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Dielectric relaxation studies of binary mixture of ethylene glycol mono phenyl ether and methanol by Time Domain Reflectometry

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ABSTRACT

The complex permittivity spectra of binary liquid mixtures of ethylene glycol mono phenyl ether (EGMPE) with methanol (MeOH) were determined in frequency range of 10 MHz to 25 GHz using Time Domain Reflectometry (TDR) at four different temperatures 283.15, 288.15, 293.15 and 298.15 K through entire concentration range. The dielectric parameters viz static permittivity (ε), high frequency limit dielectric permittivity (ε_∞) and relaxation time (τ) have been obtained by CNLS fit method. From the experimental data, parameters such as excess static permittivity (ε^E), excess inverse relaxation time $(1/\tau)^E$, Kirkwood correlation factor (g^f), effective Kirkwood correlation factor (g^{eff}) and Bruggeman's factor (f^b) and some thermo-dynamical parameters have been calculated. Excess parameters were fitted to the Redlich–Kister type polynomial equation.

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1. Introduction

Because of their different structures and varying dynamic response to an external field, each material displays a unique complex dielectric spectrum in the radio/microwave frequency range. These spectra usually include one or more dispersion. By this uniqueness, we can characterize a material by studying its dielectric dispersion [1]. Thus, the dielectric measurements have emerged as a tool for the engineers and the scientists to study the dielectric properties as a diagnostic index for material compounds in mixtures. Polar–polar liquid mixtures have been subject to a number of dielectric studies [2–10], which provided important insight into H-bond dynamics. In this paper, we have reported a detailed dielectric characterization of binary mixtures of ethylene glycol mono phenyl ether (EGMPE) and methanol (MeOH).

The aim of this study is to work out a relaxation model for describing the broadband complex permittivity spectra $\varepsilon^*(\omega) = \varepsilon'(\omega) - j\varepsilon''(\omega)$ of EGMPE–MeOH mixtures over the entire mixing range. The dynamical aspects of the EGMPE–MeOH solutions have also been examined to gain some information on the expected interactions. Dielectric dispersion data for EGMPE–MeOH solutions at various concentrations are presented at four different temperatures.

2. Materials and sample preparation

EGMPE (extra pure) and MeOH (AR grade) were obtained from HPLC Pvt. Ltd. (India) and were used without further purification. Binary mixtures of EGMPE with MeOH were prepared at nine concentrations by volume. The concentration was then converted into the mole fraction of component 1, using the following formula [10]:

$$x_1 = \frac{\rho_1 \times v_1/M_1}{\rho_1 \times v_1/M_1 + \rho_2 \times v_2/M_2} \quad (1)$$

where M is the molecular weight, v is the volume and ρ is the density. 1 represents MeOH and 2 represents EGMPE. The mole fraction is accurate to 0.1%.

3. Experimental methods

The dielectric spectra of EGMPE with MeOH mixture were obtained by Time Domain Reflectometry (TDR). The Tektronix model no. DSA8200 Digital Serial Analyzer sampling oscilloscope along with the sampling module 80E08 has been used and this module provides accurate oscilloscope measurement with user selectable bandwidth of 20 or 30 GHz. The block diagram and experimental setup of Time Domain Reflectometry is shown in Fig. 1. The coaxial cable semi-rigid copper, EZ_86/M17 (Huber + Suhner Electronics Pvt. Ltd.), with flat end was used. The outer diameter of cable is 2.2 mm. Sampling oscilloscope monitors changes in step pulse after reflection from the end of line. Reflected pulse without sample $R_1(t)$ and with sample

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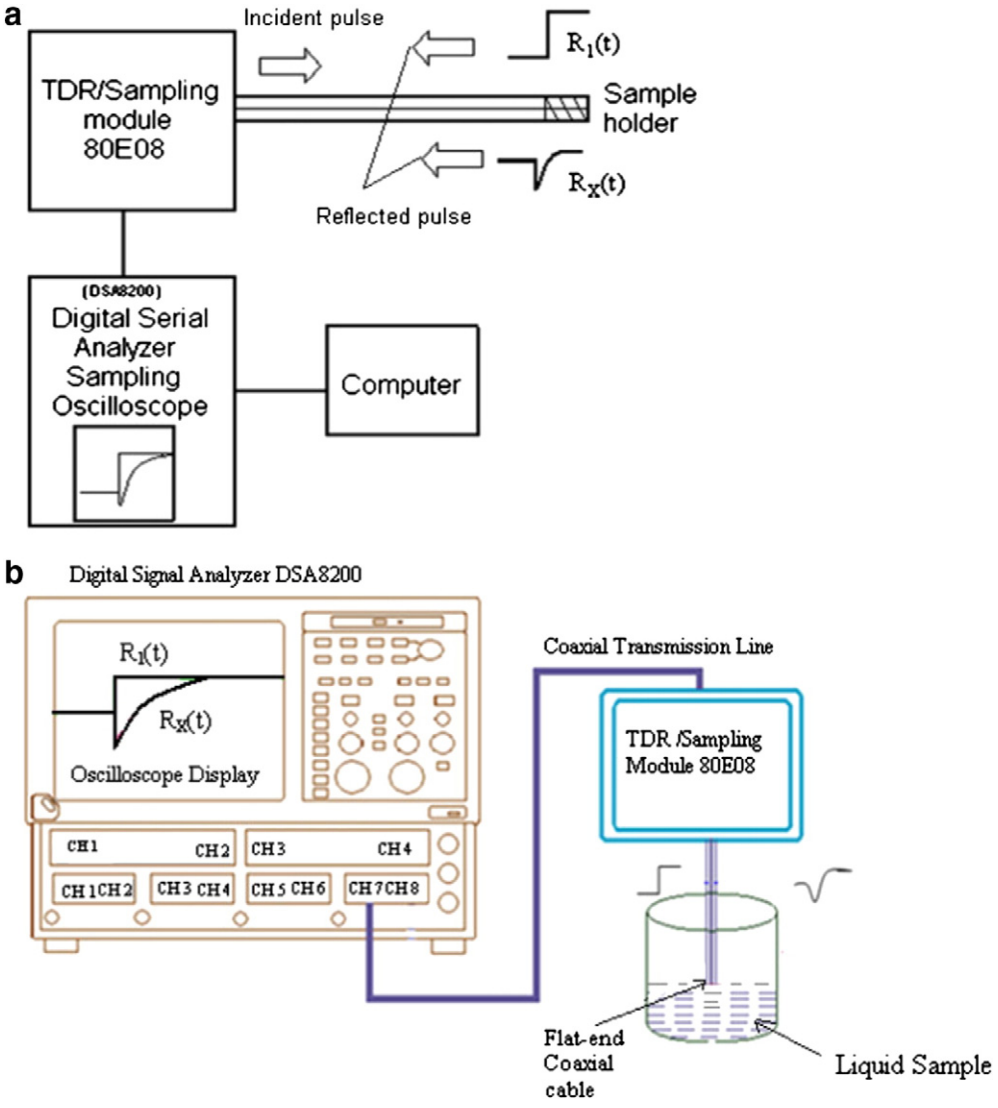

Fig. 1. (a) Block diagram of Time Domain Reflectometry (TDR). (b) Experimental setup of TDR.

$R_X(t)$ were recorded in time window of 2 ns and digitized in 2000 points in the memory of the oscilloscope and transferred to the computer. These recorded pulses are added $[q(t) = R_1(t) + R_X(t)]$ and subtracted $[p(t) = R_1(t) - R_X(t)]$. Further the Fourier transformation of p(t) and q(t) was obtained by Shannon [11] and Samulon [12] methods respectively, for the frequency range of 10 MHz to 30 GHz. The complex reflection spectra were determined [13,14] as follows:

$$\rho^*(\omega) = (c/j\omega d)[p(\omega)/q(\omega)] \quad (2)$$

where c is the speed of light, ω is the angular frequency, d is the effective pin length and p(x) and q(x) are the Fourier transforms p(t) and q(t) method respectively. The complex permittivity spectra $\varepsilon^*(\omega)$ is obtained from complex reflection coefficient q* by using bilinear calibration method [13,14]. Fig. 2 shows complex permittivity spectra $[\varepsilon^*(\omega) = \varepsilon'(\omega) - j\varepsilon''(\omega)]$ for the entire molar concentration of MeOH in the frequency range from 10 MHz to 25 GHz at 298.15 K. Where $\varepsilon'(\omega)$ is the real component known as dielectric constant and $\varepsilon''(\omega)$ imaginary component known as dielectric loss.

4. Data analysis

A dispersion is mainly characterized by two parameters, dielectric relaxation strength (Δε) and relaxation time (τ), so the relation between these two parameters and the quantitative make up of a mixture can be used for this purpose. To determine the dispersion parameters from the spectra, we have applied to Cole–Davidson function

$$\varepsilon^* = \varepsilon_{\infty 1} + \frac{\Delta\varepsilon}{[1 + (j\omega\tau)]^{\beta}} \quad (3)$$

as the dispersion models in our CNLS fitting. $\varepsilon_{\infty 1}$ is the high frequency limit dielectric permittivity, τ is the relaxation time, and ω is the angular frequency, which reaches to Debye function when β equals to unity [15].

The empirical parameter β controls the dispersion shape, which usually shows an asymptotic long tail at the high frequency end of dispersion while the low frequency part resembles Debye behavior. CNLS fitting takes both the real and imaginary parts of the complex permittivity into the objective function simultaneously to ensure a complete fit [16].

The choice of the relaxation model depends on the frequency range accessible by the experiment and on the quality of the data [17]. Since the complex permittivity spectra of EGMPE–MeOH mixtures are obtained in the frequency range of 10 MHz–25 GHz single C–D/Debye model is used to fit the experimental data and the relaxation parameters are obtained for the main relaxation process.

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