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Molecular Structure of Single DNA Complexes with Positively Charged Dendronized Polymers

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Abstract: Positively charged dendronized polymers with protonated amine groups at the periphery and different dendron generations are cylindrically shaped nanoobjects whose radii and linear charge densities can be varied systematically. These polyelectrolytes have been complexed with DNA and subsequently adsorbed on precoated mica substrates. The analysis of scanning force microscopy data indicates that DNA wraps around the dendronized polymers. The calculated pitch is 2.30 ± 0.27 and 2.16 ± 0.27 nm for DNA wrapped around dendronized polymers of generation two and four, respectively. The complex with the second generation has been shown to be negatively charged, which is consistent with the theory of spontaneous overcharging of macro-ion complexes, when the electrostatic contribution to the free energy dominates over the elastic energy. The complexes may be of interest for the development of nonviral gene delivery systems.

Introduction

In viruses and cells, DNA is organized in tightly packed structures. Much research has been carried out in order to obtain insight into the mechanisms of condensation and aggregation of DNA,[1] which both can be induced in vitro by a variety of positive ions, due to electrostatic interactions with the oppositely charged phosphate groups on the DNA backbone. DNA molecules condense into toroids and rods in the presence of multivalent cations[2] or polyamines[3] (polyplexes), but the resulting structures were not resolved on the molecular level. Also in complexes formed with cationic polymers[4,5] and cationic dendrimers,[6] the molecular structure remains unclear, while X-ray diffraction on complexes formed from DNA and cationic lipids[7] (lipoplexes) reveals multilammellar structures. Most of the synthetic cationic agents forming these complexes and aggregates are developed for potential use as DNA vectors in novel gene therapies. An example is the spherical poly(amidoamine) (PAMAM) dendrimer.[8] The structure of its complex with DNA can influence the in vivo interactions with the biological material and therefore affect the efficiency of transfection, which depends in particular on the structure, size, and charge density of the dendrimers.[9] However, again, the structure of this self-assembled nonviral gene delivery system is not well understood.[10]

On the other hand, a well-known ordered structure of compacted DNA is found in the nucleus of eukaryotic cells, where the DNA is associated with histone proteins to form the chromatin. X-ray crystallography has shown that, within the nucleosome, the smallest unit of the chromosome, 146 bp DNA wraps in 1.65 turns around the histone octamers[11] like a thread around a spool. Interestingly, the nucleosomal core particles have a net negative charge because the negative charge of the wrapped DNA is significantly larger than the total positive charge of the histone protein octamer.[12,13]

Aside from the biological and medical aspects, the molecular structure of polyelectrolyte complexes may be used to improve our general understanding of polyelectrolyte interactions. Polyelectrolyte adsorption on charged flat surfaces or spheres (i.e., layer-by-layer adsorption[14]) has been a focus in experimental and theoretical studies.[15,16] Also, theoretical models of the

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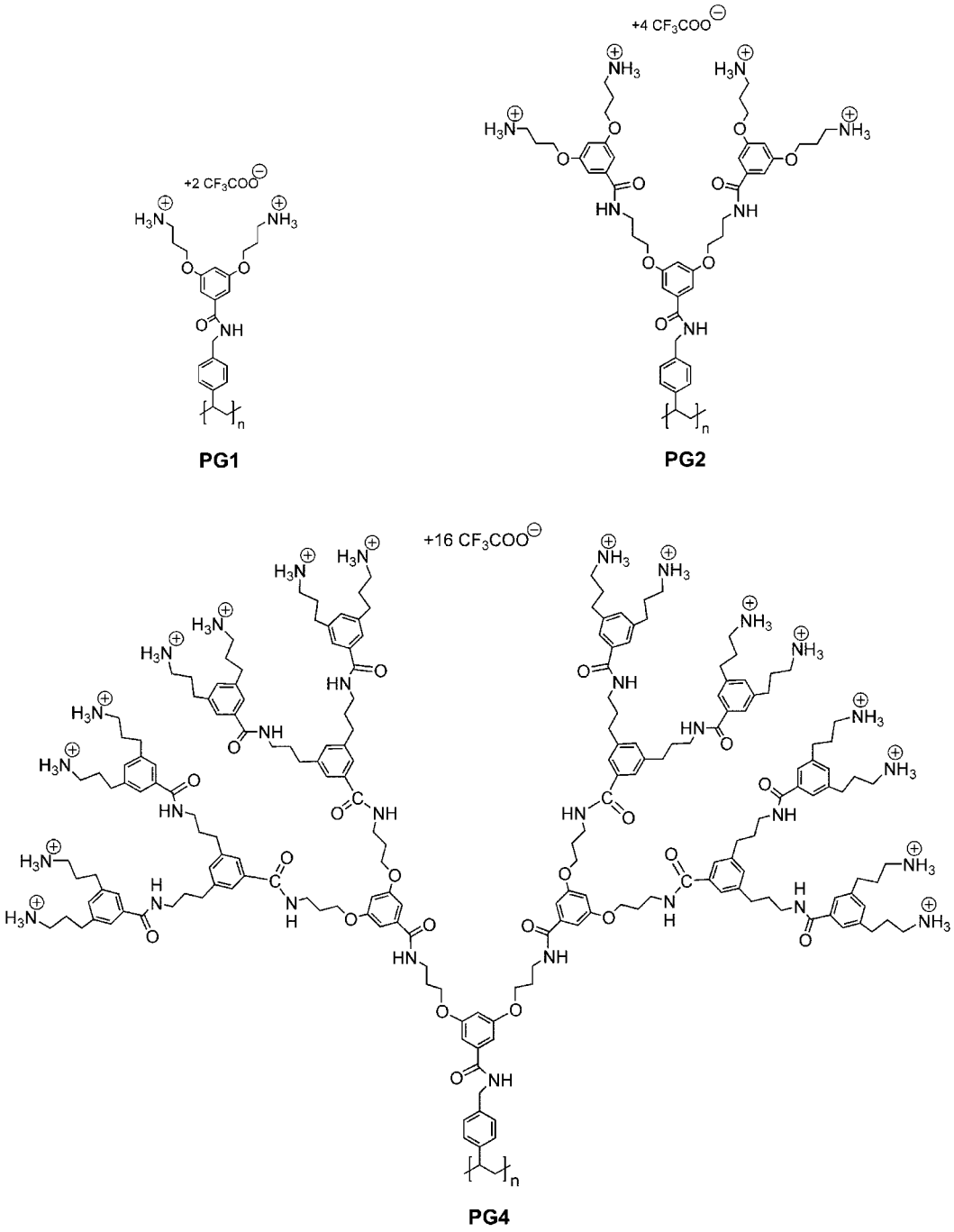
Chart 1. Chemical Structures of the Used Dendronized Polymers



structure of complexes formed between a stiff charged cylinder and an oppositely charged flexible or semiflexible polymer have been investigated.[17,18] However, experimentally the structure of single polyelectrolyte complexes remains a challenge.[19]

Here we report on the complex formation of DNA molecules with cylindrically shaped dendronized polymers. Starting with the amino-terminal dendronized polystyrene of generation one, higher generations were obtained by the so-called mixed "attach-to" approach.[20,21] Thus, the synthesized dendronized polymers of generations two, three, and four possessed approximately the same chain length and polydispersity but varying numbers of charges as well as radii. The obtained complexes were adsorbed on bare, negatively charged mica or on mica coated with a positively charged polymer, and then imaged by scanning force microscopy (SFM).

Materials and Methods

Starting with a core dendronized polymer $PG1^{P}$ of generation one (here the superscript "P" indicates the protection by trimethylsilylethyloxycarbonyl), higher generations (two, three, and four) were obtained by subsequent deprotection and dendronization steps (Chart 1).

In the present study we have used the unprotected (charged) analogues of generations two (PG2) and four (PG4), having a molar

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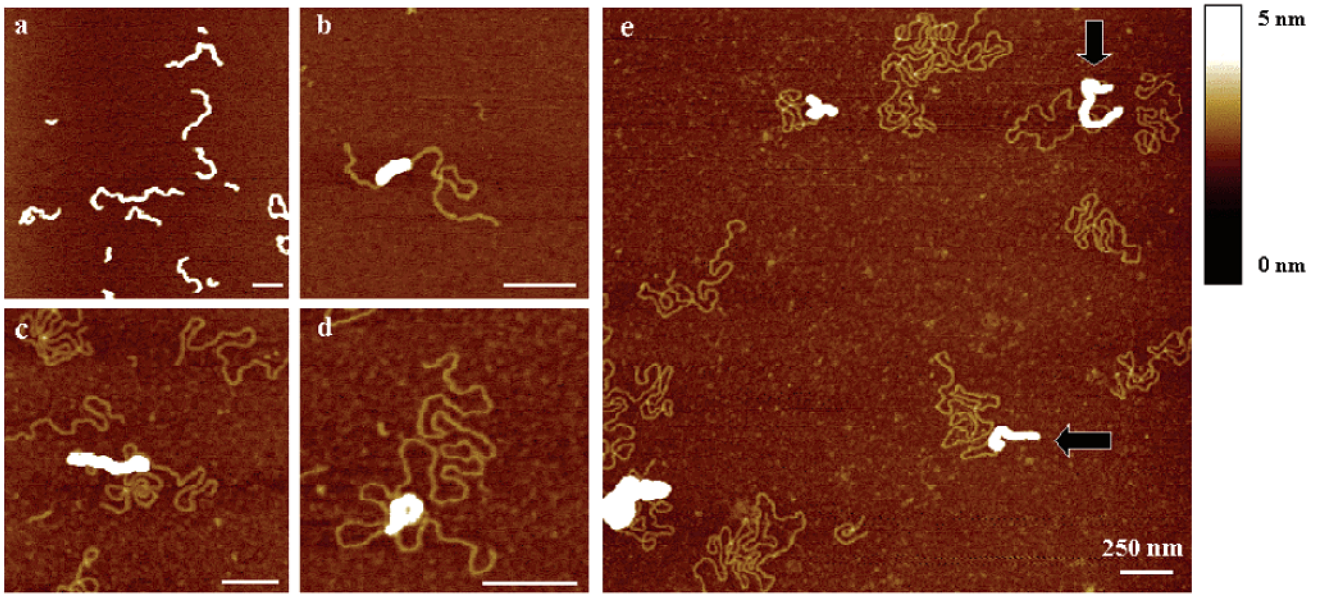

Figure 1. High-resolution SFM images. (a) Dendronized polymer PG4 deposited onto freshly cleaved mica. (b−e) DNA/PG4 complexes of charge ratio 1:0.7 precipitated onto poly-L-ornithine-coated mica. The scale bars represent 250 nm.

mass per repeat unit of 1285 and 5089 g/mol (including their counterions), respectively, with a quantified structure perfection of 97 ± 1% (for PG4).[22] The number of charges per repeat unit was 4 for PG2 and 16 for PG4, which means a 4 times higher linear charge density for PG4 compared to that for PG2. The molar mass distribution of the core dendronized polymer $PG1^P$ was characterized by analytical gel permeation chromatography (GPC) measurements. All charged dendronized polymers were soluble in water, and each generation was stored in a stock solution (5 mM *N*-[2-hydroxyethyl]piperazine-*N*′-[2-ethanesulfonic acid] (HEPES), NaOH at pH 7.5) at a concentration of 12 ng/μL. pUC19 plasmid DNA was linearized using *Bam*HI (New England Biolabs, Frankfurt, Germany). After purification with QIAprep Spin Miniprep Kit (Qiagen, Hilden, Germany), its concentration was determined by light absorbance at $\lambda = 260$ nm.

For substrates we used freshly cleaved mica (PLANO W. Plannet GmbH, Wetzlar, Germany) or poly-L-ornithine (molar mass 30 000−70 000 g/mol, Sigma, St. Louis, MO)-coated mica. For the coating, a freshly cleaved mica surface was placed onto a 5-μL droplet of 0.1 mg/mL poly-L-ornithine solution for 5 min, rinsed three times with deionized water, and then dried under a stream of N_2 gas. While freshly cleaved mica serves as a negatively charged surface,[23] the polymer-coated surface is positively charged.[5]

Isolated dendronized polymers were deposited onto freshly cleaved mica substrates similarly from diluted dendronized polymer stock solutions. With the same procedure, also DNA molecules were deposited onto poly-L-ornithine-coated mica substrates from a buffer solution (5 mM HEPES, NaOH at pH 7.5) diluted to a DNA concentration of 1 ng/μL.

DNA/dendronized polymer complexes were formed by adding an appropriately diluted dendronized polymer stock solution to a DNA buffer solution. The different dilutions resulted in DNA/dendronized polymer charge ratios of 1:10, 1:5, 1:2.5, and 1:1 through 1:0.1, taking into account that 1 ng of DNA contains 1.9×10^{12} negative charges (molar mass of 649 g/mol per bp) and 1 ng of each PG2 and PG4 contains 1.9×10^{12} positive charges. Each DNA/dendronized polymer mixture was allowed to adsorb onto freshly cleaved and poly-L-ornithine-coated mica substrates. First, the DNA/dendronized polymer solution was shaken for 1 min. Longer preparation times of about 10 min did not give different results. The sample was then prepared as described above.

Scanning force microscopy (SFM) images were recorded using a MultiMode scanning probe microscope (Digital Instruments, Inc., Santa Babara, CA) that was operated in tapping mode.[24] Olympus etched silicon cantilevers were used with a typical resonance frequency in the range of 200−400 kHz and a spring constant of 42 N/m. All samples were measured at room temperature in air environment. SFM height measurements were based on the cross-sectional profiles.

Results

Single dendronized polymers (PG2 and PG4) were deposited from buffer solutions onto freshly cleaved mica and visualized via SFM. Figure 1a shows single PG4.

The visualized polymers were analyzed by determining contour length distributions and heights. To evaluate the contour length distributions, the contour of each single molecule was divided into straight segments of 2−5 nm. For this analysis, only such dendronized polymers were selected which possessed two clearly recognizable ends. The histograms of the contour lengths of about 100 molecules of PG2 and PG4 are displayed in Figure 2a.

The data were fitted using the Schulz−Flory number distribution, since the dendronized polymers were synthesized via radical polymerization and possess a high degree of polymerization:

$$h(P) = \alpha^P \ln \alpha \qquad (1)$$

where P is the degree of polymerization and α is the probability of propagation. The number-average molecular length $\langle L_n \rangle = \sum (N_i L_i)/\sum N_i$, the weight-average molecular length $\langle L_w \rangle = \sum (N_i L_i^2)/\sum (N_i L_i)$, and the length polydispersity PD $= \langle L_w \rangle / \langle L_n \rangle$ were calculated (Table 1). Overall, no significant changes

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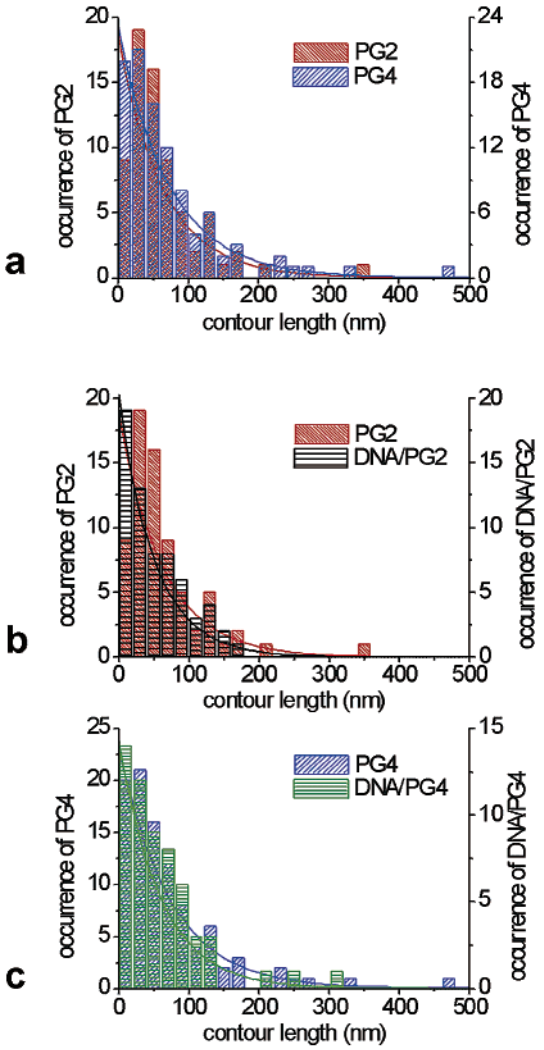

Figure 2. Contour length distributions of dendronized polymers. (a) Different generations PG2 (red) and PG4 (blue). (b) PG2 (red) and complexes formed from PG2 and DNA (black). (c) PG4 (blue) and complexes formed from PG4 and DNA (green). The data are corrected for the error due to the broadening effects of the tip, assuming the calculated heights of the dendronized polymers and complexes (eq 2) and a tip radius of 15 nm.[25]

Table 1. Number-Average Contour Length, Weight-Average Contour Length, and Length Polydispersity of Dendronized Polymers and Complexes

system	$\langle L_n \rangle$ (nm)	$\langle L_w \rangle$ (nm)	PD = $\langle L_n \rangle / \langle L_w \rangle$
PG2	64 ± 12	110 ± 24	1.98 ± 0.17
PG4	73 ± 10	152 ± 21	1.66 ± 0.11
DNA/PG2	52 ± 18	86 ± 37	1.72 ± 0.33
DNA/PG4	59 ± 13	117 ± 26	2.08 ± 0.17

in the fitted Schulz−Flory distributions and the corresponding statistical distribution averages were observed between generations two and four (Figure 2a and Table 1).

This result verifies the successful synthesis of PG2 and PG4 from the same core dendronized polymer $PG1^P$. The average height of PG2 and PG4 from tapping mode images was 1.8 ± 0.3 and 3.0 ± 0.3 nm, respectively.

The contour length distribution of linear pUC 19 showed that the molecules were almost monodisperse, with an average length of 844 ± 66 nm (data not shown). Apparently due to mechanical stress during preparation steps, a small amount of linear pUC 19 molecules was damaged. The apparent persistence length of DNA molecules adsorbed onto mica substrates precoated with positively charged poly-L-ornithine was 15.4 ± 0.5 nm, as determined by a shape analysis.[26] This is smaller than that of DNA molecules adsorbed on untreated mica from a $MgCl_2$ solution (P = 53 nm). The difference suggests that DNA molecules adsorbed on poly-L-ornithine-treated mica are kinetically trapped on the surface.[27,28] The height of the DNA molecules in tapping mode was 0.7 ± 0.1 nm, which is comparable to that reported in earlier experiments (e.g., ref 29).

When DNA was mixed with dendronized polymers, different complexes were formed upon varying the DNA/dendronized polymer concentration and with it the charge ratio from 1:10, 1:5, 1:2.5, and 1:1 through 1:0.1. DNA/dendronized polymer complexes with charge ratios of 1:10, 1:5, and 1:2.5 adsorbed only onto freshly cleaved mica and not onto the positively charged, poly-L-ornithine-coated mica substrates. However, these adsorbates were too aggregated for high-resolution imaging via SFM. In contrast, DNA/dendronized polymer complexes with charge ratios of 1:1 through 1:0.1 adsorbed only onto poly-L-ornithine-coated mica substrates. Complexes with a charge ratio of 1:1 through 1:0.1 on poly-L-ornithine-coated mica were the most suitable complexes for SFM analysis. Figure 1b−e shows common types of complexes. For their analysis, only those complexes were chosen which exhibited a constant height along their contour, and where the single DNA strands that came out of this complex belonged clearly to the complex (indicated by arrows in Figures 1e). For contour length measurements, more highly resolved images of single complexes were used (Figure 1b−d). There are different types of complexes. While for the complex in Figure 1b clearly one DNA molecule is used, in Figure 1c,d more than one DNA molecule must be involved, since the length of the DNA coming out of the complexes is longer than one DNA contour length. For further analysis, also the average heights were measured, which amounted to 4.0 ± 0.3 nm for DNA/PG2 and 5.5 ± 0.6 nm for DNA/PG4. While the absolute height value depended on the generation of the dendronized polymer, the increases in height of both the DNA/PG2 and the DNA/PG4 complex with respect to the bare dendronized polymers were the same, i.e., 2.4 ± 0.5 nm.

Discussion

We compared the contour length distributions of the bare dendronized polymers and their complexes with DNA. From Figure 2b,c one can see that the Schulz−Flory distributions of the data obtained for the DNA/PG2 and DNA/PG4 complexes compared to PG2 and PG4, respectively, and the corresponding statistical distribution averages (Table 1) showed no differences within the errors. From the similarity of the distributions of the bare polymers and the complexes, we conclude that the contour length of the complexes is defined by the bare dendronized polymers.

On the other hand, the length of the DNA that contributes to the complex (L_{DNA-C}) can be obtained by subtracting the

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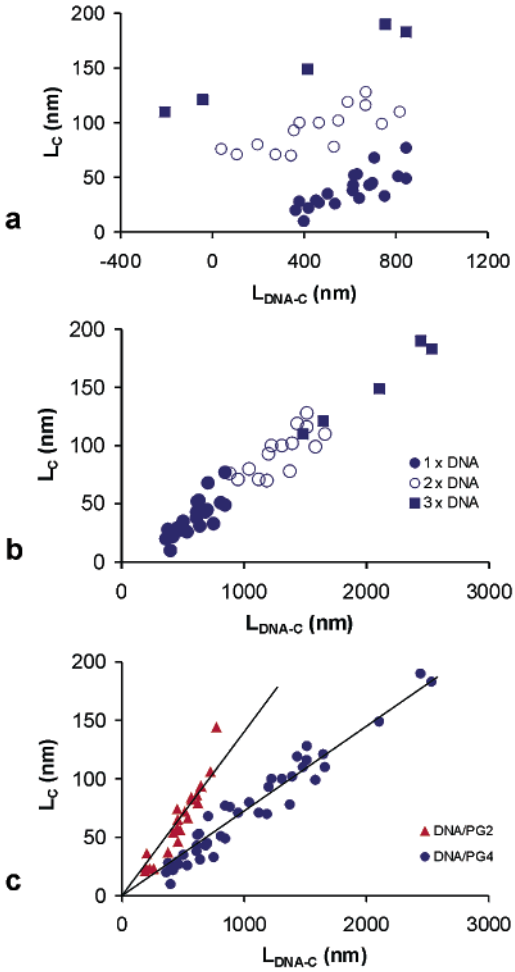

Figure 3. Linear dependence of the complex length (L_C) and DNA length used for complexes (L_{DNA-C}). (a) Contour length of the complex (L_C) versus length of DNA within the complex (L_{DNA-C}) for the PG4/DNA complex. (b) Data from (a) allowing for more than one DNA molecule involved in a complex. Dendronized polymers complexing with one (●), two (○), and three (■) DNA molecules. (c) Linear dependence of complex length (L_C) and DNA length used for complexes (L_{DNA-C}) for PG2 (▲) and PG4 (●).

measured contour length of the DNA molecule coming out of the complex (L_{out}) from the length of the monodisperse DNA (L_0) by $L_{DNA-C} = L_0 - L_{out}$. In Figure 3a, the contour lengths of the complexes (L_c) are plotted versus the DNA that contributes to the complex (L_{DNA-C}). The results can be grouped in three linear parts, which also contain negative values for L_{DNA-C}. We attribute these three parts to complexes formed with one, two, and three monodisperse DNA (L_0) strands.

Looking at Figure 1d, one can see that there were complexes formed which possessed more than one DNA molecule of the length L_0. Therefore, plotting the contour length of the complexes (L_c) versus the DNA that contributes to the complex (L_{DNA-C}), and allowing doubles and triples of the monodisperse DNA ($2L_0$ and $3L_0$) with the complex, the data exhibit an overall linear dependence (Figure 3b). The slope (m) varied depending on the generation of the dendronized polymer used (Figure 3c): for DNA/PG2 complexes it is $m = 0.14 \pm 10\%$, and for DNA/PG4 complexes it is $m = 0.08 \pm 10\%$. On the basis of this linear dependence, the height measurements, and the comparison of the contour length distributions of complexes and dendronized polymers, it is concluded that the DNA molecules wrap around the positively charged dendronized

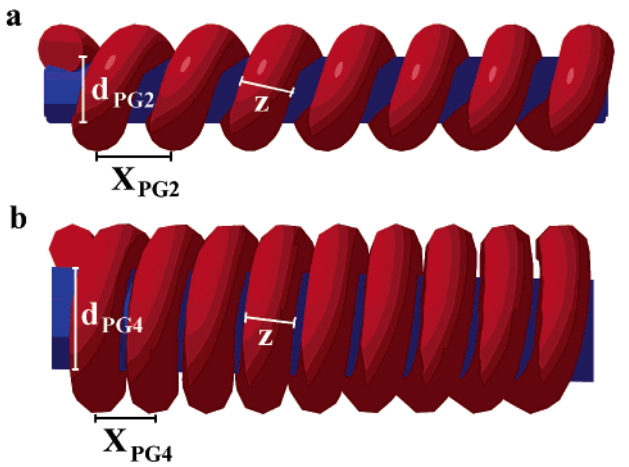

Figure 4. Model of the complex. DNA wraps around the dendronized polymer PG2 (a) and PG4 (b) resulting in pitches X_{PG2} and X_{PG4}.

polymer. For a quantitative model, one should take into account that SFM images underestimate the height of all the polymers, i.e., the dendronized polymers, the DNA molecules, and the complexes, due to tip−sample interactions (deformation of the sample).[30] On the basis of the volume of the dendronized polymers, their radii can be estimated to be 1.6 (±10%) nm for PG2 and 3.3 (±10%) nm for PG4, using

$$r = \sqrt{\frac{M}{N_A \pi \rho_g l}} \qquad (2)$$

where M is the molar mass, N_A is Avogadro's constant, ρ_g is the polymer density (assumed to be ≈1 g/cm^3), and $l = 0.25$ nm is the contour length of the repeat unit in the *all-trans* conformation. Using these radii and the theoretical diameter for DNA (2 nm), we calculate the DNA length required for one turn around the dendronized polymers (U) to be 16.3 ± 1.0 nm for PG2 and 27.0 ± 2.1 nm for PG4. With $X_{PGi} = m_i U_i$ (i stands for the different generations), this results in a DNA pitch of $X_{PG2} = 2.30 \pm 0.27$ nm for PG2 and $X_{PG4} = 2.16 \pm 0.27$ nm for PG4 (Figure 4), leaving some space for water layers. Also, N. V. Hud and K. H. Downing[31] report on DNA toroids using cryoelectron microscopy which show a minimum fringe spacing of 2.03 nm.

On the other hand, the high curvature of DNA around a diameter of 6.6 ± 0.7 nm of PG4 is practically identical to the average diameter (6.4 nm) formed by the superhelix of the nucleosome core particle.[11] The curvature for PG2 is a little bit higher.

It has been shown in previous studies[7,32] that the DNA spacing on charged lipid membranes increases when the surface charge density of the lipid membranes decreases. This increase in the pitch size can also be seen in our experiment upon decreasing the linear charge density of the dendronized polymer from PG4 to PG2.

The overall charge of the complex depended on the dendronized polymer generations involved in the complex formation. If we assume two negatively charged phosphates per 0.34 nm bp length, four positively charged amine groups on the PG2,

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and 16 on the PG4 per 0.25 nm repeat unit in the *all-trans* configuration, the DNA/PG4 complexes were nearly neutral, whereas the DNA/PG2 complexes were negatively (over)-charged. The fact that both complexes adsorbed only onto positively coated mica substrate is consistent with negatively charged complexes. While the radii and the linear charge densities of the two dendronized polymers are different, the charge ratio was kept constant near the isoelectric point (1/0.7 DNA/dendronized polymer). Evidence for the overcharging of charged macro-ions and surfaces by electrostatic interaction with oppositely charged macromolecules has been given experimentally[14,15] and theoretically.[16,33,34] Park et al.[17] provide a theoretical approach for spontaneous overcharging of a negatively charged semiflexible chain and a positively charged rigid cylinder. The theory is based on the Poisson−Boltzmann equation for macro-ions in aqueous monovalent salt solutions in the limit of low salt concentrations. It predicts an over- and undercharging of the complex depending on the flexibility (persistence length) of the chain. In our experiment, we identify the semiflexible chain with DNA and the rigid cylinder with the dendronized polymer. The flexibility of the chain (DNA) stays constant, while the radii and the linear charge densities of the cylinder (i.e., the dendronized polymer) are varied, which in the theory is associated with the elastic energy term and the electrostatic energy term of the system, respectively. Adhering to the theory of Park et al., the effective dimensionless charge density ν in the minimum energy state of the system can be calculated by minimizing the total free energy, F_{tot}, of the complex system with respect to ν. The theory predicts undercharging ($\nu > 0$) only when the elastic energy term is larger than the electrostatic energy term and overcharging for the opposite case. For the case of the DNA/PG2 complex, we can conclude that the contribution of the electrostatic energy is larger than the contribution of the elastic energy in the complex ($\nu < 0$), while for the DNA/PG4 complex these two terms are about equal ($\nu \approx 0$). This indicates that the overall charge of the complex might be selected by using polyelectrolytes with different flexibilities and linear charge densities.

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Conclusion

In summary, we propose a molecular level structural model for a DNA/dendronized polymer complex, according to which the polyelectrolyte with the smaller linear charge density (DNA) is wrapped around the more highly charged dendronized polymer. For the different dendron generations, we propose that the interplay between the electrostatic energy and elastic energy defines both the overall charge of the complex and the different pitch sizes for the wrapped DNA. The dendronized polymers together with DNA are a useful model system to test theories on the interaction of oppositely charged polyelectrolytes. Further experiments should elucidate the influence of the stiffness of the polyelectrolytes as well as the salt concentration on the complex formation. Moreover, this novel complex might be used for nonviral gene delivery systems and help to optimize the transfection efficiency based on the structure of the vector system.

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