

Abstract Submitted  
for the MAR11 Meeting of  
The American Physical Society

**First-principles study of the structure of  $\text{RuO}_2 \cdot x\text{H}_2\text{O}$**  FEI ZHOU, YONGDUO LIU, UCLA, MARK ASTA, UC Berkeley, VIDVUDS OZOLINS, UCLA — Hydrous ruthenia,  $\text{RuO}_2 \cdot x\text{H}_2\text{O}$ , is a high-performance electrode material for electrochemical supercapacitors. Two different structural models of hydrous ruthenia had been proposed. In one of them, hydrogen is incorporated in metal vacancies inside the oxide host ( “bulk model”), while in the other model structural water associated with Ru-O occupies the region between rutile nanograins ( “core + grain-boundary model”). We present a theoretical examination of the validity of the bulk model by optimizing hydrogen positions within  $\text{RuO}_2$  with proton-compensated Ru vacancies using a combination of a systematic search algorithm based on electrostatics, database searching and density-functional theory calculations. We find that all the considered bulk model structures are unstable by  $\sim 0.3 - 0.4$  eV per  $\text{H}_2\text{O}$  molecule with respect to phase separation into anhydrous  $\text{RuO}_2$  and water. Structures with hydroxyl groups or aggregate  $\text{H}_2\text{O}$  are significantly lower in energy (though still unstable with respect to phase separation), demonstrating that the water prefers to agglomerate outside  $\text{RuO}_2$ . Our results strongly disfavor the bulk model with hydrogen inside  $\text{RuO}_2$  and support the core+grain-boundary model of hydrous ruthenia.

Fei Zhou  
UCLA

Date submitted: 23 Dec 2010

Electronic form version 1.4