PL-2 PHOTOSENSITIZED ONE-ELECTRON OXIDATION OF DNA

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DNA may serve as an excellent molecule to store information in nano-electronic devices, if the electronic readout of the sequence became possible. The measurement of the kinetics of the hole transfer in DNA may open the way for it. So far, there are many experimental and theoretical studies for the mechanism of "one-dimensional"conductivity in the double helix. Gel electrophoretic analysis showed the hole transfer over 100 , while the kinetics of single-step hole transfer was studied by time-resolved measurements. Here, our results show direct observation of the long-distance hole transfer in DNA which depends upon the distance and DNA sequence [1-6].

DNA modified with naphthalimide (NI) and phenothiazine (PTZ), which worked as an electron acceptor and donor, respectively, was synthesized. Irradiation of the NI-site with a 355-nm laser flash causes charge separation between NI and adjacent adenine (A) to give NI radical anion (NI) and A radical cation (A), respectively. A part of a hole escapes from the initial charge recombination process via consecutive A-hopping to be trapped at G. Once G is generated far from NI, charge recombination proceeds by either strongly distance dependent super-exchange or A-hopping following slow A oxidation by G. Thus, in the case of six A bases, the lifetime of the charge separated state was more than several hundred microseconds, allowing the direct observation of the long-distance hole transfer process.

We clearly demonstrated the hole transfer process over long distances through DNA which occurred in the time scale of microseconds to milliseconds. Hole transfer kinetics was highly dependent upon the DNA sequence and base stacking. Given a certain sequence and length of DNA, an unique hole transfer kinetic will be obtained. The direct measurements of long-range hole transfer though DNA may allow high throughput readout of the stored information.

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