Supplementary Information

Preparation and evaluation of PAMAM dendrimer-based polymer gels physically cross-linked by hydrogen bonding

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S1. Preparation of dendrimers with ethylenediamine as the core

PAMAM dendrimer was synthesized from methyl acrylate (MA) and ethylenediamine (EDA) as reported before.[1,2] MA was added dropwise to EDA solution in 25 mL of methanol under the protection of nitrogen and the solution was stirred at 0-4 °C for 30 min. Then the solution was stirred at room temperature for additional 24 h. The mixture was transferred to a rotary evaporator at 50 °C to remove methanol and excess MA, and a half generation (0.5G) PAMAM dendrimer was obtained. Then a certain amount of EDA was added dropwise to 0.5G PAMAM dendrimer in 25 mL of methanol in a nitrogen atmosphere, and the solution was stirred at 0-4 °C for 30 min. The solution

was stirred for another 24 h at room temperature. The solution was transferred to a rotary evaporator at 55 ^{o}C to remove excess methanol and EDA, and 1.0G PAMAM was prepared. The procedure is similar to get high generation dendrimers.

Preparation of dendrimers with 1,4-diaminobutane as the core

MA was added dropwise to the 1,4-diaminobutane (DAB) solution in 25 mL of methanol under the protection of nitrogen and the solution was stirred at 0-4 ^{o}C for 30 min. Then the solution was stirred for additional 24 h at room temperature. The solution was transferred to a rotary evaporator at 50 ^{o}C to remove methanol and excess MA, and 0.5G PAMAM dendrimer was obtained. A certain amount of DAB was added dropwise to 0.5G PAMAM dendrimer in 25 mL of methanol in the nitrogen atmosphere and the solution was stirred at 0-4 ^{o}C for 30 min. The solution was stirred for 32 h at room temperature. The solution was transferred to a rotary evaporator at 65 ^{o}C to remove excess methanol and DAB. It is well-known that with the increase of carbon chain, the boiling point of aliphatic amines increase. So the excess aliphatic amines cannot be completely removed by using a rotary evaporator. The crude products were settled with hexane after evaporation, then 1.0G PAMAM was prepared. The pure product was achieved after drying. The procedure is similar to get high generation dendrimers.

Preparation of dendrimers with 1,6-hexamethylenediamine as the core

MA was added dropwise to the 1,6-hexamethylenediamine (HMD) solution in 25 mL of methanol under the protection of nitrogen, and the solution was stirred at 0-4 ^{o}C for 30 min. Then the solution was stirred for 24 h at room temperature. The products were transferred to a rotary evaporator to remove methanol and excess MA, and a half 0.5G PAMAM dendrimer was obtained. A certain amount of HMD was added dropwise to 0.5G PAMAM dendrimer in 25 mL of methanol in the nitrogen atmosphere and the solution was stirred at 0-4 ^{o}C for 30 min. The solution was stirred for 36 h at room temperature. The solution was transferred to a rotary evaporator at 70 ^{o}C to remove excess methanol and HMD. The crude products were settled with ice ether after evaporation, then 1.0G PAMAM was prepared. The pure product was achieved after

drying. The procedure is similar to get high generation dendrimers.

Preparation of dendrimers with 1,8-diaminooctane as the core

MA was added dropwise to the 1,8-diaminooctane (OMDA) solution in 25 mL of methanol under the protection of nitrogen and the solution was stirred at 0-4 ^{o}C for 30 min. Then the solution was stirred for 24 h at room temperature. The products were transferred to a rotary evaporator to remove methanol and excess MA, and 0.5G PAMAM dendrimer was obtained. A certain amount of OMDA was added dropwise to 0.5G PAMAM dendrimer in 25 mL of methanol in the nitrogen atmosphere and the solution was stirred at 0-4 ^{o}C for 30 min. The solution was stirred for 42 h at room temperature. The products were transferred to a rotary evaporator at 85 ^{o}C to remove excess methanol and OMDA. The crude products were settled with ice ether after evaporation, then 1.0G PAMAM was prepared. The pure product was achieved after drying. The procedure is similar to get high generation dendrimers.

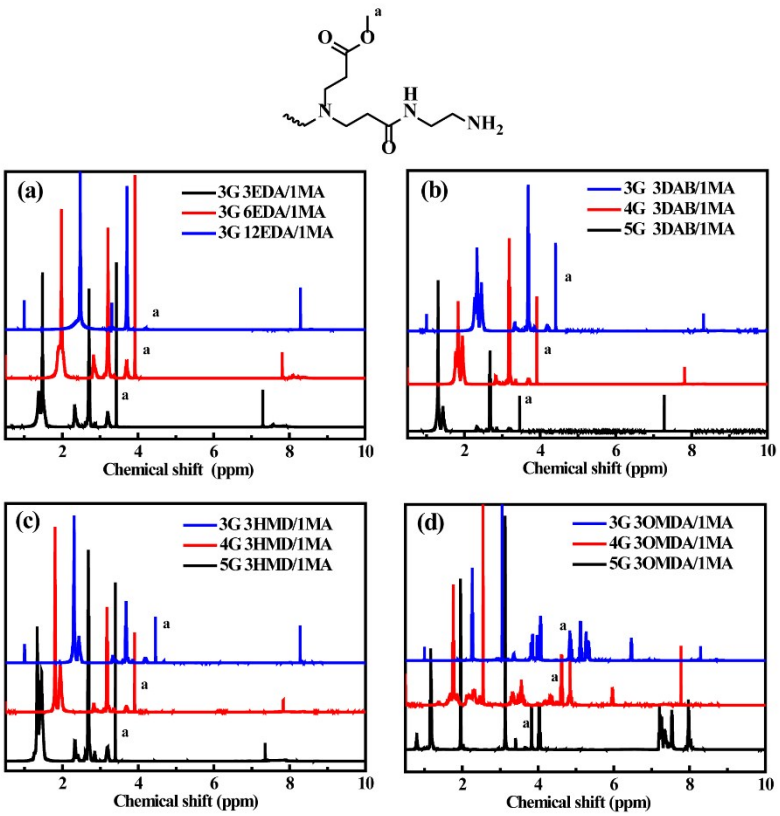

Fig. S1 ^{1}H NMR spectra of dendrimers before forming polymer gels.

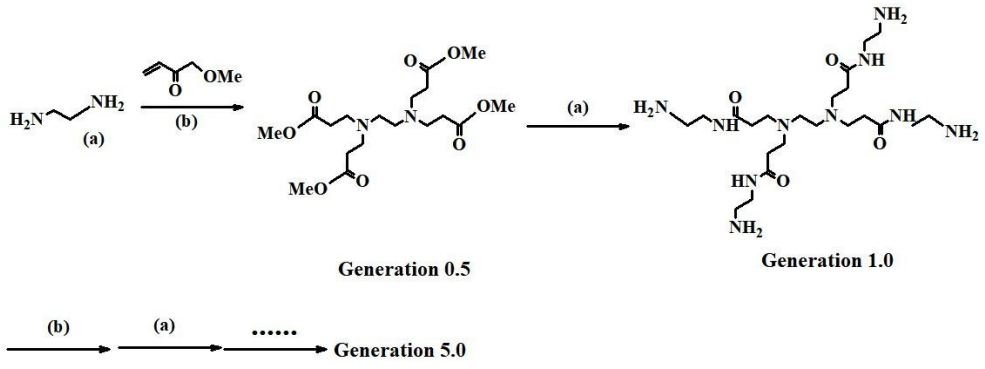

Fig. S2 Schematic diagram of synthetic dendrimers

Table S1 Synthesis conditions of dendrimers

Dendrimer	EDA			DAB			HMD			OMDA		
Generation	3G	4G	5G	3G	4G	5G	3G	4G	5G	3G	4G	5G
Reaction time (h)	12	24	36	14	26	30	16	28	42	18	32	48
Temperature (^{o}C)	25	30	30	25	30	30	30	35	35	30	35	35

Table S2 Purification conditions of dendrimers

Dendrimer	EDA			DAB			HMD			OMDA		
Generation	3G	4G	5G	3G	4G	5G	3G	4G	5G	3G	4G	5G
Rotary evaporation	2h			2.5h			3h			4h		
	55 ^{o}C			65 ^{o}C			70 ^{o}C			85 ^{o}C		
Purification	/			Hexane			Ice ether			Ice ether		

Table S3 Molar ratio of synthetic dendrimers and polymer gels

		EDA PAMAM	DAB PAMAM	HMD PAMAM	OMDA PAMAM
Dendrimer	0.5G	EDC:MA 1:4	DAB:MA 1:4	HMD:MA 1:4	OMDA:MA 1:4
	1.0G	0.5G:EDC 1:4	0.5G:DAB 1:4	0.5G:HMD 1:4	0.5G:OMDA 1:4
	1.5G	1.0G:MA 1:8	1.0G:MA 1:8	1.0G:MA 1:8	1.0G:MA 1:8
	2.0G	1.5G:EDC 1:8	1.5G:DAB 1:8	1.5G:HMD 1:8	1.5G:OMDA 1:8
	2.5G	2.0G:MA 1:16	2.0G:MA 1:16	2.0G:MA 1:16	2.0G:MA 1:16
	3.0G	2.5G:EDC 1:16	2.5G:DAB 1:16	2.5G:HMD 1:16	2.5G:OMDA 1:16
	3.5G	3.0G:MA 1:32	3.0G:MA 1:32	3.0G:MA 1:32	3.0G:MA 1:32
	4.0G	3.5G:EDC 1:32	3.5G:DAB 1:32	3.5G:HMD 1:32	3.5G:OMDA 1:32
	4.5G	4.0G:MA 1:64	4.0G:MA 1:64	4.0G:MA 1:64	4.0G:MA 1:64
	5.0G	4.5G:EDC 1:64	4.5G:DAB 1:64	4.5G:HMD 1:64	4.5G:OMDA 1:64
Gel	3.0G	2.5G:EDC 1:3	2.5G:DAB 1:3	2.5G:HMD 1:3	2.5G:OMDA 1:3
	4.0G	3.5G:EDC 1:3	3.5G:DAB 1:3	3.5G:HMD 1:3	3.5G:OMDA 1:3
	5.0G	4.5G:EDC 1:3	4.5G:DAB 1:3	4.5G:HMD 1:3	4.5G:OMDA 1:3

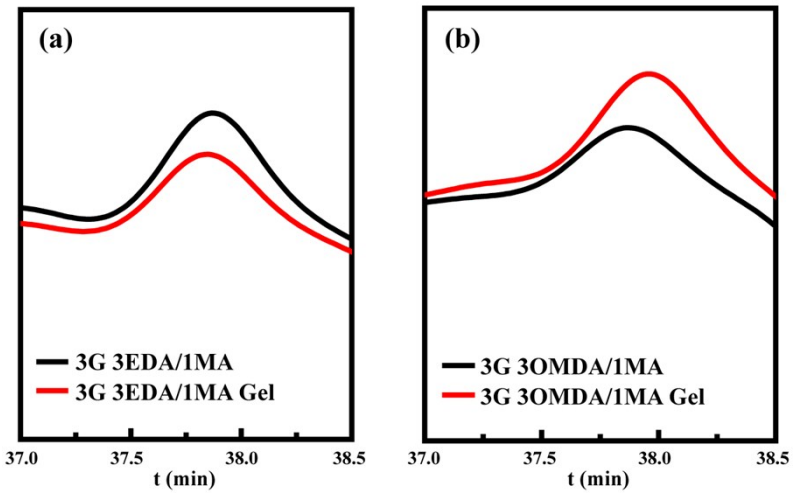

Fig. S3 GPC of dendrimers before and after forming polymer gels. To measure the molecular weight of the gels, the gels were immersed into acetate buffer (pH=5.0) until becoming a solution. Hydrogen bonds were broken due to the protonation of the amino groups. Then the solution was used for GPC.

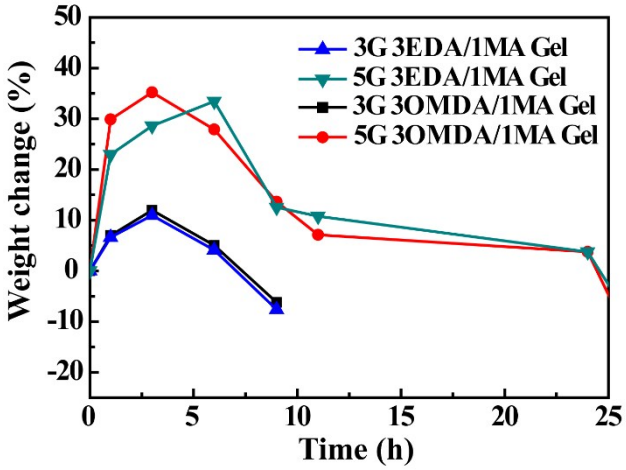

Fig. S4 Weight change of dendrimer-based polymer gels under acidic condition (acetate buffer, pH=5.0).

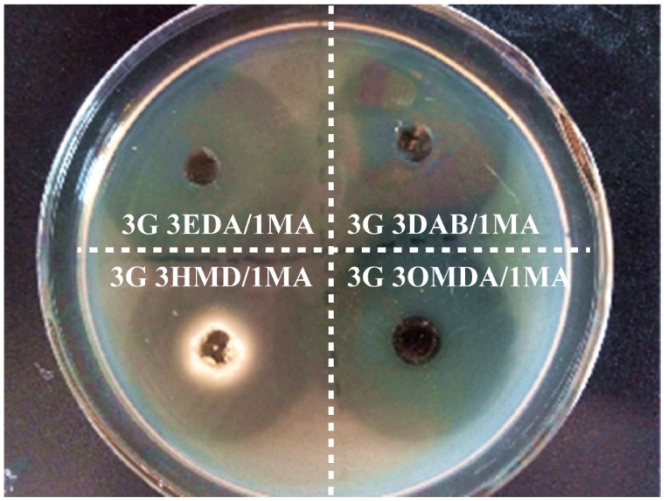

Fig. S5 Antibacterial properties of 3G dendrimers against *Escherichia coli*.

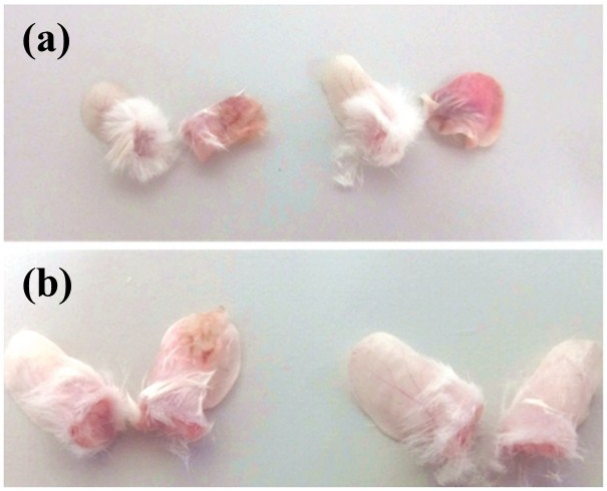

Fig. S6 Photographs of the mouse ears after being treated with (a) high concentration of dendrimers and (b) low concentration of dendrimers.

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