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Force-activated reactivity switch in a bimolecular chemical reaction at the single molecule level ROBERT SZOSZKIEWICZ, Kansas State University, SERGI GARCIA-MANYES, JIAN LIANG, TZU-LING KUO, JULIO M. FERNANDEZ, Columbia University, NY — Mechanical force is a distinct and usually less explored way to activate chemical reaction because it can deform the reacting molecules along a well-defined direction of the reaction coordinate. However, the effect of mechanical force on the free- energy surface that governs a chemical reaction is still largely unknown. The combination of protein engineering with single-molecule force-clamp spectroscopy allows us to study the influence of mechanical force on the rate at which a protein disulfide bond is reduced by some reducing agents in a bimolecular substitution reaction (so-called SN2). We found that cleavage of a protein disulfide bond by hydroxide anions exhibits an abrupt reactivity "switch" at 500 pN, after which the accelerating effect of force on the rate of an SN2 chemical reaction greatly diminishes. We propose that an abrupt force- induced conformational change of the protein disulfide bond shifts its ground state, drastically changing its reactivity in SN2 chemical reactions. Our experiments directly demonstrate the action of a force-activated switch in the chemical reactivity of a single molecule. References: S. Garcia-Manyes, J. Liang, R. Szoszkiewicz, T-L. Kuo and J. M. Fernandez, Nature Chemistry, 1, 236-242, 2009.

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