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Nanostructured polyelectrolyte complexes based on water-soluble thiacalix[4]Arene and pillar[5]arene: Self-assembly in micelleplexes and polyplexes at packaging DNA

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Abstract

© 2020 by the authors. Licensee MDPI, Basel, Switzerland. Controlling the self-assembly of polyfunctional compounds in interpolyelectrolyte aggregates is an extremely challenging task. The use of macrocyclic compounds offers new opportunities in design of a new generation of mixed nanoparticles. This approach allows creating aggregates with multivalent molecular recognition, improved binding effciency and selectivity. In this paper, we reported a straightforward approach to the synthesis of interpolyelectrolytes by co-assembling of the thiacalix[4]arene with four negatively charged functional groups on the one side of macrocycle, and pillar[5]arene with 10 ammonium groups located on both sides. Nanostructured polyelectrolyte complexes show effective packaging of high-molecular DNA from calf thymus. The interaction of co-interpolyelectrolytes with the DNA is completely different from the interaction of the pillar[5]arene with the DNA. Two different complexes with DNA, i.e., micelleplex- and polyplex-type, were formed. The DNA in both cases preserved its secondary structure in native B form without distorting helicity. The presented approach provides important advantage for the design of effective biomolecular gene delivery systems.

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Keywords

Co-assembly, DNA packing, Interpolyelectrolyte, Nanomaterials, Pillar[5]arene, Selective recognition, Thiacalix[4]arene

References

[1] Li, L.-L.; An, H.-W.; Peng, B.; Zheng, R.; Wang, H. Self-assembled nanomaterials: Design principles, the nanostructural effect, and their functional mechanisms as antimicrobial or detection agents. Mater. Hor. 2019, 6, 1794-1811.

[2] Ariga, K.; Nishikawa, M.; Mori, T.; Takeya, J.; Shrestha, L.K.; Hill, J.P. Self-assembly as a key player for materials nanoarchitectonics. Sci. Technol. Adv. Mat. 2019, 20, 51-95.

[3] Sevim, S.; Sorrenti, A.; Franco, C.; Furukawa, S.; Pané, S.; deMello, A.J.; Puigmartí-Luis, J. Self-assembled materials and supramolecular chemistry within microfluidic environments: From common thermodynamic states to non-equilibrium structures. Chem. Soc. Rev. 2018, 47, 3788-3803.

[4] Amit, M.; Yuran, S.; Gazit, E.; Reches, M.; Ashkenasy, N. Tailor-Made Functional Peptide Self-Assembling Nanostructures. Adv. Mater. 2018, 30, 1707083.

[5] Pergushov, D.V.; Müller, A.H.E.; Schacher, F.H. Micellar Interpolyelectrolyte Complexes. Chem. Soc. Rev. 2012, 41, 6888-6901.

[6] van der Gucht, J.; Spruijt, E.; Lemmers, M.; Cohen Stuart, M.A. Polyelectrolyte Complexes: Bulk Phases and Colloidal Systems. J. Colloid Interface Sci. 2011, 361, 407-422.

[7] Zhao, Q.; An, Q.F.; Ji, Y.; Qian, J.; Gao, C. Polyelectrolyte Complex Membranes for Pervaporation, Nanofiltration and Fuel Cell Applications. J. Membr. Sci. 2011, 379, 19-45.

[8] Ahmadiannamini, P.; Li, X.; Goyens, W.; Joseph, N.; Meesschaert, B.; Vankelecom, I.F.J. Multilayered Polyelectrolyte Complex Based Solvent Resistant Nanofiltration Membranes Prepared from Weak Polyacids. J. Membr. Sci. 2012, 394-395, 98-106.

[9] Delcea, M.; Möhwald, H.; Skirtach, A.G. Stimuli-Responsive LbL Capsules and Nanoshells for Drug Delivery. Adv. Drug Deliv. Rev. 2011, 63, 730-747.

[10] Kim, S.H.; Jeong, J.H.; Lee, S.H.; Kim, S.W.; Park, T.G. Local and Systemic Delivery of VEGF siRNA Using Polyelectrolyte Complex Micelles for Effective Treatment of Cancer. J. Control. Release 2008, 129, 107-116.

[11] Wang, Q.; Schlenoff, J.B. The Polyelectrolyte Complex/Coacervate Continuum. Macromolecules 2014, 47, 3108-3116.

[12] Lounis, F.M.; Chamieh, J.; Gonzalez, P.; Cottet, H.; Leclercq, L. Prediction of Polyelectrolyte Complex Stoichiometry for Highly Hydrophilic Polyelectrolytes. Macromolecules 2016, 49, 3881-3888.

[13] Fu, J.; Fares, H.M.; Schlenoff, J.B. Ion-Pairing Strength in Polyelectrolyte Complexes. Macromolecules 2017, 50, 1066-1074.

[14] Love, J.C.; Estroff, L.A.; Kriebel, J.K.; Nuzzo, R.G.; Whitesides, G.M. Self-assembled monolayers of thiolates on metals as a form of nanotechnology. Chem. Rev. 2005, 105, 1103-1169.

[15] Daniel, M.-C.; Astruc, D. Gold Nanoparticles: Assembly, Supramolecular Chemistry, Quantum-Size-Related Properties, and Applications toward Biology, Catalysis, and Nanotechnology. Chem. Rev. 2004, 104, 293-346.

[16] Chen, M.; Diao, G.W.; Li, C.H.; Zhou, X.M. Phase transition of silver nanoparticles from aqueous solution to chloroform with the help of inclusion complexes of p-sulfonated calix[4]arene and alkanethiol molecules and its application in the size sorting of nanoparticles. Nanotechnology 2007, 18, 175706.

[17] Chen, M.; Ding, W.H.; Kong, Y.; Diao, G.W. Conversion of the Surface Property of Oleic Acid Stabilized Silver Nanoparticles from Hydrophobic to Hydrophilic Based on Host-Guest Binding Interaction. Lanmuir 2008, 24, 3471-3478.

[18] Baldini, L.; Sansone, F.; Massera, C.; Casnati, A.; Ugozzoli, F.; Ungaro, R. Designing nanoporous crystalline materials by self-assembly: 2D hydrogen-bonded networks from upper rim calix[4]arene diamide derivatives. Inorg. Chim. Acta 2007, 360, 970-980.

[19] Seganish, J.L.; Santacroce, P.V.; Salimian, K.J.; Fettinger, J.C.; Zavalij, P.; Davis, J.T. Regulating supramolecular function in membranes: Calixarenes that enable or inhibit transmembrane Cl- transport. Angew. Chem. Int. Ed. 2006, 45, 3334-3338.

[20] Schafer, C.; Eckel, R.; Ros, R.; Mattay, J.; Anselmetti, D. Photochemical Single-Molecule Affnity Switch. J. Am. Chem. Soc. 2007, 129, 1488-1489.

[21] Organo, V.G.; Leontiev, A.V.; Sgarlata, V.; Dias, H.V.R.; Rudkevich, D.M. Supramolecular Features of Calixarene-Based Synthetic Nanotubes. Angew. Chem. Int. Ed. 2005, 44, 3043-3047.

[22] Atwood, J.L.; Steed, J.W. Encyclopedia of Supramolecular Chemistry; CRC Press: Boca Raton, FL, USA, 2004; pp 1-1744.

[23] Gorbatchuk, V.V.; Savelyeva, L.S.; Ziganshin, M.A.; Antipin, I.S.; Sidorov, V.A. Molecular recognition of organic guest vapor by solid adamantylcalix[4]arene. Russ. Chem. Bull. 2004, 53, 60-64.

[24] Ziganshin, M.A.; Yakimova, L.S.; Khayarov, K.R.; Gorbatchuk, V.V.; Vysotsky, M.O.; Böhmer, V. Guest exchange in dimeric capsules of a tetraurea calix[4]arene in the solid state. Chem. Commun. 2006, 37, 3897-3899.

[25] Yakimova, L.S.; Ziganshin, M.A.; Sidorov, V.A.; Kovalev, V.V.; Shokova, E.A.; Tafeenko, V.A.; Gorbatchuk, V.V. Molecular Recognition of Organic Vapors by Adamantylcalix[4]arene in QCM Sensor Using Partial Binding Reversibility. J. Phys. Chem. B 2008, 112, 15569-15575.

[26] Stoikov, I.I.; Mostovaya, O.A.; Yakimova, L.S.; Yantemirova, A.A.; Antipin, I.S.; Konovalov, A.I. Phosphorus-bridged calixarene phosphites: Dramatic influence of a tert-butyl group at the upper rim of the macrocycle upon anion binding. Mendeleev Commun. 2010, 20, 359-360.

[27] Ogoshi, T.; Hasegawa, Y.; Aoki, T.; Ishimori, Y.; Inagi, S.; Yamagishi, T.-A. Reduction of Emeraldine Base Form of Polyaniline by Pillar[5]arene Based on Formation of Poly(pseudorotaxane) Structure. Macromolecules 2011, 44, 7639-7644.

[28] Ogoshi, T.; Kitajima, K.; Aoki, T.; Yamagishi, T.A.; Nakamoto, Y. Effect of an Intramolecular Hydrogen Bond Belt and Complexation with the Guest on the Rotation Behavior of Phenolic Units in Pillar[5]arenes. J. Phys. Chem. Lett. 2010, 1, 817-821.

[29] Li, C.; Xu, Q.; Li, J.; Yao, F.; Jia, X. Complex interactions of pillar[5]arene with paraquats and bis(pyridinium) derivatives. Org. Biomol. Chem. 2010, 8, 1568-1576.

[30] Shurpik, D.N.; Yakimova, L.S.; Rizvanov, I.K.; Plemenkov, V.V.; Stoikov, I.I. Water-soluble pillar[5]arenes: Synthesis and characterization of the inclusion complexes with p-toluenesulfonic acid. Macroheterocycles 2015, 8, 128-134.

[31] Xu, Z.; Jia, S.; Wang, W.; Yuan, Z.; Ravoo, B.J.; Guo, D.-S. Heteromultivalent peptide recognition by co-assembly of cyclodextrin and calixarene amphiphiles enables inhibition of amyloid fibrillation. Nat. Chem. 2019, 86, 86-93.

[32] Chen, X.-M.; Chen, Y.; Yu, Q.; Gu, B.-H.; Liu, Y. Supramolecular assemblies with near-infrared emission mediated in two stages by cucurbituril and amphiphilic calixarene for lysosome- targeted cell imaging. Angew. Chem. Int. Ed. 2018, 57, 12519-12523.

[33] Yakimova, L.; Padnya, P.; Tereshina, D.; Kunafina, A.; Nugmanova, A.; Osin, Y.; Evtugyn, V.; Stoikov, I. Interpolyelectrolyte mixed nanoparticles fromanionic and cationic thiacalix[4]arenes for selective recognition of model biopolymers. J. Mol. Liq. 2019, 279, 9-17.

[34] Yakimova, L.S.; Gilmanova, L.H.; Evtugyn, V.G.; Osin, Y.N.; Stoikov, I.I. Self-assembled fractal hybrid dendrites from water-soluble anionic (Thia)calix[4] arenes and Ag+. J. Nanopart. Res. 2017, 19, 173-183.

[35] Khan, G.S.; Shah, A.; Zia-Ur-Rehman; Barker, D. Chemistry of DNA minor groove binding agents. J. Phtoch. Photobio. B 2012, 115, 105-118.

[36] Kapuscinski, J.; Skoczylas, B. Fluorescent complexes of DNA with DAPI 40-6-diamidine-2-phenyl indole 2HCl or DC140-6-dicarboxyamide-2-phenyl indole. Nucleic Acids Res. 1978, 5, 3775-3799.

[37] Biancardi, A.; Biver, T.; Secco, F.; Mennucci, B. An investigation of the photophysical properties of minor groove bound and intercalated DAPI through quantum-mechanical and spectroscopic tools. Phys. Chem. Chem. Phys. 2013, 15, 4596-4603.

[38] Kasyanenko, N.A.; Tikhomirov, R.A.; Bakulev, V.M.; Demidov, V.N.; Chikhirzhina, E.V.; Moroshkina, E.B. DNA Complexes with Cobalt(II) Phthalocyanine Disodium Disulfonate. ACS Omega 2019, 4, 16935-16942.

[39] Zadmard, R.; Schrader, T. DNA recognition with large calixarene dimers. Angew. Chem. Int. Ed. 2006, 45, 2703-2706.

[40] Pabbathi, A.; Samanta, A. Spectroscopic and Molecular Docking Study of the Interaction of DNA with a Morpholinium Ionic Liquid. J. Phys. Chem. B 2015, 119, 11099-11105.

[41] Banerjee, D.; Pal, S.K. Dynamics in the DNA Recognition by DAPI: Exploration of the Various Binding Modes. J. Phys. Chem. B 2008, 112, 1016-1021.

[42] Ankerfors, C.; Bertin, A.; Bouhallab, S.; Croguennec, T.; Müller, M.; Petzold, G.; Schwarz, S.; Wagberg, L. Polyelectrolyte Complexes in the Dispersed and Solid State II; Springer: Berlin, Germany, 2014; pp 1-256.

[43] Bhattacharjee, S. DLS and zeta potential What they are and what they are not? J. Control. Release 2016, 235, 337-351.

[44] Lohani, N.; Rajeswari, M.R. Preferential binding of anticancer drugs to triplex DNA compared to duplex DNA: A spectroscopic and calorimetric study. RSC Adv. 2016, 6, 39903-39917.

[45] Shahabadi, N.; Mohammadi, S. Synthesis characterization and DNA interaction studies of a new Zn (II) complex containing different dinitrogen aromatic ligands. Bioinorg. Chem. Appl. 2012, 571913-571921.

[46] Gomathi, R.; Ramu, A.; Murugan, A. Evaluation of DNA Binding, Cleavage, and Cytotoxic Activity of Cu (II), Co (II), and Ni (II) Schiff Base Complexes of 1-Phenylindoline-2, 3-dione with Isonicotinohydrazide. Bioinorg. Chem. Appl. 2014, 215392.

[47] Norden, B.; Kurucsev, T. Analysing DNA complexes by circular and linear dichroism. J. Mol. Recognit. 1994, 7, 141-156.

[48] Kaushik, M.; Kaushik, S.; Roy, K.; Singh, A.; Mahendru, S.; Kumar, M.; Chaudhary, S.; Ahmed, S.; Kukreti, S. A bouquet of DNA structures: Emerging diversity. Biochem. Biophys. Rep. 2016, 5, 388-395.

[49] Banavali, N.K.; Roux, B. Free Energy Landscape of A-DNA to B-DNA Conversion in Aqueous Solution. J. Am. Chem. Soc. 2005, 127, 6866-6876.

[50] Gubendran, A.; Palsamy Kesavan, M.; Ayyanaar, S.; Mitu, L.; Athappan, P.; Rajesh, J. Non-enolisable Knoevenagel condensate appended Schiff bases-metal (II) complexes: Spectral characteristics, DNA-binding and nuclease activities. Spectrochim. Acta A 2017, 181, 39-46.

[51] Luo, J.; Xie, Z.; Lam, J.W.Y.; Cheng, L.; Chen, H.; Qiu, C.; Kwok, H.S.; Zhan, X.; Liu, Y.; Zhu, D. et al. Aggregation-induced emission of 1-methyl-1,2,3,4,5-pentaphenylsilole. Chem. Commun. 2001, 18, 1740-1741.

[52] Yakimova, L.S.; Puplampu, J.B.; Evtugin, G.A.; Stoikov, I.I. Polyfunctional branched nitrogen-containing p-ter--butylthiacalix[4]arene derivatives as effcient agents for packaging calf thymus DNA. Russ. Chem. Bull. 2017, 66, 1515-1523.

[53] Kabanov, A.V.; Astafieva, I.V.; Chikindas, M.L.; Rosenblat, G.F.; Kiselev, V.I.; Severin, E.S.; Kabanov, V.A. DNA interpolyelectrolyte complexes as a tool for effcient cell transformation. Biopolymers 1991, 31, 1437-1443.

[54] Kabanov, A.V.; Kabanov, V.A. DNA Complexes with Polycations for the Delivery of Genetic Material into Cells. Bioconjugate Chem. 1995, 6, 7-20.

[55] Tan, Z.; Jiang, Y.; Zhang, W.; Karls, L.; Lodge, T.P.; Reineke, T.M. Polycation Architecture and Assembly Direct Successful Gene Delivery: Micelleplexes Outperform Polyplexes via Optimal DNA Packaging. J. Am. Chem. Soc. 2019, 141, 15804-15817.

[56] Jiang, Y.; Reineke, T.M.; Lodge, T.P. Complexation of DNA with Cationic Copolymer Micelles: Effects of DNA Length and Topology. Macromolecules 2018, 51, 1150-1160.

[57] Jiang, Y.; Lodge, T.P.; Reineke, T.M. Packaging pDNA by Polymeric ABC Micelles Simultaneously Achieves Colloidal Stability and Structural Control. J. Am. Chem. Soc. 2018, 140, 11101-11111.

[58] Stein, H.N. The Preparation of Dispersions in Liquids; CRC Press: Boca Raton, FL, USA, 1995; pp 1-264.