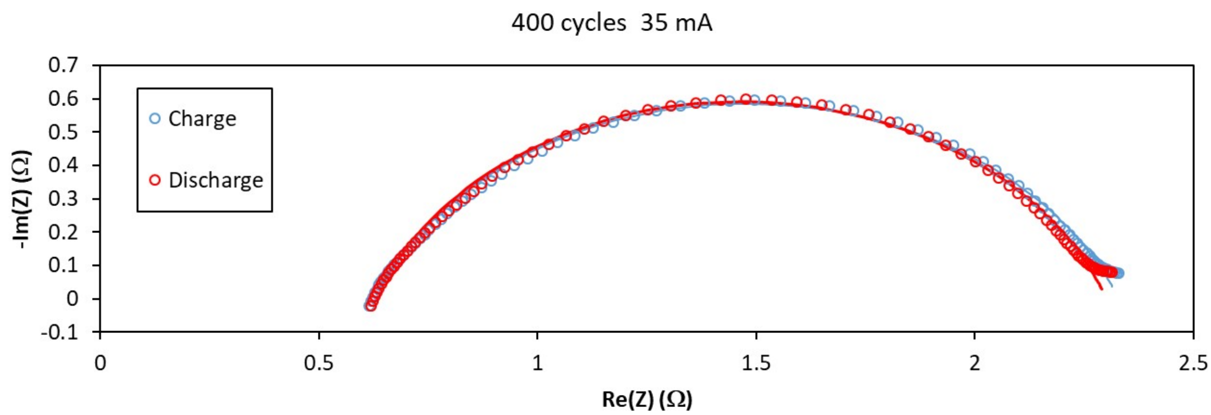


Figure S7f. Nyquist-plots obtained by EIS and the corresponding DRT spectra of a single zinc–O_2(ads) battery cell recorded after 200 cycles at a DC current of 75 mA during charging and discharging. The solid lines represent the fitted impedance data using the ECM shown in the inset of Fig. 4a in the article.



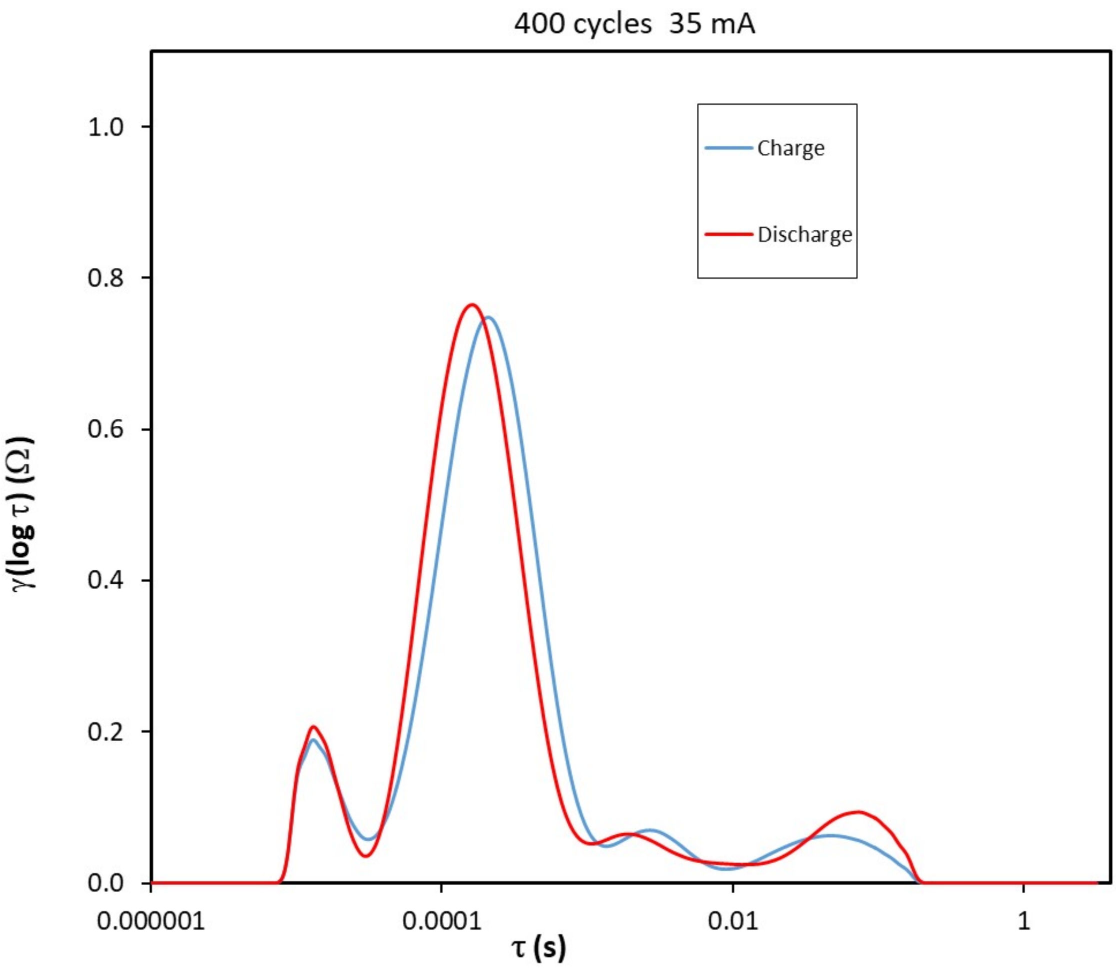
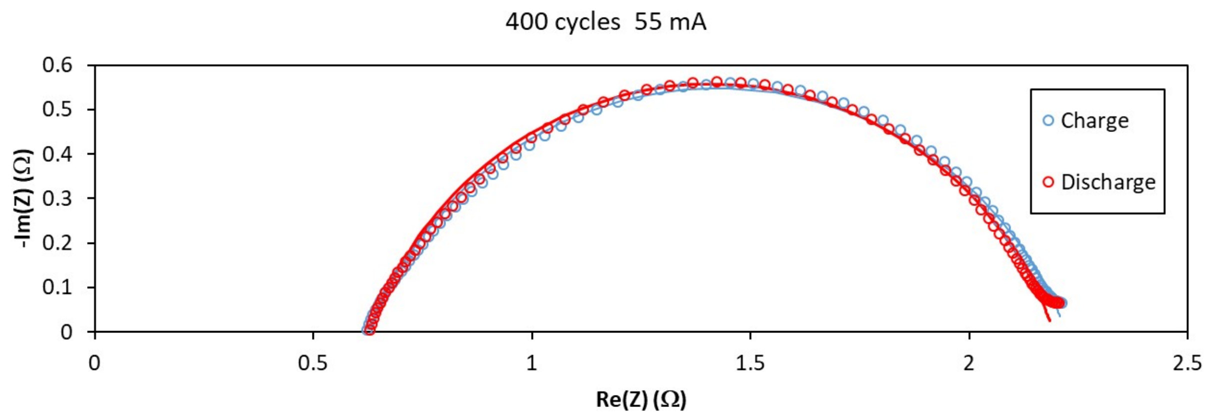


Figure S7g. Nyquist-plots obtained by EIS and the corresponding DRT spectra of a single zinc–O_2(ads) battery cell recorded after 400 cycles at a DC current of 35 mA during charging and discharging. The solid lines represent the fitted impedance data using the ECM shown in the inset of Fig. 4a in the article.



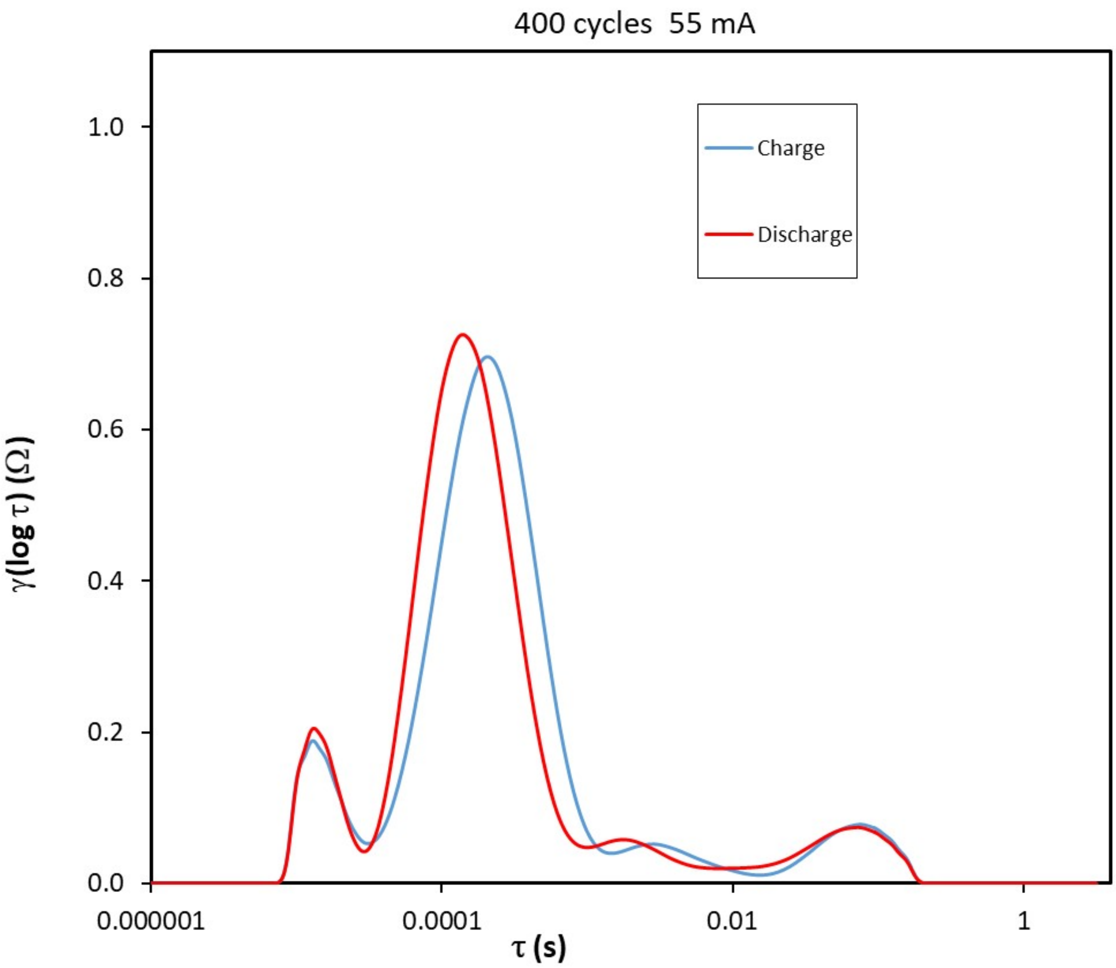
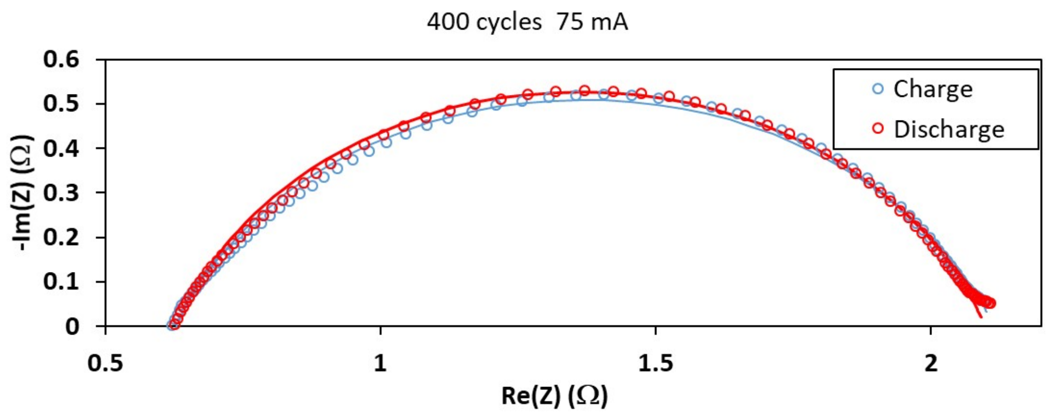


Figure S7h. Nyquist-plots obtained by EIS and the corresponding DRT spectra of a single zinc–O_2(ads) battery cell recorded after 400 cycles at a DC current of 55 mA during charging and discharging. The solid lines represent the fitted impedance data using the ECM shown in the inset of Fig. 4a in the article.



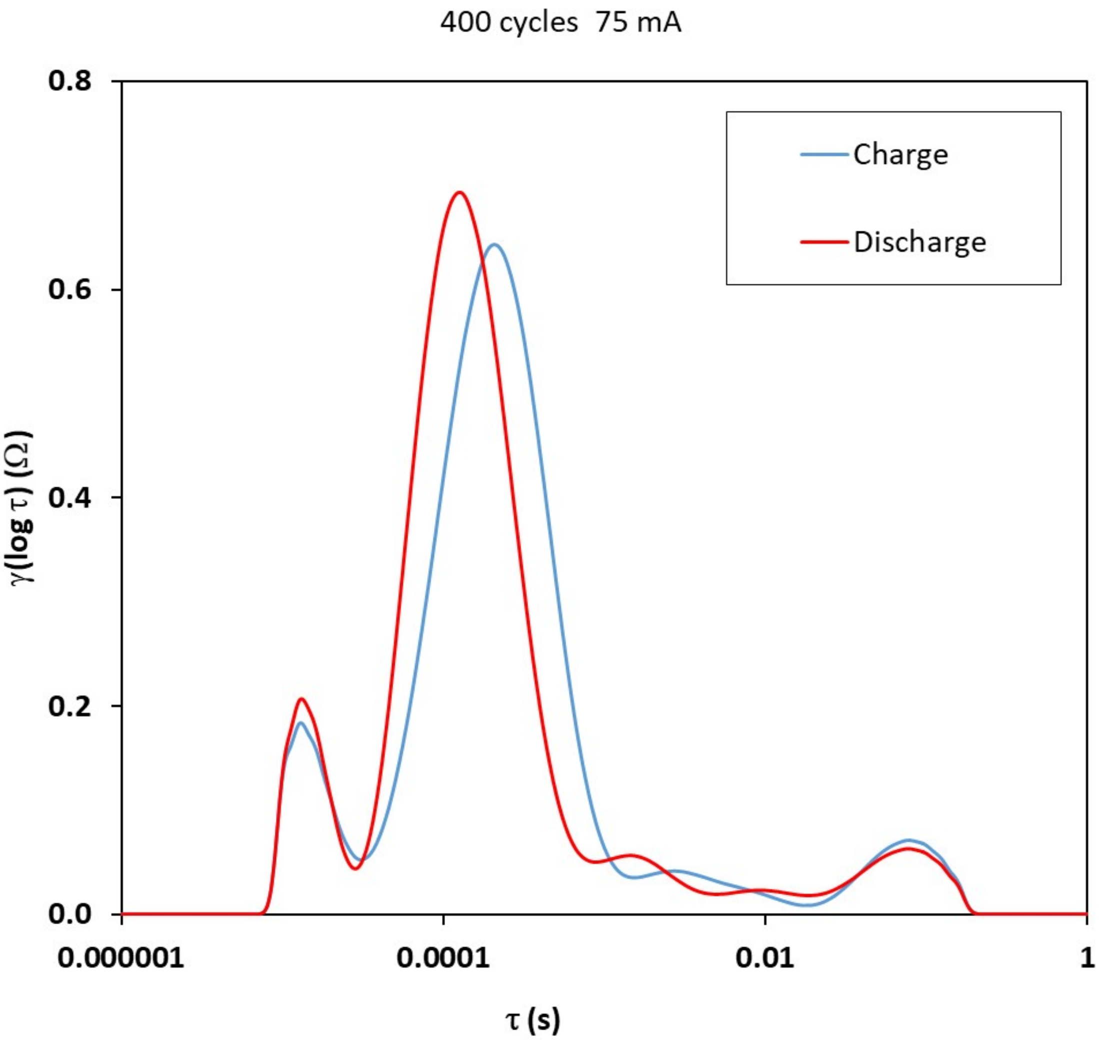
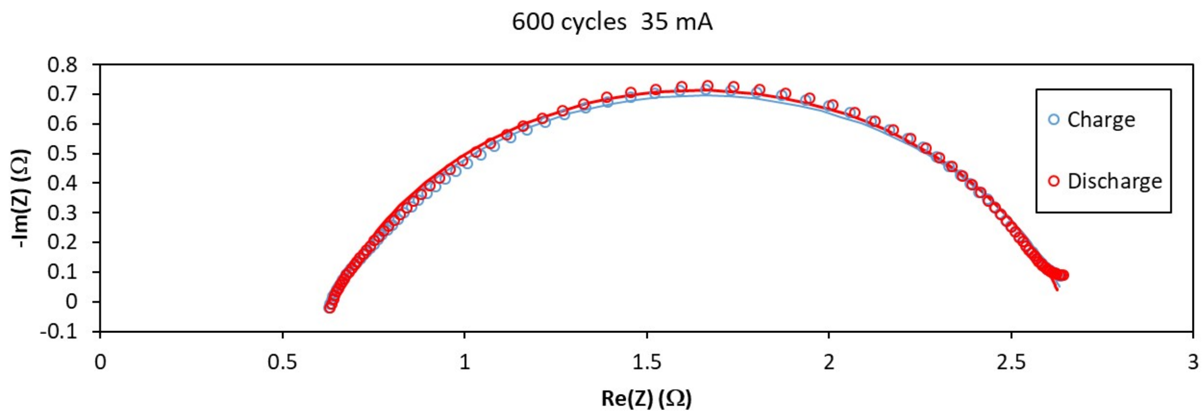


Figure S7i. Nyquist-plots obtained by EIS and the corresponding DRT spectra of a single zinc–O_2(ads) battery cell recorded after 400 cycles at a DC current of 75 mA during charging and discharging. The solid lines represent the fitted impedance data using the ECM shown in the inset of Fig. 4a in the article.



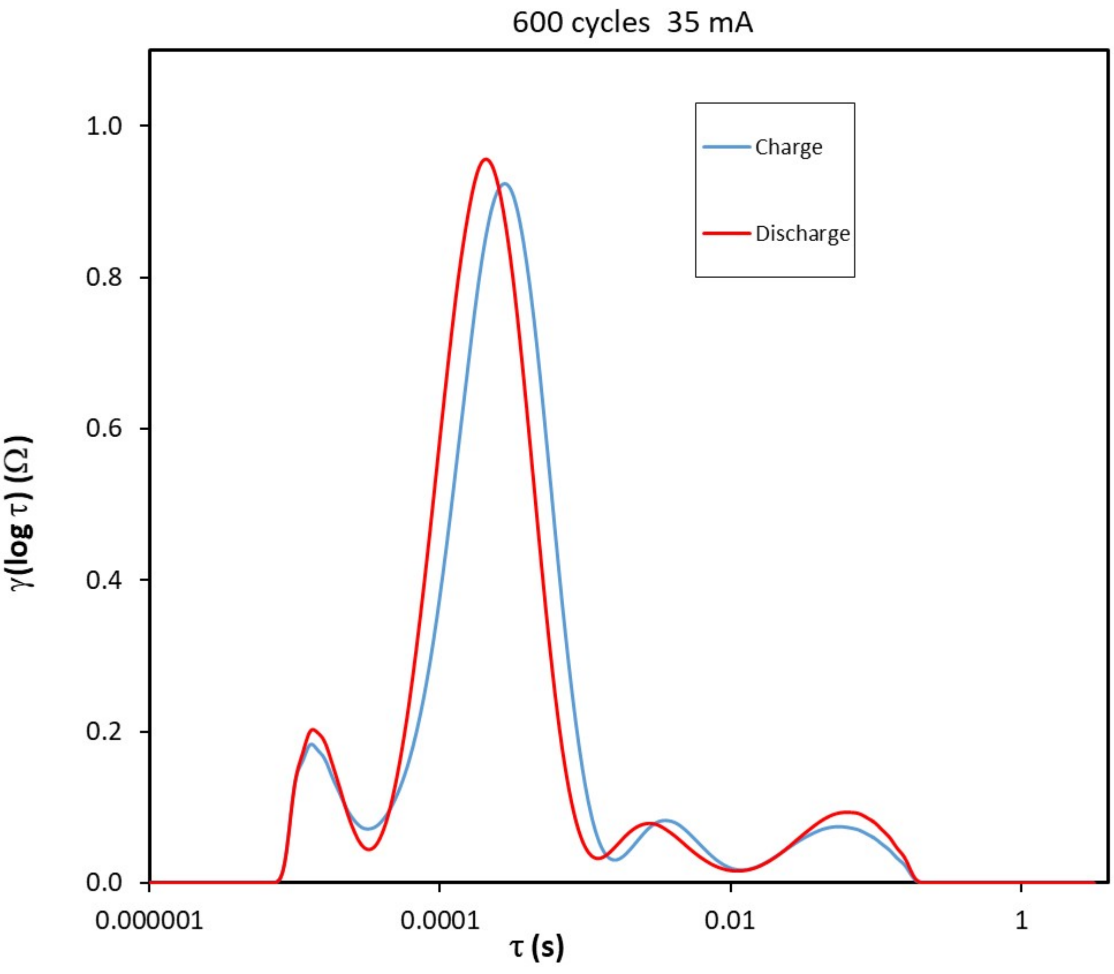


Figure S7j. Nyquist-plots obtained by EIS and the corresponding DRT spectra of a single zinc–O_2(ads) battery cell recorded after 600 cycles at a DC current of 35 mA during charging and discharging. The solid lines represent the fitted impedance data using the ECM shown in the inset of Fig. 4a in the article.

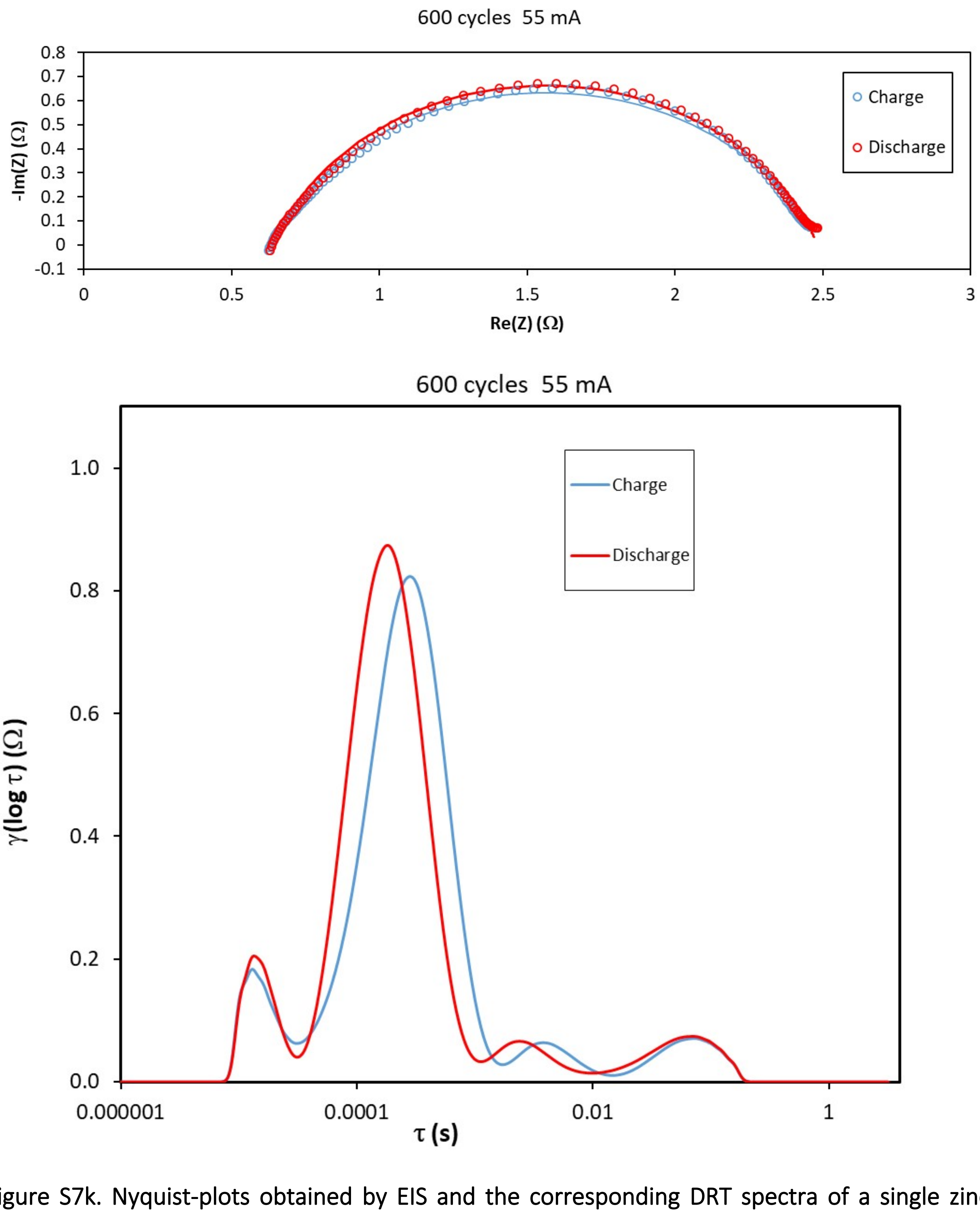
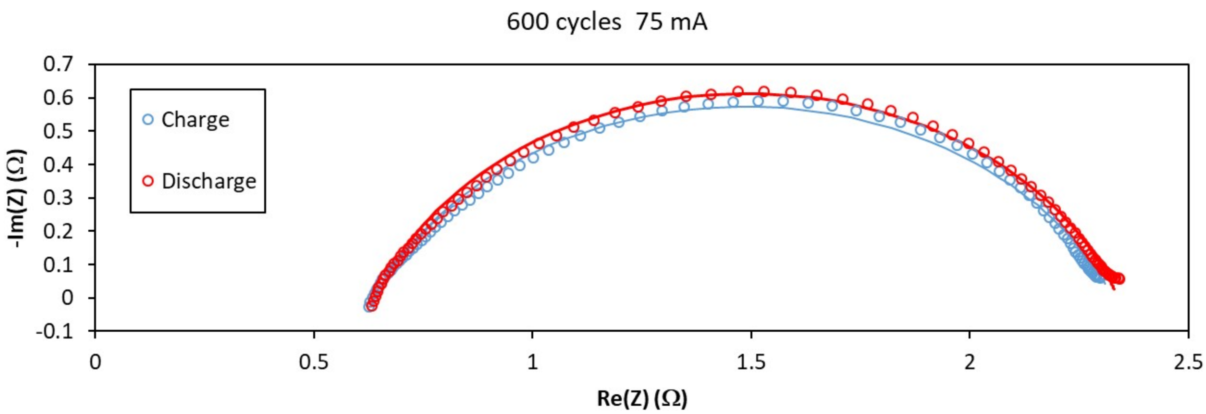


Figure S7k. Nyquist-plots obtained by EIS and the corresponding DRT spectra of a single zinc–O_2(ads) battery cell recorded after 600 cycles at a DC current of 55 mA during charging and discharging. The solid lines represent the fitted impedance data using the ECM shown in the inset of Fig. 4a in the article.



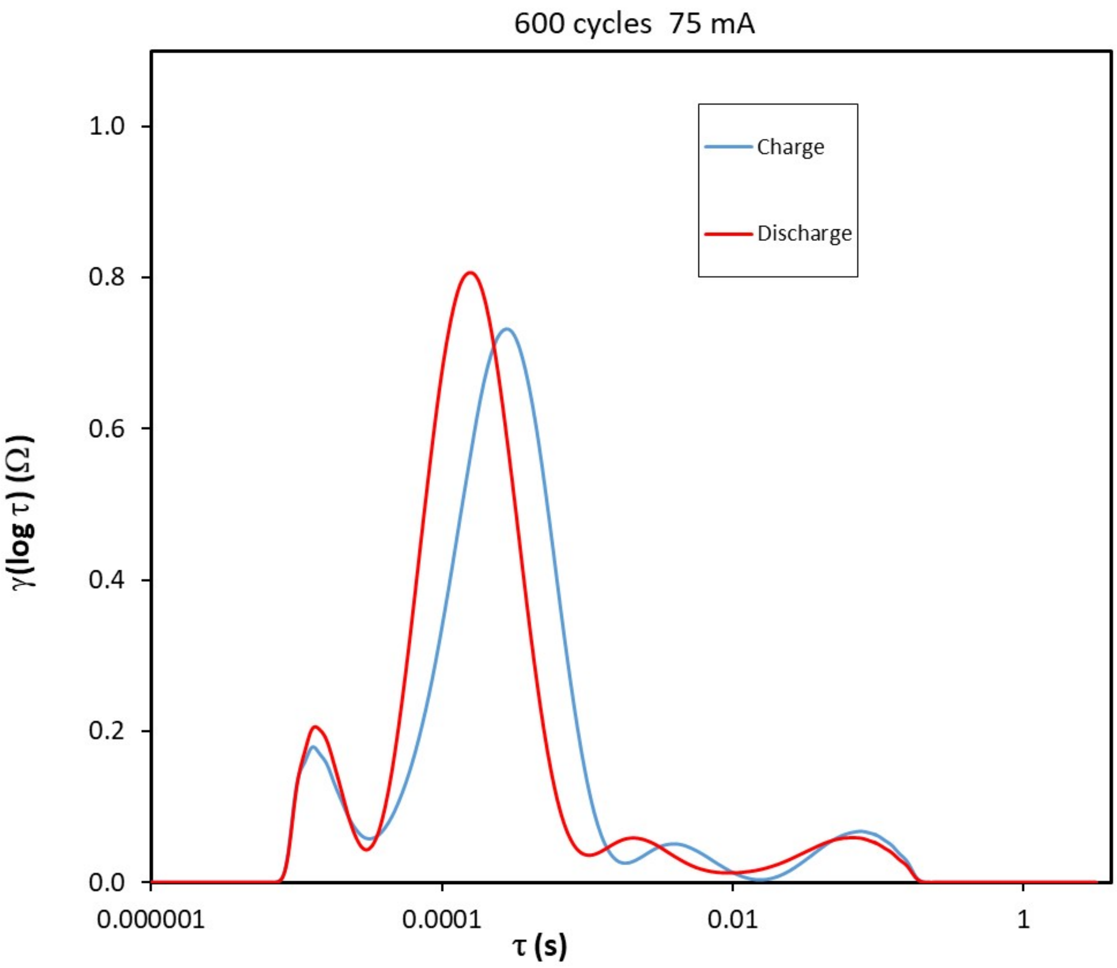


Figure S7l. Nyquist plots obtained by EIS and the corresponding DRT spectra of a single zinc–O_2(ads) battery cell recorded after 600 cycles at a DC current of 75 mA during charging and discharging. The solid lines represent the fitted impedance data using the ECM shown in the inset of Fig. 4a in the article.

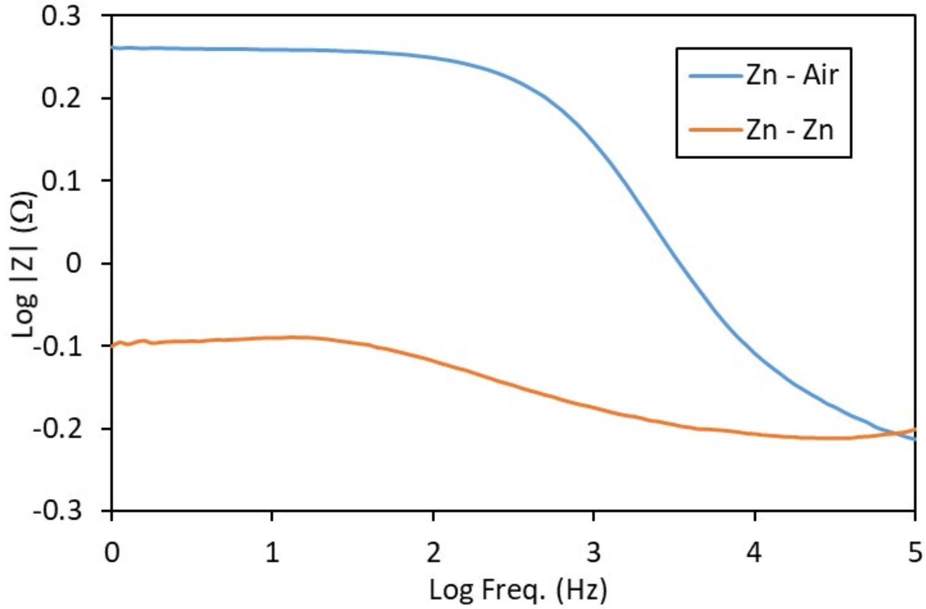


Figure S8. Amplitude Bode-plot comparing the impedances of a Zinc-air cell with that of a Zinc-Zinc cell. (New cell, at 55 mA charge DC.)

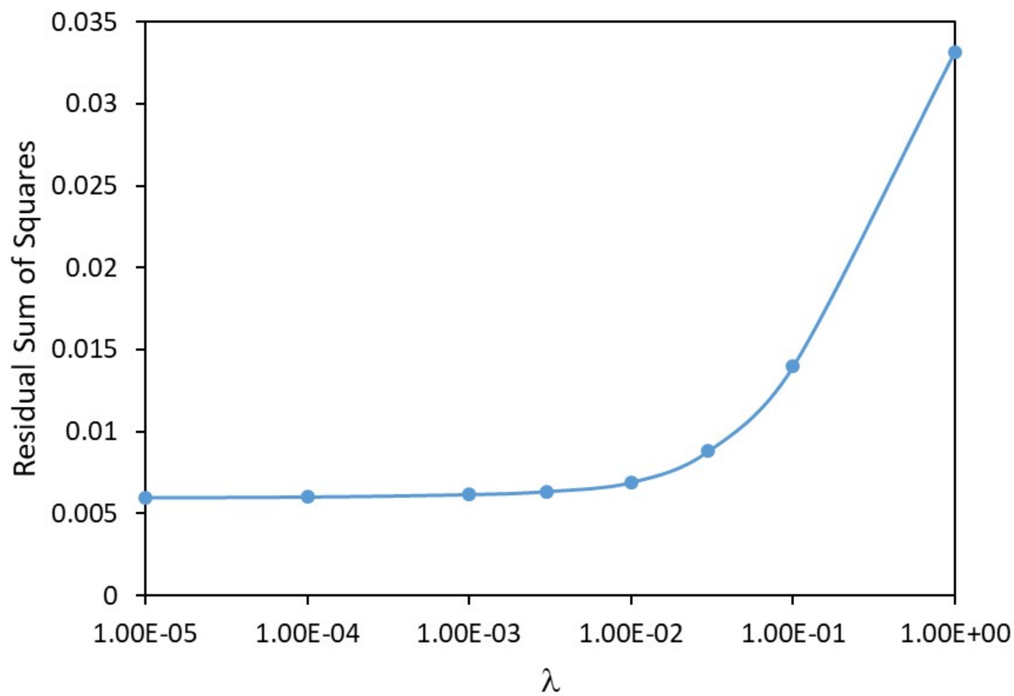


Figure S9 Sum of square residuals of the DRT regression versus the regularization parameter λ used within the DRT calculation. (Recorded after 200 cycles, at 55 mA charge DC.)

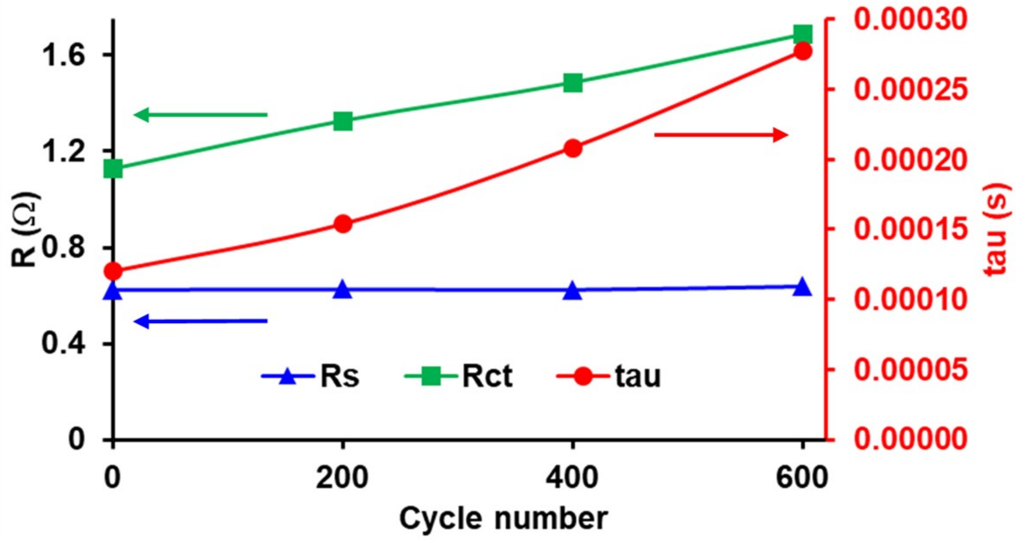


Figure S10 Plot of changing of ohmic resistance (R_s), charge transfer resistance (R_{ct}) and the time constant (tau, τ) as a function of cycle number recorded at 75 mA