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Electrochemically mediated in situ growth of electroactive polymers for highly sensitive detection of double-stranded DNA without sequence-preference

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**Abstract** 

The ability to directly detect double-stranded DNA (dsDNA) without sequence-preference continues to be a major challenge. Herein, we report an electrochemical method for the direct, highly sensitive detection of dsDNA based on the strand replacement of dsDNA by peptide nucleic acid (PNA) and the in

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situ growth of electroactive polymers through the surface-initiated electrochemically mediated atom

transfer radical polymerization (SI-eATRP). Thiolated PNA molecules are firstly self-assembled onto

gold electrode surface for the specific recognition of target dsDNA (dsDNA-T), which in turn leads to

the formation of a high density of PNA/DNA heteroduplexes on the electrode surface for the subsequent

attachment of ATRP initiators via the phosphate-Zr<sup>4+</sup>-carboxylate chemistry. By applying a negative

potential to the electrode, the air-stable Cu<sup>II</sup> deactivators can be reduced into the Cu<sup>I</sup> activators so as to

trigger the surface-initiated polymerization for the *in situ* growth of electroactive polymers. Due to the

strand replacement of dsDNA by PNA, dsDNA can be directly detected without sequence-preference.

Besides, the growth of polymers enables the modification of numerous electroactive probes, thereby

greatly improving the electrochemical signal. Under optimal conditions, a good linearity between the

electrochemical signal and the logarithm of dsDNA-T concentration over the range from 1.0 fM to 1.0

nM, with a detection limit of 0.47 fM, can be obtained. Results indicate that it is highly selective, and

holds high anti-interference capability in the presence of human serum samples. Therefore, this method

offers great promises in providing a universal and efficient solution for the direct detection of dsDNA.

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