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**Theoretical Studies of Zincate Contamination of  $\gamma$ -MnO<sub>2</sub> in Deep-Cycled Rechargeable Zn/MnO<sub>2</sub> Batteries** NIRAJAN PAUDEL, BIRENDRA ALE MAGAR, Department of Physics, New Mexico State University, Las Cruces, New Mexico 88003, USA, TIMOTHY LAMBERT, Department of Materials, Devices, and Energy Technologies, Sandia National Laboratories, Albuquerque, New Mexico 87185, USA, IGOR VASILIEV, Department of Physics, New Mexico State University, Las Cruces, New Mexico 88003, USA — Rechargeable alkaline Zn/MnO<sub>2</sub> batteries are attractive for large-scale energy storage because of their high energy density, non-toxicity, and low cost. However, efforts to develop rechargeable Zn/MnO<sub>2</sub> batteries have been hindered by a short cycle life due to the accumulation of irreversible redox reaction products in the  $\gamma$ -MnO<sub>2</sub> cathode. The penetration of Zn ions into the  $\gamma$ -MnO<sub>2</sub> electrode leads to the formation of hetaerolite (ZnMn<sub>2</sub>O<sub>4</sub>). The contamination of the  $\gamma$ -MnO<sub>2</sub> cathode material with hetaerolite reduces the battery capacity and eventually leads to the failure of the battery. We apply *ab initio* computational methods based on density functional theory to calculate the structure and formation energy of ZnMn<sub>2</sub>O<sub>4</sub> using several different gradient corrected exchange-correlation functionals, including PBE, PBESol, and PBE+U. Our calculations show that the PBE and PBESol functionals tend to underestimate the formation energy of hetaerolite, whereas the PBE+U functional significantly improves agreement with experiment. Using the results of our calculations, we analyze the influence of hetaerolite on the performance and cycle life of rechargeable Zn/MnO<sub>2</sub> batteries.

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