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Directed synthesis of $Mn_xTi_{1-x}O_2$ tunnel structured materials for energy applications TIM DROUBAY, ANNE CHAKA, SEBASTIEN KERISIT, EUGENE ILTON, Pacific Northwest National Laboratory — Mn oxides with tunnel structures are crucial in technological applications such as Li batteries, catalysis, fuel cells, electrochemical capacitors, sensors, and groundwater remediation. However, the complexity and poor quality of natural Mn oxide has hindered efforts to understand their fundamental structure-property relationships. To address this issue, we used PLD to make high-quality $Mn_xTi_{1-x}O_{2-\delta}$ single-crystal films. Attempts to synthesize pure β-MnO_2 thin films on TiO_2 substrates resulted in Mn_2O_3 dominant films. Results of ab initio thermodynamics to explain film stability as a function of growth conditions and Mn/Ti composition suggest that "protecting" the Mn in a TiO_2 matrix by co-deposition would be beneficial. This approach has met with initial success even though the resultant films have oxygen vacancies. XPS indicates that (110) oriented films are Mn-rich near the surface while (001)-oriented films are Mn-rich near the interface. XRD shows that the films are coherently strained to the substrate which may influence the oxygen non-stoichiometry. Aberration corrected TEM results corroborate the XPS and XRD results and indicate a potential Mn-dependent defect. These films will be discussed along with multilayered $(MnO_2)_m$-$(TiO_2)_n$ films. Atomistic modeling shows that alternating cation rows that only contained Ti or Mn (m,n = 1) greatly lower the activation energy for Li diffusion relative to films where Mn and Ti were homogeneously mixed.

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