Supporting Information

Computer simulations of the interactions of high-generation polyamidoamine dendrimers with electronegative membranes

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Table S1 shows the R_g values of PAMAM dendrimers as a function of generation. For comparison, we also show R_g values obtained from previous molecular simulations [1,2] and experimental measurements. [3–5] It can be seen that the R_g values in our simulations agree well with that from atomistic simulations [1,2]as well as the SAXS [3]and SANS [4,5] results for G7,G8 and G9 PAMAM dendrimers.

Table S2 shows that the values of area per lipid (APL) from our simulations and all-atom molecular dynamics. The APL has generally served as a reliable validation for membrane due to its direct relationship with the structural and dynamical properties of membrane. [6,7] It can be found from the table that our coarse-grained DMPC and DMPG bilayers, the APL of which is in good agreement with that of atomistic simulations, successfully represent the properties of a charged membrane. As for the simulation of the 3:7 DMPG/DMPC membrane bilayer, our results show that the negatively charged DMPG lipids distribute evenly in the bilayer, and there is no evident aggregation for each lipid species. This may be due to the strong repulsion among the negative charges of DMPG lipids.

Table S 1 Radius of Gyration (R_g)(Å) as a function of generation for PAMAM

Dendrimer	this work	AAMD	SAXS and SANS
G7	32.5±0.2	28.05,[1]29.09[2]	31.9,[3]34.4[4]
G8	40.1±0.1	36.48,[1]36.42[2]	40.3,[3]39.5[5]
G9	50.5±0.1	43.63,[1]46.03[2]	49.2,[3]

Table S 2 Area per lipid (Å^2) for DMPC and DMPG bilayers

	this work	AAMD[8]
DMPC	60.1±0.1	60.0±1.1
DMPG	60.2±0.1	61.8±1.2

The cumulative number of water and CL- with respect to the center of each dendrimer was computed in Fig. S 2. One can clearly see a plain region corresponds to the hydrophobic part of for each dendrisome. Also, the number of CL- within each dedrisome is smaller than that within each free dendrimer in water. This can be attributed to the neutralization of dendrimers by negatively charged lipids DMPG. The numbers of CL- are 20, 177 and 516 for G7, G8 and G9 dendrisomes, respectively. The data suggest that dendrimer terminals can fold back to the inner part of the dendrimer. The present simulations show that the number of CG water in each dendrisome only has a slight change compared with that compared with that in the free dendrimer. They are 771, 1417 and 2710 for G7, G8 and G9 dendrisomes, respectively. Note that one CG water molecule corresponds to four real water molecules in the MARTINI model. [9,10]

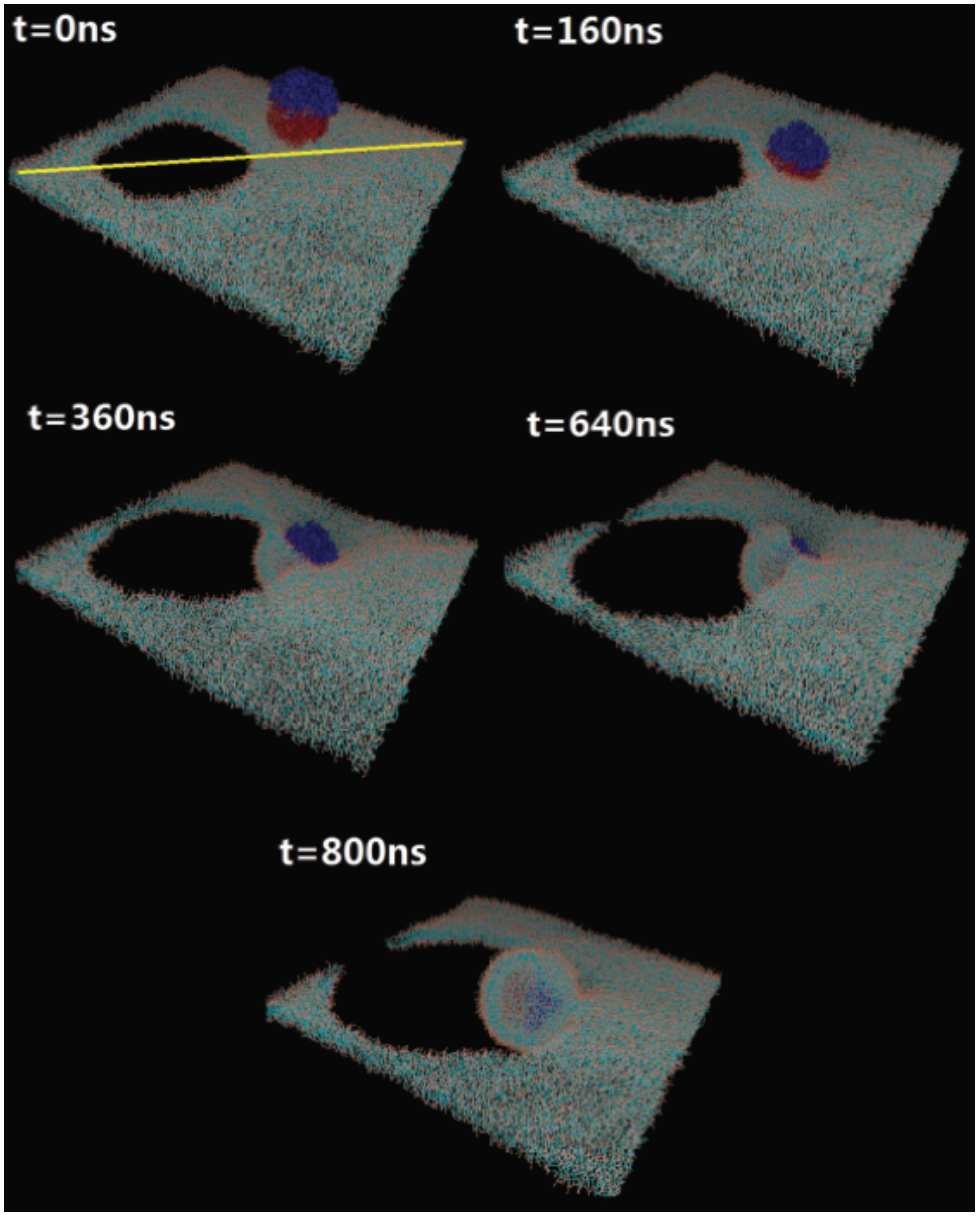

Fig. S 1 Time sequence of the G9 dendrimer interacting with the DMPG bilayer. The dendrimer was divided into two hemispheres colored blue and red independently. The yellow line was used to side view of the system in the text.

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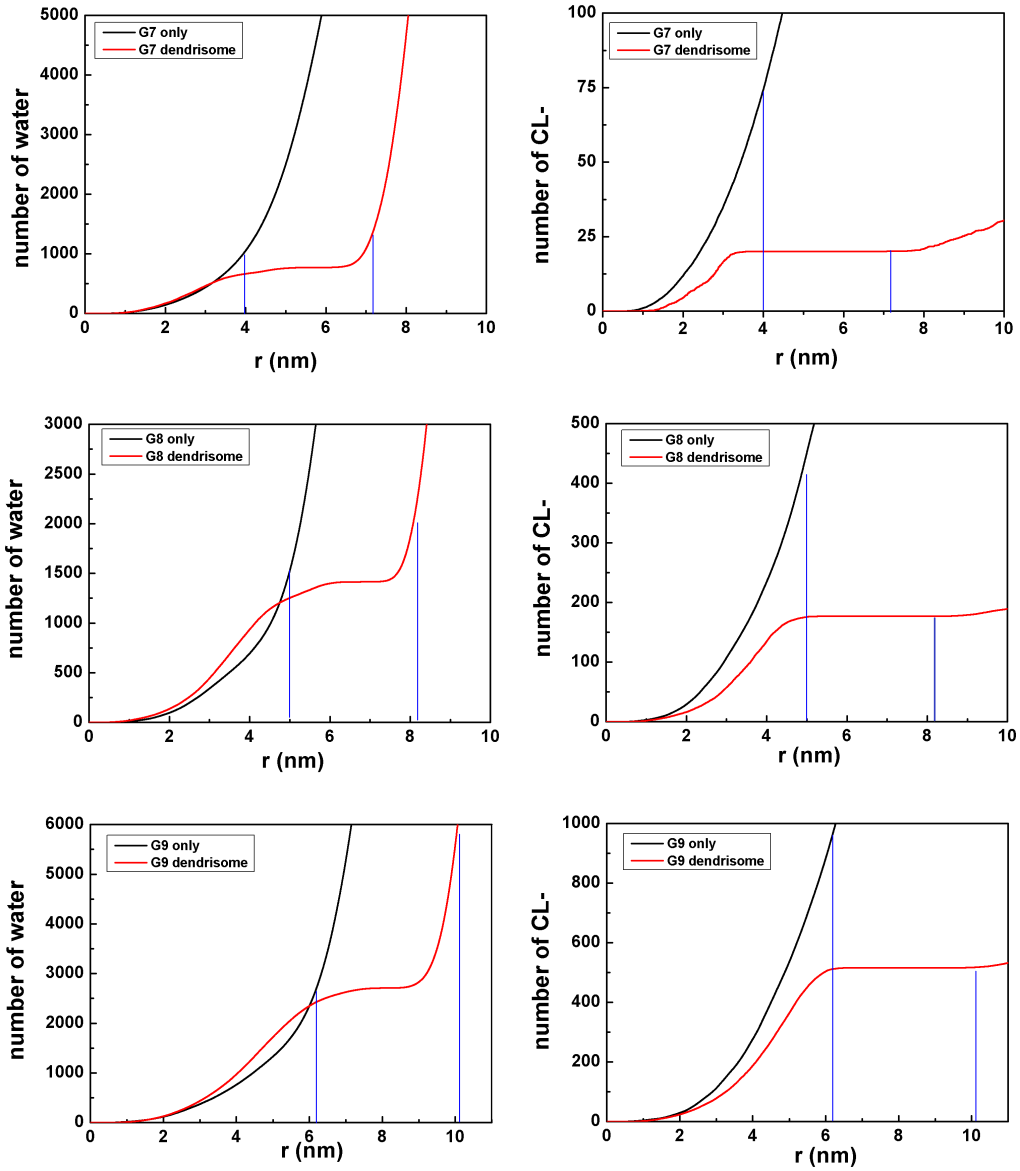

Fig. S 2 The cumulative number of water and CL- with respect to the center of mass of each dendrimer. The phase *Gn* only represents the case that the free dendrimer is submerged in water, while *Gn* dendrisome represents the case the denderimer is encased in the vesicle. The blue lines in each graph correspond to the inner and outer radii of the dendrisomes.