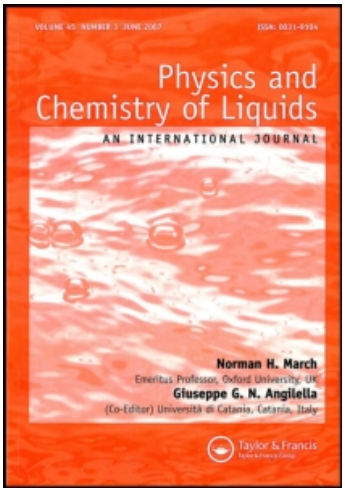

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Some Physical Properties of Binary Liquid Systems: (Ethanoic Acid or Propanoic Acid or Butanoic Acid Ethanenitrile)

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SOME PHYSICAL PROPERTIES OF BINARY LIQUID SYSTEMS: (ETHANOIC ACID OR PROPANOIC ACID OR BUTANOIC ACID + ETHANENITRILE)

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Density, viscosity and surface tension of three binary liquid systems: ethanoic acid + ethanenitrile, propanoic acid + ethanenitrile, butanoic acid + ethanenitrile have been determined at 25, 35 and 45°C, over the whole compositional range. The excess values of molar volume, viscosity, Gibbs free energy for the activation of flow and surface tension were evaluated. The excess values were fitted to a Redlich-Kister type of equation. The Grunberg-Nissan parameter, *d*, was also calculated. The binary viscosity data were fitted to the models of McAllister, Heric, Auslander and Teja and Rice. Surface tension data were fitted to the models of Zihao and Jufu, Rice and Teja, and an empirical two-constant model proposed in this study.

KEY WORDS: Excess molar volume, viscosity, activation energy, surface tension.

INTRODUCTION

As an extension to our previous work[1-5] on the measurement of transport and surface properties of non-electrolyte mixture, in the present study densities, viscosities and surface tension of the binary organic liquid mixtures of ethanoic acid or propanoic acid or butanoic acid with ethanenitrile have been found experimentally at 25, 35 and 45°C. From these experimental results, excess properties have been determined and studied in terms of sign and magnitude of the physical properties in the respective mixtures. Valuable information regarding the presence of secondary bonding forces in a particular system is derived from the study. Also the entire data on the excess properties have been fitted to the Redlich-Kister type of equation. The mixture viscosity data have been fitted to the models of McAllister, Heric, Auslander and Teja and Rice.

Binary surface tension data have been fitted to the models of: Zihao and Jufu, Rice and Teja and an empirical two parameter model.

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EXPERIMENTAL SECTION

Ethanenitrile, ethanoic acid, propanoic acid and butanoic acid (S.D's grade) were purified by the standard procedures[6]. The purity of all the components was checked by comparing their experimental density with that of literature value[6]. For each run, a fresh liquid mixture was prepared on mass basis, using a Mettler balance (precision of $1*10^{-5}$ g). The purity of each component with respect to the corresponding literature value is recorded in Table 1.

Densities of the pure components and their mixtures were measured with density meter (AP-Paar, DMA-48) calibrated at 25, 35 and 45°C with ethanol and 1,2 dichloroethane. At each temperature, the densities were measured with a precision of about $\pm 0.01\%$.

Viscosities were determined with the help of a modified Ubbelohode viscometer[7]. At each temperature the Viscometer was calibrated against the known viscosities of benzene and tetrachloromethane[8]. The constants of the viscometer at each temperature were determined from the following equation:

$$v = \eta/\rho = at - b/t \tag{1}$$

where a and b are the temperature dependent constants of the viscometer. At a particular temperature, an average value of the efflux time (t) of the mixture and corresponding values of a and b were used in Eq. (1) for the calculation of kinematic viscosity (v). Using the density values of the respective mixtures, the absolute viscosity (η) was thus calculated. The accuracy of the viscosity measurements is of the order of 0.001 cP.

Similarly, surface tension of the pure components and that of their mixtures was determined by the differential capillary rise method[8]. The difference (ΔH), in the level of liquid in two capillaries was measured with a cathetometer reading to 0.05mm. An average reproducibility of better than 0.01 mm was obtained. The difference in the levels of liquid in two capillaries of different radii is related to the surface tension of the liquid by the relation:

$$h_1 - h_2 = \Delta H = (2\sigma/\rho g)A + B - C \tag{2}$$

The constants A, B and C were calculated using the standard values of surface tension of benzene and ethylethanoate at 25°C. For each mixture of the respective systems, ΔH was recorded at three different temperatures over the entire compositional range. The surface tension of each mixture was thus calculated from Eq. (2).

Table 1 Physical constants of pure components at 25°C.

Property	*Ethanenitrile*		*Ethanoic acid*		*Propanoic acid*		*Butanoic acid*	
	Lit.[6]	*Sample used*	*Lit.*[6]	*Sample used*	*Lit.*[6]	*Sample used*	*Lit.*[6]	*Sample used*
Density (g/cm^3)	0.7766	0.7767	1.04366	1.0437	0.9880	0.9882	0.9532	0.9533

The entire experimental data for all the three systems is recorded in Table 2. All the measurements were made at a constant temperature with the help of a circulating type ultra cryostat (type MK 70, MLW, Germany) to within ± 0.02 K.

The Grunberg-Nissan parameter[9], was determined using the following expression:

$$d = \left(\frac{\ln \eta_{mix} - x_1 \ln \eta_1 - x_2 \ln \eta_2}{x_1 x_2} \right) \tag{3}$$

Table 2 Mole fraction(x_1), density(ρ), viscosity(η), surface tension(σ) and Grunberg-Nissan parameter(d) for various systems.

x_1	$\rho(g/cm^3)$	$\eta(cP)$	$\sigma(dyne/cm)$	d
System: Ethanoic acid + Ethanenitrile, 25°C				
.0000	.776700	.341800	27.54	******
.0924	.806800	.374210	26.72	−.2385
.1864	.835900	.416220	26.58	−.1720
.2820	.863800	.478550	26.54	−.0047
.3793	.890700	.557970	26.57	.1539
.4782	.916700	.650910	26.65	.2881
.5789	.942200	.753340	26.76	.4001
.6814	.967300	.860760	26.89	.4985
.7857	.992300	.968160	27.00	.5998
.8919	1.017500	1.069990	27.06	.7666
1.0000	1.043700	1.131000	27.10	******
System: Ethanoic acid + Ethanenitrile, 35°C				
.0000	.766000	.308500	26.22	******
.0924	.797200	.326670	25.75	−.5555
.1864	.826900	.362210	25.59	−.3225
.2820	.855400	.417680	25.53	−.0683
.3793	.880700	.489080	25.54	.1475
.4782	.909000	.571930	25.62	.3209
.5789	.934600	.661210	25.74	.4595
.6814	.959500	.751350	25.87	.5744
.7857	.983900	.836160	26.00	.6799
.8919	1.008300	.908780	26.08	.8137
1.0000	1.032500	.948700	26.10	******
System: Ethanoic acid + Ethanenitrile, 45°C				
.0000	.755500	.275500	24.91	******
.0924	.786600	.280840	24.60	−.8989
.1864	.816300	.316850	24.51	−.3360
.2820	.844700	.375920	24.50	.1093
.3793	.871900	.451510	24.56	.4495
.4782	.898100	.536350	24.65	.7083
.5789	.923400	.622360	24.77	.9123
.6814	.948200	.700530	24.90	1.0863
.7857	.972500	.760920	25.00	1.2577
.8919	.996800	.792500	25.06	1.4908
1.0000	1.021300	.766700	25.11	******

Table 2 *(Continued)*

System: Propanoic acid + Ethanenitrile, 25°C

.0000	.776700	.341800	27.54	******
.0727	.799200	.387070	26.72	.6551
.1499	.821700	.432590	26.34	.5499
.2322	.843800	.490030	25.04	.5835
.3199	.865400	.558840	25.81	.6373
.4136	.886500	.637810	25.66	.6899
.5141	.907200	.724670	25.57	.7370
.6221	.927500	.815640	25.51	.7794
.7383	.947500	.904820	25.44	.8214
.8640	.967600	.983280	25.32	.8782
1.0000	.988200	1.030500	25.13	******

System: Propanoic acid + Ethanenitrile, 35°C

.0000	.766000	.308500	26.22	******
.0727	.787900	.341090	25.57	.3524
.1499	.810200	.384110	25.21	.4790
.2322	.832400	.437310	24.92	.5835
.3199	.854200	.499870	24.70	.6676
.4136	.875700	.570290	24.53	.7344
.5141	.896700	.645960	24.42	.7875
.6221	.917400	.722740	24.35	.8302
.7383	.937500	.794310	24.29	.8643
.8640	.957100	.851250	24.20	.8823
1.0000	.977400	.885800	24.07	******

System: Propanoic acid + Ethanenitrile, 45°C

.0000	.755500	.275500	24.91	******
.0727	.778900	.300710	24.32	.2317
.1499	.802100	.344990	24.00	.6005
.2322	.825100	.394790	23.73	.7291
.3199	.847600	.449350	23.51	.7936
.4136	.869600	.507430	23.35	.8304
.5141	.890900	.567030	23.24	.8527
.6221	.911400	.625130	23.17	.8665
.7383	.930900	.677220	23.13	.8731
.8640	.949200	.716770	23.09	.8603
1.0000	.966600	.741200	23.10	******

System: Butanoic acid + Ethanenitrile, 25°C

.0000	.776700	.341800	27.54	******
.0598	.797900	.394380	26.73	.9508
.1252	.816800	.437730	26.37	.5459
.1971	.835700	.504360	26.07	.5935
.2763	.854600	.596810	25.85	.7180
.3641	.873200	.717000	25.71	.8442
.4621	.891300	.865260	25.64	.9525
.5720	.908700	1.038810	25.64	1.0415
.6961	.925100	1.228690	25.65	1.1199
.8375	.940300	1.414560	25.57	1.2200
1.0000	.953300	1.528300	25.31	******

System: Butanoic acid + Ethanenitrile, 35°C

.0000	.766000	.308500	26.22	******
.0598	.787300	.345170	25.57	.5152
.1252	.805700	.384910	25.25	.4276
.1971	.824200	.445470	24.98	.5872
.2763	.842800	.528540	24.77	.7679
.3641	.861400	.634890	24.62	.9265
.4621	.879600	.763210	24.54	1.0549
.5720	.897300	.908410	24.50	1.1572
.6961	.914100	1.058230	24.50	1.2435
.8375	.929700	1.187420	24.46	1.3322
1.0000	.943400	1.242100	24.33	******

System: Butanoic acid + Ethanenitrile, 45°C

.0000	.755500	.275500	24.91	******
.0598	.776900	.314650	24.36	1.0386
.1252	.796700	.368080	24.10	1.2219
.1971	.816500	.427170	23.87	1.2220
.2763	.836100	.492190	23.68	1.1825
.3641	.855400	.563200	23.54	1.1311
.4621	.873900	.639820	23.45	1.0766
.5720	.891500	.720910	23.40	1.0219
.6961	.907600	.803860	23.39	.9672
.8375	.921900	.883620	23.37	.9057
1.0000	.933500	.956200	23.35	******

From the experimental data, excess properties viz; V^E, η^E, G^E and σ^E were calculated using the following expressions[10]:

$$V^E = x_1 M_1 [\rho_{mix}^{-1} - \rho_1^{-1}] + x_2 M_2 [\rho_{mix}^{-1} - \rho_2^{-1}] \tag{4}$$

$$\log(1/\eta^0) = x_1 \log(1/\eta_1) + x_2 \log(1/\eta_2)$$

$$\eta^E = \eta_{mix} - \eta^0 \tag{5}$$

$$G^E/RT = [\ln \eta_{mix} V_{mix} - \{x_1 \ln \eta_1 V_1 + x_2 \ln \eta_2 V_2\}] \tag{6}$$

$$\sigma^E = \sigma_{mix} - [x_1 \sigma_1 + x_2 \sigma_2] \tag{7}$$

The values of V^E, η^E, G^E and σ^E were fitted with a Redlich-Kister type of Eq.[11], using least squares method.

$$P^E = x_1 x_2 \sum_{1}^{n} A_{j-1} (2x_1 - 1)^{j-1} \tag{8}$$

where P^E represents the excess property, A_j is the polynomial coefficient and n the polynomial degree. The results thus obtained are recorded in Table 3.

The following theoretical models for mixture viscosities have been tested with the experimental data. McAllister's[12] equation derived on the basis of Eyring's absolute reaction rate theory is:

$$\ln \eta_{mix} = x_1^3 \ln \eta_1 + x_2^3 \ln \eta_2 + 3x_1^2 x_2 \ln \eta_{12} + 3 x_1 x_2^2 \ln \eta_{21} - \ln[x_1 + x_2(M_2/M_1)] + 3 x_1^2 x_2 \ln[\tfrac{2}{3} + (M_2/3M_1)] + 3 x_1 x_2^2 \ln[\tfrac{1}{3} + 2M_2/3M_1)] + x_2^3 \ln[M_2/M_1], \quad (9)$$

Table 3 Coefficients of Eq. (8) and standard deviations (SD) determined by the method of least squares.

System: Ethanoic acid + Ethanenitrile

Parameter (Eq. 8)	V^E *(cm³/mol)*	G^E *(J/mol)*	η^E *(cP)*	σ^E *(dyne/cm)*
	Temp. 25.0°C			
A_0	−1.5529	690.338	−0.2625	−2.4605
A_1	1.4933	1621.95	0.4447	3.7714
A_2	0.0212	−329.566	0.1910	−3.0868
SD	0.0029	9.26497	0.00242	0.0665
	Temp. 35.0°C			
A_0	−2.4309	789.342	−0.1536	−2.0240
A_1	1.3881	2129.44	0.4893	2.6798
A_2	−0.2729	−954.789	0.0852	−0.9608
SD	0.0025	8.16051	0.0011	0.0229
	Temp. 45.0°C			
A_0	−2.5099	1895.40	0.1346	−1.302
A_1	1.5655	3455.43	0.7763	2.018
A_2	−0.1728	−1831.01	0.0625	−0.928
SD	0.0014	24.9803	0.0017	0.014

System: Propanoic acid + Ethanenitrile

	Temp. 25.0°C			
A_0	−1.0470	1902.23	0.1013	−3.058
A_1	0.7773	567.227	0.3430	5.418
A_2	0.3066	−2.42700	0.0907	−4.514
SD	0.0038	8.73959	0.0016	0.278
	Temp. 35.0°C			
A_0	−1.2033	2114.82	0.1537	−2.763
A_1	0.3036	696.697	0.3292	2.746
A_2	0.6545	−531.276	−0.0228	−1.504
SD	0.0069	4.17604	0.0004	0.032
	Temp. 45.0°C			
A_0	−2.6717	2338.99	0.2067	−2.925
A_1	0.0133	428.221	0.2353	1.984
A_2	0.2127	−841.152	−0.0983	−1.581
SD	0.0026	20.3828	0.0018	0.031

System: Butanoic acid + Ethanenitrile

	Temp. 25.0°C			
A_0	−2.7152	2687.12	−0.0526	−3.004
A_1	−0.1909	1076.51	0.7751	4.702
A_2	−0.8023	−254.503	0.2674	−2.669
SD	0.0272	24.2879	0.0047	0.067
	Temp. 35.0°C			
A_0	−2.3397	3101.87	0.1479	−2.909
A_1	−0.2437	1392.09	0.7797	3.417
A_2	−1.0470	−924.281	0.1110	−2.046
SD	0.0386	11.9302	0.0026	0.0151
	Temp. 45.0°C			
A_0	−3.9704	3078.54	0.2141	−2.718
A_1	−0.4557	−677.478	0.1390	2.446
A_2	−0.4457	−114.033	−0.0736	−1.810
SD	0.0205	13.4337	0.0015	0.0443

where $\eta_{12}$and η_{21} are the interaction parameters. Heric's[13] equation has the following form:

$$\ln \eta_{mix} = x_1 \ln \eta_1 + x_2 \ln \eta_2 + x_1 \ln M_1 + x_2 \ln M_2 - \ln(x_1 M_1 + x_2 M_2) + \delta_{12} \quad (10)$$

where δ_{12} is a deviation function expressed as:

$$\delta_{12} = x_1 x_2 [\beta_{12} + \beta_{21}(x_1 - x_2)] \quad (11)$$

Auslander's[14] model is represented by:

$$x_1(x_1 + B_{12}x_2)(\eta_{mix} - \eta_1) + A_{21}x_2(B_{21}x_1 + x_2)(\eta_{mix} - \eta_2) = 0 \quad (12)$$

where B_{12}, A_{21} and B_{21} are parameters representing binary interactions.

For a binary mixture, the Teja and Rice[15] equation based on theory of corresponding states can be written as:

$$\ln(\eta_{mix}\, \varepsilon_{mix}) = x_1 \ln(\eta_1 \varepsilon_1) + x_2 \ln(\eta_2 \varepsilon_2) \quad (13)$$

Where,

$$\varepsilon_i = V_{ci}^{2/3}/(T_{ci} M_i)^{1/2} \quad \text{for} \quad i = 1, 2 \text{ or mix}$$

$$V_{cmix} = x_1^2 V_{c1} + x_2^2 V_{c2} + 2x_1 x_2 [V_{c1}^{1/3} + V_{c2}^{1/3}]/8$$

$$T_{cmix} = \{x_1^2 T_{c1} V_{c1} + x_2^2 T_{c2} V_{c2} + 2x_1 x_2 \psi_{12} [T_{c1} V_{c1}\, T_{c2} V_{c2}]^{1/2}\}/V_{cmix}$$

η_1 to be evaluated at a temperature $T(T_{c1}/T_{cmix})$

η_2 to be evaluated at a temperature $T(T_{c2}/T_{cmix})$

T being the system temperature and ψ_{12} the interaction parameter having a value around unity.

Various models used for testing the binary surface tension data are given below.

The Zihao and Jufu[16] model equation based on the work of Hildebrand and Scott[9] can be written as:

$$\sigma_{mix} = \frac{x_1\sigma_1}{x_1 + Ax_2} + \frac{x_2\sigma_2}{x_2 + Bx_1} \tag{14}$$

Where A and B are interaction parameters.

The model of Rice and Teja[17] based on theory of corresponding states can be expressed as:

$$\sigma_{mix}/\phi_{mix} = x_1\sigma_1\phi_1 + x_2\sigma_2\phi_2 \tag{15}$$

Where,

$$\phi = V_{ci}^{2/3}/T_{ci} \quad \text{for } i = 1,2 \text{ or mix}$$

$$V_{cmix} = x_1^2V_{c1} + x_2^2V_{c2} + 2x_1x_2[V_{c1}^{1/3} + V_{c2}^{1/3}]/8$$

$$T_{cmix} = \{x_1^2T_{c1}V_{c1} + x_2^2T_{c2}V_{c2} + 2x_1x_2\psi_{12}[T_{c1}V_{c1}T_{c2}V_{c2}]^{1/2}\}/V_{cmix}$$

Here σ_1 is to be evaluated at a temperature $= T(T_{c1}/T_{cmix})$ and σ_2 at a temperature $= T(T_{c2}/T_{cmix})$. The surface tension data has also been fitted to an empirical two constant model expressed as:

$$\sigma_{mix} = x_1\sigma_1 + x_2\sigma_2 + x_1x_2[A' + B'(x_1 - x_2)] \tag{16}$$

Where A' and B' are binary interaction parameters. The parameters corresponding to Eqs (9), (11)–(16) have been determined using a least squares method and are reported in Table 4.

RESULTS AND DISCUSSION

The experimental values of physical properties (ρ, η and σ) of all the three systems along with Grunberg-Nissan parameter, d, are reported in Table 2. The value of parameter, d, changes its sign from $-$ive to $+$ive with the increase in concentration of ethanoic acid in ethanoic acid + ethanentrile system, whereas, for all other systems, d, is $+$ive; indicating the presence of specific interactions between the two components[18].

The model of Heric produces satisfactory result; whiule the models of McAllister and Auslander predict very well the viscosity data of all the three systems. Teja and Rice model, which is based on theory of corresponding states, also predicts the binary viscosity data to within reasonable accuracy.

Surface tension data is well predicted by the proposed empirical two-parameter model (Eq. 16) as well as by Rice and Teja model. The Zihao and Jufu model based on the Hilderbrand-Scott[19] equation, also predicts satisfactory results for the system studied.

Table 4 Interaction parameters for various models and standard deviations (SD) determined by least squares method.

System: Ethanoic acid + Ethanenitrile

		Temp. --->		
Model	*Constants*	*25°C*	*35°C*	*45°C*
McAllister	η_{12}	1.03468	0.94887	1.04958
(Eq. 9)	η_{21}	0.45889	0.38186	0.31753
	SD	0.00343	0.00662	0.01067
Heric	δ_{12}	0.35640	0.36830	0.73210
(Eq. 11)	δ_{21}	0.61920	0.80160	1.27960
	SD	0.25128	0.31181	0.42417
Auslander	B_{12}	0.64139	0.28964	0.50404
(Eq. 12)	A_{21}	1.67220	1.10713	1.54080
	B_{21}	0.38024	0.48422	−0.23995
	SD	0.00320	0.00300	0.00800
Teja&Rice	ψ_{12}	1.05	1.05	1.05
(Eq. 13)	SD	0.00685	0.00966	0.01845
Zihao&Jufu	A	−1.02449	−4.07649	−1.30074
(Eq. 14)	B	0.05129	0.73774	0.60871
	SD	0.2680	0.3427	0.1507
Two Parameter model	A′	−0.03520	−0.01130	0.01430
(Eq. 16)	B′	0.13260	0.09520	0.0733
	SD	0.11284	0.03544	0.0310
Rice&Teja	ψ_{12}	0.95	0.95	0.95
(Eq. 15)	SD	0.1237	0.05113	0.0302

System: Propanoic acid + Ethanenitrile

		Temp. --->		
Model	*Constants*	*25°C*	*35°C*	*45°C*
McAllister	η_{12}	0.98721	0.89071	0.7528
(Eq. 9)	η_{21}	0.60025	0.52699	0.4873
	SD	0.00136	0.00371	0.0047
Heric	δ_{12}	0.89520	0.92640	0.9938
(Eq. 11)	δ_{21}	0.18710	0.25250	0.1499
	SD	0.31167	0.37315	0.4537
Auslander	B_{12}	1.43412	0.79632	0.5415
(Eq. 12)	A_{21}	1.79879	1.06907	0.6665
	B_{21}	0.16776	0.28333	0.4647
	SD	0.00130	0.00090	.00220
Teja&Rice	ψ_{12}	1.10	1.10	1.10
(Eq. 13)	SD	0.0080	0.00426	0.0025
Zihao&Jufu	A	−7.86706	984.375	−8.3627
(Eq. 14)	B	−0.95292	196.024	29.489
	SD	0.289	0.038	1.470
Two Parameter model	A′	0.03500	0.05690	0.0428
(Eq. 16)	B′	0.18220	0.07570	0.0493
	SD	0.29189	0.04625	0.0484
Rice&Teja	ψ_{12}	0.90	0.90	0.90
(Eq. 15)	SD	0.11895	0.08996	0.06853

System: Butanoic acid + Ethanenitrile

Model	*Constants*	*Temp. --->* 25°C	35°C	45°C
McAllister	η_{12}	1.50752	1.36848	0.84291
(Eq. 9)	η_{21}	0.69801	0.59790	0.67622
	SD	0.00535	0.00806	0.00154
Heric	δ_{12}	1.24180	1.32930	1.34910
(Eq. 11)	δ_{21}	0.39040	0.52990	−0.30740
	SD	0.34509	0.31342	0.39305
Auslander	B_{12}	0.93397	0.75671	0.50023
(Eq. 12)	A_{21}	1.68470	1.36049	0.50168
	B_{21}	0.19892	0.09921	1.17389
	SD	0.00400	0.00280	0.00130
Teja&Rice	ψ_{12}	1.10	1.10	1.10
(Eq. 13)	SD	0.01093	0.01917	0.00890
Zihao&Jufu	A	989.889	989.973	0.94805
(Eq. 14)	B	129.806	123.595	0.13368
	SD	0.1078	0.0431	0.0228
Two Parameter				
model	A′	0.16250	0.16290	0.16540
(Eq. 16)	B′	0.11210	0.06700	0.0327
	SD	0.08201	0.06041	0.0529
Rice&Teja	ψ_{12}	0.90	0.90	0.90
(Eq. 15)	SD	0.0434	0.04831	0.0501

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