Hydrophilic and nanocrystalline carbon quantum dots enable highly reversible zinc-ion batteries

Shuhua Yang[a,*], Zenglong Xu[a], Song Wang[a], Jinfeng Sun[a], Degang Zhao[a], Bingqiang Cao[a], Xiutong Wang[b]

[a] Materials Center for Energy and Photoelectrochemical Conversion, School of Material Science and Engineering, University of Jinan, Jinan 250022, China

[b] Key Laboratory of Marine Environmental Corrosion and Bio-fouling, Institute of Oceanology and Center for Ocean Mega-Science, Chinese Academy of Sciences, Qingdao 266071, China.

*Corresponding author:

E-mail: yangshuhua78@163.com (Shuhua Yang)

1. Experimental Details

Preparation of MnO_2 Cathode

MnO_2 cathode was prepared by DH7000 electrochemical workstation. Using stainless steel mesh as the substrate, saturated calomel electrode and platinum sheet are used as reference electrode and counter electrode, respectively. A constant potential of -1.8 V is applied to the working electrode to prepare a MnO_2 cathode in a solution consisting of 0.1 M $MnC_4H_6O_4$ and 0.1 M $NaSO_4$.[1]

2. Supplementary Figures

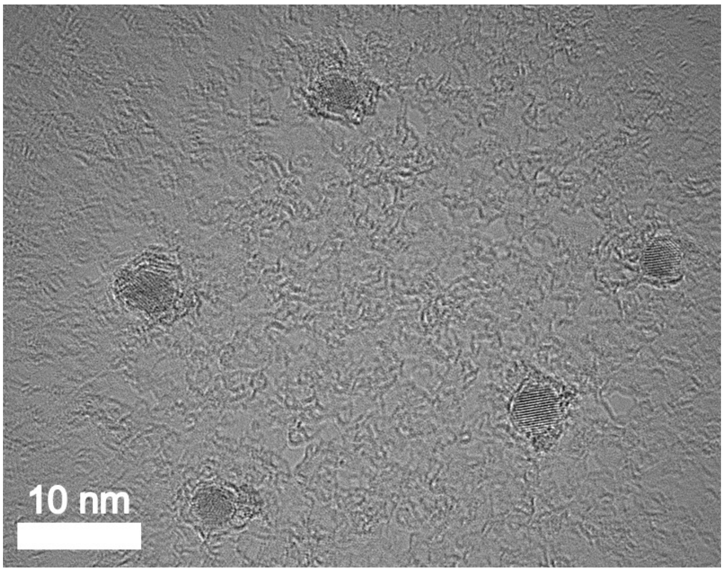

Fig. S1 TEM of CQDs.

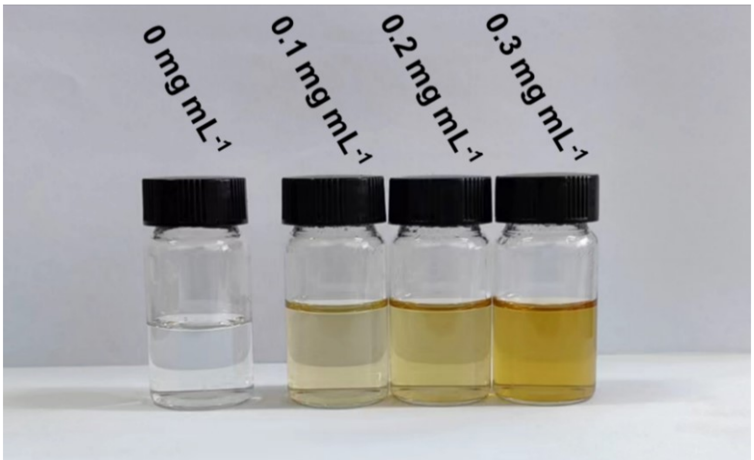

Fig. S2 Optical images of 2 M $ZnSO_4$ electrolytes at different concentrations of CQDs.

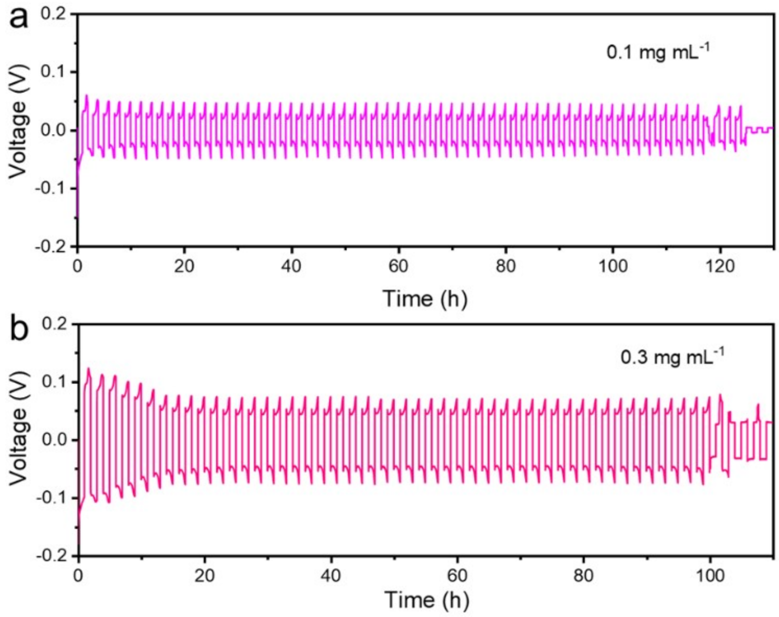

Fig. S3 cycling performance of Zn//Zn cells with (a) 0.1 mg mL^{-1} and (b) 0.3 mg mL^{-1}.

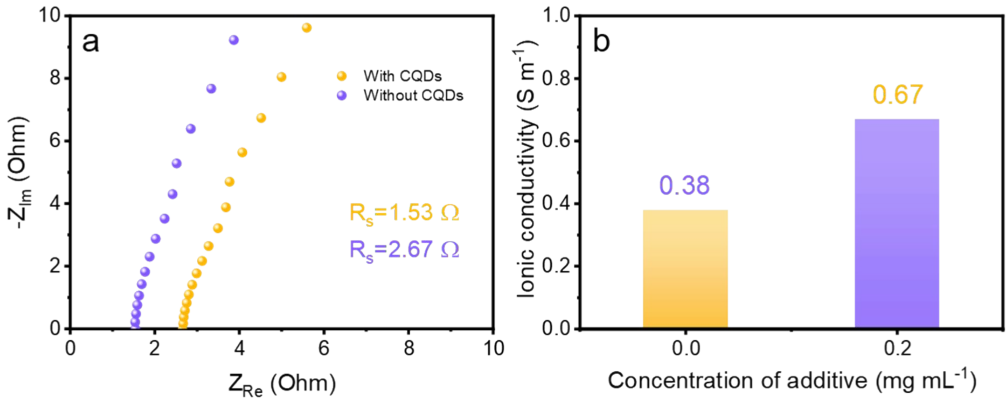

Fig. S4 (a) Nyquist plots of the SS symmetric cells in the $ZnSO_4$ electrolytes with/without CQDs. (b)The ionic conductivities of $ZnSO_4$ electrolytes with/without CQDs calculated from EIS results.

Ionic conductivities of various electrolytes were tested by the stainless steel (SS) symmetric cells and calculated from the equation[2]

$\sigma = L/(R_s \cdot S)$.

Where L represents the distance between two SS electrodes, R_s represents the resistance from EIS results, and S is the area of electrodes. In this work, the contact area between electrode and electrolyte is ~1.96 cm^{-2} and the distance between two SS electrodes is ~200 μm.

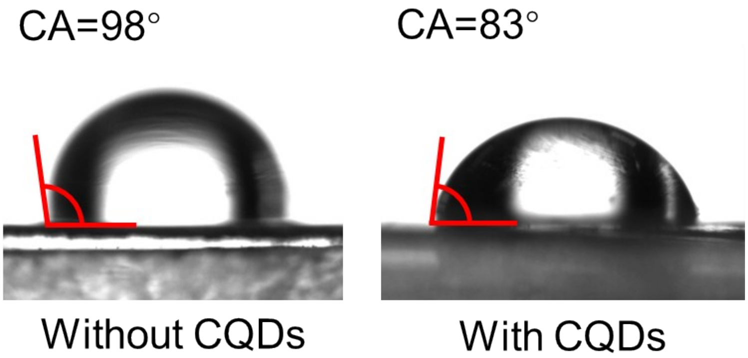

Fig. S5 The contact angle between $ZnSO_4$ electrolyte with/without CQDs and Zn foil.

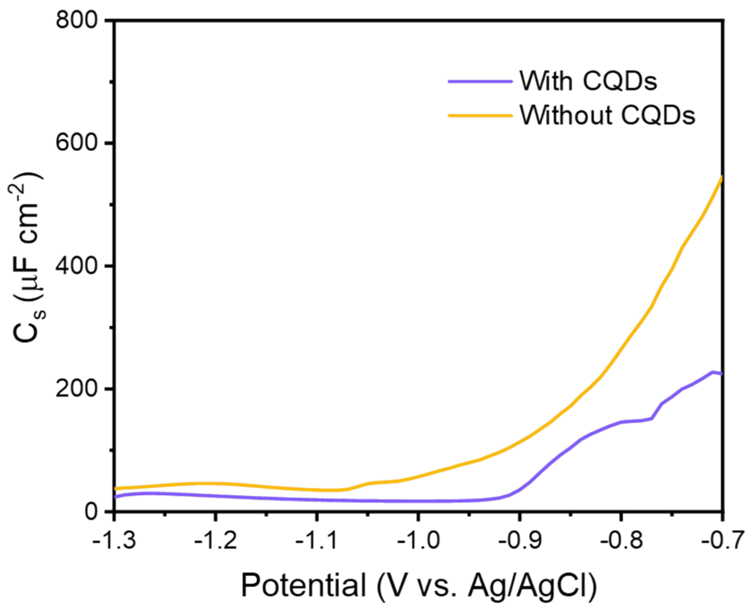

Fig. S6 Differential capacitance curve for Zn in Na_2SO_4 solution with/without CQDs additive.

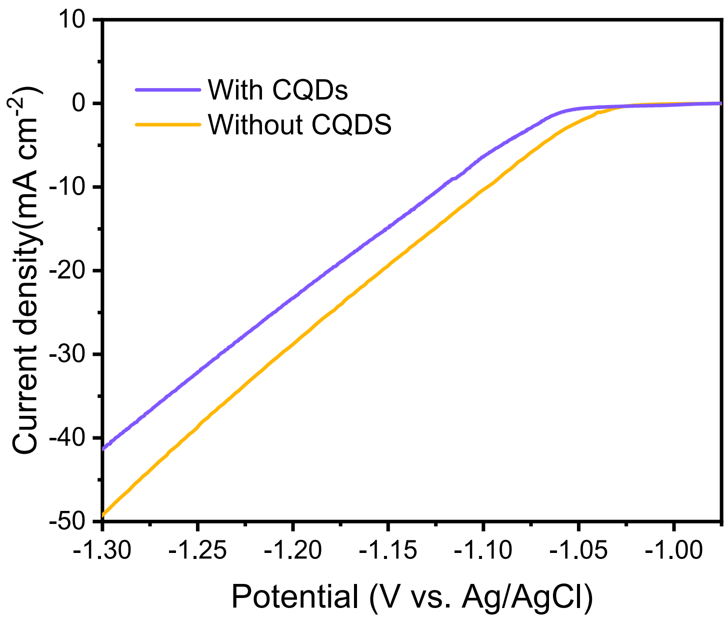

Fig. S7 LSV curves of Zn foil tested in $ZnSO_4$ electrolyte with/without CQDs.

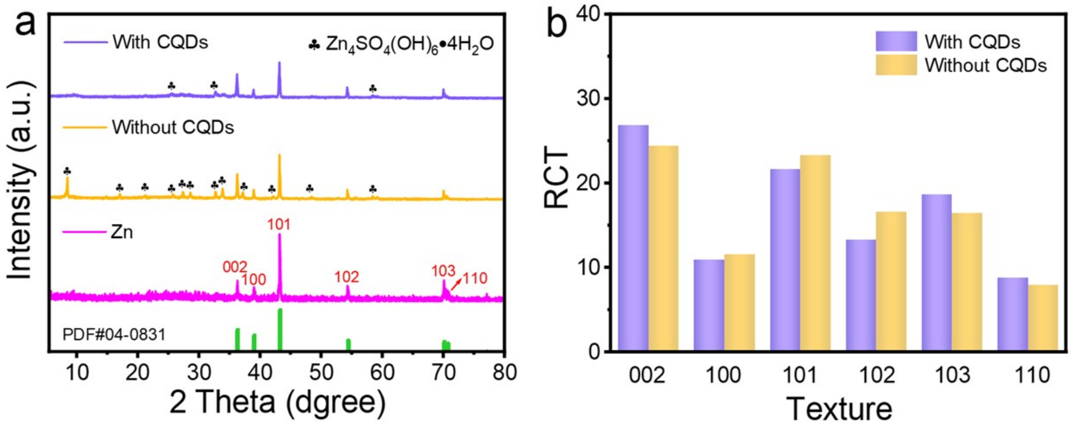

Fig. S8 (a)The X-ray diffraction (XRD) patterns of Zn anodes before cycling and after 100 hours of cycling and (b) Histograms for the corresponding fitted RTC.

The relative texture coefficients (RTCs) of each Zn lattice plane are calculated using the following formula[3]

$$RCT_{(hkl)} = \frac{I_{(hkl)}/I_{0(hkl)}}{\Sigma\left(I_{(hkl)}/I_{0(hkl)}\right)} * 100$$

where $I_{(hkl)}$ is the intensity obtained from the textured sample, and $I_{0(hkl)}$ is the intensity of the standard sample.

The relative texture coefficients (RTCs) of the Zn lattice planes of the Zn anode after cycling were calculated. The results indicate that the optimal crystallization direction after adding CQDs is along (002) crystal face.

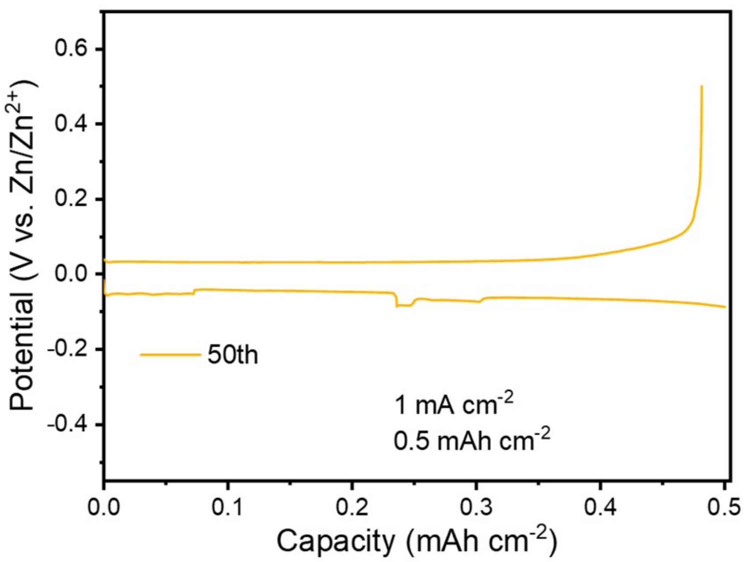

Fig. S9 Voltage-capacity curves of Zn//Cu cell without CQDs during the 50th cycle at 1 mA cm^{-2} and 0.5 mAh cm^{-2}.

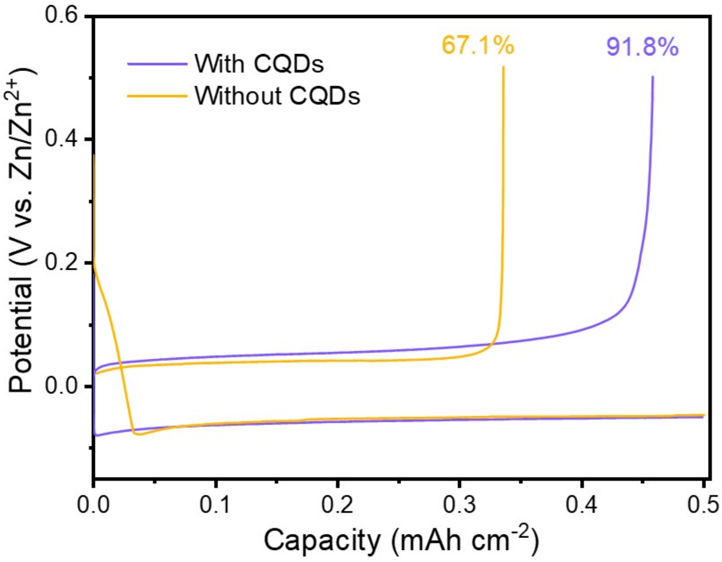

Fig. S10 Initial Coulomb efficiency of Zn//Cu cell with and without CQDs at 1 mA cm^{-2} and 0.5 mAh cm^{-2}.

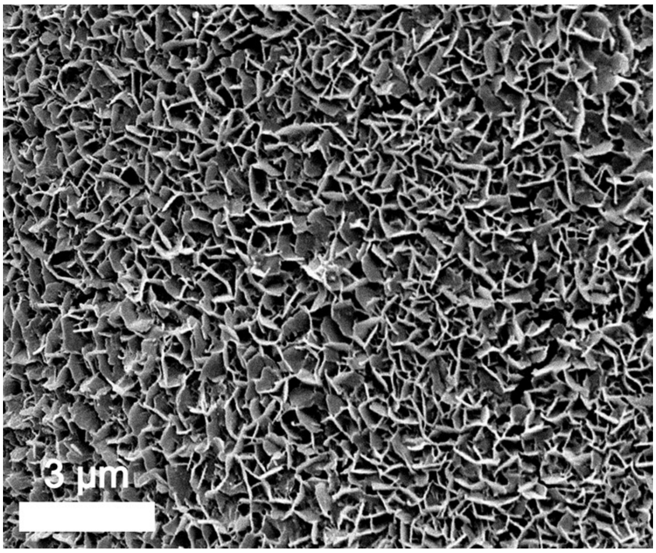

Fig. S11 SEM of MnO_2

The MnO_2 electrode is obtained by the electrochemical deposition method. The mass of MnO_2 loaded on the cathode is ~1.0 mg, and the current collector is a stainless-steel mesh.

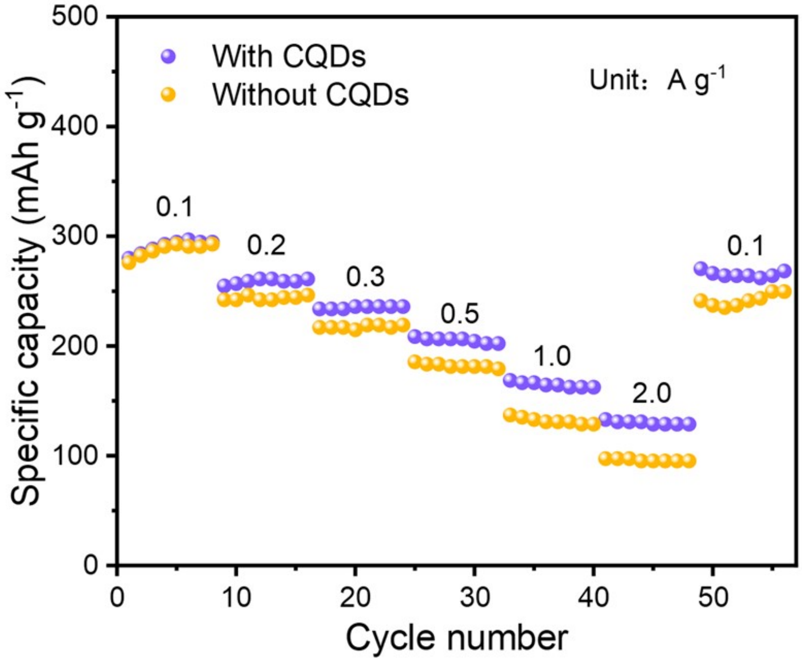

Fig. S12 Rate performances of Zn//MnO_2 battery at different current densities range from 0.1to 2.0 A g^{-1}.

Table S1. Comparison between our Zn//Zn batteries with previous reported Zn//Zn batteries with various electrolyte additives.

Electrolyte additives	Electrolyte	Current density	Capacity	Life	Reference
CQDs	2 M $ZnSO_4$	1 mA cm^{-2}	1 mAh cm^{-2}	1070 h	**This work**
		4 mA cm^{-2}	4 mAh cm^{-2}	810 h	
Na_2SO_4	2 M $ZnSO_4$	0.2 mA cm^{-2}	-	300 h	[4]
$MgSO_4$	2 M $ZnSO_4$	1 mA cm^{-2}	0.25 mAh cm^{-2}	600 h	[5]
NaF	1 M $ZnSO_4$	0.25 mA cm^{-2}	0.5 mAh cm^{-2}	368 h	[6]
TH	3 M $ZnSO_4$	1 mA cm^{-2}	1 mAh cm^{-2}	580 h	[7]
		5 mA cm^{-2}	1 mAh cm^{-2}	700 h	
Nano-Si	1 M $ZnSO_4$	5 mA cm^{-2}	1 mAh cm^{-2}	500 h	[8]
GO	2 M $ZnSO_4$	1 mA cm^{-2}	0.5 mAh cm^{-2}	650 h	[9]
Et_2O	3 M $Zn(CF_3SO_3)_2$	0.2 mA cm^{-2}	0.2 mAh cm^{-2}	250 h	[10]
TBA_2SO_4	0.2 M $ZnSO_4$	5 mA cm^{-2}	2 mAh cm^{-2}	456 h	[11]
NSQDs	2 M $ZnSO_4$	20 mA cm^{-2}	1 mAh cm^{-2}	600 h	[12]
LiCl	3 M $ZnSO_4$	0.2 mA cm^{-2}	-	170 h	[13]

Table S2. Fitting results of the Nyquist plots of Zn||Zn cells without and with CQDs.

Electrolyte	R_s (Ω)	R_{ct} (Ω)
Without CQDs	17.8	322
With CQDs	15.7	220.1

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