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| Abstract |  |
| Manganese (Mn)-based cathode materials have garnered huge research  interest for rechargeable aqueous zinc-ion batteries (AZIBs) due to the  abundance and low cost of manganese and the plentiful advantages of  manganese oxides including their dierent structures, wide range of phases,  and various stoichiometries. A novel in situ generated Mn-deﬁcient  ZnMn    O    @C (Mn-d-ZMO@C) nanoarchitecture cathode material from  self-assembly of ZnO-MnO@C for rechargeable AZIBs is reported. Analytical  techniques conﬁrm the porous and crystalline structure of ZnO-MnO@C and  theinsitugrowthofMndeﬁcientZnMn    O    @C. The Zn/Mn-d-ZMO@C cell  displays a promising capacity of  mAh g  −  at a current density of   mA g  −  with % of capacity retained after  cycles (at  mA g  −  rate). The improved performance of this cathode originates from in situ  orientation, porosity, and carbon coating. Additionally, ﬁrst-principles  calculations conﬁrm the high electronic conductivity of Mn-d-ZMO@C  cathode. Importantly, a good capacity retention (%) is obtained with a  year-old cell (after  cycles) at  mA g  −  current density. This study,  therefore, indicates that the in situ grown Mn-d-ZMO@C nanoarchitecture  cathode is a promising material to prepare a durable AZIB  Manganese (Mn)-based cathode materials have garnered huge research  interest for rechargeable aqueous zinc-ion batteries (AZIBs) due to the  abundance and low cost of manganese and the plentiful advantages of  manganese oxides including their dierent structures, wide range of phases,  and various stoichiometries. A novel in situ generated Mn-deﬁcient  ZnMn    O    @C (Mn-d-ZMO@C) nanoarchitecture cathode material from  self-assembly of ZnO-MnO@C for rechargeable AZIBs is reported. Analytical  techniques conﬁrm the porous and crystalline structure of ZnO-MnO@C and  theinsitugrowthofMndeﬁcientZnMn    O    @C. The Zn/Mn-d-ZMO@C cell  displays a promising capacity of  mAh g  −  at a current density of   mA g  −  with % of capacity retained after  cycles (at  mA g  −  rate). The improved performance of this cathode originates from in situ  orientation, porosity, and carbon coating. Additionally, ﬁrst-principles  calculations conﬁrm the high electronic conductivity of Mn-d-ZMO@C  cathode. Importantly, a good capacity retention (%) is obtained with a  year-old cell (after  cycles) at  mA g  −  current density. This study,  therefore, indicates that the in situ grown Mn-d-ZMO@C nanoarchitecture  cathode is a promising material to prepare a durable AZIB  Manganese (Mn)-based cathode materials have garnered huge research interest for rechargeable aqueous zinc-ion batteries (AZIBs) due to the abundance and low cost of manganese and the plentiful advantages of manganese oxides including their different structures, wide range of phases, and various stoichiometries. A novel in situ generated Mn-deficient ZnMn2O4@C (Mn-d-ZMO@C) nanoarchitecture cathode material from self-assembly of ZnO-MnO@C for rechargeable AZIBs is reported. Analytical techniques confirm the porous and crystalline structure of ZnO-MnO@C and the *in situ* growth of Mn deficient ZnMn2O4@C. The Zn/Mn-d-ZMO@C cell displays a promising capacity of 194 mAh g−1 at a current density of 100 mA g−1 with 84% of capacity retained after 2000 cycles (at 3000 mA g−1 rate). The improved performance of this cathode originates from in situ orientation, porosity, and carbon coating. Additionally, first-principles calculations confirm the high electronic conductivity of Mn-d-ZMO@C cathode. Importantly, a good capacity retention (86%) is obtained with a year-old cell (after 150 cycles) at 100 mA g−1 current density. This study, therefore, indicates that the in situ grown Mn-d-ZMO@C nanoarchitecture cathode is a promising material to prepare a durable AZIB. | |