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## Prediction of excess molar volumes of selected binary mixtures from refractive index data

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**Abstract:** The excess molar volumes of twenty two binary mixtures containing various groups of organic compounds: alcohols (ethanol, 1-propanol, 1,2-propanediol, 1,3-propanediol and glycerol), ketone (acetone), ester (butyl lactate), lactam (*N*-methyl-2-pyrrolidone), polyethylene glycol (PEG 200 and PEG 400) and aromatics (benzene, toluene and pyridine) were predicted from the refractive index data, using three types of equations coupled with several mixing rules for refractive index calculations: the Lorentz–Lorenz, Dale–Gladstone, Eykman, Arago–Biot, Newton and Oster. The obtained results were analysed in terms of the applied equation and mixing rule and the nature of the interactions between the components of the mixtures.

**Keywords:** excess molar volume; mixing rules; prediction; organic solvents.

### INTRODUCTION

Knowledge of the thermophysical properties (volumetric properties, refractive index, viscosity, *etc.*) of pure organic compounds and their mixtures as a function of composition and temperature is of considerable interest for industrial applications. It is necessary for the investigation of non-ideality of mixtures, caused by molecular interactions and intermolecular forces between the components of a mixture, as well as for the design of processes and process equipment.

This work represents the results obtained for excess molar volume  $V^E$  prediction from refractive index  $n_D$  data for 22 binary mixtures in the temperature range 288.15–323.15 K (in some cases 288.15–333.15 K) and at atmospheric pressure. Binary systems containing alcohols (ethanol, 1-propanol, 1,2-propane-

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diol, 1,3-propanediol, and glycerol), ketone (acetone), ester (butyl lactate), lactam (*N*-methyl-2-pyrrolidone), polyethylene glycol (PEG 200 and PEG 400) and aromatics (benzene, toluene and pyridine) were chosen, due to their: *i*) structural variety causing non-ideal behaviour in the mixtures (mixtures exhibiting positive, negative and S-shaped  $V^E$  and  $\Delta n_D$  vs. composition curves) and *ii*) industrial importance.

The alcohols examined in this work are colourless liquids miscible with water and are mainly used as solvents. In addition, ethanol is a potential clean fuel to substitute for petroleum fuels due to its low emissions, particularly the significantly low level of soot.<sup>1-3</sup> 1-Propanol is an important industrial chemical that has been used in various industrial products, such as paints and cosmetics,<sup>4</sup> and is considered a better biofuel than ethanol. One of the major uses of 1,2-propanediol is as an industrial antifreeze due to lower toxicity than ethylene glycol.<sup>5,6</sup> 1,3-Propanediol can be used in the production of polymers, cosmetics, lubricants or drugs.<sup>7-9</sup> 1,3-Propanediol-based polymers have better properties and greater stability in comparison to polymers produced from 1,2-propanediol, butanediols or ethylene glycol.<sup>10</sup> Glycerol is used in foods and cosmetics, tobacco, wrapping and packaging materials, lubricants, urethane polymers, cleaning materials, detergents, wetting agents, emulsifiers, skin protectives, as well as in the pulp and paper, and leather and textile industries.<sup>11,12</sup>

Acetone is a colourless, mobile, flammable liquid, and is the simplest ketone. The main application of this compound is as a solvent, also as an intermediate to produce many important chemical substances, such as methyl methacrylate (MMA), bisphenol-A (BPA) and methacrylic acid, and in cosmetics.

Lactate esters are non-toxic and biodegradable, with excellent solvent properties, such as the ability to dissolve organic compounds, *e.g.*, nitro and ethyl cellulose, gums, oils, dyes, polymers and paints, and as high-boiling solvents.<sup>13</sup> As a “green” solvent, butyl lactate is an excellent candidate to replace halogenated solvents, such as ozone-depleting chlorofluorocarbons (CFCs), carcinogenic methylene chloride, toxic ethylene glycol ethers, and chloroform.<sup>14</sup>

*N*-Methyl-2-pyrrolidone (NMP) is a widely used compound due to its strong and selective power as a solvent.<sup>15</sup> For example, NMP is used in the petrochemical industry, in the microelectronics fabrication industry and in the manufacture of various compounds, such as pigments, cosmetics, drugs and pesticides.<sup>16</sup>

Polyethylene glycol is a non-toxic, highly biodegradable polymer.<sup>17</sup> PEG is well known as a food-additive<sup>18,19</sup> and is used for various biological and biomedical applications in aqueous solutions with biological macromolecules.<sup>20</sup>

Aromatics were analysed as a last class of chemical compounds in this work. Pyridine, as an industrial solvent and raw material in herbicide synthesis, is widely used in the chemical, pharmaceutical and oil industries.<sup>21</sup> Considering its

toxicity, there is a constant effort to find a way to remove this substance from wastewater and environment. Benzene is one of the basic petrochemicals, which is usually used as an intermediate in the processing of other substances. A large proportion of benzene is used in the production of three chemicals, cyclohexane,<sup>22</sup> cumene<sup>23</sup> and ethylbenzene.<sup>24</sup> Toluene has wide industrial application as a feedstock and as a solvent able not only to dissolve paints, rubber, adhesives and lacquers, but also some inorganic chemicals. It is much less toxic than benzene; hence it represents a good replacement for benzene as a solvent in industry. A number of works consider its removal from industrial wastewaters,<sup>25</sup> the treatment of toluene vapours<sup>26,27</sup> and the removal of toluene from air.<sup>28</sup>

For the prediction of  $V^E$  from  $n_D$  data, three types of equations were tested: *i*) the equation proposed by Nakata and Sakurai,<sup>29</sup> based on the use of mass fraction-based mixing functions, was applied for the first time in the work of Arancibia and Katz<sup>30</sup> (Model I), *ii*) the equation obtained by the first order expansion of Model I<sup>29</sup> (Model II) and *iii*) a particular case of Model II developed for iso-refractive mixtures<sup>29</sup> (Model III), coupled with different mixing rules for refractive indices, *i.e.*, the Lorentz–Lorenz, Dale–Gladstone, Eykman, Arago–Biot, Newton and Oster<sup>31</sup> rules.

#### THEORETICAL

The different mixing rules used in this paper for refractive indices calculation are presented by the following equations:

Lorentz–Lorenz (L–L):<sup>32,33</sup>

$$\frac{n_{Di}^2 - 1}{n_{Di}^2 + 2} = \sum_{i=1}^N \left[ \varphi_i \left( \frac{n_{Di}^2 - 1}{n_{Di}^2 + 2} \right) \right] \quad (1)$$

Dale–Gladstone (D–G):<sup>34</sup>

$$n_{Di} - 1 = \sum_{i=1}^N [\varphi_i (n_{Di} - 1)] \quad (2)$$

Eykman (Eyk):<sup>35</sup>

$$\frac{n_{Di}^2 - 1}{n_{Di}^2 + 0.4} = \sum_{i=1}^N \left[ \varphi_i \left( \frac{n_{Di}^2 - 1}{n_{Di}^2 + 0.4} \right) \right] \quad (3)$$

Arago–Biot (A–B):<sup>34</sup>

$$n_D = \sum_{i=1}^N [\varphi_i (n_{Di})] \quad (4)$$

Newton (New):<sup>36</sup>

$$n_D^2 - 1 = \sum_{i=1}^N [\varphi_i (n_{Di}^2 - 1)] \quad (5)$$

Oster (Ost):<sup>37</sup>

$$\frac{(n_D^2 - 1)(2n_D^2 + 1)}{n_D^2} = \sum_{i=1}^N \left[ \varphi_i \left( \frac{(n_{Di}^2 - 1)(2n_{Di}^2 + 1)}{n_{Di}^2} \right) \right] \quad (6)$$

In the above equations,  $\varphi_i$  represents the volume fraction of the component in the mixture:

$$\varphi_i = \frac{x_i V_i}{\sum_{i=1}^N x_i V_i} \quad (7)$$

where  $V_i$  is the molar volume of component  $i$ , and  $x_i$  is its mole fraction.

The first type of equation (Model I), establishing the relation between the  $V^E$  and  $n_D$  data of the mixtures is based on the employment of a mass fraction-based mixing function for the specific refraction  $f(n_D)/\rho$ :

$$\frac{f(n_D)}{\rho} = \sum_{i=1}^2 \omega_i \frac{f(n_{Di})}{\rho_i} \quad (8)$$

and the definition of the excess molar volume (Model I) could be written as:

$$V^E = \sum_{i=1}^2 \left[ (f(n_{Di}) - f(n_D)) \left( \frac{x_i V_i}{f(n_D)} \right) \right] \quad (9)$$

where  $w_i$ ,  $\rho_i$ ,  $n_{Di}$  and  $x_i$  are the mass fraction, density, refractive index, and mole fraction of component  $i$ , respectively,  $f(n_{Di})$  and  $f(n_D)$  represent functions of the refractive index of pure component  $i$  and of a mixture, respectively, and  $V_i$  is the pre-mixing molar volume of component  $i$ .

Typical  $f(n_D)$  equations for the refractive index calculations are given by Eqs. (1)–(6).

Considering  $V^E$  in Eq. (9) as a function of  $n_D$  and expanding to the first order at  $n_{D\varphi} = n_{D1}\varphi_1 + n_{D2}\varphi_2$  provides the second type of equation (Model II) for the calculation of the excess molar volume:

$$V^E = \sum_{i=1}^2 x_i V_i \left( \frac{f(n_{Di})}{f(n_{D\varphi})} - 1 \right) - \Delta_\varphi n_D \left[ \frac{f'(n_{D\varphi})}{f^2(n_{D\varphi})} \sum_{i=1}^2 x_i V_i f(n_{Di}) \right] \quad (10)$$

where  $f(n_{D\varphi})$  is the function of the refractive index of a mixture as in Eq. (9),  $f'(n_{D\varphi})$  denotes the value of the first derivative of  $f(n_{D\varphi})$  and  $\Delta_\varphi n_D$  represents the deviation of the refractive index values of a binary mixture from the ideal binary mixture

$$\Delta_\varphi n_D = n_D - (n_{D1}\varphi_1 + n_{D2}\varphi_2) \quad (11)$$

where  $n_{D1}$  and  $n_{D2}$  are the refractive indices, and  $\varphi_1$  and  $\varphi_2$  are the components volume fractions based on the molar volumes of the pure components.

The third type of equation (Model III) is related to iso-refractive mixtures; when  $n_{D1} = n_{D2}$ , Eq. (10) is reduced to:

$$V^E = -\Delta_\phi n_D \left[ \frac{f'(n_{D1})}{f^2(n_{D1})} \sum_{i=1}^2 x_i V_i \right] \quad (12)$$

where  $f(n_{D1})$  represents the function of the refractive index and  $f'(n_{D1})$  denotes the value of the  $f(n_{D1})$  derivative, as in the above equations.

#### RESULTS AND DISCUSSION

The predictions of  $V^E$  from  $n_D$  data in the temperature range 288.15–323.15 K (or 288.15–333.15 K) and at atmospheric pressure for 22 binary systems (the experimental data were taken from the literature<sup>38–42</sup> and from ongoing investigations) were realised using the three chosen Models I–III with incorporated L–L, D–G, Eyk, A–B, New and Ost mixing rules for the determination the refractive index. The results of  $V^E$  calculation were assessed by the absolute maximum percentage average deviation  $PD_{\max}$ :

$$PD_{\max} = \frac{100}{N} \sum_{i=1}^N \left| \frac{V_{\text{exp}}^E - V_{\text{cal}}^E}{(V_{\text{exp}}^E)_{\max}} \right|_i \quad (13)$$

where  $V_{\text{exp}}^E$  and  $V_{\text{cal}}^E$  represent the experimental and calculated  $V^E$  values, respectively,  $N$  is the number of experimental data points, while  $(V_{\text{exp}}^E)_{\max}$  denotes the absolute maximum of the experimental  $V^E$  value.

The results are summarized in Table S-1 of the Supplementary material to this paper and are graphically presented in Figs. 1–4.

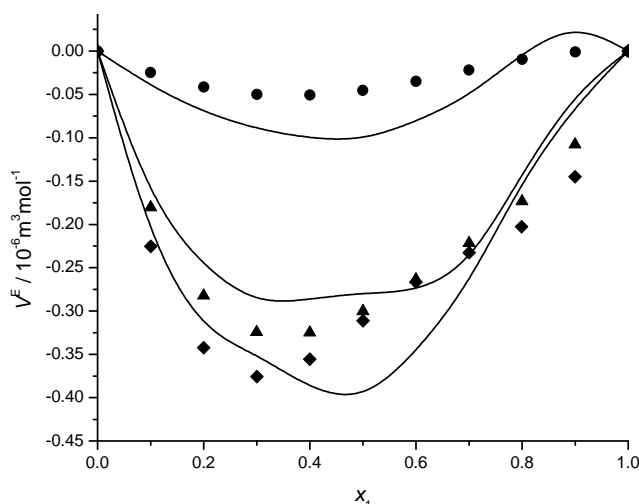


Fig. 1. Graphical presentation of the results obtained for the prediction of  $V^E$  from  $n_D$  at 298.15 K for the systems 1-propanol with: acetone (●), pyridine (▲) and NMP (◆), in which the symbols represent the experimental values and lines the predictions obtained using the appropriate Eykman mixing rule in combination with the Model I.

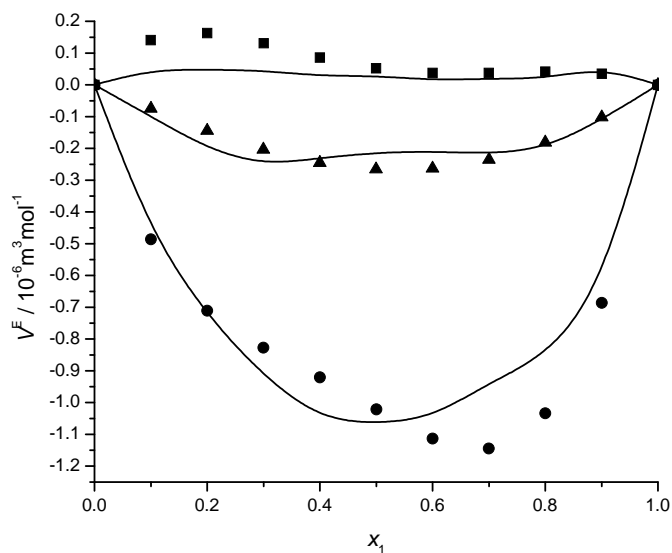


Fig. 2. Graphical presentation of the results obtained for the prediction of  $V^E$  from  $n_D$  at 298.15 K for the systems 1,2-propanediol with: (■) butyl lactate, (▲) pyridine and (●) acetone, in which the symbols represent the experimental values and the lines the predictions obtained using the appropriate Dale–Gladstone mixing rule in combination with the Model II.

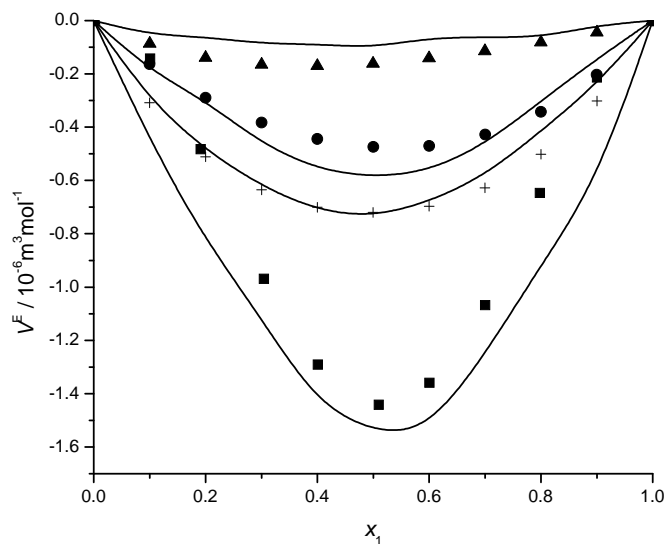


Fig. 3. Graphical presentation of the results obtained for the prediction of  $V^E$  from  $n_D$  at 313.15 K for the systems 1,3-propanediol with: (+) PEG200, (▲) pyridine, (■) butyl lactate and (●) acetone, in which the symbols represent the experimental values and lines the predictions obtained using the appropriate Newton mixing rule in combination with the Model III.

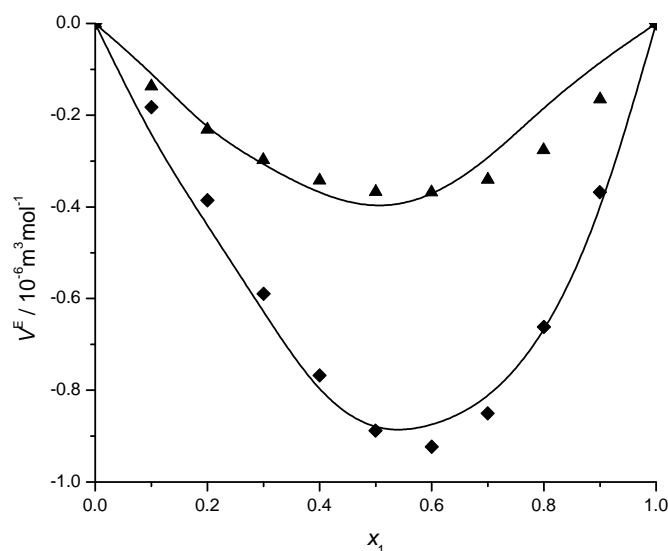


Fig. 4. Graphical presentation of the results obtained for the prediction of  $V^E$  from  $n_D$  at 323.15 K for the systems glycerol with: NMP (♦) and pyridine (▲), in which the symbols represent the experimental values and lines the predictions obtained using the appropriate Lorentz–Lorenz mixing rule in combination with the Model III.

For easier analysis, the investigated systems are classified in six groups of binary mixtures: acetone + alcohols, butyl lactate + alcohols, *N*-methyl-2-pyrrolidone + alcohols, PEGs + alcohols, pyridine + alcohols and PEGs + aromatics.

#### Acetone + alcohols

Three binary systems were investigated in this group. For the acetone+1-propanol binary mixture (Fig. 1) both  $V^E-x_1$  and  $\Delta n_D-x_1$  were S-shaped with the difference that the excess molar volume changed from negative to positive in the acetone rich region while the refractive index deviation was positive for lower and negative for higher acetone mole fractions.<sup>38</sup> For this system, it was observed (Table S-I of the Supplementary material) that the mixing rule influenced the quality of the results more than the chosen excess molar volume equation did. The best, but still unsatisfactory, fit of the experimental data was obtained using the Arago–Biot mixing rule with maximum percentage deviations ranging from 18 to 42 %, depending on the temperature. The reason for such poor prediction results could be the very small absolute  $V^E$  values and sigmoid shape of the  $V^E-x_1$  and  $\Delta n_D-x_1$  curves.

For the remaining two systems in this group, the changes in the refractive indices were positive, while the excess molar volumes were negative over the whole temperature and composition ranges. The best results for the acetone+1,2-propanediol binary mixture (Fig. 2), with values of  $PD_{\max}$  below 10 % at

all temperatures, were obtained with Model II in combination with the Dale–Gladstone mixing rule and with Model I in combination with the Newton mixing rule. For the acetone+1,3-propanediol mixture (Fig. 3), Model I in combination with the Oster mixing rule gave the best prediction results.

#### *Butyl lactate+alcohols*

The binary systems analyzed in this group were butyl lactate+1-propanol, butyl lactate+1,2-propanediol and butyl lactate+1,3-propanediol.<sup>39</sup>

For the butyl lactate+1-propanol mixture, similar results were obtained with all three types of equations and different mixing rules with the exception of the Arago–Biot mixing rule, which again gave higher values of  $PD_{\max}$ . None of the relations considered could be generally emphasized as superior since the prediction results varied with temperature. At lower temperatures, the best results were obtained using the Newton mixing rule in combination with Model II and at higher temperatures, the most successful was the Lorentz–Lorenz mixing rule coupled with Model I. The butyl lactate+1,2-propanediol binary mixture (Fig. 2) is characterized with small absolute  $V^E$  values and S-shaped  $V^E$  vs. composition curves, which is probably the reason for unsatisfactory results obtained with all applied mixing rules and models for the calculation of  $V^E$ . The possible explanation of the better results achieved for the butyl lactate+1,3-propanediol mixture (Fig. 3) in comparison with the other systems from this group might be because this system yielded the most symmetric curve  $V^E-x_1$  and positive values of both the excess molar volumes and the changes in the refractive indices. The best fit of the experimental data was obtained with the Newton mixing rule coupled with Model III or, at higher temperatures, with Model I.

#### *N-Methyl-2-pyrrolidone+ alcohols*

The systems consisting of *N*-methyl-2-pyrrolidone with mono-, di- and tri-hydroxyl alcohols, from ongoing research, showed negative excess molar volume data and positive experimental changes in the refractive indices with relatively symmetric  $\Delta n_D$  curves. For systems with ethanol or 1,3-propanediol, none of the applied mixing rules and equations for the calculation of  $V^E$  gave satisfactory prediction results. Better fits of the experimental data for the system *N*-methyl-2-pyrrolidone+1-propanol (Fig. 1) were obtained with the Dale–Gladstone mixing rule in combination with Model III. For system with glycerol, the Lorentz–Lorenz mixing rule in combination with Model III showed the best prediction capability at all the investigated temperatures.

#### *PEGs+alcohols*

Experimental changes of the refractive indices were positive for all systems in this group<sup>41</sup>, while the  $V^E-x_1$  curves were negative for the systems with PEG 400 and S-shaped with very small absolute values of excess molar volume for



systems with PEG 200. The prediction of  $V^E$  from the refractive index data was unsatisfactory irrespective of the excess molar volume equation and the mixing rule applied, where for the systems PEGs+1,2-propanediol,  $PD_{\max}$  had the highest values, for all models above 100 %.

#### *Pyridine+alcohols*

Systems containing pyridine<sup>40</sup> showed negative excess molar volume data and positive experimental changes in the refractive indices with relatively symmetric  $\Delta n_D$  curves. In the case of the pyridine+1-propanol mixture (Fig. 1), the best results at all temperatures were achieved with the Eykman mixing rule in combination with Model I or III. At some temperatures, a relatively good fit of the experimental data was also obtained with the Dale–Gladstone mixing rule coupled with Model I or the Newton mixing rule and Model II. For the remaining binary systems in this group, the Eykman mixing rule usually coupled with Model II in general gave somewhat better results than the other equations and mixing rules. In some cases, the Dale–Gladstone and the Lorentz–Lorenz mixing rule also showed satisfactory fits of the experimental data. Similar to the previous groups, the obtained results were more influenced by the mixing rule than by the equation for the calculation of the excess molar volume.

#### *PEGs+aromatics*

In this group, four systems consisting of benzene or toluene with PEG 200 or PEG 400 were included.<sup>42</sup> All of the analyzed systems showed negative excess molar volumes  $V^E$  and negative  $\Delta n_D$  values. For benzene+PEG 200 binary mixture, all the mixing rules and equations for the calculation of the excess molar volume showed unsatisfactory fits of the experimental data. In the case of benzene+PEG 400 and toluene+PEG 200 mixtures the best results at most of the investigated temperatures were obtained with Newton mixing rule coupled with Model II. At higher temperatures, satisfactory prediction results for both systems were also achieved with the Dale–Gladstone mixing rule in combination with Model I. For the toluene+PEG 400 binary system, Dale–Gladstone mixing rule in combination with Model II or III and the Eykman mixing rule coupled with Model I or III showed the best prediction capabilities, especially at higher temperatures.

Graphical presentations of the results obtained at 298.15 K for the systems with 1-propanol using the Eykman mixing rule and Model I are shown in Fig. 1. It could be seen that satisfactory fits of the experimental data were not achieved for any of the presented systems. A slightly better result than the presented ones was obtained only for the *N*-methyl-2-pyrrolidone+1-propanol mixture using the Dale–Gladstone mixing rule.

Results obtained at 298.15 K for the systems with 1,2-propanediol using the Dale–Gladstone mixing rule and Model II are given at Fig. 2. Both PEGs (PEG 200 and PEG 400) showed very poor prediction results and were omitted from Fig. 2. From the remaining three systems, the best result was obtained for the acetone+1,2-propanediol mixture with values of the percentage deviation  $PD_{\max}$  of around 7 %.

The values of the excess molar volume for the systems with 1,3-propanediol at 313.15 K, calculated from the experimental data and using the Newton mixing rule coupled with Model III are presented in Fig. 3. The best result in this case was obtained for system with butyl lactate with a value of the percentage deviation  $PD_{\max}$  of below 5 %. For the pyridine+1,3-propanediol mixture, a better fit of the experimental data than the one presented was achieved with the Eykman mixing rule, while for the acetone+1,3-propanediol binary system, the Oster mixing rule was the most appropriate.

The results presented in Fig. 4 for the systems with glycerol, obtained at 323.15 K using the Lorentz–Lorenz mixing rule in combination with Model III show that quite good fits of the experimental data were achieved for both analyzed systems.

#### CONCLUSIONS

Conclusions reached in a previous paper<sup>43</sup> were confirmed by the investigations presented in this work:

- 1) unsatisfactory fit of experimental data was achieved for systems exhibiting asymmetric or S-shaped experimental  $V^E$  vs. composition curves and for systems with very small absolute  $V^E$  values;
- 2) the obtained results were more influenced by the mixing rule employed for refractive index determination than by the model used for excess molar volume calculation;
- 3) although none of the relations considered could be generally considered as being superior, the Arago–Biot mixing rule offered the worst experimental data fit for most of the investigated systems.

#### SUPPLEMENTARY MATERIAL

Results, of the  $V^E$  prediction of the binary liquid mixtures are available electronically from <http://www.shd.org.rs/JSCS/>, or from the corresponding author on request.

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## ИЗВОД

## ПРЕДВИЂАЊЕ ДОПУНСКИХ МОЛАРНИХ ЗАПРЕМИНА ИЗАБРАНИХ БИНАРНИХ СМЕША ИЗ ПОДАТАКА ЗА ИНДЕКСЕ РЕФРАКЦИЈЕ

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Предвиђање допунских моларних запремина 22 бинарне смеше које садрже различите групе органских једињења: алкохоле (етанол, 1-пропанол, 1,2-пропандиол, 1,3-пропандиол и глицерол), кетон (ацетон), естар (бутил-лактат), лактам (*N*-метил-2-пирилодон), полимере од етилен-гликола (PEG 200 и PEG 400), као и ароматична једињења (бензен, толуен и пиридин) извршено је коришћењем података о индексима рефракције, уз примену три типа модела за прорачун допунских моларних запремина, комбинованих са правилима мешања за израчунавање индекса рефракције: Lorentz–Lorenz, Dale–Gladstone, Еукман, Arago–Biot, Newton и Oster. Добијени резултати су анализирани у зависности од примењеног модела и правила мешања, као и од природе интеракција између компонената смеше.

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