

## HYDROGEN PERMEATION THROUGH SURFACE OXIDES OF TITANIUM IRON ALLOYS

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The complex and heterogenous native surface oxide that forms on titanium and titanium-iron alloys (TiFe) is acknowledged to be critical for determining rates of initial hydride formation in hydrogen-rich environments.<sup>1</sup> However, the precise nature of its interactions with hydrogen, as well as the specific features that promote or inhibit hydrogen penetration into the underlying matrix, remain subjects of controversy. Here, we use *ab initio* simulations of diverse compositions and configurations of the surface oxide as the basis for multiscale modeling to study the hydrogen permeability of the oxide layer. We explicitly consider stoichiometries that have been observed experimentally, such as  $\text{TiFeO}_3$ ,  $\text{Ti}_2\text{FeO}_5$ ,  $\text{TiFe}_2\text{O}_4$ , and  $\text{Ti}_4\text{Fe}_2\text{O}$ .<sup>2</sup> Along with simulations of the bulk crystalline phases, we use *ab initio* molecular dynamics to construct disordered and highly defective structures to capture a wide variety of local environments for hydrogen. We take various effects, such as strain, oxygen deficiency, and the presence of such minority alloying elements as Mn and V, into consideration, and we identify how they impact the energetics and kinetics of hydrogen incorporation.

Our work provides a comprehensive analysis of the key factors governing hydrogen permeation through the surface oxide layer and, ultimately, metal hydride formation kinetics in TiFe with and without minority alloying elements. In addition, we demonstrate a fundamental change in the behavior of hydrogen as it passes through the oxide, from protonic character in higher atomic layers to hydridic character closer to the TiFe interface. By characterizing the role of the surface oxide from the atomic scale, we provide insights into the factors most important for dictating kinetics of hydriding of the underlying TiFe matrix.

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