

Description of Additional Supplementary Files

File Name: Supplementary Movie 1

Description: **MD simulation trajectory showing the interaction of SNO with gluconolactone and FADH₂.** MD trajectory highlights the conformational dynamics of FADH₂ before it approaches the SNO surface. The spheres here represent nickel (green), samarium (purple), oxygen (red), nitrogen (blue), carbon (grey) and hydrogen (white).

File Name: Supplementary Movie 2

Description: **AIMD movie showing the spontaneous hydrogen transfer mechanism.** The hydrogen transfer is facile and occurs within a fraction of a picosecond. Note that the SNO slab is held fixed to reduce the computational cost. The spheres here represent nickel (green), samarium (purple), oxygen (red), nitrogen (blue), carbon (grey) and hydrogen (white)

File Name: Supplementary Movie 3

Description: **AIMD movie showing the absence of hydrogen transfer due to steric effects.** The conformation of the FADH₂ molecule plays a key role in dictating the hydrogen transfer. As one example, in this movie, we observe that the molecule undergoes conformational change and does not dehydrogenate with the AIMD timescales. Note that the SNO slab is held fixed to reduce the computational cost. The spheres here represent nickel (green), samarium (purple), oxygen (red), nitrogen (blue), carbon (grey) and hydrogen (white).