Supporting Information

Elucidating the influence of electrostatic force on the re-arrangement of H-bonds of protein polymer in the presence of salts

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Section S1. OPLS-AA forcefield parameters

The OPLS-AA forcefield parameters are: force constants k, equilibrium bond r_o and equilibrium angle θ_o, the Fourier coefficients V, the partial charges on each atom q, and the Lennard−Jones radii and well depths, σ and ε. The geometric combining rules utilized for the Lennard−Jones coefficients are: $\sigma_{ij} = (\sigma_{ii}\sigma_{jj})^{1/2}$ and $\varepsilon_{ij} = (\varepsilon_{ii}\varepsilon_{jj})^{1/2}$.[10] To retain compatibility, all the parameters were used without any modifications.

$$E_{bonds} = \sum_i k_{b,i}(r_i - r_{o,i})^2 \quad (1)$$

$$E_{angles} = \sum_i k_{b,i}(\theta_i - \theta_{o,i})^2 \quad (2)$$

$$E_{torsion} = \sum_i \left[\begin{array}{c} \frac{1}{2}V_{1,i}(1 + \cos\varphi_i) + \frac{1}{2}V_{2,i}(1 + \cos 2\varphi_i) \\ + \frac{1}{2}V_{3,i}(1 + \cos 3\varphi_i) + \frac{1}{2}V_{4,i}(1 + \cos 4\varphi_i) \end{array} \right] \quad (3)$$

$$E_{nonbond} = \sum_{i}\sum_{j>i}\left\{\frac{q_i q_j e^2}{r_{ij}} + 4\varepsilon_{ij}\left[\left(\frac{\sigma_{ij}}{r_{ij}}\right)^{12} - \left(\frac{\sigma_{ij}}{r_{ij}}\right)^{6}\right]\right\} \quad (4)$$

All the other parameters like, partial charges on each atoms, sigma, epsilon, angle, dihedral, and improper coefficients were taken from the original OPLS-AA forcefield parameter files present in the Gromacs directory.

Table T1: Hydration radius of kosmotropes and chaotropes.

Type of anions	Anions	Hydration Radius: R_H of anions (* 10^{-10} m)
Kosmotropes	SO^{2-}_4	2.57
	CH_3COO^-	2.50
Chaotropes	Cl^-	1.58
	NO^-_3	1.43

Table T2: Values of R_g at pH 7 from MD simulations. The R_g for kosmotropes is slightly lower than the chaotropes due to the higher hydration radius of kosmotropes.

Pure Gelatin	Salt Conc (%)	Na_2SO_4	CH_3COONa	NaCl	$NaNO_3$
1.79	0.25	1.78	1.79	1.80	1.81
	1	1.77	1.78	1.81	1.82
	2	1.8	1.81	1.89	1.84

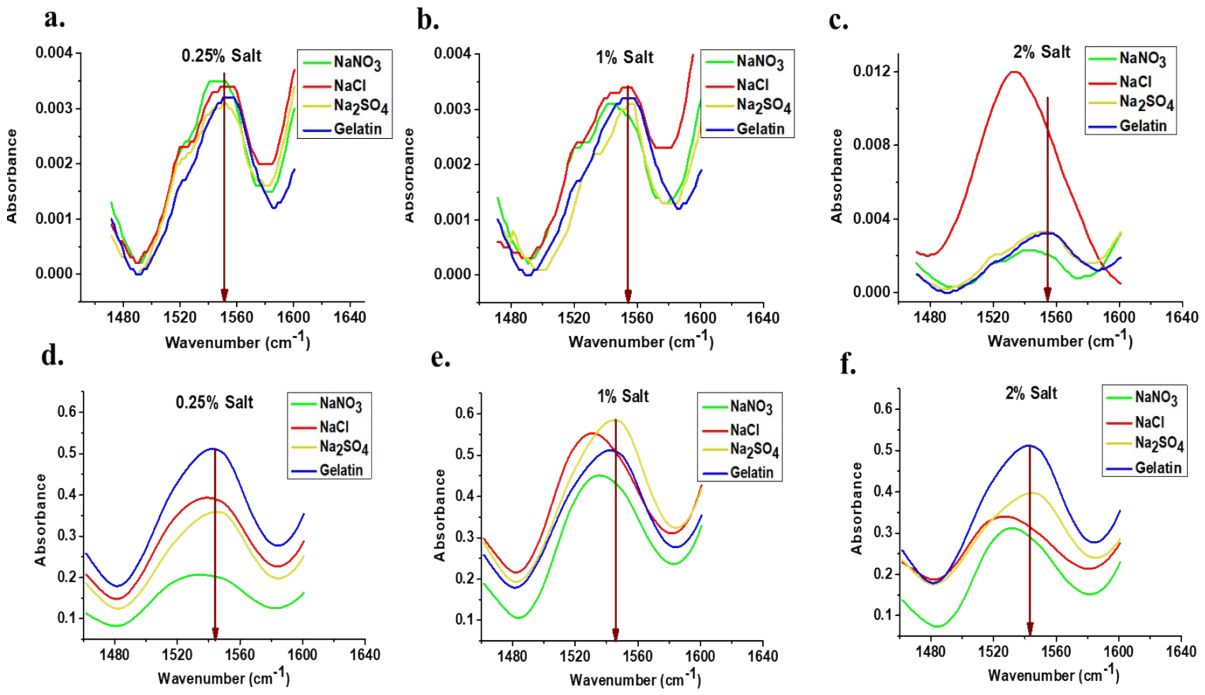

Fig S1: ATR/FTIR amide II bands of gelatin Type A with kosmotropes and chaotropes for (a) 0.25% salt concentration, (b) 1% salt concentration, and (c) 2% salt concentration. ATR/FTIR amide II bands of gelatin Type B with kosmotropes and chaotropes for (d) 0.25% salt concentration, (e) 1% salt concentration, and (f) 2% salt concentration. The chaotropes show a shift in the amide II band at 1% and 2% salt concentration as they are able to interact with gelatin.

Table T3: Values of R_g at pH 3 from MD simulations.

Pure Gelatin	Salt Conc (%)	Na_2SO_4	CH_3COONa	NaCl	$NaNO_3$
2.26	0.25	2.19	2.17	2.12	2.13
	1	2.07	2.03	1.98	1.97
	2	1.98	1.97	1.9	1.92

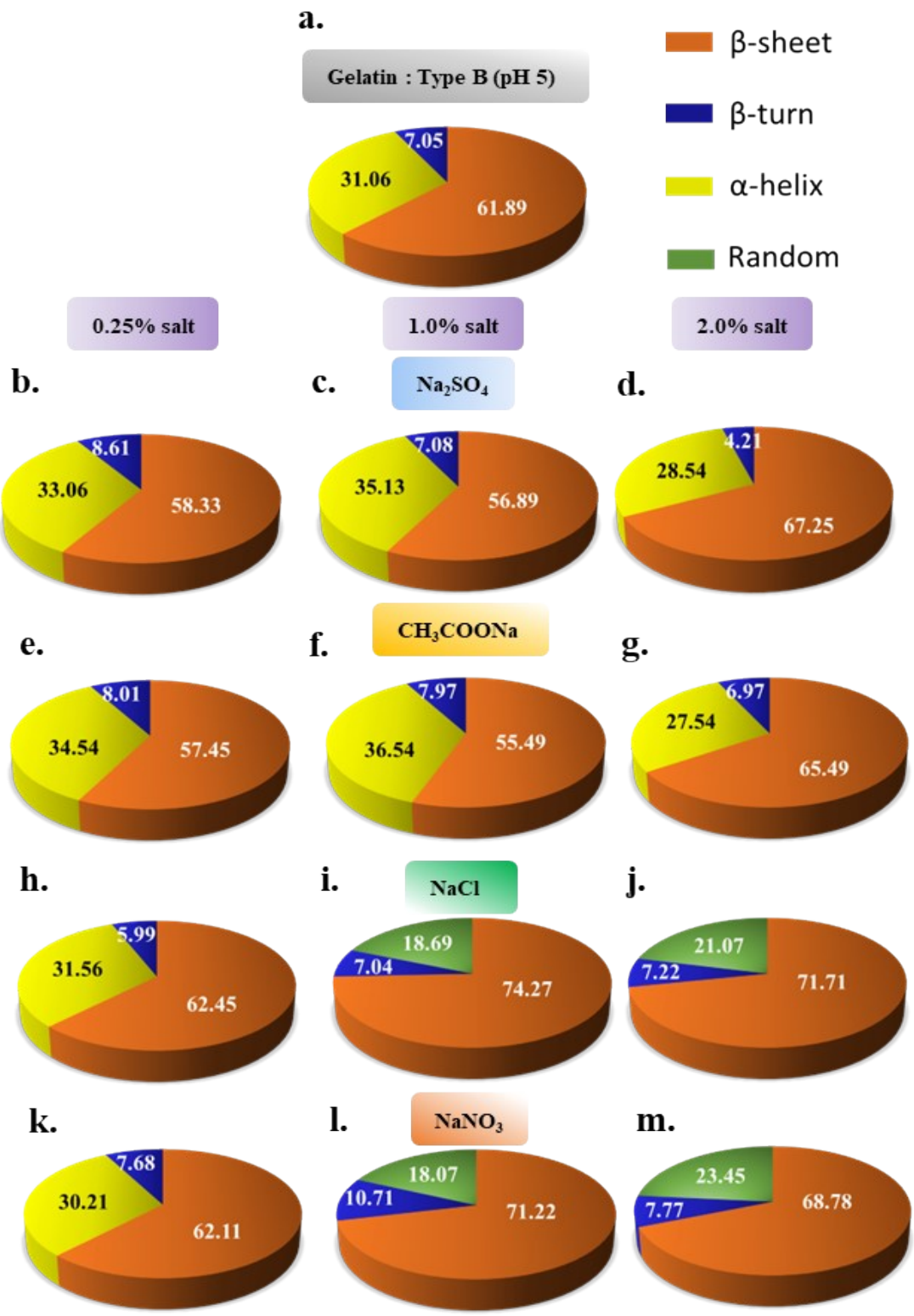

Fig. S2: Secondary structures of Gelatin (Type B) with different salts: 0.25%, 1%, and 2% salt concentrations at pH 5 (IEP). Not much change was observed for 0.25% salt. Kosmotropes: α-helix increases at 1% salt due to the collapse of chains and breaks at 2% salt due to screening of electrostatic attractions. Chaotropes: α-helix breaks into random structures.

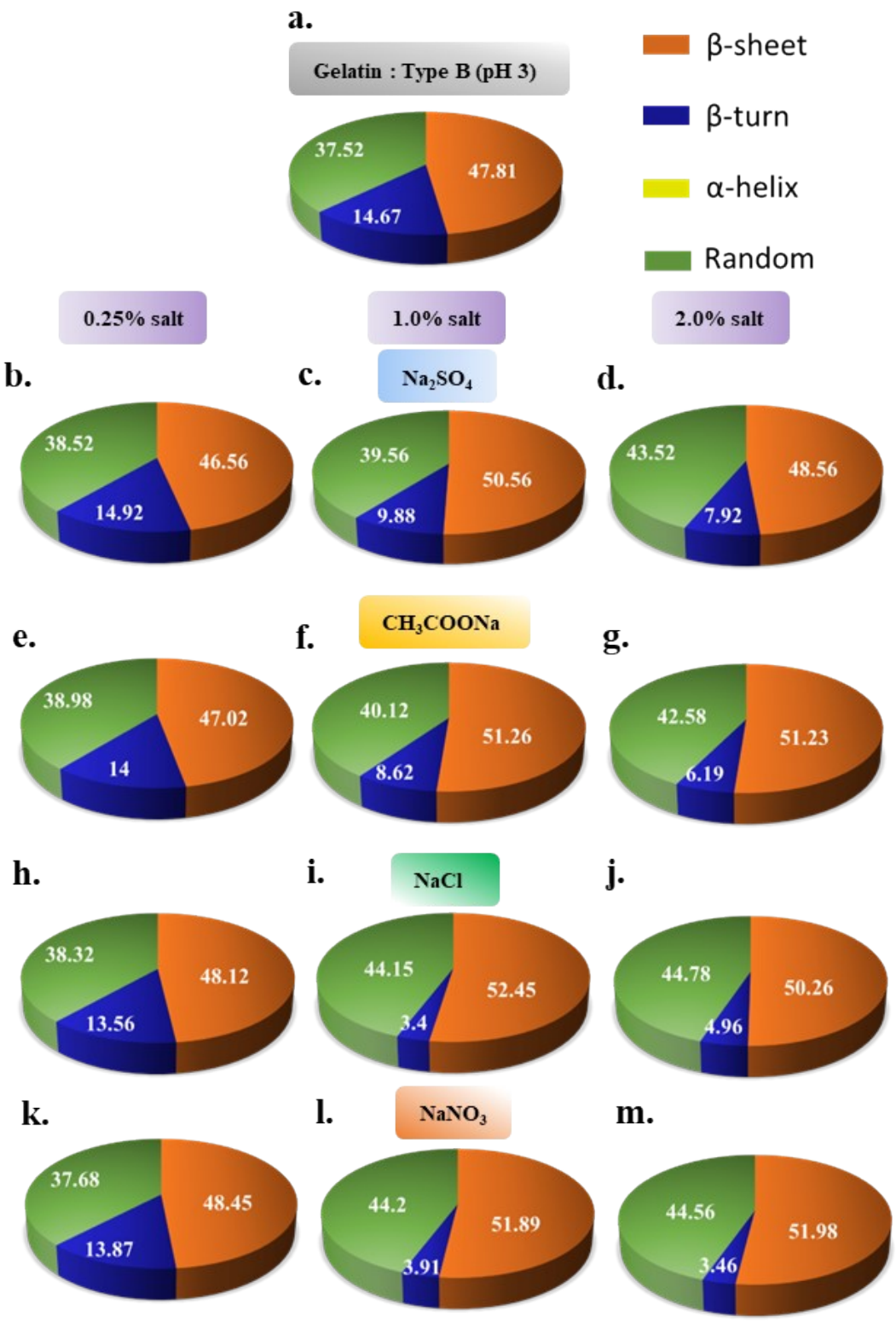

Fig. S3: Secondary structures of Gelatin (Type B) with different salts: 0.25%, 1%, and 2% salt concentrations at pH 3. At 1% and 2% salt β-sheets and random structure increases. Chaotropes interact more thus the chains are collapsed, which increases β-sheets compared to kosmotropes.

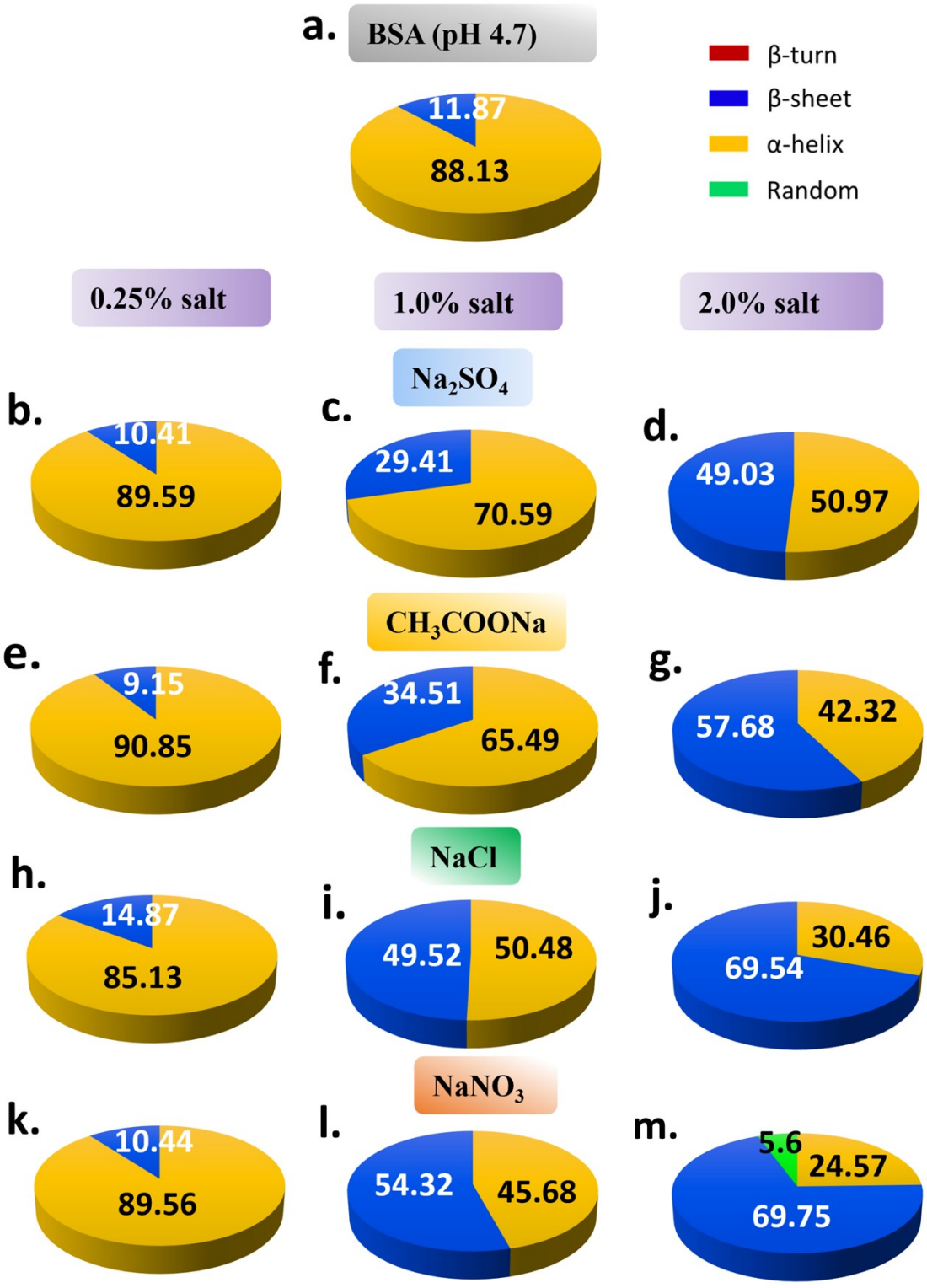

Fig. S4: Secondary structures of Bovine Serum Albumin (BSA) with different salts: 0.25%, 1%, and 2% salt concentrations at pH 4.7 (IEP). At 1% and 2% salt α-helix breaks to form β-sheets. Chaotropes interact more thus increasing β-sheets compared to kosmotropes.

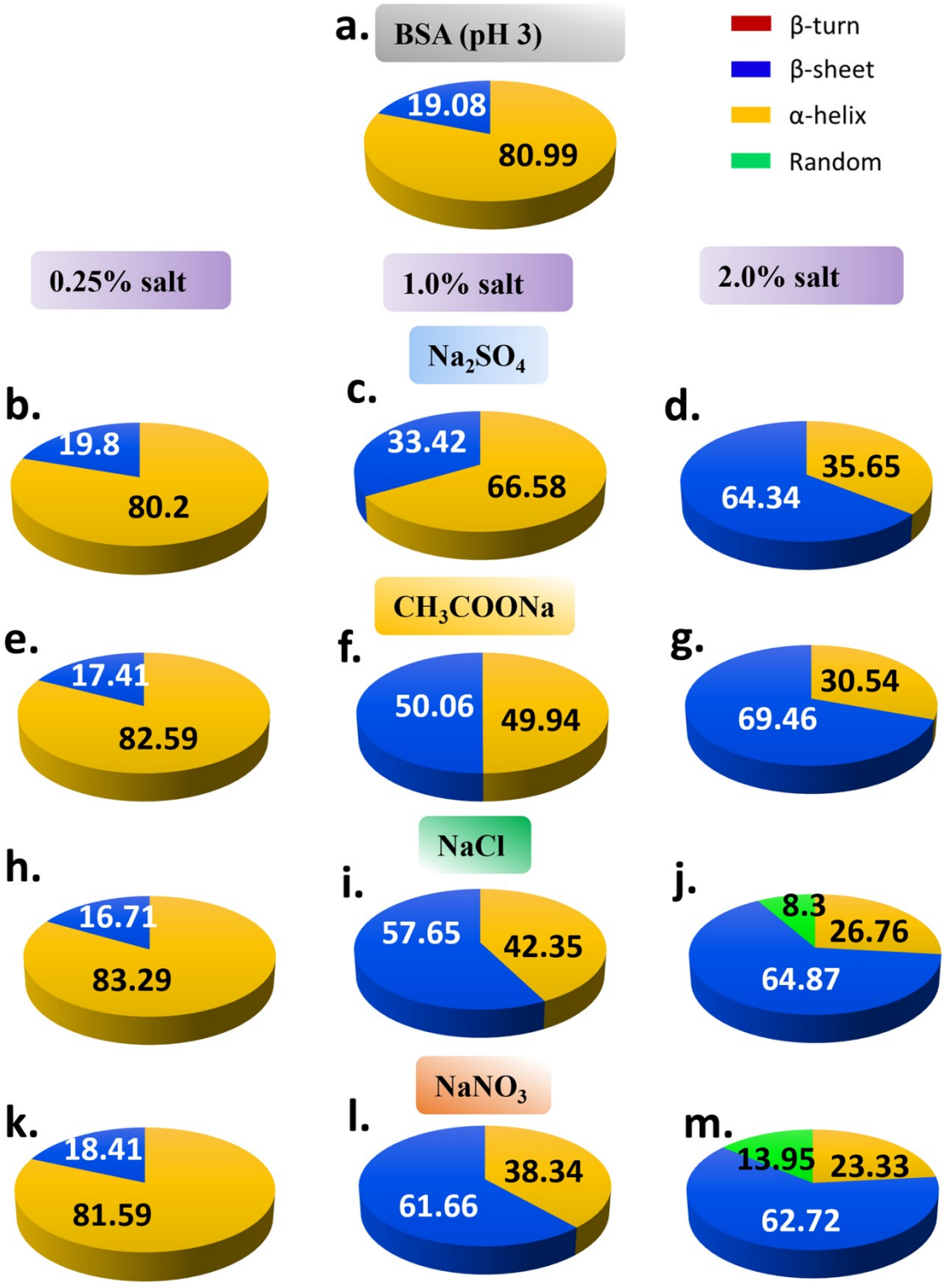

Fig. S5: Secondary structures of Bovine Serum Albumin (BSA) with different salts: 0.25%, 1%, and 2% salt concentrations at pH 3. Not much change was observed for 0.25% salt. Kosmotropes: α-helix breaks to form β-sheets. Chaotropes: α-helix breaks into random structures.

Table T4: Variation in gelatin secondary structure with time (Gelatin Type A with 2% NaCl).

Sample	β- sheet (%)	Random (%)	α- helix (%)	β - turn (%)
Gelatin	59.46	12.28	19.38	8.88
Gelatin & salt @1 min	66.61	28.33	-	5.05
Gelatin & salt @6 min	-	86.123	-	13.87
Gelatin & salt @11 min	8.14	75.45	-	16.40
Gelatin & salt @16 min	66.34	23.97	-	9.69
Gelatin & salt @21 min	68.31	24.01	-	7.68
Gelatin & salt @26 min	66.47	25.63	-	7.9