Electronic Supplementary Material (ESI) for CrystEngComm.

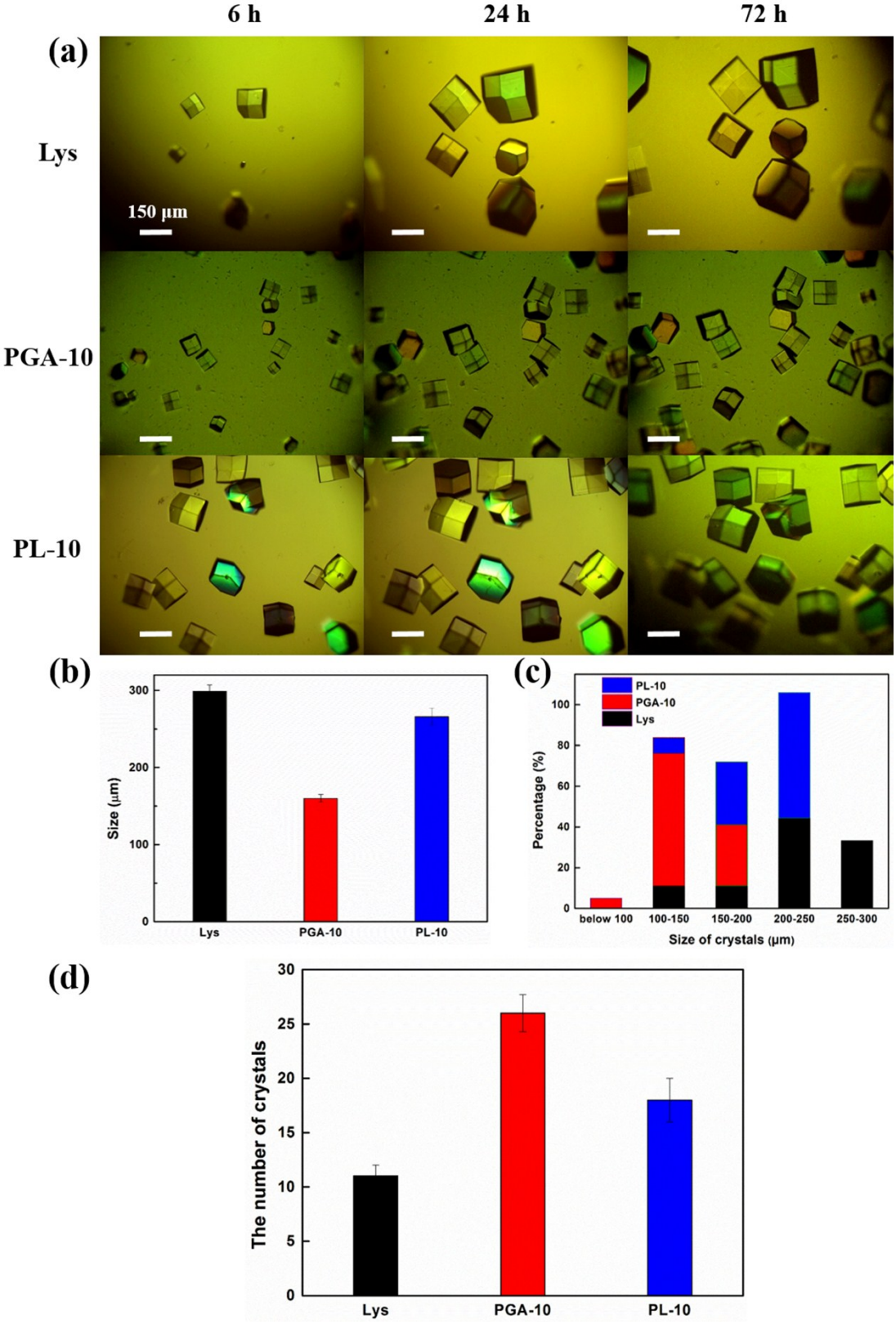


Fig. S1 (a) Photographs of lysozyme crystals grown at 10 °C for 6 h, 24 h, 72 h under the conditions of 15 mg/mL lysozyme and 0.6 M NaCl; together with the largest

length of lysozyme crystals (b), the size distribution of lysozyme crystals (c) and the number of lysozyme crystals (d) for 72h. Scale bar is 150 μm.

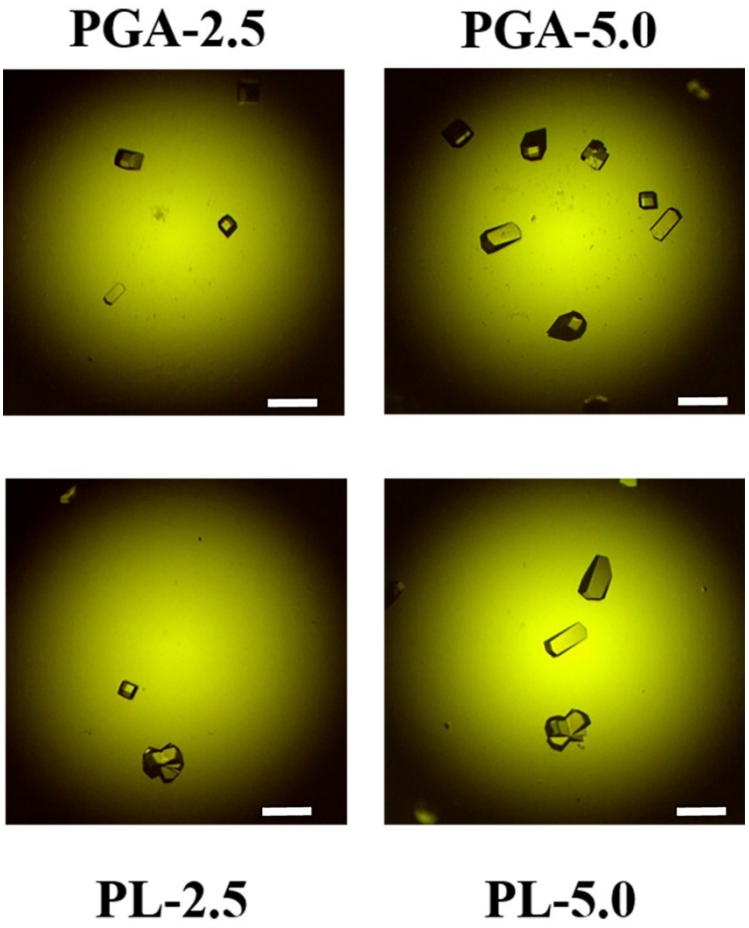


Fig. S2 Photographs of lysozyme crystals grown at 10 °C for 72 h in the presence of the additive (PGA-2.5, PGA-5.0, PL-2.5 and PL-5.0), under the conditions of 5 mg/mL lysozyme and 0.6 M NaCl. Scale bar is 150 μm.

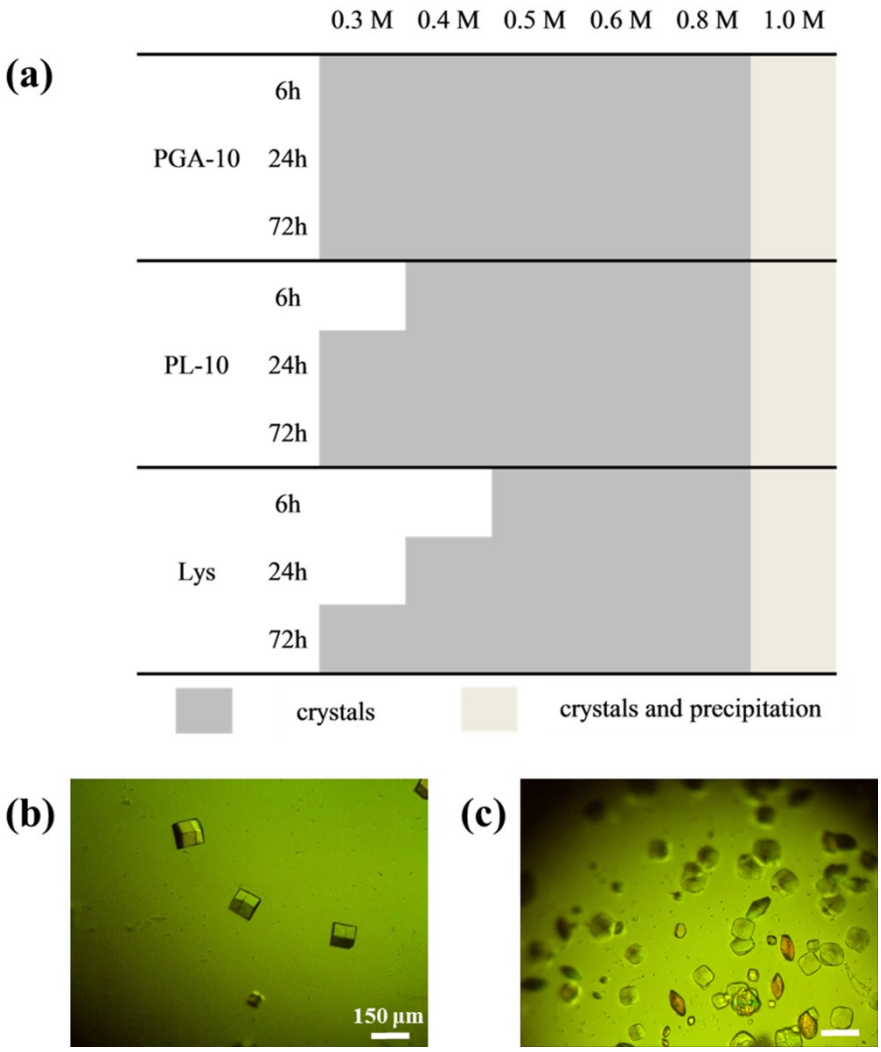


Fig. S3 (a) Results of lysozyme crystals (15 mg/mL) grown at 10 °C for 6 h, 24 h, 72 h with the different concentrations of NaCl in the absence of additives and in the presence of the additive with 10 mg/mL PGA and 10 mg/mL PL. (b) Photographs of lysozyme crystals grown in the presence of 10 mg/mL PL and 0.4 mg/mL NaCl at 10 °C for 6 h, and those grown in the presence of 10 mg/mL PGA and 0.3 mg/mL NaCl (c). Scale bar is 150 µm.

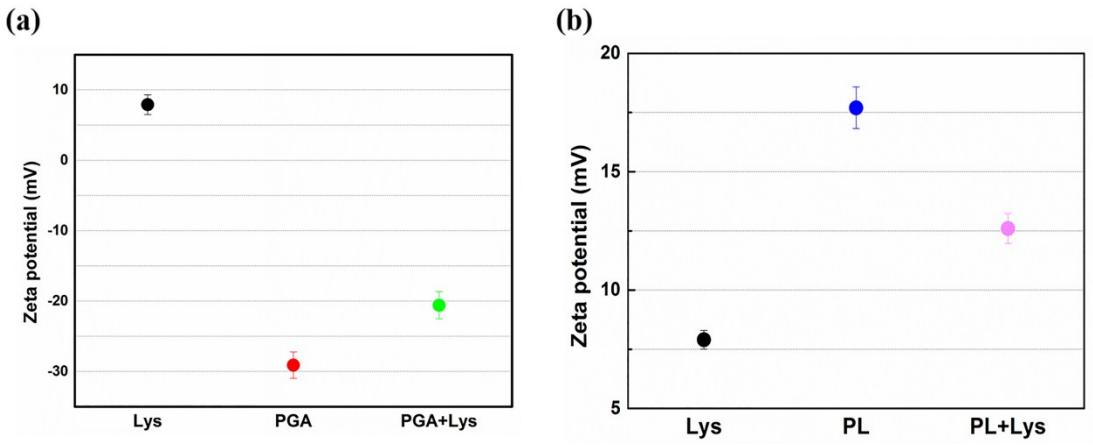


Fig. S4 Zeta potential results measured in solutions of 15 mg/mL protein, of 10 mg/mL PGA or 10 mg/mL PL and the mixture of the protein and the addition of PGA (a) or PL (b).

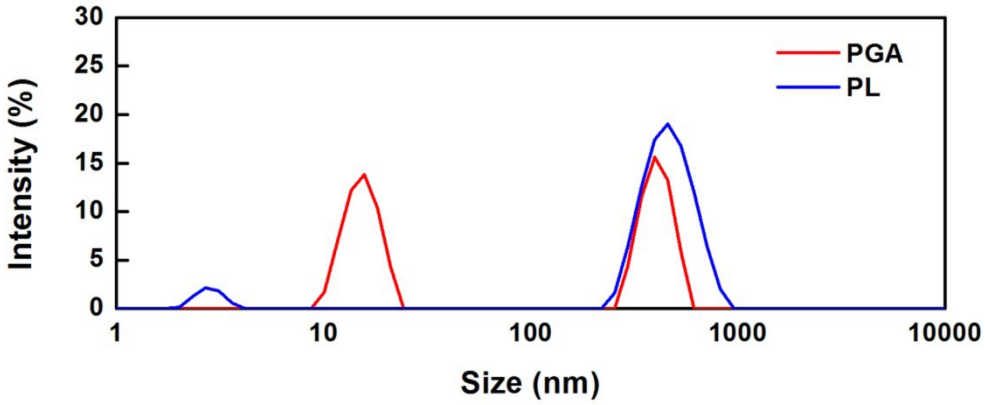


Fig. S5 Particle size measurement results in 10 mg/mL PGA and 10 mg/mL PL containing 0.1 M sodium acetate buffer and 0.6 M NaCl.

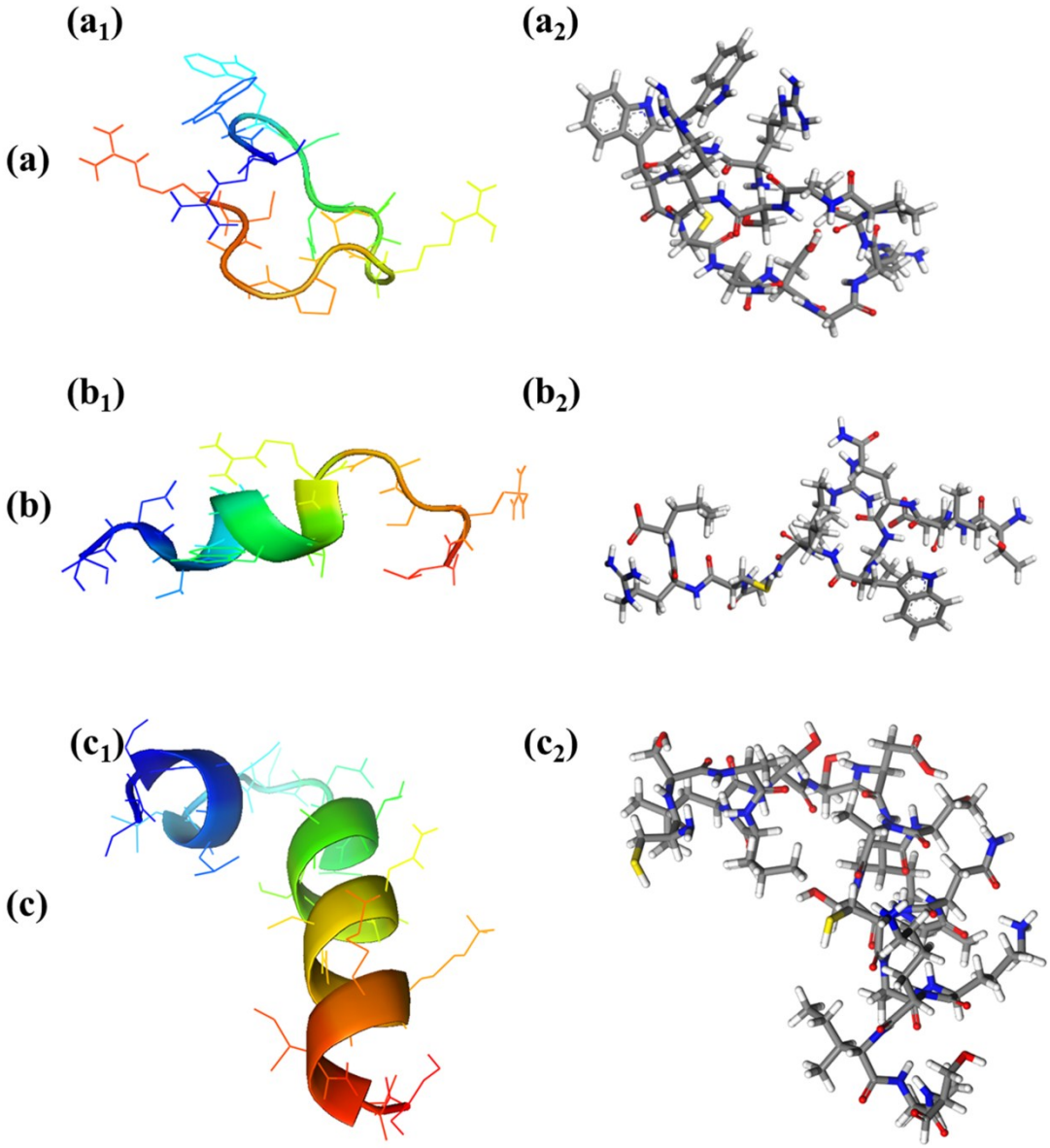
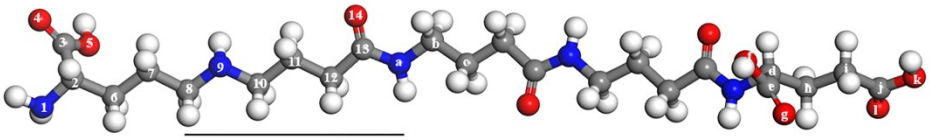


Fig. S6 The cartoon model of lysozyme segments with the random coil SEG1 (a_1), the helix-like structure SEG2 (b_1) and α-helical structure SEG3 (c_1), together with the corresponding stick model of the optimized structures after calculation using COMPASS force fields (a_2, b_2 and c_2).

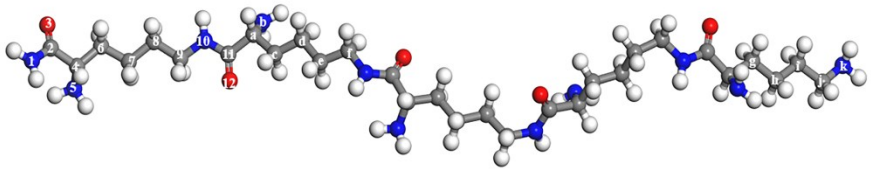
(a)



	group	charge (e)
1	-N*H$_2$-C-	-0.319
2	-NH$_2$-C*-	0.091
3	-C-C*OOH-	0.275
4	-C-CO*OH-	-0.265
5	-C-COO*H-	-0.331
6	-C-C*H$_2$-CH$_2$-	-0.027
7	-C-CH$_2$-C*H$_2$-	-0.039
8	-CH$_2$-C*H$_2$-	-0.005
9	-CH$_2$-N*H-CH$_2$-	-0.317
10	-CH$_2$-NH-C*H$_2$-	-0.004
11	-CH$_2$-C*H$_2$-CH$_2$-	-0.032
12	-CH$_2$-CH$_2$-C*H$_2$-	0.028
13	-CH$_2$-C*O-	0.220
14	-CH$_2$-CO*-	-0.275

	group	charge (e)
a	-CO-N*H-	-0.356
b	-NH-C*H$_2$-CH$_2$-	0.018
c	-NH-CH$_2$-C*H$_2$-	-0.027
d	-C*H-COOH-	0.115
e	-CH-C*OOH-	0.281
f	-CH-COO*H-	-0.264
g	-CH-CO*OH-	-0.330
h	-CH-C*H$_2$-CH$_2$-	-0.014
i	-CH-CH$_2$-C*H$_2$-	0.036
j	-CH$_2$-C*OOH-	0.258
k	-CH$_2$-COO*H-	-0.332
l	-CH$_2$-CO*OH	-0.267

(b)



	group	charge (e)
1	-N*H$_2$-CO-	-0.368
2	-NH$_2$-C*O-	0.234
3	-NH$_2$-CO*-	-0.274
4	-CO-C*H-NH$_2$-	0.084
5	-CO-CH-N*H$_2$-	-0.320
6	-C-C*H$_2$-CH$_2$-	-0.029
7	-C-CH$_2$-C*H$_2$-	-0.050
8	-CH$_2$-C*H$_2$-CH$_2$-	-0.036
9	-CH$_2$-CH$_2$-C*H$_2$-	0.017
10	-CH$_2$-N*H$_2$-CO-	-0.355
11	-CH$_2$-NH$_2$-C*O-	0.236
12	-CH$_2$-NH$_2$-CO*-	-0.273

	group	charge (e)
a	-CO-C*H-NH$_2$-	0.084
b	-CO-CH-N*H$_2$-	-0.320
c	-C-C*H$_2$-CH$_2$-	-0.029
d	-C-CH$_2$-C*H$_2$-	-0.050
e	-CH$_2$-C*H$_2$-CH$_2$-	-0.036
f	-CH$_2$-CH$_2$-C*H$_2$-	0.017
g	-C-C*H$_2$-CH$_2$-	-0.029
h	-C-CH$_2$-C*H$_2$-	-0.050
i	-CH$_2$-C*H$_2$-CH$_2$-	-0.041
j	-CH$_2$-CH$_2$-C*H$_2$-	-0.008
k	-CH$_2$-N*H$_2$	-0.330

Fig. S7 Structures of the optimized models of PGA (a) and PL (b) and their charging characteristics.

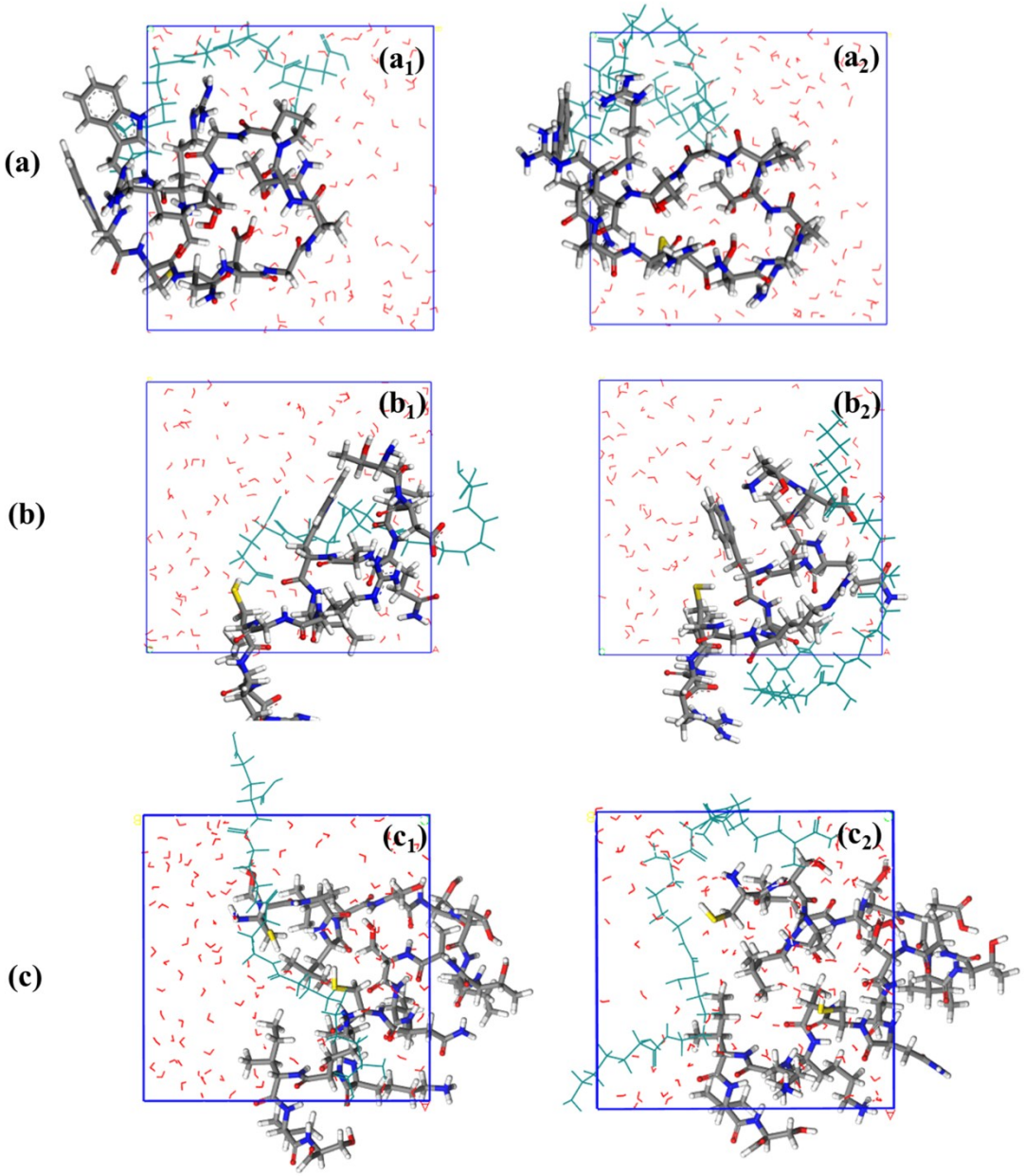


Fig. S8 Snapshot of three lysozyme segments with the random coil structure (a:SEG1), the helix structure (b:SEG2) and the α-helical structure (c:SEG3) interacted with PGA (with the subscript of 1) and PL (with the subscript of 2).