THERMODYNAMIC STUDIES IN TERNARY MIXTURES CONTAINING WATER, 2-METHOXYETHANOL AND T- BUTANOL AT 298.15 K

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Abstract

Ultrasonic speeds and isentropic compressibilities of aqueous solutions of Water + 2-methoxyethanol (2ME) + t-butanol have been determined at 298.15 K. The concentrations of t-butanol at which ultrasonic speed becomes maximum and isentropic compressibility becomes minimum are found to decrease with increase in the concentration of 2ME in the cosolvent (Aqueous 2ME). This behaviour indicates that the aqueous ternary solutions are less structured than aqueous t-butanol. This behaviour is also well reflected in the concentration dependence of excess ultrasonic speed and excess isentropic compressibility. This behaviour is explained as due to a decrease in the ability of t-butanol to form clathrate hydrates owing to the presence of 2ME. When the concentration of 2ME in the cosolvent (x_{2ME}) > 0.11, ultrasonic speed decreases and isentropic compressibility increases with concentration of t-butanol indicating that the ternary solution behaving as normal solution where in any further addition of 2ME or t-butanol leads to destabilization of the hydrogen bonded structure of water and t-butanol looses its ability to form clathrate hydrates in aqueous solutions.

Key words: Ternary solution; ultrasonic speed; isentropic compressibility ;clathrate hydrates; hydrophobic hydration

1. Introduction

In recent years, there has been considerable advancement in the experimental investigation of excess thermodynamic properties of liquid mixtures. These properties have been adequately employed in understanding the nature of molecular interactions in binary and ternary liquid mixtures. In chemical industry, knowledge of the thermodynamic properties of non-electrolyte solutions is essential in the design involving chemical separation, heat transfer, mass transfer and fluid flow. Ultrasonic speed and compressibility behaviour of binary and ternary mixtures containing water and nonelectrolytes have been studied extensively, particularly at very low concentration of nonelectrolytes [1-14]. The peak observed in ultrasonic speed versus concentration of nonelectrolyte in water + nonelectrolyte mixture has been attributed to stabilization of the three dimensional hydrogen

bonded structure of water by the nonelectrolyte leading to the formation of clathrate hydrates or enhancement in the long range order of water due to hydrophobic interactions or stabilization of the hydrogen bonded clusters of water against thermal collapse or complex formation depending on the nature of the solute. The ultrasonic behaviour of aqueous t-butanol in the water rich region has been studied extensively in view of highly hydrophobic propensity of t-butanol. The maximum enhancement in the hydrogen structure of water due to the hydrophobic interactions between t-butanol percent of t-butanol as evidenced by the existence of extrema in many of the physical properties. Baumgartner and Atkinson[2] proposed the formation of clathrate hydrates well characterized by 17 hydrate structures from the observation of ultrasonic speed peak at 0.0556 molefraction of t-butanol. The strength of the hydrophobic interaction of t-butanol is also sensitive to temperature as revealed by the hydroxyl – proton chemical shift observed as a function of temperature by Atkinson and Symons [9] and neutron diffraction studies by Bowron et al [10] in aqueous t-butanol solutions.

It would be interesting to study how the hydrophobic hydration in aqueous t-butanol in the water rich region gets influenced by the presence of small amounts of another completely miscible nonelectrolyte. The authors have studied the ultrasonic behaviour of ternary mixtures of water + t-butanol + 2-ethoxyethanol [20] and water + t-butanol + dimethylformamide [21] and found to be useful in understanding the molecular interactions.The studies on the ultrasonic behaviour of water + t-butanol + 2-methoxyethanol are presented and discussed in this paper.

2. Experimental

2.1. Materials

Analytical reagent grade 2-methoxyethanol (2ME) and t-butanol were purified according to the standard procedures[15]. The densities of the pure liquids determined using a bicapillary type pycnometer with an accuracy of 2 parts in 10^5 are 780.92 kg m^{-3} and 960.03 kg m^{-3} for t-butanol and 2ME respectively at 298.15 K. These values are in good agreement with literature values[3,13,16,17].

Triple distilled degassed water was used to prepare stock solutions (varied concentrations) of aqueous 2ME. This stock solution was used to prepare ternary solution of t-butanol of desired concentration. The concentration of t-butanol in the ternary solution was calculated by considering aqueous 2ME binary stock solution as a cosolvent of effective molecular weight M_{12} given by $x_wM_w + x_sM_s$ where M_w, M_s and x_w, x_s represent the molecular weights and molefractions of water and 2ME respectively. All solutions were prepared by weight with an accuracy of 0.1 mg. The buoyancy corrected weights were used for calculating the molefractions. The error involved in the estimation of mole fractions of the samples is of the order of 1×10^{-4}.

2.2. Measurements

Ultrasonic speeds in pure liquids and aqueous binary (water + 2ME) and ternary (water + 2ME + t-butanol) solutions at 298.15 K were determined using a single crystal variable path interferometer designed and fabricated in our laboratory. A tri-tet crystal controlled oscillator with frequency stability of ±1 Hz was used to excite the quartz transducer. The quartz transducer whose fundamental frequency is 1 MHz was excited at its third harmonic. The frequency was measured with a digital frequency meter with an accuracy of one part per million. The voltage variations across the transducer were observed using a difference amplifier followed by an electronic voltmeter.

The mechanical assembly of the interferometer was immersed in a thermostatic water bath whose temperature can be controlled to ±0.01ºC using suitable permanent heaters followed by " On and Off" low wattage heaters. The temperature gradients inside the experimental liquid were minimized by stirring it periodically. The temperature of the interferometric liquid measured using a bead type thermistor, which forms one arm of the constant current Wheatstone bridge followed by chopper-stabilized operational amplifier as null detector, was found to be controlled to within ±0.005ºC. The thermostat and the electronic assembly were housed in the air-conditioned room whose temperature was maintained at 20 ±1º C. The path length was measured for 50 dips. For each solution six to eight measurements were made and the average was chosen as the ultrasonic speed which was found to be accurate to ±0.05 m s^{-1}. The densities of the solutions were also determined using a bicapillary type pycnometer with an accuracy of 2 in 10^5.

3. Results and Discussion

Ultrasonic speeds (u_{ts}) in aqueous 2ME at different concentrations of t-butanol are presented graphically in Fig.1. The densities (ρ_{ts}) of aqueous 2ME at different concentrations of t-butanol are presented in Table 1. The pure liquid parameters used to evaluate $(\beta_{ts})_{id}$ and hence $(u_{ts})_{id}$ are presented in Table 2. The isentropic compressibility (β_{ts}) of the solutions have been evaluated using the standard relation $\beta_{ts} = [u_{ts}^2 \rho_{ts}]^{-1}$. where u_{ts}, and ρ_{ts} represents ultrasonic speed and density of the ternary solution respectively.

The excess isentropic compressibilities of the ternary solutions (β_{ts}^{E}) have been evaluated using the relation

$$\beta_{ts}^{E} = \beta_{ts} - [\Phi_{t\text{-}B}\beta_{t\text{-}B} + \Phi_{cs}\beta_{cs}] \quad (1)$$

Where β_{ts} represents the compressibility of the ternary solution and $\beta_{t\text{-}B}$, β_{cs} represent the compressibilities of t-butanol and cosolvent (aqueous 2ME) respectively.$\Phi_{t\text{-}B}$ and Φ_{cs} represent the volume fractions of t-butanol and cosolvent respectively in the ternary solution where

$$\Phi_{t\text{-}B}+\Phi_{cs}=1 \quad (2)$$

The excess ultrasonic speeds in the ternary solutions have been determined using the following expressions [18]

$$u_{ts}^{E}=(u_{ts})_{exp}-(u_{ts})_{id} \quad (3)$$

where

$$(u_{ts})_{id}= [[(\beta_{ts})_{id}(\rho_{ts})_{id}]^{-1}]^{1/2} \quad (4)$$

and

$$(\beta_{ts})_{id} = (K_{ts})_{id} - [\, T\,(V_{ts})_{id}\,(\alpha_{ts})_{id}^{2} / ((C_P)_{ts})_{id})\,] \quad (5)$$ here

$$(\rho_{ts})_{id} = \Phi_{t\text{-}B}\rho_{t\text{-}B} + \Phi_{cs}\rho_{cs} \quad (6)$$

$$(K_{ts})_{id}=\Phi_{t\text{-}B}K_{t\text{-}B} + \Phi_{cs}\,K_{cs} \quad (7)$$

$$(V_{ts})_{id} = X_{t\text{-}B}V_{t\text{-}B} + X_{cs}\,V_{cs} \quad (8)$$

$$(\alpha_{ts})_{id} = \Phi_{t\text{-}B}\,\alpha_{t\text{-}B} + \Phi_{cs}\,\alpha_{cs} \quad (9)$$

$$[(C_P)_{ts}]_{id} = X_{t\text{-}B}(C_P)_{t\text{-}B} + X_{cs}\,(C_P)_{cs} \quad (10)$$

where β, K, ρ, α, V and C_P represent the isentropic compressibility, isothermal compressibility, density, thermal expansion coefficient, molar volume and molar heat capacity at constant pressure respectively. The suffixes ts, t-B and cs refer to ternary solution, t-Butanol and cosolvent respectively. The K_{cs}, α_{cs},$(C_P)_{cs}$ of cosolvent of different concentrations of 2ME have been taken from the experimental data available in literature [18,19] and are presented in Table 2.

The variation of isentropic compressibility (β_{ts}), excess sound speed (u_{ts}^{E}) and excess isentropic compressibility (β_{ts}^{E}) with molefraction of t-butanol[19] in the ternary solution (Water + 2ME + t-butanol) at different concentrations of 2ME in the cosolvent (water + 2ME) are presented graphically in Figs. 2- 4 respectively.

An examination of the data presented in Fig.1 indicates that the concentration of t-butanol at which ultrasonic speed reaches maximum, decreases with increase in concentration of 2-methoxyethanol. This behavior indicates that the ternary system is less structured than the aqueous t-butanol. Possibly, 2-methoxyethanol destabilizes clathrate hydrate formation of water-t-butanol system. At a concentration $x_{2ME} > 0.093$, ultrasonic speed decreases with increase in the concentration of t-butanol in the ternary solution. This behaviour indicates that the hydrophobic interactions between water and t-butanol molecules leading to clathrate hydrate structure as proposed [2,3,6,7] or the hydrophobic hydration induced local density enhancement in the solvent water by t-butanol as proposed by Bowron et al [10] are influenced by the presence of 2-methoxyethanol in the ternary solution. The ultrasonic speed of the ternary system when the concentration of x_{2ME} is 0.11, decreases

with increasing concentration of t-butanol. In the low concentration range the 2-methoxyethanol forms micelles with water. The growth patterns terminate with the formation of highly labile aggregates [17] and hence there will be significant reduction of direct contact between smaller tail and water.

An examination of the data presented in Fig. 2 indicates that the isentropic compressibility decreases with the concentration of t-butanol up to a particular concentration $(x_{t\text{-}B})_{opt}$ and there after increases. When the concentration of $x_{2ME} > 0.11$ the isentropic compressibility of the ternary system increases linearly with increasing concentration of t-butanol.

As seen from the Fig. 3 the excess ultrasonic speed reaches a maximum up to a particular concentration of $x_{t\text{-}B}$ and then remains almost constant with increase in $x_{t\text{-}B.}$

Fig. 4 indicates that excess isentropic compressibility decreases linearly up to a particular concentration of $x_{t\text{-}B}$ and then takes a change in slope in the curve for the higher concentrations of $x_{t\text{-}B}$. However, a similar trend is observed when the concentration of $x_{2ME} > 0.11$ the β_{ts}^{E} linearly decreases.

In Table 3 are presented $(x_{t\text{-}B})_{opt}$ versus x_{2ME} in the cosolvent observed in u_{ts}, β_{ts} and u_{ts}^{E} versus $X_{t\text{-}B}$ for the ternary system of water + 2-methoxyethanol + t-butanol. An examination of the data presented in Table 2 indicates that when x_{2ME} in the cosolvent exceeds 0.11, the ternary solution looks like a normal solution wherein any further addition of t-butanol leads to destabilization of the hydrogen bonded structure of water and t-butanol looses its ability to form clathrate hydrates in aqueous solutions.

These observations show that 2-methoxyehtanol preserves the clathrate structure of water-t-butanol mixture. As the concentration of 2-methoxyethanol increases more than 0.11, the ternary system behaves like a normal system. This may be due to the fact that 2-methoxyethanol forms a miceller solutions with water.

Conclusions

The ultrasonic speed maximum and isentropic compressibility minimum shift towards lower concentrations of t-butanol as the concentration of 2ME increases in the ternary solution. When the concentration of 2ME in the cosolvent is more than 0.11 molefraction, the ultrasonic speed decreases linearly and isentropic compressibility increases linearly with concentration of t-butanol, the solution exhibiting the behaviour of a normal solution. The concentration dependences of excess ultrasonic speed and excess isentropic compressibility also reflect this behaviour of the ternary solution. When

$x_{2ME} > 0.11$ both $u^E$and β^E vary linearly with molefraction of t-butanol may be due to the fact that 2-methoxyethanol forms a miceller solutions with water.

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Figure Legends

Fig.1. Variation of Ultrasonic speed (u_{ts}) with mole fraction of t-Butanol ($x_{t\text{-}B}$) in the ternary system (water + 2-Methoxyethanol + t-Butanol) at different concentrations of 2-Methoxyethanol (x_{2ME}) in the cosolvent
(water + 2-Methoxyethanol)

x_{2ME} = A : 0.0000, B: 0.0112, C : 0.0346, D : 0.0521,
E : 0.0724, F : 0.0914, G : 0.1105

Fig.2. Variation of isentropic compressibility (β_{ts}) with mole fraction of t-Butanol ($x_{t\text{-}B}$) in the ternary system (water + 2-Methoxyethanol + t-Butanol) at different concentrations of 2-Methoxyethanol (x_{2ME}) in the cosolvent
(water + 2-Methoxyethanol)

x_{2ME} = A : 0.0000, B: 0.0112, C : 0.0346, D : 0.0521,
E : 0.0724, F : 0.0914, G : 0.1105

Fig.3. Variation of excess ultrasonic speed (u_{ts}^{E}) with mole fraction of t-Butanol ($x_{t\text{-}B}$) in the ternary system (water + 2-Methoxyethanol + t-Butanol) at different concentrations of 2-Methoxyethanol (x_{2ME}) in the cosolvent
(water + 2-Methoxyethanol)

x_{2ME} = A : 0.0000, B: 0.0112, C : 0.0346, D : 0.0521,
E : 0.0724, F : 0.0914, G : 0.1105

Fig.4. Variation of excess isentropic compressibility (β_{ts}^{E}) with mole fraction of t-Butanol ($x_{t\text{-}B}$) in the ternary system (water + 2-Methoxyethanol + t-Butanol) at different concentrations of 2-Methoxyethanol (x_{2ME}) in the cosolvent (water + 2-Methoxyethanol)

x_{2ME} = A : 0.0000, B: 0.0112, C : 0.0346, D : 0.0521,
E : 0.0724, F : 0.0914, G : 0.1105

Table 1 - u_{ts}, ρ_{ts}, β_{ts}, $(u_{ts})_{id}$, $(\beta_{ts})_{id}$ and u_{ts}^{E}, β_{ts}^{E} versus mole fraction of t-butanol ($X_{t\text{-}B}$) in the cosolvent (water + 2-methoxyethanol) at 298.15 K

$X_{t\text{-}B}$	u_{ts}	ρ_{ts}	β_{ts}	$(\beta_{ts})_{id}$	β_{ts}^{E}	$(u_{ts})_{id}$	u_{ts}^{E}
			X_{2ME} = 0.0112				
0.0000	1543.1	996.48	42.1	-	-	-	-
0.0105	1562.2	996.19	41.1	49.3	-8.2	1434.8	127.4
0.0218	1583.6	994.61	40.1	52.9	-12.8	1393.7	189.9
0.0326	1600.0	993.11	39.3	55.9	-16.6	1361.6	238.4
0.0472	1604.7	991.02	39.2	59.7	-20.5	1326.5	278.2
0.0593	1594.6	988.86	39.8	62.5	-22.7	1302.7	291.9
0.0726	1575.4	985.56	40.9	65.2	-24.3	1280.9	294.5
0.0834	1556.8	980.51	42.1	67.3	-25.2	1265.8	291.0
			X_{2ME} = 0.0346				
0.0000	1580.7	988.42	40.5	-	-	-	-
0.0086	1595.4	988.82	39.7	50.1	-10.4	1425.1	170.3
0.0174	1607.8	987.33	39.2	52.6	-13.4	1395.7	212.1
0.0271	1614.4	986.82	38.9	55.2	-16.3	1368.0	246.4
0.0408	1612.6	985.14	39.0	58.6	-19.6	1335.5	277.1
0.0512	1606.3	980.16	39.5	60.9	-21.4	1314.9	291.4
0.0578	1598.1	976.14	40.1	62.3	-22.2	1303.3	294.8
0.0664	1578.3	953.07	42.1	64.0	-21.9	1289.6	288.7
			X_{2ME} = 0.0521				
0.0000	1610.1	988.91	39.0	-	-	-	-
0.0084	1619.4	988.01	38.6	50.2	-11.6	1423.3	196.1
0.0175	1626.3	987.39	38.3	52.1	-13.8	1400.5	225.8
0.0294	1630.9	986.89	38.1	54.9	-16.8	1371.0	259.9
0.0432	1627.1	968.71	39.0	58.0	-19.0	1341.0	286.1
0.0541	1618.6	944.29	40.4	60.1	-19.7	1321.9	296.7
0.0662	1603.4	939.04	41.4	62.3	-20.9	1303.3	300.1
0.0763	1586.4	931.05	42.7	64.0	-21.3	1289.6	296.8

$X_{t\text{-}B}$	u_{ts}	ρ_{ts}	β_{ts}	$(\beta_{ts})_{id}$	β_{ts}^{E}	$(u_{ts})_{id}$	u_{ts}^{E}
			X_{2ME} = 0.0724				
0.0000	1632.6	980.42	38.3	-	-	-	-
0.0054	1638.4	979.81	38.0	50.9	-12.9	1413.2	225.2
0.0115	1646.4	975.25	37.8	52.5	-14.7	1395.4	251.0
0.0209	1651.5	968.97	37.8	54.8	-17.0	1371.0	280.5
0.0284	1644.6	961.56	38.5	56.5	-18.0	1353.8	290.8
0.0379	1631.5	953.95	39.4	58.6	-19.2	1334.4	297.1
0.0462	1616.4	946.89	40.4	60.3	-19.9	1319.4	297.0
0.0542	1606.9	936.64	41.4	61.9	-20.5	1306.3	300.6
			X_{2ME} = 0.0914				
0.0000	1646.4	978.56	37.7	-	-	-	-
0.0042	1654.5	975.53	37.5	51.4	-13.9	1407.2	247.3
0.0108	1659.6	970.22	37.4	53.0	-15.6	1389.5	270.1
0.0168	1653.3	967.54	37.8	54.4	-16.6	1374.7	278.6
0.0234	1645.5	951.56	38.8	55.9	-17.1	1359.9	285.6
0.0315	1631.4	940.56	40.0	57.6	-17.6	1343.4	288.0
0.0421	1618.5	932.66	40.9	59.7	-18.8	1324.2	294.3
0.0514	1601.4	916.66	42.5	61.5	-19.0	1309.3	292.1
			X_{2ME} = 0.1105				
0.0000	1653.9	966.81	37.8	-	-	-	-
0.0029	1649.4	965.14	38.1	51.8	-13.7	1402.3	247.1
0.0103	1644.5	963.12	38.4	53.5	-15.1	1383.9	260.6
0.0174	1634.4	959.15	39.0	55.0	-16.0	1367.9	266.5
0.0256	1620.7	956.21	39.8	56.7	-16.9	1351.2	269.5
0.0344	1609.7	951.12	40.6	58.5	-17.9	1335.0	274.7
0.0453	1596.4	949.12	41.3	60.5	-19.2	1317.2	279.2
0.0595	1577.3	940.66	42.7	62.9	-20.2	1297.0	280.3

Table 2. u, ρ, V, C_P, α, β and K of pure liquids at 298.15 K

Liquid	U ($m\ s^{-1}$)	ρ ($Kg\ m^{-3}$)	V x 10^6 ($m^3\ mol^{-1}$)	C_P ($J\ mol^{-1}\ K^{-1}$)	α x 10^4 (K^{-1})	βx10^{11} ($N^{-1}m^2$)	Kx10^{11} ($N^{-1}m^2$)	Ref.
Water	1496.7	997.07	18.08	75.30	2.59	44.77	45.25	4
t-butanol	1123.2	780.95	94.91	224.9	12.68	101.50	121.72	4
2-methoxyethanol	1344.8	960.03	79.25	176.4	8.23	57.52	66.60	3,19

Table 3. $(x_{t-B})_{opt}$ versus X_{2BE} in the cosolvent observed in u, β,u^E and β^E versus (x_{t-B}) in the ternary solutions of water + 2ME + t-butanol

X_{ME}	$(X_{t-B})_{opt}$		
	u_{ts}	**β_{ts}**	**U_{ts}^E**
0.0000	0.051	0.048	0.065
0.0112	0.048	0.045	0.062
0.0346	0.038	0.035	0.051
0.0521	0.030	0.029	0.048
0.0724	0.020	0.016	0.035
0.0914	0.011	0.007	0.025
0.1105	-	-	-

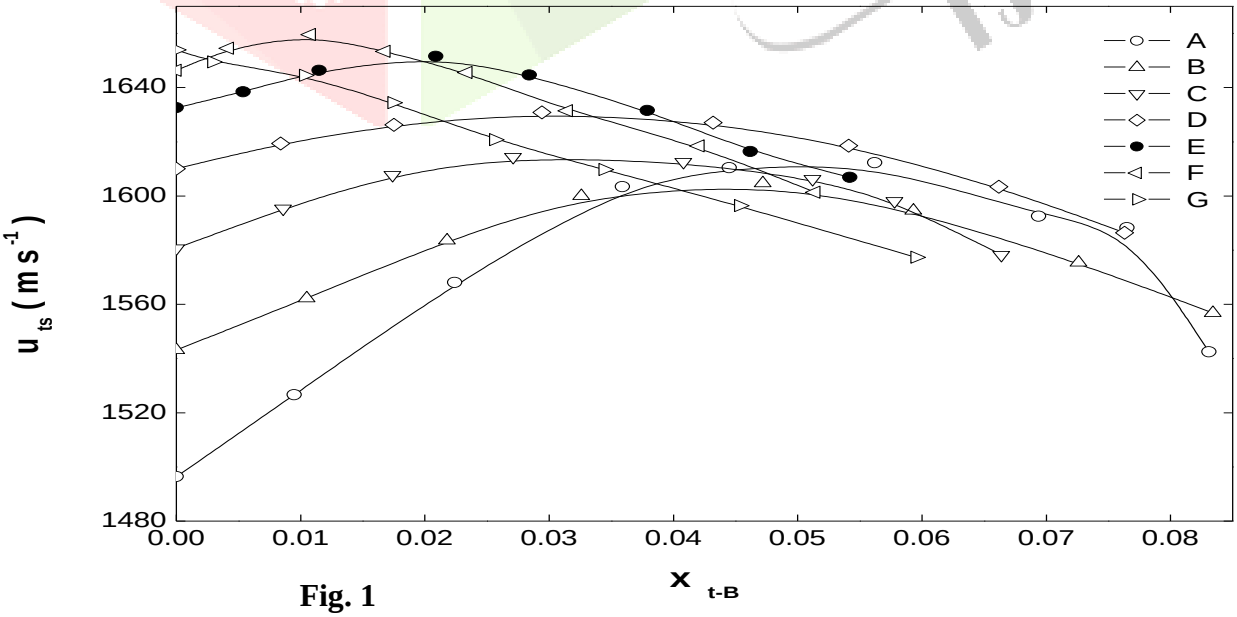
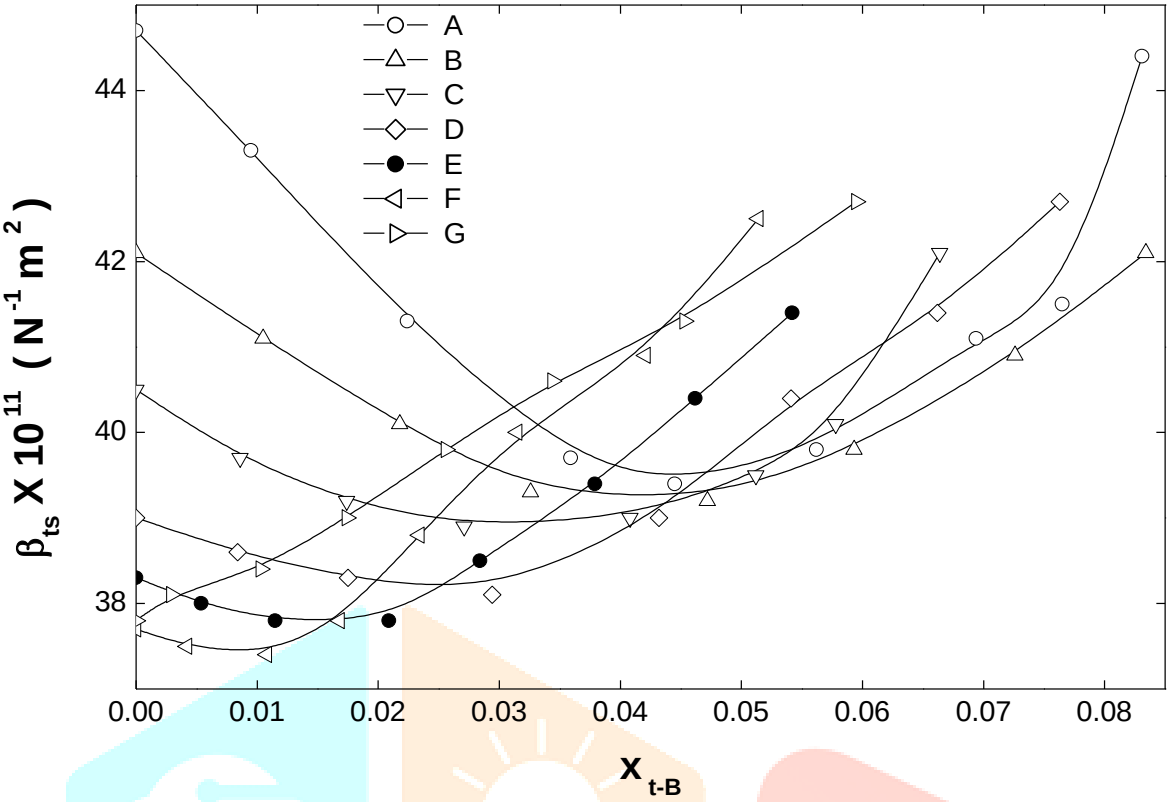


Fig. 1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38
0.00
0.01
0.02
0.03
0.04
0.05
0.06
0.07
0.08
β_{ts} X 10^{11} (N^{-1} m^2)
X_{t-B}

Fig. 2

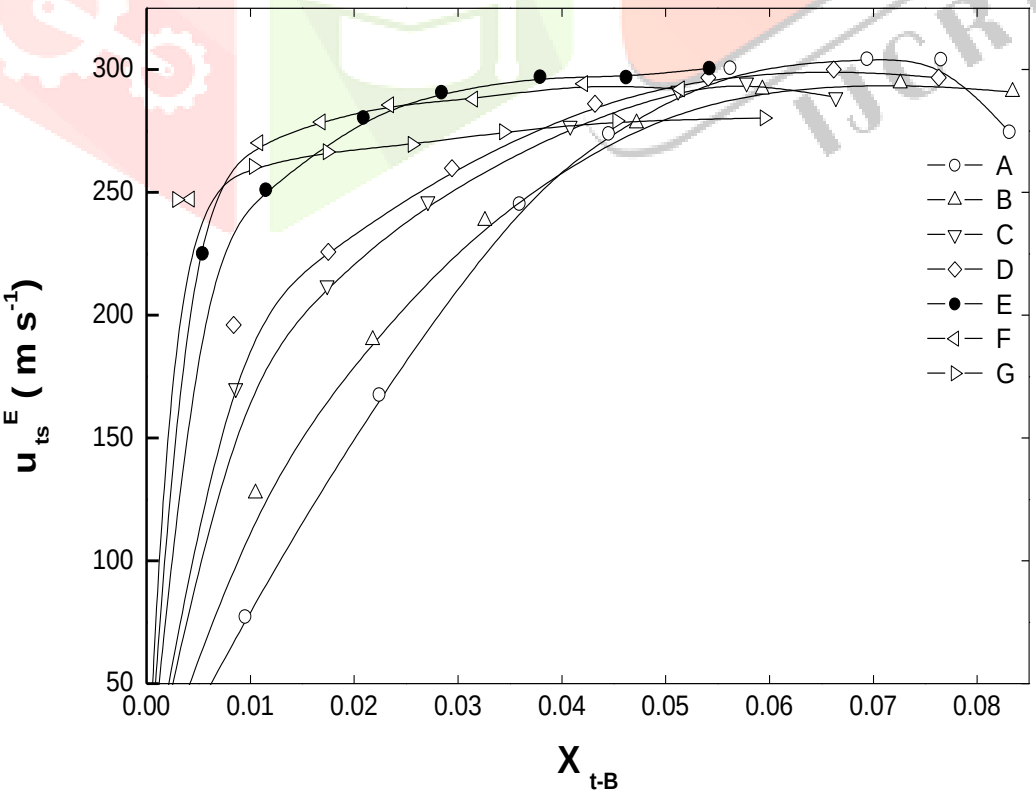
A
B
C
D
E
F
G
300
250
200
150
100
50
0.00
0.01
0.02
0.03
0.04
0.05
0.06
0.07
0.08
u_{ts}^{E} (m s^{-1})
X_{t-B}

Fig. 3

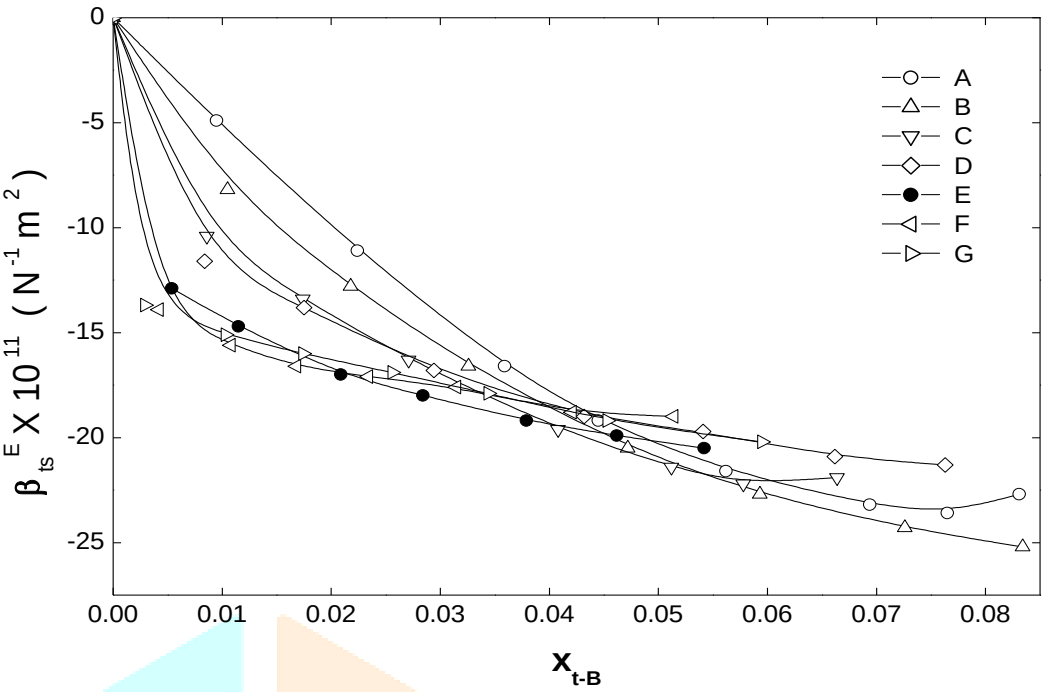
0
-5
-10
-15
-20
-25
0.00
0.01
0.02
0.03
0.04
0.05
0.06
0.07
0.08
$\beta^{E}_{ts} \times 10^{11}$ ($N^{-1} m^{2}$)
x_{t-B}
A
B
C
D
E
F
G

Fig. 4