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First-principles study of oxygen reduction reaction on the tetragonal ZrO_2(101) surface YOSHIYUKI YAMAMOTO, SHUSUKE KASAMATSU, OSAMU SUGINO, The Institute for Solid State Physics, The University of Tokyo — ZrO_2 has attracted attention as a durable and inexpensive electrocatalyst for future polymer electrolyte fuel cells [1]. Its oxygen reduction reaction (ORR) activity has been made comparable to platinum by introducing oxygen vacancies (Vos) and impurities, such as C and N atoms, but their role is unidentified experimentally. Theoretically, elucidating the ORR activity at a defective oxide surface is a major challenge. In this context, we attacked this problem with first-principles density functional calculations of the tetragonal ZrO_2(101) surface adsorbed with water molecules [2]. By investigating various adsorbed configurations, we have found that the surface structure is much more complex than that thought based on a previous calculation [3]. We also found that the pristine surface is not reactive against ORR because of too weak adsorptions of intermediates. On the contrary, the surface becomes reactive when introduced with a Vo and two N atoms at the surface according to our free-energy diagram of ORR. Our calculation also suggests that the defective surface is activated at the surface Vo but not at the surface Zr site. [1] K. Ota et al., ECS Transaction, 45 27(2012). [2] Y. Yamamoto et al., in preparation. [3] A. Hofmann et al., J. Phys. Chem. B, 108 14652(2004).

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