TABLE S I: Concentration and temperature (T) dependent phase behavior of G[a,b]

T/K	[G]					
	5%	4%	2.5%	1.0%	0.5%	0.05%
288	GL(GL)	GL(GL)	GL(GL)	$L_1(L_1)$	L(L)	L(L)
290	GL(GL)	GL(GL)	GL(GL)	L(L)	L(L)	L(L)
293	GL(GL)	GL(GL)	GL(M_2)	L(L)	L(L)	L(L)
296	GL(GL)	GL(GL)	GL(L)	L(L)	L(L)	L(L)
298	GL(GL)	M_1(L)	M_1(L)	L(L)	L(L)	L(L)
300	GL(L_1)	M_2(L)	L_1(L)	L(L)	L(L)	L(L)
303	L(L)	L(L)	L(L)	L(L)	L(L)	L(L)
308-320	L(L)	L(L)	L(L)	L(L)	L(L)	L(L)

[a]GL:Gel; L:Liquid; L_1:viscous liquid; M_1: upper surface slightly mobile; M_2: upper surface mobile only.

[b] Results shown in parenthesis were obtained under cooling conditions.

TABLE S II: Surface tension (γ_G) of G at different concentrations in pH9 buffer solution at 303K (A) and that of 0.05% G at different temperatures (B)

(A)

[G]/%	0	0.002	0.005	0.025	0.05	0.10	0.20	0.36	0.52	0.67	0.80
γ_G /mN m^{-1}	66.0	65.6	63.5	60.3	58.3	57.1	56.2	55.2	54.2	50.8	50.6

(B)

T/K	293	298	303	308	313
γ_G /mN m^{-1}	56.2	57.9	58.3	58.6	59.5

TABLE S III: Micellization and other physicochemical parameters of ATAB in pH9 boric acid-borax buffer at 303K

ATAB	cmc[a,e]			Interfacial Adsorption Parameters[a]				Bulk properties[a]		
	Tensio-metry	Conducto-metry	Microcal-orimetry	γ_{cmc} [d,e]	$10^7 \Gamma_{max}$ [f]	A_{min} [f]	$-\Delta G^0_{ad}$ [h]	f [g]	$-\Delta G^0_m$ [g]	$-\Delta H_m$ [e]
OTAB	0.28	0.24	0.30	33.1	10.9	1.53	76.3	0.48	46.0	15.1
CTAB[b]	0.83	0.89	0.95[c]	33.2	11.9	1.40	74.4	0.68	46.7	8.81
TTAB	3.47	3.91	3.56	32.8	12.4	1.34	67.0	0.67	40.2	5.71
DTAB	13.2	13.5	12.4	32.9	13.5	1.23	58.5	0.63	34.0	3.79

[a] cmc, γ_{cmc}, Γ_{max} and A_{min}, are in mM, mN m^{-1}, mol m^{-2} and nm^2 $molecule^{-1}$ units respectively; ΔG^0_{ad}, ΔG^0_m and ΔH_m are in kJ mol^{-1}.

[b]Cmc of CTAB determined from fluorimetry was 0.84mM.

[c] cmc, ΔH_m (Temp) for CTAB: 1.07, -3.02 (293), 1.00, -4.23 (298), 0.95, -8.81 (303), 1.17, -10.77 (308) and 1.19, -12.48 (313). The cmcs are in mM, ΔH_m in kJ mol^{-1} and temperature in K. The van't Hoff enthalpy, ΔH_m^{VH} (calculated following earlier procedures with $f = 0.68$) at 293, 298, 303, 308 and 313K were found to be -22.7, -39.7, -56.1, -71.9 and -87.3kJ mol^{-1} respectively.[53b,56] The specific heat capacity at one atmospheric pressure (calculated from the slope of the linear fit between ΔH_m and T) obtained from calorimetry (ΔC_{p_m}) and van't Hoff method ($\Delta C_{p_m}^{VH}$) were -510±50 and -3230±30J K^{-1} mol^{-1} respectively.

[d] γ_{cmc} for pure CTAB in aqueous solution at 298, 303, 308 and 313K are 33.1, 33.5, 34.2 and 34.7 mN m^{-1} respectively.[8]

[e]Standard deviations (SD) for cmc are $\pm 3\%$, $\pm 5\%$, $\pm 2\%$, $\pm 4\%$ for tensiometric, conductometric, microcalorimetric, and fluorimetric methods respectively.

[f] Γ_{max} is calculated following the Gibbs adsorption equation $\Gamma_{max} = -1/(2.303nRT) \underset{[ATAB]\sim cmc}{Lt} d\gamma/dlog[ATAB]$, where n, R, T and [ATAB] are the number of species formed per ATAB monomer in solution (it was 2 for the surfactants by ionization), the universal gas constant, absolute temperature and the molar concentration of ATAB in solution respectively. Here concentration was used in place of activity, as the solutions in use were fairly dilute. A_{min} was obtained from the relation $A_{min} = 10^{18}/N_A \Gamma_{max}$ in nm^2 $molecule^{-1}$, where N_A is the Avogadro number.[8,17,50,51]

[g] The parameter f was obtained using the relation, $f = 1-(S_2/S_1)$.[54a] ΔG^0_m was obtained from $\Delta G^0_m = (1+f)RTlnX_{cmc}$, where X_{cmc} is the cmc of pure ATAB in the mole fraction scale at temperature T.[17,50,51,54]

[h] ΔG^0_{ad} is calculated from the equation $\Delta G^0_{ad} = \Delta G^0_m - (\pi/\Gamma_{max})$, where π is the surface pressure at the saturated air/solution interface [$\pi = \gamma_0 - \gamma_{cmc}$, where γ_0 and γ_{cmc} are the surface tensions of the buffer solution (66.0mN m^{-1}) and that of ATAB at cmc respectively].[8,17,50,51]

TABLE S IV: Interfacial adsorption and other thermodynamic parameters[a] for the interaction of (a) G-CTAB at varying [G], (b) 0.05% G interaction with varying ATAB and (c) 0.05% G-CTAB interaction at varying temperature

System	γ_{T_4}	10^7 $\Gamma_{max}^{T_4}$	$A_{min}^{T_4}$	f_i		$-\Delta G_i^{0\,b}$		$-\Delta G^0_{ad,T_4}$
				f_{C_T}	f_{T_4}	$-\Delta G^0_{C_T}$	$-\Delta G^0_{m,T_4}$	
(a)								
0.005%	34.1	6.65	2.50	-	0.70	-	47.5	91.7
0.025%	34.2	5.78	2.87	-	0.63	-	44.9	90.1
0.05%	34.2	5.52	3.01	0.19	0.62	56.6	43.8	87.4
0.1%	34.2	5.48	3.03	0.40	0.54	65.4	41.2	82.9
0.2%	34.4	4.58	3.63	0.48	0.50	68.3	39.3	81.4
(b)								
OTAB	34.2	9.96	1.67	0.30	0.56	42.5	45.5	70.4
CTAB	34.2	5.52	3.01	0.19	0.62	56.6	43.8	87.4
TTAB	32.6	-	-	-	0.66	-	39.7	-
DTAB	32.4	-	-	-	0.63	-	33.6	-

(c)

T/K	γ_{T_4}	10^7 $\Gamma_{max}^{T_4}$	$A_{min}^{T_4}$
293	34.7	7.37	2.25
298	34.5	5.68	2.92
303	34.2	5.52	3.01
308	33.6	5.36	3.10
313	32.9	5.08	3.27

[a] γ_{T_4}, $\Gamma_{max}^{T_4}$ and $A_{min}^{T_4}$ are in mN m^{-1}, mole m^{-2} and in nm^2 $molecule^{-1}$ respectively; ΔG_i^0 and $\Delta G^0_{ad,T_4}$ are in kJ $mole^{-1}$.

[b]Putting f and X for the respective states ΔG_i^0 s were calculated. X refers to the mole fraction corresponding to C_T and T_4 (vide. footnote g, Table SI for relevant equations).

Legend to figure

Figure S I. Tensiometric, conductometric and thermometric profiles of pure ATAB dilution in pH9 buffer solution at 303K.

Main diagram displays tensiometric isotherms of DTAB and TTAB. Arrowheads indicate cmc points.

Inset: Conductometric and enthalpy profiles for pure DTAB indicating the ways to get cmc, f and ΔH_m. For pure OTAB, cmc and ΔH_m in the enthalpy profile were estimated following Sigmoidal-Boltzmann fitting procedure.[1]

References and Notes

1. Hait, S. K.; Moulik, S. P.; Palepu, R. *Langmuir* **2002**, *18*, 2471.

Figure

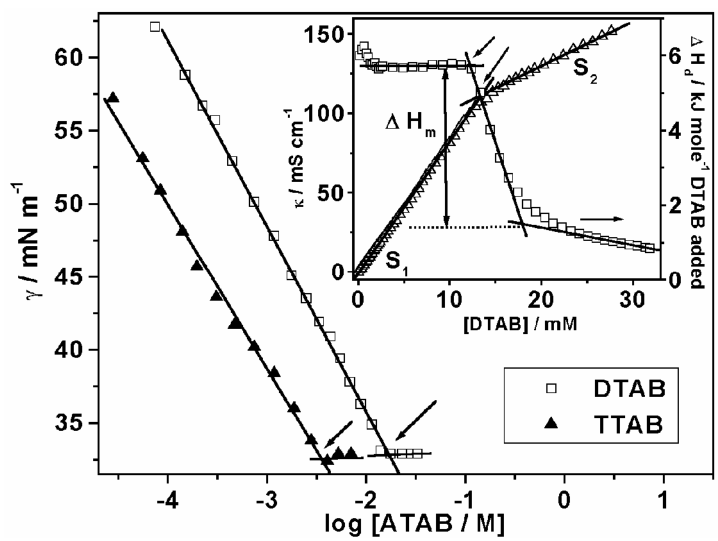

Figure S I.