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First-principles analysis on enzymatic degradation of nylon

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Nylon-6 is a long polymer generally consisting of more than one hundred units of 6-aminohexanoate (Ahx). The byproducts Ahx-linear and Ahx-cyclic oligomers, generated during the production, represent wastes to be disposed and possibly recovered as raw material. In this respect, the Ahx-dimer hydrolase (NylB) has been found to be a viable possibility to realize the desired degradation process. Recently, the enzymatic hydrolysis of the Ahx-linear dimer (Ald) into Ahx has become the target of increasing research efforts, with the purpose of developing an environmentally friendly way of disposing nylon wastes. However the detailed reaction mechanism is still largely unknown. Our extensive simulations aimed at understanding the fundamental issue of the binding structure upon mutations and at unraveling the detailed reaction mechanism occurring in NylB, which is one of the nylon oligomer hydrolase taken from bacteria. By means of the classical molecular dynamics, we first determined the substrate-binding structures, for which accurate X-ray data are not yet available. Then we analyzed their related reaction mechanism via reactive QM/MM approaches. The selected QM region includes a catalytic site composed of Ser112, Lys115, Tyr170, and Tyr215, and nearby amino acids plus H₂O molecules. Initially, Ser-OH attacks the amide compounds, forming a tetrahedral intermediate with a free energy barrier of 0.7 eV, where nucleophilicity of Ser112 is enhanced by the NH₃⁺ of Lys115. Then Tyr215 donates an H⁺ to the intermediate state during the conversion to acyl-enzyme, thus breaking the N-C bond in Ald and releasing Ahx.

Biography

Yasuteru Shigeta is a theoretical chemist who graduated from Department of Chemistry, Osaka University and obtained Doctor of Science degree at there in 2000. During his doctoral course, he was awarded a JSPS Research Fellowship for Young Scientists, and after graduation, again was awarded a JSPS Research Fellowship for Young Scientists. He worked at The University of Tokyo during 2004-2007, Tsukuba University during 2007-2008, University of Hyogo during 2008-2010, Osaka University 2010-2014 and is now the Professor of Tsukuba University majored in computational science and biophysics. He has published more than 100 scientific papers in reputed journals and was awarded several prestigious prizes.

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