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Enantioselective cleavage of supercoiled plasmid DNA catalyzed by chiral macrocyclic lanthanide(III) complexes.

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Streszczenie

The enantiomers of the Sm (III), Eu (III) and Yb (III) complexes $[LnL(NO_3)_2](NO_3)$ of a chiral hexaazamacrocycle were tested as catalysts for the hydrolytic cleavage of supercoiled plasmid DNA. The catalytic activity was remarkably enantioselective; while the $[LnL^{SSSS}(NO_3)_2](NO_3)$ enantiomers promoted the cleavage of plasmid pBR322 from the supercoiled form (SC) to the nicked form (NC), the $[LnL^{RRRR}(NO_3)_2](NO_3)$ enantiomers were inactive. Kinetics of plasmid DNA hydrolysis was also investigated by agarose electrophoresis and it indicated typical single-exponential cleavage reaction. The hydrolytic mechanism of DNA cleavage was confirmed by the successful ligation of hydrolysis product by T4 ligase. The NMR study of the solutions of the complexes in various buffers indicated that the complexes exist as monomeric cationic complexes $[LnL(H_2O)_3]^{3+}$ in slightly acidic solutions and as dimeric cationic complexes $[Ln_2L_2(\mu\text{-}OH)_2(H_2O)_2]^{4+}$ in slightly basic 8 mM solutions, with the latter form being a possible catalyst for hydrolysis of phosphodiester bonds.

Słowa kluczowe

DNA cleavage, Dinuclear lanthanide complex, Enantioselectivity, Chiral macrocycle

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