

Abstract Submitted  
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**Carbon-based nanostructured materials for enhanced H<sub>2</sub> production** M.K. KOSTOV, E.E. SANTISO, A.M. GEORGE, K.E. GUBBINS, M.B. NARDELLI, North Carolina State University — A key fundamental limit of the thermal splitting of bulk water is the fact that the ground state of oxygen is paramagnetic, whereas the ground state of water is diamagnetic. Here, we propose to explore a new paradigm in H<sub>2</sub> production: a process in which the system remains on the spin singlet potential surface throughout the reaction, by exploiting the catalytic role of defective carbon substrates. Using first principles modeling techniques, we found evidence that mono-vacancy defects in graphite and carbon nanotubes give rise to a rich chemistry, yielding many possible water dissociation pathways, some of which have activation barriers lower than half the value for the dissociation of bulk water. This reduction is caused by spin selection rules that allow the system to remain on the same spin surface throughout the reaction. These novel reactions enhance the hydrogen yield and the reaction rate. In the presence of water only, this reaction is self-limiting: when all of the defects are oxidized, the reaction is complete, and no further H<sub>2</sub> is produced. There are several possibilities to achieve regeneration of the active surface sites, such as photo-excitation, vibrational excitations or further reaction with other molecules. We will discuss this exploration in the context of a complete cycle of energy storage and release through the production of H<sub>2</sub>.

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