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Optical properties of binary and ternary liquid mixtures containing tetralin, isobutylbenzene and dodecane

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ABSTRACT

Refractive indices of binary and ternary mixtures formed by tetralin (1,2,3,4-tetrahydronaphthalene), isobutylbenzene (2-methyl-1-propyl benzene) and *n*-dodecane are presented over a wide range of compositions. All measurements of the refractive index have been conducted at 298.15 K and atmospheric pressure using two light sources: one in the visible ($\lambda = 670$ nm) and the other in the infrared ($\lambda = 925$ nm) spectrum. The concentration derivatives of the refractive index have been determined. The mixture compositions, where these two wavelengths are applicable for the measurements of mass transport coefficients by interferometry, are estimated and discussed.

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1. Introduction

Optical properties and especially the variation of the refractive index with temperature and concentration are the basis of different optical methods aimed at studying convection and heat and mass transfer in transparent media [1,2]. Among the existing methods for measurement of thermodiffusion and Soret coefficients, the optical techniques play an important role – such as Thermal Lens Technique [3], Beam Deflection [4,5], Thermal Diffusion Forced Rayleigh Scattering [6,7], Optical Digital Interferometry [8,9]. All of these techniques require the accurate knowledge of the refractive index derivatives with respect to concentration and temperature (the so-called "contrast factors") $\partial n_{\lambda_i}/\partial c_j,\ i,\ j = 1,2$.

The study of binary mixtures can be performed without serious problems, but to extend the above mentioned techniques to *N*-component mixtures, $N-1$ beams of different wavelengths are required. Correspondingly, the wavelengths (or compositions) should be chosen in such a way that the matrix of the refractive index derivatives with respect to concentration (its elements are $a_{ij} = \partial n_{\lambda_i}/\partial C_j,\ i,\ j = 1, N-1$) has not be ill-conditioned [10]. The literature data on refractive index and its compositional dependence for ternary mixtures of liquids at different wave lengths are rather poor. There exists the measurements of the refractive index for the mixtures of associate liquids [12] and mixtures containing hydrocarbons and alcohols [11]. Looking forward to investigate mass transport coefficients in ternary mixtures of hydrocarbons we examine the optical properties at two different wave lengths: red ($\lambda = 670$ nm) and infrared ($\lambda = 925$ nm).

The previous studies of optical properties for the mixtures containing hydrocarbons and alcohols [11] and mixtures of associated liquids [12] indicate that applying different mixing rules [13–18] does not allow to obtain accurate values of the concentration derivatives of the refractive index $\partial n_{\lambda_i}/\partial C_j,\ i,\ j = 1,2$. Therefore, the best way to obtain information about concentration dependence of the refractive index in mixtures with complicated behavior is to perform measurements within the whole concentration range. Currently, the knowledge of matrix a_{ij} for *THN-IBB-nC*$_{12}$ mixture is especially important because it was used in the recent experiments on the International Space Station in the frame of DSC experiment. To take benefit from in-flight tests, the post-flight laboratory investigations with this mixture will actively continue.

We report measured refractive indices in mixtures formed by 1,2,3,4-tetrahydronaphthalene (*THN*), 2-methyl-1-propyl benzene (*IBB*) and *n*-dodecane (nC_{12}) at two different wavelengths. Possibility of using different mixing rules for the determination of refractive indices and their derivatives with respect to compositions is also discussed.

2. Experimental section

The origin and some properties of the chemical compounds used in the experiments are given in table 1. Seventy three compositions have been studied to cover the whole range of concentrations for mixtures (see figure 1). The mass transport coefficients for considered mixture were measured only at the point with equal

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TABLE 1
Samples information.

Short name	Source	Mole fraction purity	Analysis method	Density, g/cm^3 at 298.15 K	CAS number
THN	Acros Organics	0.98	GC[a]	0.973	119-64-2
IBB	Acros Organics	0.995	GC	0.85	538-93-2
nC_{12}	Acros Organics	0.99	GC	0.753	112-40-3

[a] Gas–liquid chromatography.

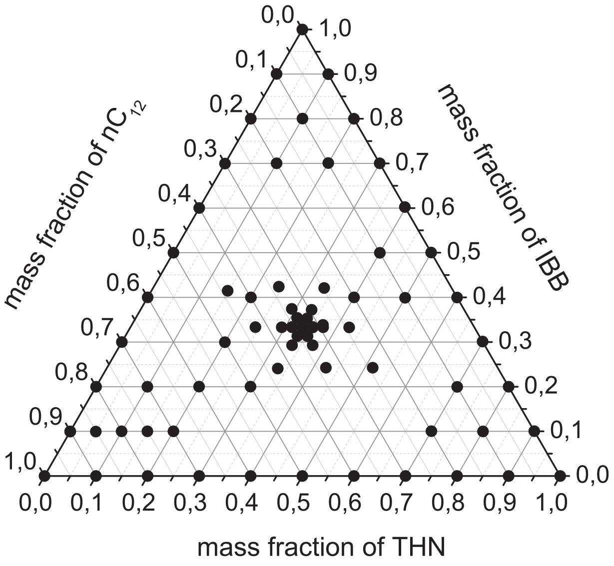

FIGURE 1. Concentration sampling.

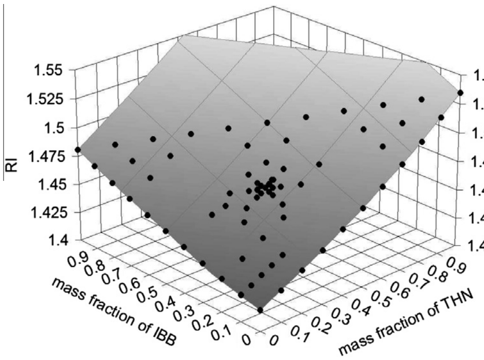

FIGURE 2. Refractive index of the $THN/IBB/nC_{12}$ mixture at $\lambda = 670$ nm.

mass fractions (1:1:1). Because the reported results in [5,19] displayed large discrepancy the knowledge of accurate values of all physical properties at this point is important. Correspondingly, the refractive indices around this point were measured with a smaller sampling than other once.

Detailed description of the experimental procedure for the refractive index measurements was done previously [11,12]. The mixtures were prepared by weighing each component using an electronic balance manufactured by Sartorius with resolution 10^{-2} mg/30 g. The possible error in mass fraction is estimated to be around ±0.0001. The refractive indices were measured with an Abbe refractometer manufactured by Bellingham and Stanley model 60/LR with a resolution of $5 \cdot 10^{-5}$ units of index of refraction. Beams of two laser diodes with $\lambda = 670$ nm and $\lambda = 925$ nm were used as a light source. Temperature during experiments was controlled by high precision refrigerated circulating bath NESLAB RTE-300. The current temperature of measurement prism of the refractometer was monitored using the build-in temperature sensor with uncertainty ±0.1 K. The possible uncertainty in the refractive index measurements is estimated to be less than ±0.00007 units.

3. Results and discussion

Refractive indices of binary and ternary mixtures containing *THN*, *IBB* and nC_{12} were studied and identified by notations (C_1, C_2, C_3) correspondingly. Experimental data on the refractive index of all investigated mixtures are shown at figure 2 and presented in table 2.

On the basis of the measured values the fitting polynomial functions were constructed, $n_{1f} = n_f(C_1, C_2)$, $n_{2f} = n_f(C_2, C_3)$, $n_{3f} = n_f(C_1, C_3)$. In general form these equations can be written e.g. for (C_1, C_2) as

$$\begin{aligned} n_{1f}(670\ \mathrm{nm}) = {} & 0.012653\,C_1^4 + 0.022242\,C_1^3 C_2 - 0.012809\,C_1^3 \\ & + 0.024506\,C_1^2 C_2^2 - 0.0093941\,C_1^2 C_2 \\ & + 0.029931\,C_1^2 + 0.025903\,C_1 C_2^3 \\ & - 0.023488\,C_1 C_2^2 + 0.040227\,C_1 C_2 \\ & + 0.086586\,C_1 + 0.008583\,C_2^4 - 0.016358\,C_2^3 \\ & + +0.023176\,C_2^2 + 0.04696\,C_2 + 1.4183, \end{aligned} \tag{1}$$

$$\begin{aligned} n_{1f}(925\ \mathrm{nm}) = {} & 0.0040518\,C_1^4 + 0.013177\,C_1^3 C_2 \\ & + 0.0029917\,C_1^3 + 0.011976\,C_1^2 C_2^2 \\ & - 0.00028537\,C_1^2 C_2 + 0.020123\,C_1^2 \\ & + 0.0033914\,C_1 C_2^3 + 0.0003478\,C_1 C_2^2 \\ & + 0.033512\,C_1 C_2 + +0.085095\,C_1 \\ & + 0.0034824\,C_2^4 - 0.0061261\,C_2^3 \\ & + +0.016368\,C_2^2 + 0.045621 C_2 + 1.414, \end{aligned} \tag{2}$$

where C_i is the mass fraction of the mixture component *i*.

For calculations of the coefficients of equations (1) and (2) a polynomial regression model with linear least squares techniques was used. The polynomials for the other functions (n_{2f}, n_{3f}) were constructed in a similar way. On average the differences between the measured points and the fitting function at these points do not exceed 0.025%, i.e.

$$\frac{n_{exp}(c_i) - n_f(c_i)}{n_{exp}(c_i)} \cdot 100\% < 0.025\%.$$

3.1. Concentration derivatives of the refractive index

The derivatives $(\partial n / \partial C_i)_{p,T,C_{j \neq i}}$ were determined by analytical differentiation of the polynomial fitting equations for the concentration dependence of the refractive index: $n_{1f} = n_f(C_1, C_2)$, $n_{2f} = n_f(C_2, C_3)$, $n_{3f} = n_f(C_1, C_3)$:

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