Pyrite (FeS2) is the most abundant and widespread sulfide mineral in the Earth’s surface and in deep-see vents. In the Iron-Sulfur World scenario,1 metal sulfides (including pyrite) under hydrothermal connditions were suggested to play an important catalytic role in the synthesis of simple building blocks of life from simple molecules. Among a variety of simple chemical systems that have been investigated as prebiotic precursor, formamide (NH2CHO) has attracted a lot of interest due to several reasons. First, formamide has four important elements (N, C, O, H) for the synthesis of biomolecules. Second, formamide is an abundant molecule in the universe and its chemistry is closely related to the chemistry of HCN, a famous prebiotic precursor.2 And last but not least, formamide can decompose to form a wide range of low-molecular-weight products which facilitates the diversity of the synthesis pathways.3 Answer to the need of theoretical investigations of catalytic effects at molecular level, we set out to perform theoretical calculations of reactions of formamide on the (100) surface of pyrite. The density-functional theory (DFT) method with a plane wave-pseudopotential basis was used to study possible adsorption complexes of formamide on the ideal and sulfur-vacancy defect surfaces. Several hydrogen transfer reactions transforming formamide into its tautomers, formimic acid (NHCHOH) and aminohydroxylmethylene (NH2COH), were investigated in details. The DFT results suggest that the unimolecular hydrogen transfer reactions of formamide have high energy barrier (44-78 kcal/mol) on both ideal and defect surface. However, the reaction barriers significantly reduce with the presence of one water molecule. Reaction barriers of less than 20 kcal/mol were found for several water-assisted tautomerizations on the ideal and defect surfaces.

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