

How Do Amino Acids Transport Electrons Through Peptides?



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Cuvillier Verlag Mai 2008, 2008. Taschenbuch. Book Condition: Neu. 147x20x17 mm. Neuware - A peptide model was designed, which allows the investigation of amino acid side chain participation in ET through peptides. Aromatic amino acids function as oxidizable spectroscopic sensors for the direct observation of charged intermediates during the ET process. Tyrosine as electron donor, situated at the N-terminus of the peptide, provides driving force for the ET process and is irreversibly oxidized to a long-lived phenoxyl radical with a sharp absorption band. Two methoxysubstituted phenylalanine derivatives were chosen as additional spectroscopic sensors, yielding oxidized transients with different absorption spectra. They were synthesized in their enantiopure form and investigated with respect to their electrochemical and spectroscopical properties. In the peptide model, they function as C-terminal electron acceptor precursor and central relay, separated from each other and the donor by a proline matrix. The electron acceptor can be generated by laser irradiation of an injection unit, containing a tbutyl ketone as chromophore. Transient absorption spectra recorded 40 ns after the laser flash were used for the examination of intramolecular ET efficiencies between the redox sites. The observation of intramolecular ET from the N-terminus to the C-terminus of a nonapeptide, which consisted of two triproline spacers and the three spectroscopic sensors was possible. This peptide showed a well-defined polyproline II helical structure, leading to a donor/acceptor separation of 20 Å. The aromatic relay, functioning as spectroscopic sensor for the detection of oxidized intermediates in donor to acceptor ET, could be exchanged by a number of different amino acids, without changing the overall structure of the peptide. Thus, the peptide model allowed us to determine the influence of the central amino acid - separated from both, donor and acceptor, by 10 Å - on ET. The simultaneous occurrence of electron acceptor, oxidized donor...



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