Study and analysis of various mixing rules of refractive index for some liquid mixtures

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Abstract.

In present paper we have study and discussion about relative validity and importance of various mixing rules viz. Weiner (W), Gladstone–Dale (G–D, Lorentz–Lorentz (L–L), Arago-Biot (A-B), Heller (H) and Newton's realtion (N) with following organic binary liquid mixtures of Cetane (C), + 1-Butanol (B),+ 1-Pentanol (P),+ 1-Hexanol (H), + 1-Heptanol (HP)at (293,298,303) K over the entire mole fraction range. We have also made Comparison between various mixing rules and result has been expressed in terms of average percentage deviation.

Keywords: *Refractive index, Lorentz–Lorentz relation; Arago-Biot, Heller relation; Weiner relation, Newton's relation, Gladstone–Dale relation.*

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1. INTRODUCTION

Refractive index is an important characteristic constant of materials for transparent materials. This characteristic is fundamental to determine the changes in amplitudes of reflected and transmit light beams also. Measurement of refractive index of a mixture of liquids of different refractive indices gives information about the proportion in which they are mixed. These measurements can be used to determine the purity of liquid samples. In fact the measurement of refractive index is used as an important tool of investigation in the field of Analytical Chemistry. This study can predict the molecular rearrangement due to mixing of organic liquids. It is very well known that inside a matter there are small atomic dipoles or atomic oscillators and since refractive index is closely related with the ordering and orientations of these oscillators that is why it is expected that variation of refractive index of liquid mixtures with temperature and composition can give unmatched information about molecular rearrangement arises due to mixing. Many workers have examined the mixing rules of refractive index for pure liquids and binary liquid mixtures [1-7]. There exist the definite expansion or/and contraction of liquid due to mixing. Hence densities become changed. Due to this change in density, we find considerable variation in refractive index which was firstly examined by Laplace. Gladstone-Dale gave a formula for the determination of refractive index of a mixture using the properties of pure components [11]. There are many numbers of theoretical mixing rules out of these the rules due to Lorentz-Lorentz and Weiner are extensively used [11-14].Various empirical and semi-empirical relations have been formulated earlier and tested. Relative merits of these mixing rules have been discussed by Pandey et.al. [10]. Their exists serious drawback of these mixing rules in their inability to account for changes in volume and refractivity during mixing as they are based only on volume additivity. From above description, it is good to make discussion over these mixing rules for the mixtures taken in this study, given as follows, for which we have already report [5] the various properties such as refractive index, density, excess molar volumes etc.

Cetane (C) + 1-Butanol (B)

Cetane (C) + 1-Pentanol (P)

Cetane (C) + 1-Hexanol (H)

Cetane (C) + 1-Heptanol (HP)

2. THEORY

Lorentz-Lorentz relation (L-L) is given by

$$\frac{\left(n_m^2-1\right)}{\left(n_m^2-2\right)}=\phi_1\frac{\left(n_1^2-1\right)}{\left(n_1^2+2\right)}+\phi_2\frac{\left(n_2^2-1\right)}{\left(n_2^2+2\right)} \quad \text{........................ (1)}$$

This is most frequently used mixing rule in analysis of refractive index.

Gladstone-Dale relation (G-D) is given as

$$\left(n_m-1\right)=\phi_1\left(n_1-1\right)+\phi_2\left(n_2-1\right) \quad \text{.................... (2)}$$

Weiner relation (W) is given by

$$\frac{\left(n_m^2-n_1^2\right)}{\left(n_m^2-2n_1^2\right)}=\phi_2\frac{\left(n_2^2-n_1^2\right)}{\left(n_2^2+2n_1^2\right)} \quad \text{.................... (3)}$$

It applies to isotropic bodies of spherically symmetrical shape and proposes volume additivity.

Heller (H) equation is given by –

$$\frac{n_m-n_1}{n_1}=\frac{3}{2}\phi_2\frac{\left(n_2^2-n_1^2\right)}{\left(n_2^2+2n_1^2\right)} \quad \text{.................... (4)}$$

This relation is limiting case of Weiner’s relation.

Arago - Biot relation (A-B) is given by

$$n_m=\phi_1 n_1+\phi_2 n_2 \quad \text{.................... (5)}$$

Newton relation (N) is given by

$$\left(n_m^2-1\right)=\left(n_1^2-1\right)\phi_1+\left(n_2^2-1\right)\phi_2 \quad \text{.................... (6)}$$

In above equations n_m, n_1, n_2 respectively represents the refractive index of mixture, solvent and solute respectively ϕ_1 and ϕ_2 are the volume fractions of solvent and solute respectively.

3. Results and Discussion

Values of refractive index calculated with different mixing rule equations for system (C+B), (C+P), (C+H) and (C+HP) at all given temperature range viz. 293,298,303 K are given in table-1 to table-4 respectively and average percentage deviation for liquid mixtures under study at temperatures 293,298,303 K for all the mixing rules equations are given in Table5. If we make close observation of table1-5, we can make following discussion. First important point to be mentioned here is that all the theoretical relations for the prediction of refractive indices are in good agreement with the corresponding values of refractive index, find out experimentally for all the binary mixtures under consideration in present study. For system (C+B) (C+P) (C+H) (C+HP) all the average percentage deviation values are negative for all the temperature range under study and is very close to each other and also have very low values. Thus it may be concluded that these relations shows best result for these system, in comparison to other binary liquid mixtures. Minimum value of APD (average percentage deviation) are given by Lorenz – Lorenz (L-L) relation for system (C+B) and also APD values becomes decreases as temperature increases for this system, other relations gives no systematic dependency on temperature. For system (C+P) APD values due to L-L is very close to each other. However minimum values of APD are due to Heller's relation at 293K and at 298K it is due to A-B while at 303K for Wiener. Similar trends are obtained due to (C+H) and (C+HP) systems as a final remark we can say that all the relations for mixing rules give good results and are in good agreement. However in present study Newton's relation gives minimum values of APD for most cases.

There are number of reasons for the deviation of theoretical values from experimental one. When we make the mixture of an organic liquid then the various physical properties becomes changed and they are significantly different from the properties of the original components. As we know that very low compressibility and lack of shear rigidity are the two fundamental properties of liquid phase of matter hence we can say that liquid phase of the matter shows both type of nature as shows by gases and solids because the lack of shear rigidity and very low compressibility are the fundamental properties of gases and solids respectively. The limitation to these mixing rules theories is responsible for it. Normally it is considered that all the molecules are perfectly spherical in shape but this is not true every time. In Nomoto theory [9] it is assumed that the volume does not change on mixing of liquid mixtures, hence no interaction takes place. In the same way the assumption for the formation of ideal mixing relation is that the ratio of specific heats of the components is equal to the ratio of specific heats of ideal mixtures and the volumes are also equal, this again shows that no molecular interaction is taken in account [15]. But actually, due to the presence of various types of forces such as dispersion forces, charge transfer, dipole-dipole, hydrogen bonding, and dipole-induced dipole interactions, the interaction between molecules of liquids takes place on mixing of two liquids [8]. In this way the observed difference between theoretical values and experiment values of refractive index shows that there exists molecular interaction between the molecules in liquid mixture.

4. CONCLUSION

In the present study we have attempted to find out the relative validity and importance of various mixing rules taken here, for the prediction of refractive index of binary liquid mixture. Here we have also make discussion about the temperature dependence of these relations. With the help of above discussion it is clear that various mixing rules are interrelated. The reason for the particular relation gives very good agreement in some systems but deviates in others is due to different size and nature of molecules taken under consideration.

Table-1 Refractive index of (C+B) mixture at various temperatures by different mixing rules.

X_1	n_m	G-D	A-B	L-L	W	H	N
293K							
0.0000	1.3985	1.3985	1.3985	1.3985	1.3985	1.3984	1.3985
0.0540	1.4039	1.4040	1.4040	1.4039	1.4040	1.4039	1.4041
0.1460	1.4111	1.4111	1.4111	1.4110	1.4111	1.4110	1.4112
0.2650	1.4175	1.4176	1.4176	1.4175	1.4175	1.4175	1.4177
0.3550	1.4212	1.4212	1.4212	1.4211	1.4212	1.4212	1.4213
0.4630	1.4245	1.4246	1.4246	1.4245	1.4246	1.4246	1.4247
0.5580	1.4270	1.4270	1.4270	1.4270	1.4270	1.4270	1.4271
0.6690	1.4293	1.4293	1.4293	1.4293	1.4293	1.4293	1.4294
0.7750	1.4311	1.4311	1.4311	1.4311	1.4311	1.4311	1.4312
0.8590	1.4324	1.4324	1.4324	1.4323	1.4324	1.4324	1.4324
0.9450	1.4335	1.4335	1.4335	1.4335	1.4335	1.4335	1.4335
1.0000	1.4341	1.4341	1.4341	1.4341	1.4341	1.4341	1.4341
298K							
0.0000	1.3978	1.3978	1.3978	1.3978	1.3978	1.3977	1.3978
0.0540	1.4032	1.4032	1.4032	1.4032	1.4032	1.4031	1.4033
0.1460	1.4102	1.4103	1.4103	1.4102	1.4102	1.4102	1.4104
0.2650	1.4168	1.4167	1.4167	1.4166	1.4167	1.4167	1.4168
0.3550	1.4203	1.4203	1.4203	1.4202	1.4203	1.4203	1.4204
0.4630	1.4237	1.4237	1.4237	1.4237	1.4237	1.4237	1.4238
0.5580	1.4261	1.4261	1.4261	1.4261	1.4261	1.4261	1.4262
0.6690	1.4285	1.4284	1.4284	1.4284	1.4284	1.4284	1.4285
0.7750	1.4302	1.4302	1.4302	1.4302	1.4302	1.4302	1.4303
0.8590	1.4315	1.4315	1.4315	1.4314	1.4315	1.4315	1.4315
0.9450	1.4324	1.4324	1.4324	1.4324	1.4324	1.4324	1.4324
1.0000	1.4332	1.4332	1.4332	1.4332	1.4332	1.4332	1.4332
303K							
0.0000	1.3971	1.3971	1.3971	1.3971	1.3971	1.397	1.3971
0.0540	1.4024	1.4025	1.4025	1.4024	1.4025	1.4024	1.4025
0.1460	1.4094	1.4095	1.4095	1.4094	1.4094	1.4094	1.4096
0.2650	1.4158	1.4158	1.4158	1.4157	1.4158	1.4158	1.4159
0.3550	1.4194	1.4194	1.4194	1.4193	1.4194	1.4194	1.4195
0.4630	1.4227	1.4228	1.4228	1.4227	1.4227	1.4227	1.4229
0.5580	1.4251	1.4251	1.4251	1.4251	1.4251	1.4251	1.4252
0.6690	1.4274	1.4274	1.4274	1.4274	1.4274	1.4274	1.4275
0.7750	1.4291	1.4292	1.4292	1.4291	1.4292	1.4292	1.4292
0.8590	1.4304	1.4304	1.4304	1.4304	1.4304	1.4304	1.4304
0.9450	1.4315	1.4315	1.4315	1.4315	1.4315	1.4315	1.4315
1.0000	1.4321	1.4321	1.4321	1.4321	1.4321	1.4321	1.4321

Table-2 Refractive index of (C+P) mixture at various temperatures by different mixing rules.

X_1	n_m	G-D	A-B	L-L	W	H	N
293K							
0.0000	1.4100	1.4100	1.4100	1.4100	1.4100	1.4099	1.4100
0.0683	1.4140	1.4140	1.4140	1.4140	1.4140	1.4139	1.4140
0.1693	1.4186	1.4186	1.4186	1.4185	1.4186	1.4185	1.4186
0.2483	1.4213	1.4214	1.4214	1.4213	1.4214	1.4213	1.4214
0.3648	1.4246	1.4247	1.4247	1.4246	1.4246	1.4246	1.4247
0.4519	1.4266	1.4266	1.4266	1.4266	1.4266	1.4266	1.4267
0.5739	1.4289	1.4289	1.4289	1.4289	1.4289	1.4289	1.4289
0.6508	1.4301	1.4301	1.4301	1.4301	1.4301	1.4301	1.4301
0.7492	1.4314	1.4314	1.4314	1.4314	1.4314	1.4314	1.4315
0.8734	1.4329	1.4329	1.4329	1.4329	1.4329	1.4329	1.4329
0.9622	1.4338	1.4338	1.4338	1.4338	1.4338	1.4338	1.4338
1.0000	1.4341	1.4341	1.4341	1.4341	1.4341	1.4341	1.4341
298K							
0.0000	1.4081	1.4081	1.4081	1.4081	1.4081	1.408	1.4081
0.0683	1.4122	1.4122	1.4122	1.4122	1.4122	1.4122	1.4123
0.1693	1.4170	1.4170	1.4170	1.4170	1.4170	1.4170	1.4171
0.2483	1.4199	1.4199	1.4199	1.4199	1.4199	1.4199	1.4200
0.3648	1.4233	1.4234	1.4234	1.4233	1.4234	1.4233	1.4234
0.4519	1.4254	1.4254	1.4254	1.4254	1.4254	1.4254	1.4255
0.5739	1.4278	1.4278	1.4278	1.4278	1.4278	1.4278	1.4278
0.6508	1.4290	1.4290	1.429	1.4290	1.4290	1.4290	1.4291
0.7492	1.4304	1.4304	1.4304	1.4304	1.4304	1.4304	1.4305
0.8734	1.4319	1.4319	1.4319	1.4319	1.4319	1.4319	1.4319
0.9622	1.4328	1.4328	1.4328	1.4328	1.4328	1.4328	1.4328
1.0000	1.4332	1.4332	1.4332	1.4332	1.4332	1.4332	1.4332
303K							
0.0000	1.4069	1.4069	1.4069	1.4069	1.4069	1.4068	1.4069
0.0683	1.4111	1.4111	1.4111	1.4110	1.4110	1.4110	1.4111
0.1693	1.4158	1.4159	1.4159	1.4158	1.4158	1.4158	1.4159
0.2483	1.4188	1.4188	1.4188	1.4187	1.4188	1.4188	1.4189
0.3648	1.4222	1.4222	1.4222	1.4222	1.4222	1.4222	1.4223
0.4519	1.4243	1.4243	1.4243	1.4243	1.4243	1.4243	1.4244
0.5739	1.4266	1.4267	1.4267	1.4266	1.4267	1.4267	1.4267
0.6508	1.4279	1.4279	1.4279	1.4279	1.4279	1.4279	1.4280
0.7492	1.4293	1.4293	1.4293	1.4293	1.4293	1.4293	1.4293
0.8734	1.4308	1.4308	1.4308	1.4308	1.4308	1.4308	1.4308
0.9622	1.4317	1.4317	1.4317	1.4317	1.4317	1.4317	1.4317
1.0000	1.4321	1.4321	1.4321	1.4321	1.4321	1.4321	1.4321

Table-3 Refractive index of (C+H) mixture at various temperatures by different mixing rules.

X_1	n_m	G-D	A-B	L-L	W	H	N
293K							
0.0000	1.4175	1.4175	1.4175	1.4175	1.4175	1.4175	1.4175
0.0711	1.4200	1.4200	1.4200	1.4200	1.4200	1.4200	1.4200
0.1735	1.4229	1.4230	1.4230	1.4229	1.4229	1.4229	1.4230
0.2592	1.4249	1.4250	1.4250	1.4249	1.4250	1.4249	1.4250
0.3430	1.4266	1.4266	1.4266	1.4266	1.4266	1.4266	1.4266
0.4467	1.4283	1.4283	1.4283	1.4283	1.4283	1.4283	1.4284
0.5741	1.4301	1.4301	1.4301	1.4301	1.4301	1.4301	1.4301
0.6507	1.4310	1.4310	1.4310	1.4310	1.4310	1.4310	1.4310
0.7493	1.4320	1.4320	1.4320	1.4320	1.4320	1.4320	1.4320
0.8527	1.4330	1.4330	1.4330	1.4329	1.4330	1.4330	1.4330
0.9311	1.4336	1.4336	1.4336	1.4336	1.4336	1.4336	1.4336
1.0000	1.4341	1.4341	1.4341	1.4341	1.4341	1.4341	1.4341
298K							
0.0000	1.4163	1.4163	1.4163	1.4163	1.4163	1.4163	1.4163
0.0711	1.4189	1.4189	1.4189	1.4188	1.4189	1.4188	1.4189
0.1735	1.4218	1.4219	1.4219	1.4218	1.4218	1.4218	1.4219
0.2592	1.4239	1.4239	1.4239	1.4239	1.4239	1.4239	1.4239
0.3430	1.4256	1.4256	1.4256	1.4256	1.4256	1.4256	1.4256
0.4467	1.4273	1.4273	1.4273	1.4273	1.4273	1.4273	1.4274
0.5741	1.4291	1.4291	1.4291	1.4291	1.4291	1.4291	1.4291
0.6507	1.4300	1.4300	1.4300	1.4300	1.4300	1.4300	1.4301
0.7493	1.4311	1.4311	1.4311	1.4311	1.4311	1.4311	1.4311
0.8527	1.4320	1.4320	1.4320	1.4320	1.4320	1.4320	1.4320
0.9311	1.4327	1.4327	1.4327	1.4327	1.4327	1.4327	1.4327
1.0000	1.4332	1.4332	1.4332	1.4332	1.4332	1.4332	1.4332
303K							
0.0000	1.4149	1.4149	1.4149	1.4149	1.4149	1.4149	1.4149
0.0711	1.4175	1.4175	1.4175	1.4175	1.4175	1.4175	1.4175
0.1735	1.4205	1.4206	1.4206	1.4205	1.4205	1.4205	1.4206
0.2592	1.4227	1.4226	1.4226	1.4226	1.4226	1.4226	1.4227
0.3430	1.4243	1.4243	1.4243	1.4243	1.4243	1.4243	1.4244
0.4467	1.4261	1.4261	1.4261	1.4261	1.4261	1.4261	1.4262
0.5741	1.4279	1.4279	1.4279	1.4279	1.4279	1.4279	1.4280
0.6507	1.4289	1.4289	1.4289	1.4289	1.4289	1.4289	1.4289
0.7493	1.4299	1.4299	1.4299	1.4299	1.4299	1.4299	1.4300
0.8527	1.4309	1.4309	1.4309	1.4309	1.4309	1.4309	1.4309
0.9311	1.4316	1.4316	1.4316	1.4316	1.4316	1.4316	1.4316
1.0000	1.4321	1.4321	1.4321	1.4321	1.4321	1.4321	1.4321

Table-4 Refractive index of (C+HP) mixture at various temperatures by different mixing rules.

X_1	n_m	G-D	A-B	L-L	W	H	N
293K							
0.0000	1.4175	1.4175	1.4175	1.4175	1.4175	1.4175	1.4175
0.0711	1.4200	1.4200	1.4200	1.4200	1.4200	1.4200	1.4200
0.1735	1.4229	1.4230	1.4230	1.4229	1.4229	1.4229	1.4230
0.2592	1.4249	1.4250	1.4250	1.4249	1.4250	1.4249	1.4250
0.3430	1.4266	1.4266	1.4266	1.4266	1.4266	1.4266	1.4266
0.4467	1.4283	1.4283	1.4283	1.4283	1.4283	1.4283	1.4284
0.5741	1.4301	1.4301	1.4301	1.4301	1.4301	1.4301	1.4301
0.6507	1.4310	1.4310	1.4310	1.4310	1.4310	1.4310	1.4310
0.7493	1.4320	1.4320	1.4320	1.4320	1.4320	1.4320	1.4320
0.8527	1.4330	1.4330	1.4330	1.4329	1.4330	1.4330	1.4330
0.9311	1.4336	1.4336	1.4336	1.4336	1.4336	1.4336	1.4336
1.0000	1.4341	1.4341	1.4341	1.4341	1.4341	1.4341	1.4341
298K							
0.0000	1.4163	1.4163	1.4163	1.4163	1.4163	1.4163	1.4163
0.0711	1.4189	1.4189	1.4189	1.4188	1.4189	1.4188	1.4189
0.1735	1.4218	1.4219	1.4219	1.4218	1.4218	1.4218	1.4219
0.2592	1.4239	1.4239	1.4239	1.4239	1.4239	1.4239	1.4239
0.3430	1.4256	1.4256	1.4256	1.4256	1.4256	1.4256	1.4256
0.4467	1.4273	1.4273	1.4273	1.4273	1.4273	1.4273	1.4274
0.5741	1.4291	1.4291	1.4291	1.4291	1.4291	1.4291	1.4291
0.6507	1.4300	1.4300	1.4300	1.4300	1.4300	1.4300	1.4301
0.7493	1.4311	1.4311	1.4311	1.4311	1.4311	1.4311	1.4311
0.8527	1.4320	1.432	1.4320	1.4320	1.4320	1.4320	1.4320
0.9311	1.4327	1.4327	1.4327	1.4327	1.4327	1.4327	1.4327
1.0000	1.4332	1.4332	1.4332	1.4332	1.4332	1.4332	1.4332
303K							
0.0000	1.4149	1.4149	1.4149	1.4149	1.4149	1.4149	1.4149
0.0711	1.4175	1.4175	1.4175	1.4175	1.4175	1.4175	1.4175
0.1735	1.4205	1.4206	1.4206	1.4205	1.4205	1.4205	1.4206
0.2592	1.4227	1.4226	1.4226	1.4226	1.4226	1.4226	1.4227
0.3430	1.4243	1.4243	1.4243	1.4243	1.4243	1.4243	1.4244
0.4467	1.4261	1.4261	1.4261	1.4261	1.4261	1.4261	1.4262
0.5741	1.4279	1.4279	1.4279	1.4279	1.4279	1.4279	1.4280
0.6507	1.4289	1.4289	1.4289	1.4289	1.4289	1.4289	1.4289
0.7493	1.4299	1.4299	1.4299	1.4299	1.4299	1.4299	1.4300
0.8527	1.4309	1.4309	1.4309	1.4309	1.4309	1.4309	1.4309
0.9311	1.4316	1.4316	1.4316	1.4316	1.4316	1.4316	1.4316
1.0000	1.4321	1.4321	1.4321	1.4321	1.4321	1.4321	1.4321

Table-5 Values of APD for various mixtures

Mixture	T (K)	GD	AB	LL	W	H	N
C+B	293	-0.0001	0.0001	-0.0002	-0.0001	-0.0004	-0.0001
	298	-0.0003	-0.0034	-0.0012	-0.0023	-0.0012	-0.0005
	303	-0.006	-0.0074	-0.0002	-0.0045	-0.0003	-0.002
C+P	293	0.0012	-0.0013	-0.0004	0.0034	-0.0027	-0.006
	298	0.0023	0.0012	-0.0012	-0.0003	-0.0012	0.0002
	303	-0.0012	-0.0012	-0.0053	0.0023	-0.0034	-0.004
C+H	293	-0.0045	-0.0023	-0.0034	-0.0043	-0.0001	-0.0073
	298	-0.0024	-0.0012	-0.0023	-0.0021	-0.003	-0.0033
	303	-0.0003	-0.0014	-0.0024	-0.0026	-0.0024	-0.0032
C+HP	293	-0.0034	-0.0003	0.0023	0.0002	0.0003	0.0063
	298	-0.0002	-0.0012	-0.0023	-0.0023	-0.00343	-0.0038
	303	-0.0002	-0.0002	-0.0004	-0.0023	-0.0024	-0.0032

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