

Figure 1: (a) Typical evolution of turbidity as a function of surfactant concentration for complexes of oppositely charged surfactant and polyelectrolyte. (b) Illustration of packing parameter P_s in the case of a single molecule of surfactant; V_c and l_c represent the volume and the length of the surfactant tail, a_0 is the equilibrium area per polar group at the self-assembly surface. (c) Expected modes of self-assembly according to the value of packing parameter. (d) Typical ternary phase diagram of oppositely charged surfactants in pure water. (e) Possible shapes of surfactant/humic component associations that lead to a change in packing parameter with surfactant amount.

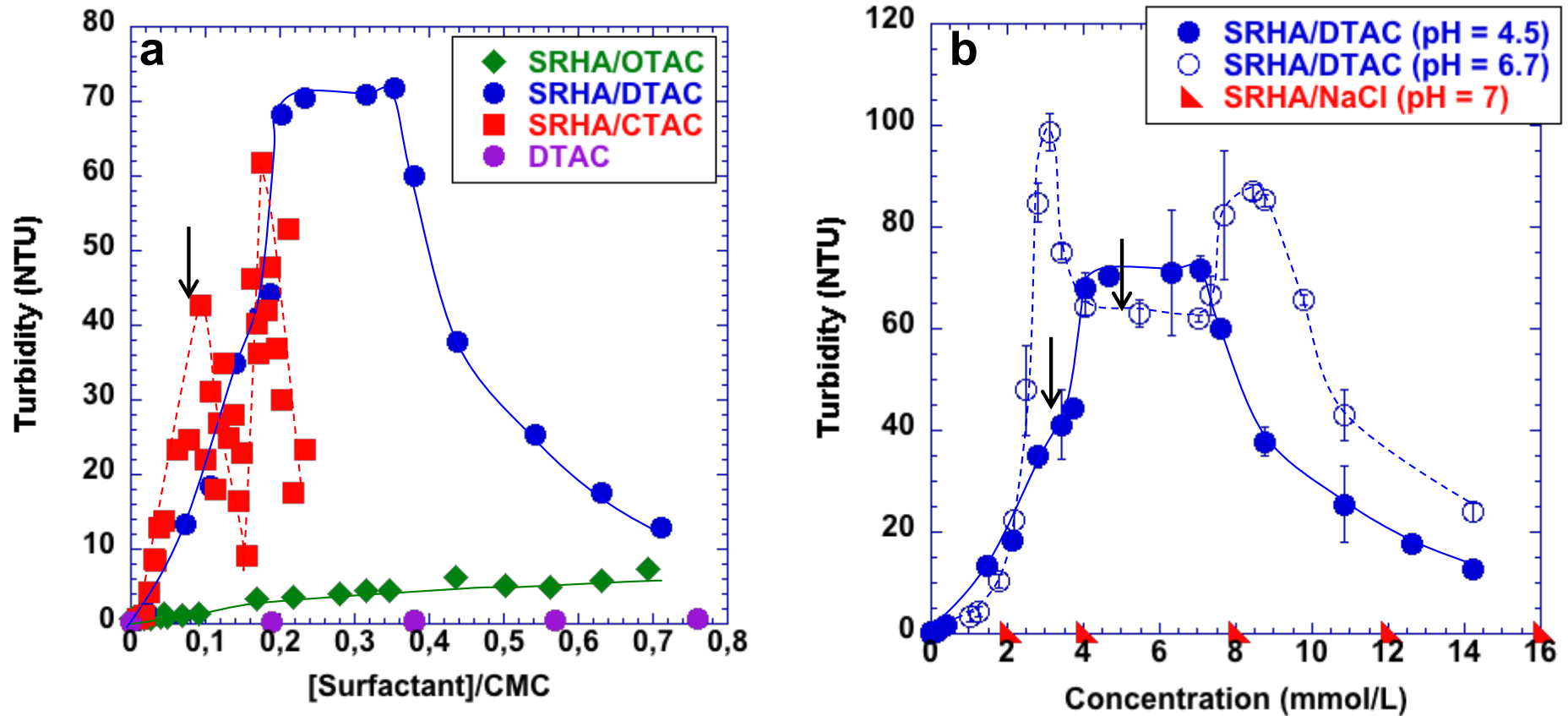


Figure 2: Turbidity of SRHA/surfactant suspensions vs concentration 1h after mixing ([SRFA] = 20 mg/L: (a) pH = 4.5 - The surfactant concentration is normalized to the critical micelle concentration (CMC) to take into account the difference in solubility of OTAC,DTAC, and CTAC. The DTAC addition in pure water (control) only induces a slight increase in turbidity. For sake of clarity, the error bars are not shown in fig 1a. (b) Influence of pH on the turbidity of SRHA/DTAC suspensions – The addition of NaCl provides an equivalent increase in solution conductivity as DTAC without any change in turbidity. The addition of DTAC to pure water leads to a negligible increase in turbidity. The error bars correspond to one standard deviation. The arrows indicate the Points of Zero Mobility. The lines in the figures are only a guide to the eye.

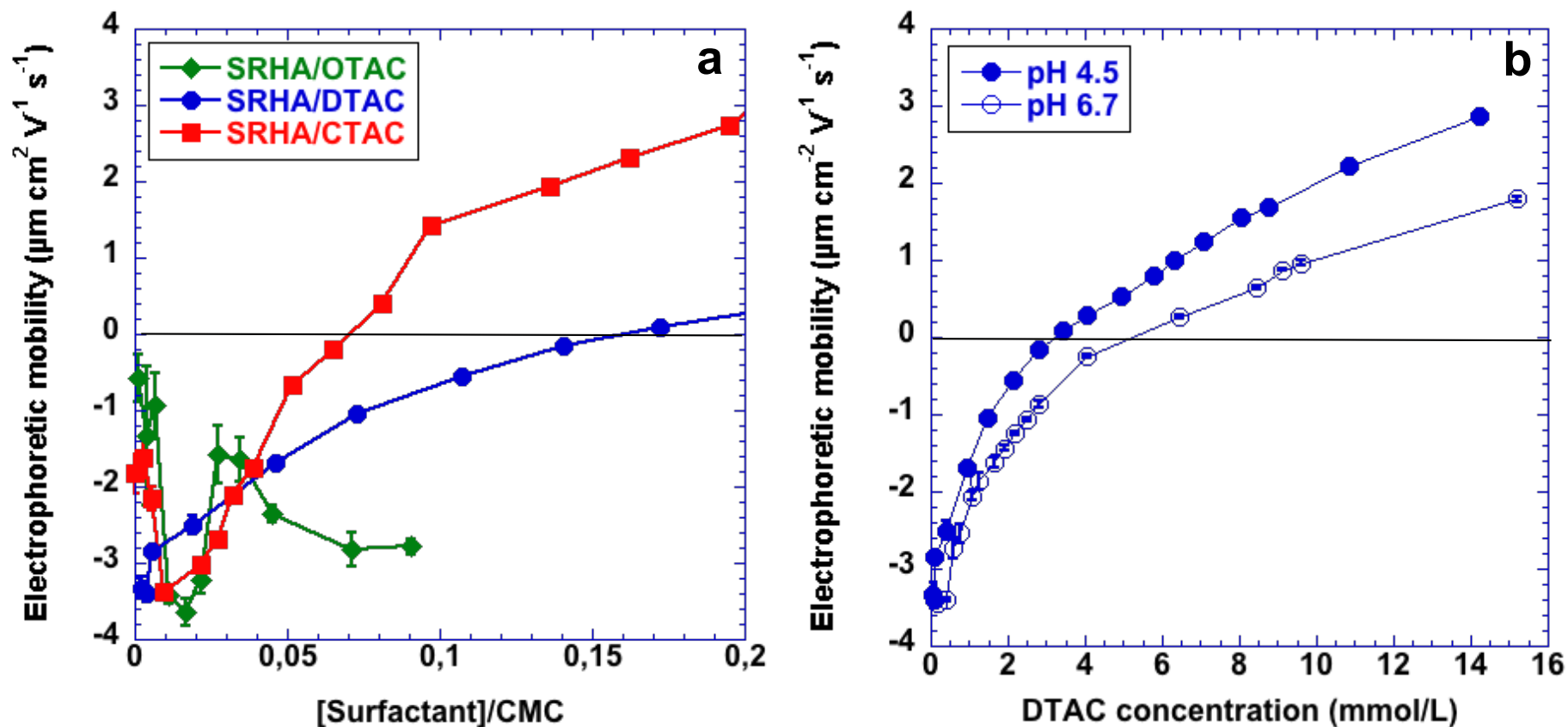


Figure 3: Electrophoretic mobility of SRHA/surfactant complexes: (a) pH 4.5 - The surfactant concentration is normalized to the critical micelle concentration (CMC) to take into account the difference in solubility of OTAC,DTAC, and CTAC. (b) Influence of pH on the electrophoretic mobility of SRHA/DTAC complexes versus DTAC concentration. Most error bars (one standard deviation) are smaller than the symbol size. The lines in the figures are only a guide to the eye.

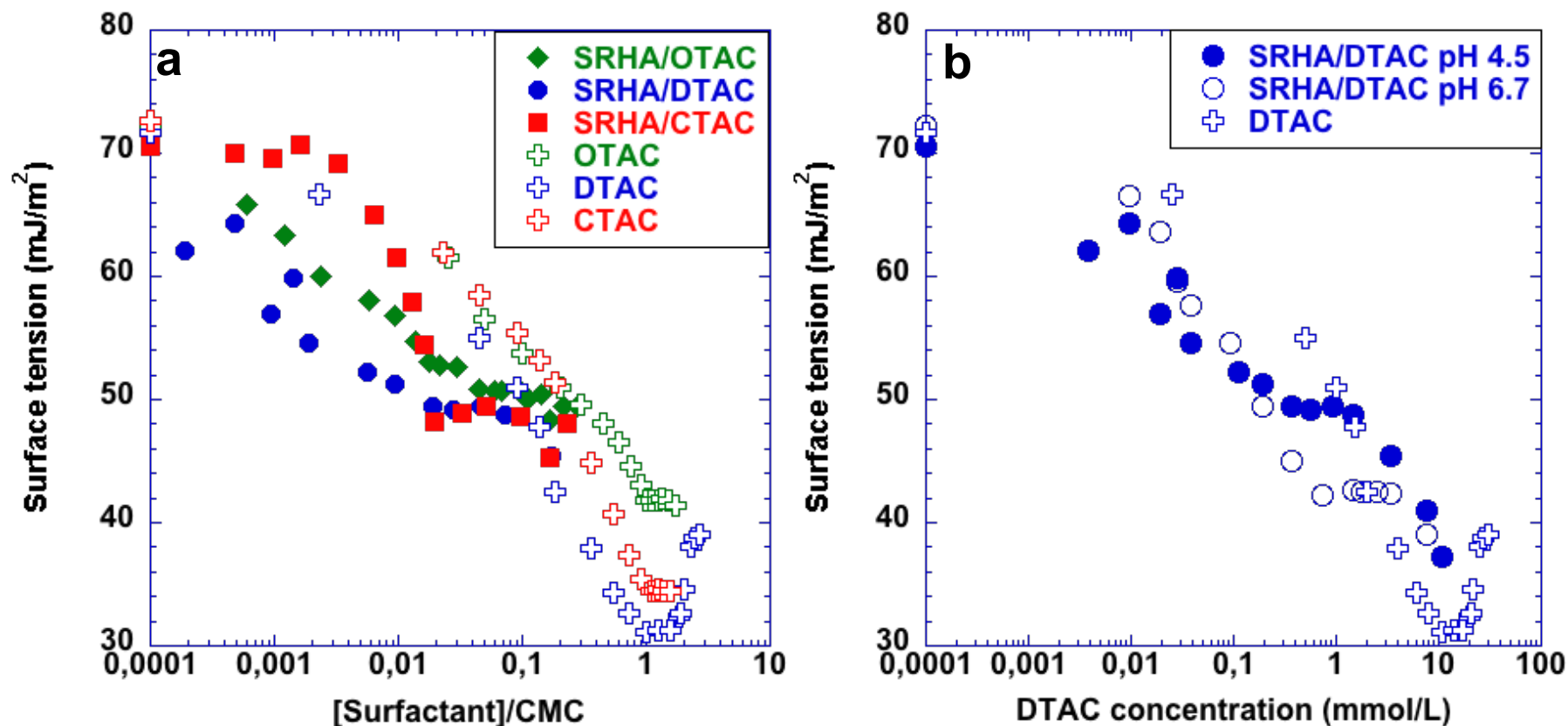


Figure 4: (a) Equilibrium surface tension in the system SRHA/alkyltrimethylammonium chloride as a function of surfactant concentration normalized to the respective CMC of surfactants at pH 4.5. (b) Effect of pH on the equilibrium surface tension of SRHA/DTAC suspensions. The surface tension curves of pure surfactants are given for comparison. Each data point is the average of three measurements.

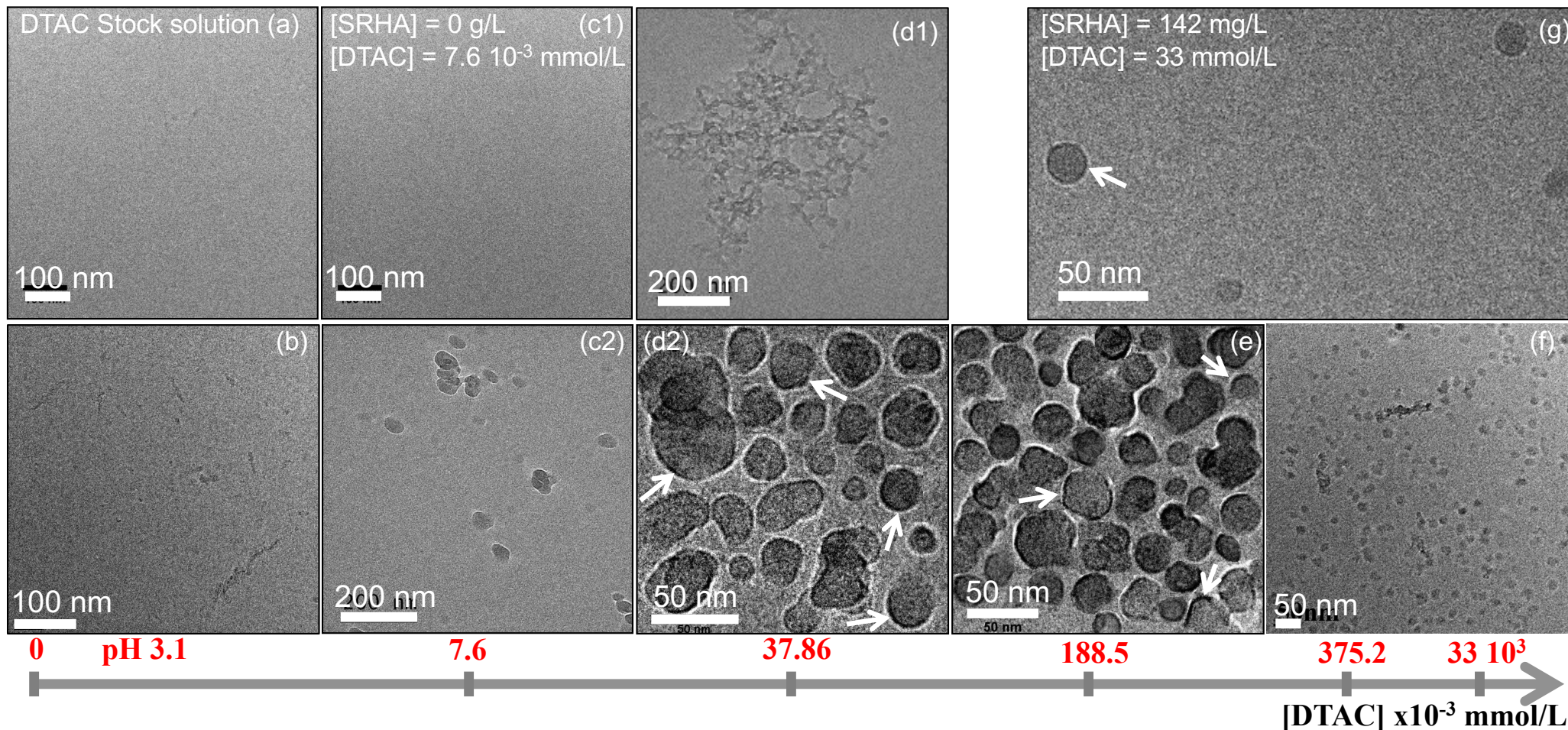


Figure 5: CryoTEM micrographs of SRHA/DTAC molecular structures obtained at pH 4.5 for various DTAC concentrations. (a) DTAC stock solution; (b) SRHA suspension of 20 mg/L at pH 4.5; (c1) DTAC solution diluted with deionised water to $[DTAC] = 7.6 \cdot 10^{-3}$ mmol/L; (c2) SRHA/DTAC complexes at $[DTAC] = 7.6 \cdot 10^{-3}$ mmol/L; (d1) Spongy network structure formed at $[DTAC] = 37.86 \cdot 10^{-3}$ mmol/L; (d2) Dense packing of unilamellar vesicles formed at $[DTAC] = 37.86 \cdot 10^{-3}$ mmol/L; (e); vesicular structures formed at $[DTAC] = 188.5 \cdot 10^{-3}$ mmol/L; (f) SRHA/DTAC molecular structures formed at $[DTAC] = 375.2 \cdot 10^{-3}$ mmol/L; (g) SRHA/DTAC well formed unilamellar vesicles at $[DTAC] = 33$ mmol/L; the white arrows indicate the higher electron density at the edges of vesicular structures. See text for details.

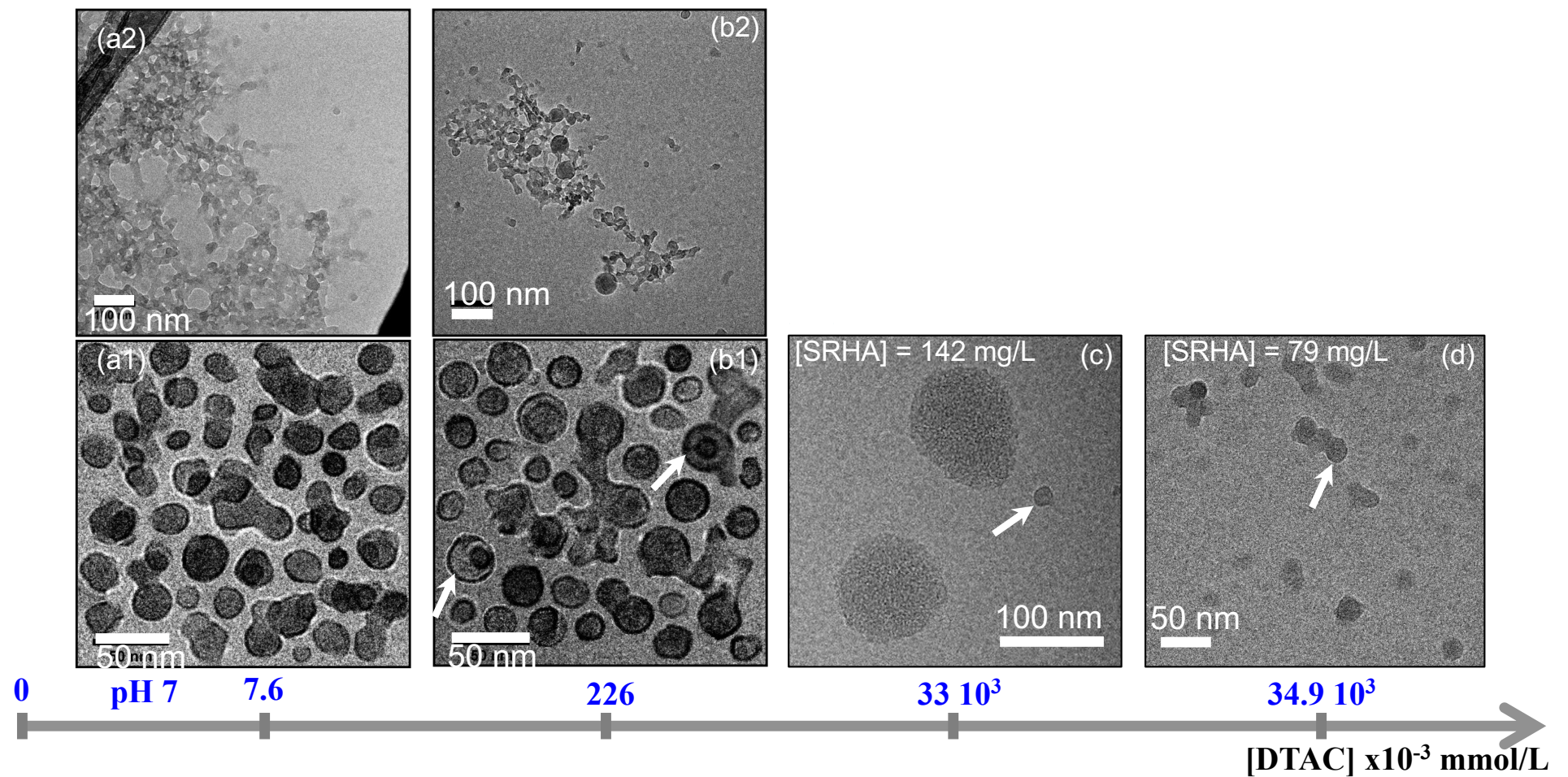


Figure 6: CryoTEM micrographs of SRHA/DTAC molecular structures obtained at pH 6.7 for various DTAC concentrations. (a1) SRHA/DTAC unilamellar vesicular structures formed at $[DTAC] = 7.6 \times 10^{-3}$ mmol/L; (a2) Spongy network structure at $[DTAC] = 7.6 \times 10^{-3}$ mmol/L; (b1) Branched spongy network structure with budding vesicles at $[DTAC] = 226 \times 10^{-3}$ mmol/L; (c) SRHA/DTAC multilamellar vesicular structures formed at $[DTAC] = 226 \times 10^{-3}$ mmol/L; aggregated SRHA/DTAC complexes and vesicles formed at 33 mmol/L; (d) Partially aggregated SRHA/DTAC vesicles at $[DTAC] = 34.9$ mmol/L. The white arrows indicate the higher electron density at the edges of vesicular structures.

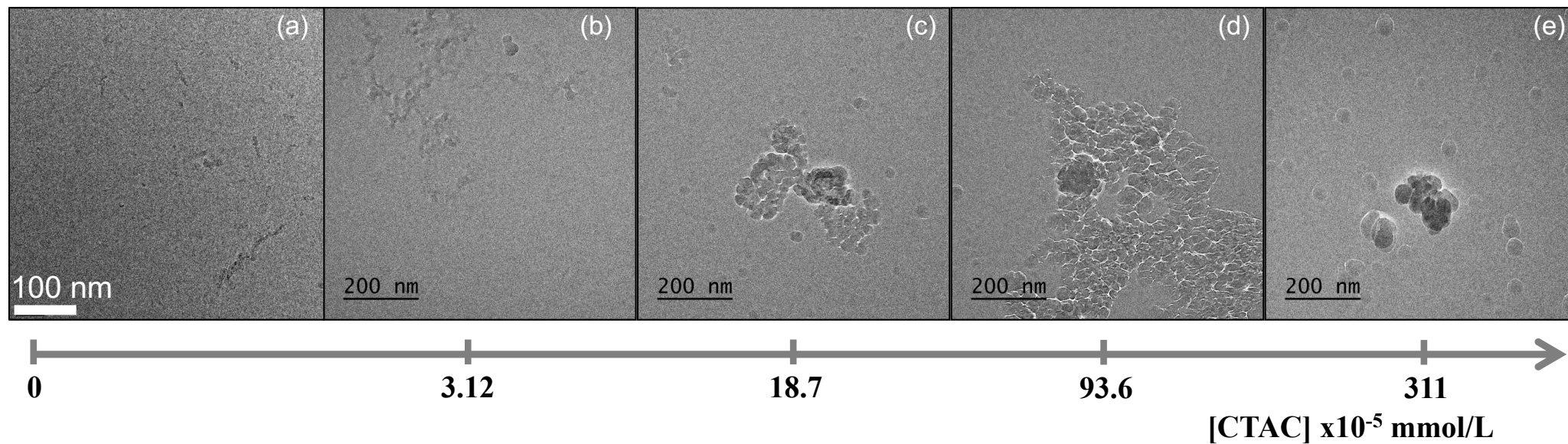


Figure 7: CryoTEM micrographs of SRHA/CTAC molecular structures obtained at pH 4.5 for various CTAC concentrations. (a) [CTAC] = 0 mmol/L; (b) [CTAC] = 3.12×10^{-5} mmol/L; (c) [CTAC] = 18.7×10^{-5} mmol/L; (d) [CTAC] = 93.6×10^{-5} mmol/L; (e) [CTAC] = 311×10^{-5} mmol/L. See text for details.