Study of static permittivity and hydrogen bonded structures in amide–alcohol mixed solvents

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ABSTRACT

Relative static permittivity of the binary mixtures of amides (formamide (FA), N-methylformamide (NMF), N,N-dimethylformamide (DMF) and N,N-dimethylacetamide (DMA)) with alcohols (ethyl alcohol (EA) and ethylene glycol (EG)) over the entire composition range have been investigated at 1 MHz and 30 °C. The concentration-dependent relative permittivity increment and the excess permittivity of these binary mixtures reveal that the strength of heterogeneous H-bonded molecular interactions, the dipolar ordering and the molar ratio of stable adduct of amides with EA and EG are influenced by number of hydroxyl groups of the alcohol molecules and the type of substitution in amide molecules.

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1. Introduction

Design of mixed solvents of required solvating power with suitable liquid state range to predict the solubility and chemical stability of the solute is a highly interdisciplinary field of research in condensed matter physics, chemistry, biology, pharmaceutical and soft material sciences. The relative static permittivity (dielectric constant) of the solvents is a fundamental thermodynamical property, which is significantly influenced by H-bond molecular interactions and its measurement provides the path to understand the molecular structures of the pure solvents and their mixtures. The static permittivity of a dipolar liquid is mainly governed by its dipole moment, Kirkwood correlation factor and the number of molecules per unit volume. Due to these facts the activity coefficient, ion interactions and solvation, drug solubility and many more physico-chemical properties of the solvents are the function of their static permittivity. Among polar solvents, amides have high polarity, strong solvating power, and a large liquid state range which makes them an important class of organic solvents.

Conformation of H-bonding molecular structures and dynamics of amides has remained intensive subject of investigations [1–6], which is owing to the fact that amides are models for studying H-bonded peptide linkages [7] and also used as electrolyte solvents [8,9]. The strong H-bonded self-association in alcohols [10,11] has made commonly used solvents from the early stage of solu-

tion chemistry. Therefore, it is interesting to investigate the static permittivity of amides mixed with alcohols for their molecular conformations and potential applications in the field of science and technology. Recently, we have investigated the dielectric constants of amide–amide mixtures [12], amide–amine mixtures [13,14], amide–water mixtures [15], amide–glycerol mixtures [16] and amide–cosolvent mixtures [17–20] in relation to their molecular conformations.

In continuation of our ongoing research work on mixed solvents, purpose of this study is to provide the precise static permittivity data of the amides (FA, NMF, DMF and DMA) mixed with ethyl alcohol (EA) (monohydric alcohol) and ethylene glycol (EG) (dihydric alcohol) over the entire concentration range at 30 °C. Further, the aim of this contribution is to confirm the H-bonded network formation in amide–alcohol mixtures in regard to their structural conformation, and to reveal the effect of number of hydroxyl groups of alcohol molecules on the H-bonded hetero-molecular interactions with unsubstituted, mono-substituted (N-alkylamide) and di-substituted (N,N-dialkylamide) amides.

2. Experimental

Reagent-grade formamide (FA), N,N-dimethylformamide (DMF), N,N-dimethylacetamide (DMA), ethyl alcohol (EA) and ethylene glycol (EG) were purchased from E. Merck and Loba Chemie of India. N-methylformamide (NMF) is obtained from Sigma–Aldrich, USA. The mass fraction purities of these samples as reported by the manufacturers are: FA > 99.5%, NMF > 99.5%, DMF > 99.5%, DMA > 99.5%, EA > 99.9% and EG > 99.5%. The binary

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Table 1
Values of relative static permittivity ε_m and limiting high frequency permittivity $\varepsilon_{\infty m}$ of the binary mixtures of amide–EA and amide–EG at different mole fraction of EA, X_{EA} and EG, X_{EG} at 30 °C.

Amide–EA binary mixtures											
FA–EA			NMF–EA			DMF–EA			DMA–EA		
X_{EA}	ε_m	$\varepsilon_{\infty m}$	X_{EA}	ε_m	$\varepsilon_{\infty m}$	X_{EA}	ε_m	$\varepsilon_{\infty m}$	X_{EA}	ε_m	$\varepsilon_{\infty m}$
0.000	106.14	2.0635	0.000	169.76	2.0221	0.000	36.55	2.0126	0.000	37.72	2.0403
0.046	100.82	2.0520	0.101	144.63	2.0022	0.087	35.57	2.0028	0.102	36.31	2.0255
0.095	95.19	2.0383	0.202	123.72	1.9853	0.169	34.72	1.9912	0.197	35.21	2.0121
0.146	89.27	2.0246	0.302	104.17	1.9662	0.249	33.96	1.9799	0.285	34.16	1.9983
0.199	82.58	2.0095	0.402	85.99	1.9460	0.325	32.98	1.9662	0.367	32.95	1.9825
0.254	76.43	1.9943	0.502	71.06	1.9265	0.399	32.16	1.9547	0.444	32.15	1.9695
0.312	70.03	1.9790	0.602	57.80	1.9072	0.469	31.39	1.9446	0.515	31.25	1.9586
0.374	64.23	1.9662	0.702	46.91	1.8865	0.537	30.57	1.9324	0.583	30.30	1.9438
0.438	57.92	1.9491	0.802	37.60	1.8659	0.603	29.63	1.9204	0.646	29.54	1.9321
0.506	51.93	1.9349	0.901	29.54	1.8469	0.666	28.78	1.9072	0.705	28.71	1.9185
0.577	46.28	1.9149	1.000	23.87	1.8333	0.726	27.80	1.8934	0.761	27.78	1.9019
0.652	41.17	1.8991				0.785	27.05	1.8826	0.814	27.00	1.8884
0.732	36.15	1.8826				0.841	26.22	1.8725	0.864	26.20	1.8742
0.816	31.37	1.8632				0.896	25.39	1.8553	0.912	25.49	1.8605
0.905	27.60	1.8469				0.949	24.65	1.8436	0.957	24.64	1.8450
1.000	23.87	1.8333				1.000	23.87	1.8333	1.000	23.87	1.8333
Amide–EG binary mixtures											
FA–EG			NMF–EG			DMF–EG			DMA–EG		
X_{EG}	ε_m	$\varepsilon_{\infty m}$	X_{EG}	ε_m	$\varepsilon_{\infty m}$	X_{EG}	ε_m	$\varepsilon_{\infty m}$	X_{EG}	ε_m	$\varepsilon_{\infty m}$
0.000	106.14	2.0635	0.000	169.76	2.0221	0.000	36.55	2.0126	0.000	37.72	2.0403
0.048	103.75	2.0621	0.105	139.93	2.0221	0.090	37.69	2.0164	0.106	38.21	2.0420
0.099	100.44	2.0595	0.209	118.26	2.0246	0.176	38.57	2.0198	0.204	38.76	2.0443
0.151	96.49	2.0563	0.312	102.12	2.0246	0.258	39.36	2.0218	0.294	39.26	2.0446
0.206	92.28	2.0532	0.413	87.49	2.0246	0.335	39.93	2.0232	0.378	39.64	2.0449
0.263	88.70	2.0506	0.514	76.46	2.0246	0.410	40.56	2.0246	0.455	39.95	2.0446
0.322	84.61	2.0480	0.613	67.14	2.0246	0.481	40.98	2.0255	0.527	40.24	2.0437
0.384	80.30	2.0449	0.711	59.59	2.0221	0.548	41.23	2.0275	0.594	40.45	2.0420
0.449	75.79	2.0420	0.809	52.60	2.0221	0.613	41.38	2.0281	0.656	40.59	2.0400
0.517	70.99	2.0392	0.905	46.26	2.0221	0.676	41.42	2.0281	0.715	40.62	2.0383
0.588	66.04	2.0363	1.000	39.84	2.0221	0.735	41.35	2.0288	0.769	40.51	2.0366
0.662	61.55	2.0339				0.792	41.24	2.0282	0.821	40.49	2.0338
0.740	56.35	2.0306				0.847	41.01	2.0272	0.870	40.46	2.0315
0.823	51.05	2.0281				0.900	40.71	2.0262	0.916	40.29	2.0303
0.909	46.12	2.0269				0.951	40.23	2.0249	0.959	40.06	2.0275
1.000	39.84	2.0221				1.000	39.84	2.0221	1.000	39.84	2.0221

mixtures of FA, NMF, DMF and DMA with EA and EG were prepared by volume percent using exact volume amounts of the pure liquids by syringing into stoppered bottles to prevent evaporation and environmental contamination at room temperature. Mole fractions of the mixture constituents were determined by weighing the used volume amounts of the pure liquids to reduce possible errors in mole-fraction calculations. The binary mixtures of FA–EA/EG, DMF–EA/EG and DMA–EA/EG were prepared at 16 different volume-fraction concentrations in the range 0–1 and in the steps of 0.06666 of each of the pure components. The binary mixtures of NMF–EA/EG were prepared at 11 different volume-fraction concentrations in the range 0–1 and in the steps of 0.1 of each of the pure components.

The precise values of the static permittivity ε of pure solvents and ε_m of the binary mixtures were determined by capacitive measurement at 1 MHz. Agilent 4284A precision LCR meter and Agilent 16452A liquid dielectric test fixture were used for the capacitance measurement without and with sample. The dielectric constant measurement accuracy of the dielectric test fixture is ±0.3%, which was estimated by the calibration of the cell with standard dipolar liquids. The limiting high frequency permittivity ε_∞ of pure solvents and $\varepsilon_{\infty m}$ of the binary mixtures, were taken as the square of refractive index n_D, which was measured with an Abbe refractometer at the wavelength of sodium-D line. The maximum measurement error in the ε_∞ values is ±0.02%. All measurements were made at a temperature 30 ± 0.01 °C, which was maintained by

a microprocessor based Thermo-Haake DC10 controller. The experimental ε_m and $\varepsilon_{\infty m}$ values of the amide–alcohol binary mixtures at various mole fractions of the alcohol are recorded in Table 1.

The relative permittivity increment, $\Delta \varepsilon$, also called the deviations of relative permittivity of the binary mixture was determined by the mole-fraction-weighted additive mixture law Eq. (1):

$$\Delta \varepsilon = \varepsilon_m - (\varepsilon_1 X_1 + \varepsilon_2 X_2) \tag{1}$$

where X is the mole fraction and subscripts 1 and 2 represent the constituents of binary mixture. The evaluated $\Delta \varepsilon$ values along with ε_m and $\varepsilon_{\infty m}$ values of the binary mixtures of amide–EA and amide–EG are plotted against the mole fraction of EA, X_{EA} and EG, X_{EG} in Figs. 1 and 2, respectively.

The excess permittivity ε^E of amide–EA and amide–EG mixtures was determined from the volume-fraction-weighted additive mixture law Eq. (2):

$$\varepsilon^E = \varepsilon_m - (\varepsilon_1 \phi_1 + \varepsilon_2 \phi_2) \tag{2}$$

where ϕ_1 and ϕ_2 are the volume fractions of liquids 1 and 2 of binary mixtures, respectively. The evaluated ε^E values of the amide–alcohol mixed solvents using Eq. (2) are plotted against volume fraction ϕ_{EA} and ϕ_{EG} in Fig. 3.
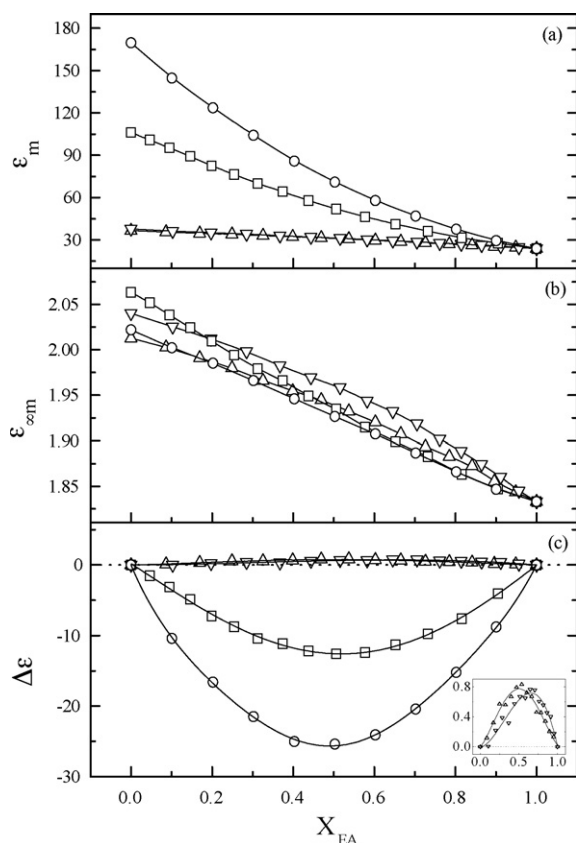
Fig. 1. Plots of ε_m, $\varepsilon_{\infty m}$ and $\Delta\varepsilon$ against mole fraction of ethyl alcohol (EA) X_{EA}, of FA–EA (□), NMF–EA (○), DMF–EA (△) and DMA–EA (▽) binary mixtures at 30 °C. In (a) and (b) lines are smooth joining through the data points, whereas in (c) lines are polynomial fits of data points drawn using Origin® non-linear curve fitting tool. Inset of (c) shows the enlarged view of $\Delta\varepsilon$ values for DMF–EA and DMA–EA mixtures.

Fig. 2. Plots of ε_m, $\varepsilon_{\infty m}$ and $\Delta\varepsilon$ against mole fraction of ethylene glycol (EG) X_{EG}, of FA–EG (□), NMF–EG (○), DMF–EG (△) and DMA–EG (▽) binary mixtures at 30 °C. In (a) and (b) lines are smooth joining through the data points, whereas in (c) lines are polynomial fits of data points drawn using Origin® non-linear curve fitting tool. Inset of (c) shows the enlarged view of $\Delta\varepsilon$ values for FA–EG, DMF–EG and DMA–EG mixtures.

3. Results and discussion

The measured ε values of amides and alcohols in pure liquid state are found in good agreement with the literature values [1,4,12–25]. The variation of ε_m of amide–EA and amide–EG mixtures against mole fraction of EA, X_{EA} and EG, X_{EG} (Figs. 1a and 2a) are more or less non-linear, which suggests the formation of H-bonded complexes between amides and alcohols over the entire concentration range. The non-linear behaviour of $\varepsilon_{\infty m}$ values of the investigated mixtures (Figs. 1b and 2b), reflects the change in electronic polarization due to unlike molecules H-bond interactions [11,23–25].

The Kirkwood correlation factor g is a measure of H-bonded dipolar ordering in the polar molecules [10,11]. The g values of FA, NMF, DMA, EA and EG are 2.22, 4.19, 1.50, 2.73 and 2.35, respectively, established the existence of self-associated H-bonded linear chain type molecular structures with parallel dipolar alignments in their pure liquid state, whereas unity g value of DMF reveals its non-associative H-bonded molecular behaviour [12–20]. Among the members of amide family, the higher g value of NMF molecules (mono-substituted N-alkylamide) suggests a strong and large range H-bonded ordering in its pure liquid state as compared to FA (unsubstituted amide) and DMA (di-substituted N,N-dialkylamide) molecules [12]. In comparison to EG (dihydric alcohol), EA (mono-hydric alcohol) molecules have higher range dipolar ordering and strong H-bond association as evidenced from the higher g value of EA molecules [23]. On mixing of different self-associated amides with mono and dihydric alcohols, there may be a formation of large range 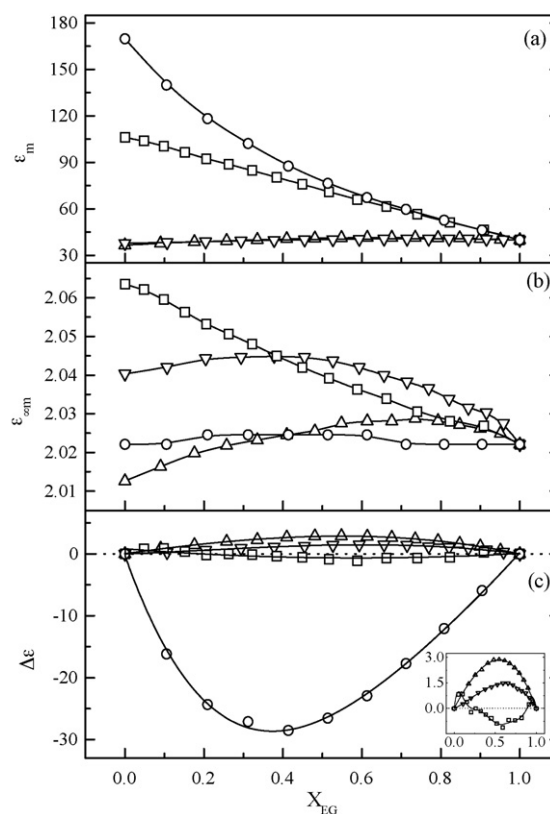amide–alcohol hetero-molecular structures with variation in their self-associated H-bonded structures. In amide–alcohol mixtures, there may be three kinds of H-bonded cooperative domains, namely, amide–amide and alcohol–alcohol homomolecular structures, and amide–alcohol hetero-molecular structures.

The investigations of concentration-dependent $\Delta\varepsilon$ and ε^E values of the mixed dipolar solvents established the following information regarding H-bond molecular interactions between unlike molecules [12–21,23–32].

(i) $\Delta\varepsilon$ and $\varepsilon^E = 0$ indicates that the mixture constituents do not interact and thus have an ideal mixing behaviour;

(ii) $\Delta\varepsilon$ and $\varepsilon^E < 0$ indicates that the mixture constituents act as structure-breaker for the homogeneous H-bonded pure liquid structures, and hence there is decrease in the total number of parallel aligned effective dipoles that contributed to the mixture dielectric polarization;

(iii) $\Delta\varepsilon$ and $\varepsilon^E > 0$ indicates that the constituents of the binary mixture interact in such a way that they act as structure-makers and results in an increase of effective number of parallel aligned dipoles contributing to the mixture dielectric polarization;

(iv) The magnitude of $\Delta\varepsilon$ and ε^E values is the evidence of the strength of unlike molecules H-bond interactions. The large $\Delta\varepsilon$ values suggest the stronger H-bond hetero-molecular connectivities and *vice versa*; and

(v) The molar concentration corresponding to the pronounced maximum of $\Delta\varepsilon$ values represent the molar ratio of a stable adduct in the mixture.

The X_{EA} and X_{EG} values corresponding to the maximum magnitude of the deviation of relative permittivity $\Delta\varepsilon_{max}$ of amide–EA

Table 2

Maximum values of the relative permittivity increment, $\Delta\varepsilon_{max}$ and the excess permittivity, $\varepsilon^{E}_{(max)}$ and corresponding mole-fraction (X) and volume-fraction (ϕ) concentrations of the alcohols of amide–EA and amide–EG binary mixtures at 30 °C.

Amide–EA mixtures					Amide–EG mixtures				
	$\Delta\varepsilon_{max}$	X_{EA}	$\varepsilon^{E}_{(max)}$	ϕ_{EA}		$\Delta\varepsilon_{max}$	X_{EG}	$\varepsilon^{E}_{(max)}$	ϕ_{EG}
FA–EA	−12.59	0.52	−12.58	0.61	FA–EG	−0.92	0.55	−0.96	0.63
NMF–EA	−25.66	0.49	−25.34	0.48	NMF–EG	−28.66	0.36	−28.27	0.37
DMF–EA	0.77	0.50	0.76	0.46	DMF–EG	2.86	0.55	2.86	0.48
DMA–EA	0.72	0.65	0.72	0.55	DMA–EG	1.46	0.60	1.45	0.49

and amide–EG binary mixtures were determined by fitting the $\Delta\varepsilon(X_{EA})$ and $\Delta\varepsilon(X_{EG})$ data plots to the polynomial equation using Origin® non-linear curve fitting tool, which are shown by continuous lines in Figs. 1c and 2c. The values of $\Delta\varepsilon_{max}$ and corresponding X_{EA} and X_{EG} of these binary mixtures are recorded in Table 2. The values of maximum excess permittivity, $\varepsilon^{E}_{(max)}$ and corresponding volume-fraction concentration of the alcohols were determined by fitting the $\varepsilon^{E}(\phi_{EA})$ and $\varepsilon^{E}(\phi_{EG})$ data plots to the polynomial equation as shown by continuous lines in Fig. 3, and these values are also recorded in Table 2.

3.1. Molecular interactions in amide–EA mixtures

Fig. 1c shows that the FA–EA and NMF–EA mixtures have negative $\Delta\varepsilon$ values of large magnitude, which confirms the formation of heterogeneous molecular pairs with decrease in total number of parallel aligned dipoles. The magnitude of $\Delta\varepsilon$ values of NMF–EA

Fig. 3. Plots of ε^{E} against volume fraction of ethyl alcohol (EA) ϕ_{EA} (a) and ethylene glycol (EG) ϕ_{EG} (b), of FA–EA/EG (□), NMF–EA/EG (○), DMF–EA/EG (△) and DMA–EA/EG (▽) binary mixtures at 30 °C. In (a) and (b) lines are polynomial fits of data points drawn using Origin® non-linear curve fitting tool. Inset of (a) shows the enlarged view of ε^{E} values for DMF–EA and DMA–EA and inset of (b) shows the enlarged view of ε^{E} values for FA–EG, DMF–EG and DMA–EG mixtures.

mixtures is almost twice that of FA–EA mixtures, which reveals the formation of strong H-bonded hetero-molecular interactions of mono-substituted N-alkylamide as compared to the unsubstituted amide with ethyl alcohol molecules. The DMF and DMA (N,N-dialkylamide) mixed with EA have positive $\Delta\varepsilon$ values but of small magnitude, which shows the formation of weak H-bond molecular interactions between these unlike molecules with some increase in number of parallel aligned dipoles contributing to the dielectric polarization. The ε values of pure DMF and DMA are nearly same although there is significant difference in their molecular size. The almost equal $\Delta\varepsilon$ values of these mixtures (Fig. 1c) confirm that DMF and DMA have nearly same H-bonding interaction strength with EA molecules.

The pronounced maximum in $\Delta\varepsilon$ values (Fig. 1c) around $X_{EA} \sim 0.5$ for the FA–EA, NMF–EA and DMF–EA mixtures (Table 2) confirms the formation of a stable adduct of 1:1 molar ratio in these mixtures. In case of DMA–EA mixture, the $X_{EA} \sim 0.65$ suggests the formation of 2:1 complexes of EA to the DMA molecules. Although the DMF and DMA have nearly equal H-bond interaction strength with EA, but their molar ratio of stable adduct is influenced by the molecular size of these N,N-dialkyl-substituted amides.

3.2. Molecular interactions in amide–EG mixtures

Fig. 2c shows that the FA–EG and NMF–EG mixtures have negative $\Delta\varepsilon$ values, whereas these values are positive for DMF–EG and DMA–EG mixtures. Large magnitude of $\Delta\varepsilon$ values of NMF–EG mixtures conclude the formation of strong H-bond interactions between NMF and EG molecules, with significant decrease in number of parallel aligned effective dipoles contributing to the dielectric polarization. Comparative small magnitude of $\Delta\varepsilon$ values of FA–EG mixtures suggest the formation of weak H-bonded interactions between these molecules with some decrease in the number of parallel aligned effective dipoles. The small positive $\Delta\varepsilon$ values of DMF–EG and DMA–EG mixtures indicate the formation of weak H-bonded interactions between unlike molecules in these mixtures with some increase in number of effective parallel aligned dipoles. Comparative higher $\Delta\varepsilon$ values of DMF–EG mixture shows that the H-bond interactions of DMF and DMA with EG molecules are influenced by the size of N,N-dialkylamides.

The pronounced maximum in $\Delta\varepsilon$ values at $X_{EG} \sim 0.5$ of FA–EG and DMF–EG mixtures (Fig. 2c and Table 2) suggest the formation of 1:1 complexation in these mixtures. The maxima in $\Delta\varepsilon$ values at $X_{EG} \sim 0.6$ for the DMA–EG mixture reveals the formation of 2:1 complexes of EG to DMA molecules. The NMF–EG mixture shows the maximum $\Delta\varepsilon$ values at $X_{EG} \sim 0.36$, which suggest the stable adduct of 1:2 molar ratio of EG to NMF molecules.

Recently, Iglesias et al. [29,31,32] generalized various thermodynamical quantities of different dipolar liquid mixtures and concluded that the mixture permittivity is a volume-fraction-weighted sum of the mixture pure component relative permittivities given by Eq. (2). Considering this fact, the volume-fraction-dependent excess permittivity ε^{E} of the amide–alcohol binary mixtures were analyzed and compared with the mole-fraction-dependent values
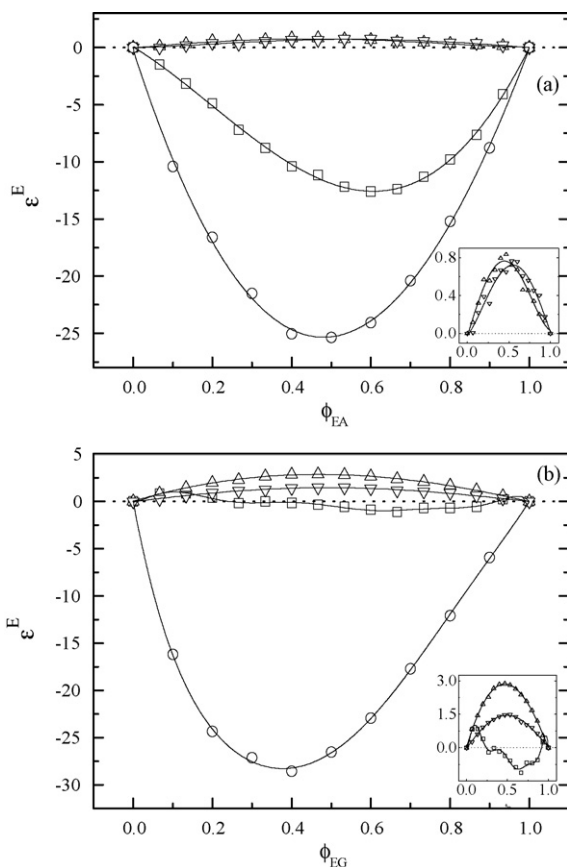
of the $\Delta\varepsilon$. The comparison of the shape and magnitude of the $\Delta\varepsilon$ and ε^E plots (Figs. 1c with 3a; and 2c with 3b), and also the $\Delta\varepsilon_{max}$ and $\varepsilon^E_{(max)}$ values of same amide–alcohol mixtures (Table 2) reveals that the values of these parameters have good resemblance. From these observations it can be concluded that both Eqs. (1) and (2) provide the same information on the heterogeneous molecular interactions in these amide–alcohol mixtures. Earlier [19], similar results were also observed for amides mixed with dimethylsulphoxide and 1,4-dioxane.

Further, the comparative magnitude of $\Delta\varepsilon$ and ε^E values of the amide–alcohol mixtures (Figs. 1c with 2c; and 3a with c), it is inferred that the H-bonded interaction strength of NMF molecules with EA and EG is strongest, and the molar ratio of their stable adduct is influenced by the number of hydroxyl groups of the alcohols molecules. It seems that the large difference in g values of NMF and alcohols is responsible for the formation of strong hetero-molecular H-bond interactions in their binary mixtures. Comparative $\Delta\varepsilon$ and ε^E values of FA–EA and FA–EG mixtures suggest that the FA form strong H-bond interaction with monohydric alcohol, which may be due to significant difference in g values of FA and EA molecules, whereas nearly equal g values of FA and EG molecules is responsible for the formation of weak H-bond interactions between these unlike molecules. Both the DMA and DMF have weak H-bond interactions with the EA and EG molecules, which confirms that the N,N-dialkyl substitution in the molecules of amide family prevents the formation of strong H-bond interactions with the mono- and dihydric alcohols.

4. Conclusions

The paper reports the precisely measured static permittivity values of the binary mixtures of amides family with EA and EG molecules over the entire composition range. The excess permittivity values conclude that these mixtures have non-ideal mixing behaviour. The FA molecules form strong H-bond interactions with EA and comparatively weak interactions with EG, whereas NMF have strongest H-bond interactions with both the EA and EG molecules with decrease of effective parallel aligned dipoles contributed in their dielectric polarization. The DMF and DMA molecules have weak H-bond interactions with EA and EG molecules but increase their parallel aligned effective dipolar ordering. The EA form complexes of 1:1 molar ratio with FA, NMF and DMF, and 2:1 with DMA. The molar ratio of stable adduct of EG with FA and DMF is 1:1, whereas 1:2 and 2:1 complexes are formed in EG–NMF and EG–DMA mixtures, respectively. A close resemblance observed in mole-fraction-dependent relative permittivity increment and volume-fraction-dependent excess permittivity values of amide–alcohol mixtures confirms the suitability of both the mole-fraction-weighted and volume-fraction-weighted additive values of static permittivity for the analysis of the heterogeneous H-bonded molecular interactions in these binary systems.

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