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PREDICTION OF MOLECULAR INTERACTIONS BASED ON THEORETICAL EVALUATION OF ULTRASONIC VELOCITY AND EXCESS ACOUSTICAL PARAMETERS FROM 288.15- 318.15K

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Abstract-Ultrasonic velocity was computed from collision factor theory (CFT), Free length theory (FLT), Nomoto (NOM) and Van dael (VAN) liquid state model for 2-butanol and dodecane from 288.15-318.15K over the whole composition range and atmospheric pressure and compared with the literature values. Excess isentropic compressibility (β_s^E) and Excess acoustic impedance (Z^E) were also calculated for aforesaid binary system at different temperatures. Average absolute % deviation was the criteria of the success of these liquid state models. R^2 values were also calculated to test the accuracy and applicability of aforementioned liquid state models at different temperatures. Degree of molecular interactions (α) were computed from ideal mixing relation to determine the extent of molecular association between the binary components. Collision factor theory deals a fair agreement with experimental findings in comparison to other liquid state models.

Keywords- Free length theory, CFT, Van dael, Nomoto, molecular interaction.

I. INTRODUCTION

In recent years ultrasonic velocity has become a subject of deep interest in the determination of various thermodynamic and acoustical properties of pure liquid and liquid mixture. Which plays a significant role in the development of various liquid state models and to analyze the molecular interactions present in the liquid mixture. The variation of ultrasonic velocity with concentration of binary mixture provides a very fruitful information regarding the molecular interactions. An exhaustive literature survey reveals that various researcher [1-4] have evaluated ultrasonic velocity with help of collision factor theory (CFT), free length theory (FLT), Nomoto and Van dael ideal mixing relation and compared with measured values at different temperatures and predict the molecular interactions. In the continuation of previously published work

[5], this paper is concerned with the theoretical evaluation of ultrasonic velocity and excess acoustical parameters for binary liquid mixture of 2-butanol with dodecane by aforementioned liquid state models such as collision factor theory [6-7] based on space filling factor, Jacobson's Free length theory [8] which relates intermolecular free length and ultrasonic velocity, Nomoto relation [9] based on linear dependence of molar sound velocity and ideal volume of mixing and Van dael [10] ideal mixing relation based on adiabatic compressibility, volume fraction and ratio of specific heat. The theoretical evaluated values of ultrasonic velocity were compared and tested with the measured work of J. Peleterio [11]. Excess isentropic compressibility (β_s^E) and Excess acoustic impedance (Z^E) were also calculated by theoretically evaluated ultrasonic velocity at different temperatures. Extent of molecular interaction was determined by ideal mixing relation over the entire composition range from temperature 288.15-318.15K. Average absolute % deviation (AAPD) computed for all the mentioned liquid state models was the criteria of the success of results. The main aim of this work was to evaluate the ultrasonic velocity and their excess acoustical parameters with the help of above-mentioned liquid state models for weakly interacting liquids and predict the molecular interactions of binary liquid components at different temperatures.

II. MODELING

A. Collision Factor theory (CFT)

Schaaf's [6] derived a relation between ultrasonic velocity and space filling factor using collision factor and U_∞ in pure liquid is given below:

$$U = \left(\frac{B}{V_m}\right) U_\infty S \quad (1)$$

Where $\left(\frac{B}{V_m}\right)$ is space filling factor and V_m is molar volume.



Nutsch-Kuhkies [7] modified the above Scharff's equation of ultrasonic velocity for binary system as

$$U_{CFT} = U_{\infty} \left\{ \sum_{i=1}^2 X_i S_i \right\} \left\{ \frac{X_1 B_1 + X_2 B_2}{V_m} \right\} \quad (2)$$

S is collision factor, $U_{\infty} = 1600$ m/s

B. Free Length Theory (FLT)

Jacobson [8] theoretical model which relates intermolecular free length (L_f), density and ultrasonic velocity

$$U = \frac{K}{(L_f \rho^{1/2})} \quad (3)$$

Where K is temperature dependent constant.

$$L_f = \left\{ \frac{2V_a}{Y} \right\} \quad (4)$$

Where Y is surface area of molecule, $V_a = (V_m - V_0)$ is available volume, V_m and V_0 are molar volume of pure liquid at experimental temperature and absolute zero temperature respectively.

Put the value of L_f in equation (3)

$$U = \left\{ \frac{KY}{(2V_a \rho^{1/2})} \right\} \quad (5)$$

For binary system equation (5) can be written as:

$$U_{FLT} = \left[\frac{K(X_1 Y_1 + X_2 Y_2)}{2\{V_{M,T} - (X_1 V_1^0 + X_2 V_2^0)\} \rho_{Mix}^2} \right] \quad (6)$$

Where $V_{M,T}$, T is molar volume of binary system can be calculated by the following relation:

$$V_{M,T} = \frac{(X_1 M_1 + X_2 M_2)}{\rho_{Mix}} \quad (7)$$

Where M_1 and M_2 are molecular weight of pure liquids.

C. Nomoto relation

Nomoto [9] proposed an empirical relation, which was used to determine the ultrasonic velocity of liquid mixtures by assuming the linear dependence of molar sound velocity (R) and ideal volume of mixing.

$$U^{NOMO} = \left\{ \left(\sum_{i=1}^2 \frac{X_i R_i}{X_i V_i} \right)^3 \right\} \quad (8)$$

Where R is molar sound velocity can be determined by the following relation:

$$R = (M_i / \rho_i) U^{1/3} \quad (9)$$

M_i and ρ_i are molecular weight and density of pure liquid component.

D. VAN DEAL RELATION

Van deal [10] proposed an empirical relation to determine the ultrasonic velocity in ideal system, based on adiabatic compressibility, volume fraction (Φ_i) and ratio of specific heat (γ_i). If binary system is ideal and $\gamma_i = \gamma_{IM}$ and $V_1 = V_2$ than ultrasonic velocity can be determined the following relation:

$$U^{VAN} = \left\{ \left(\sum_{i=1}^2 \frac{1}{\sqrt{X_i M_i}} \right) \left(\sum_{i=1}^2 \left(\frac{X_i}{M_i U_i} \right)^{-1/2} \right) \right\} \quad (10)$$

Where X_i is the mole fraction of individual component.

III. RESULT AND DISCUSSION

Table1 represents density of binary liquid mixture, experimental and theoretical ultrasonic velocity along with their percentage deviation calculated from collision factor theory (CFT), free length theory (FLT), Nomoto and Van dael ideal mixing relation at different temperatures. Excess isentropic compressibility and excess acoustic impedance computed from aforementioned liquid state models over the entire composition range (0.04602-0.95177) from temperature 288.15-318.15 K are presented in table2. Degree of molecular interaction computed from ideal mixing relation over the entire composition range are presented in table 3. Table 4 represents the average absolute % deviation and R^2 values computed for various liquid state models from temperature 288.15-318. A close observation of table 1 reveals that the ultrasonic velocity calculated by CFT, FLT and Nomoto decreases linearly and density of the binary liquid mixture increases with increase in the concentration of 2-butanol for all the temperatures but ultrasonic velocity calculated by van dael ideal mixing relation decreases with increase in the concentration up to 0.59429 of 2-butanol than start increasing.

$$\beta_s = \frac{1}{\rho U^2} \quad (11)$$

$$Z = \rho \times U \quad (12)$$

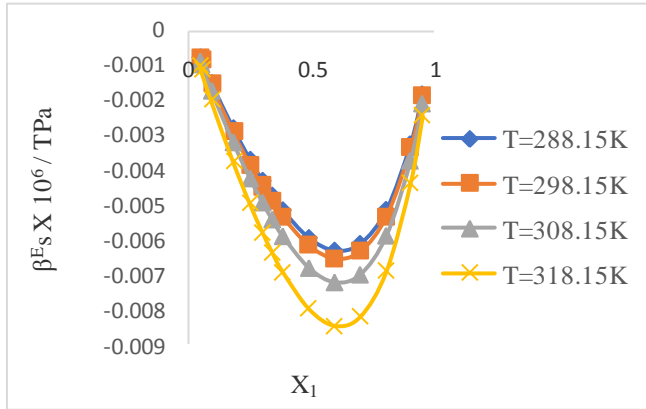


Fig. 1. Variation of β_s^E with mole fraction (X_1)

The nonlinear variation of ultrasonic velocity with increase in the concentration of 2-butanol clearly indicate the occurrence of molecular association [12] due to dipole-dipole, dipole-induced dipole interaction and dispersion forces. Similar behaviour was observed for all the temperatures. Positive % deviation in ultrasonic velocity is result of molecular association and negative % deviation indicate the molecular dissociation. Excess acoustical parameters were calculated by following relation:

$$\beta_s^E = \beta_s - \{X_1(\beta_s)_1 + X_2(\beta_s)_2\} \quad (13)$$

$$Z^E = Z - (X_1Z_1 + X_2Z_2) \quad (14)$$

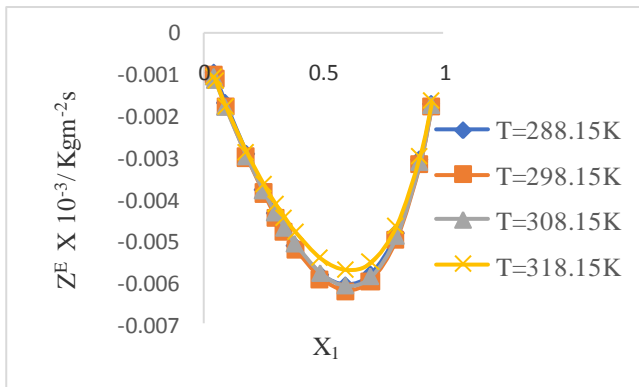


Fig. 2. Variation of Z^E with mole fraction (X_1)

A perusal of table 2 indicate that excess isentropic compressibility (β_s^E) and excess acoustic impedance (Z^E) computed by all discussed liquid state models firstly decreases than increases with increase in the concentration of 2-butanol. Similar trends were observed for all the temperatures. Negative isentropic compressibility indicate that system is highly packed [13-14]. Negative value of β_s^E increases with increase in concentration of 2-butanol up to specific concentration point than decreases with increase in concentration as shown in figure 1. Which indicate that

dipole-induce dipole type of interaction predominates over dipole-dipole interaction responsible for making system less compressible [15]. Similar trends were also observed in case of excess acoustic impedance (Z^E) for all the models from temperatures 288.15-318.15K as shown in figure 2. Which confirm the dissociation of dipolar interaction and formation of weak dipole-induced dipole interactions [16]. These results can be confirmed on the basis of degree of molecular interaction (a) presented in table 3.

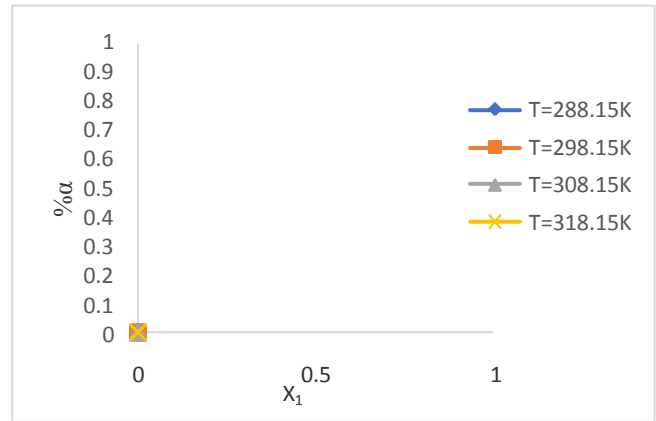


Fig. 3. Variation of % α with mole fraction (X_1)

Which indicate that molecular interaction increases with increase in the concentration of 2-butanol up to 20.74%, 20.31%, 19.99% and 19.81% at 288.15, 298.15, 308.15 and 318.15K respectively than decreases as shown in figure 3. AAPD presented in table 4 varies from (0.453-1.357), (0.976-1.346), (0.623-1.753) and (5.534-6.106) and R^2 values varies from (0.9996-1), (0.9997-0.9999), (0.9995-0.9999) and (0.9942-0.9947) for CFT, FLT, Nomoto and Van dael relation respectively at different temperatures. Increasing order of AAPD of concerning liquid state model is as follows: CFT < FLT < Nomoto < Van dael ideal mixing relation. R^2 values computed for various liquid state models verify the results obtained from AAPD. Which indicate that collision factor theory (CFT) deals a fair agreement with experimental findings in comparison to other liquid state models.

IV. CONCLUSION

Ultrasonic velocity and Excess acoustical parameters play a vital role in understanding of various kinds of interaction during mixing of liquids having similar or different behaviour. In above discussion it can be concluded that for liquids having polar and non-polar behaviour, when mixed excess acoustical parameters calculated from ultrasonic velocity by various liquid state models increases negatively with increase in mole fraction than decreases. Which confirm the dissociation of dipole-dipole interaction and formation of new dipole-induced dipole interaction. Formation of such dipole-induced dipole interaction is responsible for reducing the space



between the liquids which make the binary liquid mixture less compressible. AAPD and R^2 value shows that collision factor theory was found to be more consistent and accurate with experimental finding for all the temperatures than other liquid state models.

V. ACKNOWLEDGEMENT

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Table-1 Experimental and theoretical ultrasonic velocity from 288.15-318.15K

X_1	ρ_{MIX}	U_{Exp}	U_{CFT}	U_{FLT}	U_{NOM}	U_{VAN}	$\% \Delta U_{CFT}$	$\% \Delta U_{FLT}$	$\% \Delta U_{NOM}$	$\% \Delta U_{VAN}$
T=288.15K										
0.04602	0.75279	1312.74	1311.65	1316.45	1280.27	1289.14	0.08	-0.28	2.47	1.80
0.0507	0.75285	1312.32	1311.08	1316.35	1280.14	1286.51	0.09	-0.31	2.45	1.97
0.09287	0.75361	1309.26	1306.03	1315.26	1278.98	1264.51	0.25	-0.46	2.31	3.42
0.18128	0.75547	1302.82	1295.73	1312.66	1276.33	1227.22	0.54	-0.76	2.03	5.80
0.24985	0.75715	1297.76	1288.07	1310.36	1274.05	1205.24	0.75	-0.97	1.83	7.13
0.30044	0.75857	1294.05	1282.62	1308.46	1272.22	1192.36	0.88	-1.11	1.69	7.86
0.33974	0.75979	1291.23	1278.53	1306.85	1270.71	1184.12	0.98	-1.21	1.59	8.30
0.38271	0.76121	1288.14	1274.20	1304.98	1268.95	1176.76	1.08	-1.31	1.49	8.65
0.48939	0.76543	1280.62	1264.23	1299.57	1264.05	1165.46	1.28	-1.48	1.29	8.99
0.59429	0.77068	1273.68	1255.78	1293.01	1258.30	1163.46	1.41	-1.52	1.21	8.65
0.69831	0.77727	1267.06	1249.13	1284.98	1251.41	1170.26	1.41	-1.41	1.23	7.64
0.80469	0.78596	1260.19	1244.77	1274.65	1242.71	1186.75	1.22	-1.15	1.39	5.83
0.90245	0.79636	1253.5	1243.81	1262.61	1232.65	1211.46	0.77	-0.73	1.66	3.35
0.95177	0.80284	1250.22	1244.81	1255.27	1226.57	1227.94	0.43	-0.40	1.89	1.78
T=298.15K										
0.04602	0.74546	1273.46	1272.99	1277.54	1246.06	1251.01	0.04	-0.32	2.15	1.76
0.0507	0.74551	1273.1	1272.47	1277.45	1245.94	1248.46	0.05	-0.34	2.13	1.94
0.09287	0.74619	1269.82	1267.78	1276.46	1244.82	1227.20	0.16	-0.52	1.97	3.36
0.18128	0.74797	1263.38	1258.22	1274.02	1242.27	1191.16	0.41	-0.84	1.67	5.72
0.24985	0.74959	1258.36	1251.09	1271.85	1240.08	1169.91	0.58	-1.07	1.45	7.03
0.30044	0.75094	1254.67	1246.02	1270.08	1238.33	1157.46	0.69	-1.23	1.30	7.75
0.33974	0.75215	1251.84	1242.19	1268.53	1236.88	1149.50	0.77	-1.33	1.20	8.18
0.38271	0.75351	1248.76	1238.14	1266.77	1235.19	1142.40	0.85	-1.44	1.09	8.52
0.48939	0.75762	1241.13	1228.78	1261.63	1230.49	1131.52	0.99	-1.65	0.86	8.83
0.59429	0.76272	1234.11	1220.77	1255.42	1224.97	1129.65	1.08	-1.73	0.74	8.46
0.69831	0.7692	1227.66	1214.36	1247.73	1218.36	1136.30	1.08	-1.63	0.76	7.44
0.80469	0.7778	1221.36	1209.97	1237.80	1210.00	1152.36	0.93	-1.35	0.93	5.65
0.90245	0.78821	1215.73	1208.62	1226.13	1200.35	1176.40	0.58	-0.86	1.27	3.23
0.95177	0.79474	1213.24	1209.26	1218.99	1194.52	1192.42	0.33	-0.47	1.54	1.72
T=308.15K										
0.04602	0.73806	1234.92	1234.95	1239.31	1212.33	1213.44	0.00	-0.36	1.83	1.74
0.0507	0.73811	1234.48	1234.46	1239.23	1212.21	1210.97	0.00	-0.38	1.80	1.90
0.09287	0.73872	1231.07	1230.06	1238.31	1211.12	1190.33	0.08	-0.59	1.62	3.31



0.80469	-0.0051	-1.1737	-1.1710	-1.0915	-0.0048	-0.0283	-0.0299	-0.0739
0.90245	-0.0033	-1.1453	-1.1306	-1.1014	-0.0030	-0.0180	-0.0269	-0.0438
0.95177	-0.0018	-1.1341	-1.1100	-1.1119	-0.0017	-0.0101	-0.0247	-0.0236
				T=298.15K				
0.04602	-0.0008	-1.3479	-1.3118	-1.3186	-0.0010	-0.6543	-0.0245	-0.0208
0.0507	-0.0009	-1.3456	-1.3100	-1.3135	-0.0011	-0.6516	-0.0246	-0.0227
0.09287	-0.0016	-1.3249	-1.2938	-1.2688	-0.0018	-0.6263	-0.0254	-0.0385
0.18128	-0.0029	-1.2819	-1.2601	-1.1842	-0.0030	-0.5726	-0.0267	-0.0650
0.24985	-0.0038	-1.2492	-1.2340	-1.1268	-0.0038	-0.5305	-0.0277	-0.0803
0.30044	-0.0045	-1.2253	-1.2146	-1.0890	-0.0044	-0.4991	-0.0283	-0.0890
0.33974	-0.0049	-1.2071	-1.1997	-1.0625	-0.0048	-0.4745	-0.0286	-0.0943
0.38271	-0.0053	-1.1873	-1.1832	-1.0362	-0.0052	-0.4474	-0.0290	-0.0989
0.48939	-0.0062	-1.1400	-1.1424	-0.9832	-0.0059	-0.3787	-0.0295	-0.1045
0.59429	-0.0065	-1.0962	-1.1022	-0.9485	-0.0062	-0.3091	-0.0294	-0.1021
0.69831	-0.0063	-1.0564	-1.0622	-0.9312	-0.0060	-0.2372	-0.0286	-0.0917
0.80469	-0.0053	-1.0211	-1.0211	-0.9311	-0.0050	-0.1597	-0.0266	-0.0714
0.90245	-0.0034	-0.9951	-0.9831	-0.9469	-0.0031	-0.0834	-0.0235	-0.0423
0.95177	-0.0019	-0.9852	-0.9638	-0.9607	-0.0017	-0.0423	-0.0212	-0.0229
				T=308.15K				
0.04602	-0.0009	-1.1787	-1.1453	-1.1469	-0.0010	-0.5979	-0.0210	-0.0201
0.0507	-0.0010	-1.1765	-1.1435	-1.1416	-0.0011	-0.5953	-0.0211	-0.0220
0.09287	-0.0017	-1.1563	-1.1281	-1.0956	-0.0018	-0.5723	-0.0219	-0.0372
0.18128	-0.0032	-1.1146	-1.0958	-1.0086	-0.0030	-0.5235	-0.0232	-0.0627
0.24985	-0.0042	-1.0828	-1.0708	-0.9500	-0.0038	-0.4851	-0.0242	-0.0774
0.30044	-0.0049	-1.0598	-1.0524	-0.9118	-0.0043	-0.4564	-0.0247	-0.0858
0.33974	-0.0054	-1.0421	-1.0381	-0.8851	-0.0047	-0.4340	-0.0251	-0.0909
0.38271	-0.0059	-1.0230	-1.0224	-0.8587	-0.0051	-0.4092	-0.0254	-0.0953
0.48939	-0.0068	-0.9773	-0.9833	-0.8066	-0.0058	-0.3466	-0.0260	-0.1007
0.59429	-0.0072	-0.9351	-0.9449	-0.7740	-0.0061	-0.2831	-0.0260	-0.0985
0.69831	-0.0070	-0.8971	-0.9067	-0.7601	-0.0059	-0.2175	-0.0252	-0.0885
0.80469	-0.0059	-0.8636	-0.8675	-0.7649	-0.0049	-0.1465	-0.0232	-0.0690
0.90245	-0.0037	-0.8396	-0.8312	-0.7865	-0.0031	-0.0766	-0.0200	-0.0409
0.95177	-0.0021	-0.8307	-0.8129	-0.8038	-0.0017	-0.0389	-0.0177	-0.0221
				T=318.15K				
0.04602	-0.0010	-1.0070	-0.9769	-0.9725	-0.0010	-0.5444	-0.0176	-0.0195
0.0507	-0.0011	-1.1799	-0.9753	-0.9669	-0.0011	-0.6038	-0.0177	-0.0213
0.09287	-0.0020	-1.1528	-0.9603	-0.9189	-0.0018	-0.5803	-0.0185	-0.0360
0.18128	-0.0038	-1.0963	-0.9292	-0.8287	-0.0029	-0.5304	-0.0199	-0.0606



0.24985	-0.0050	-1.0529	-0.9051	-0.7683	-0.0036	-0.4911	-0.0207	-0.0747
0.30044	-0.0058	-1.0213	-0.8873	-0.7291	-0.0041	-0.4619	-0.0212	-0.0827
0.33974	-0.0064	-0.9968	-0.8735	-0.7019	-0.0044	-0.4389	-0.0216	-0.0877
0.38271	-0.0069	-0.9704	-0.8584	-0.6752	-0.0048	-0.4137	-0.0219	-0.0919
0.48939	-0.0080	-0.9059	-0.8207	-0.6234	-0.0054	-0.3499	-0.0224	-0.0970
0.59429	-0.0085	-0.8452	-0.7836	-0.5925	-0.0057	-0.2853	-0.0224	-0.0949
0.69831	-0.0082	-0.7886	-0.7466	-0.5817	-0.0055	-0.2188	-0.0216	-0.0853
0.80469	-0.0069	-0.7363	-0.7087	-0.5913	-0.0046	-0.1472	-0.0197	-0.0665
0.90245	-0.0044	-0.6954	-0.6737	-0.6188	-0.0029	-0.0768	-0.0166	-0.0395
0.95177	-0.0024	-0.6782	-0.6560	-0.6396	-0.0016	-0.0390	-0.0143	-0.0213

Table -3 Degree of molecular association at different temperatures

X_1	$\alpha^{288.15K}$	$\alpha^{298.15K}$	$\alpha^{308.15K}$	$\alpha^{318.15K}$
0.04602	3.70%	3.62%	3.57%	3.58%
0.0507	4.05%	3.99%	3.92%	3.93%
0.09287	7.20%	7.07%	6.96%	6.93%
0.18128	12.70%	12.49%	12.33%	12.25%
0.24985	15.94%	15.69%	15.49%	15.40%
0.30044	17.79%	17.50%	17.29%	17.17%
0.33974	18.91%	18.60%	18.37%	18.24%
0.38271	19.83%	19.49%	19.23%	19.09%
0.48939	20.74%	20.31%	19.99%	19.81%
0.59429	19.84%	19.35%	18.96%	18.71%
0.69831	17.23%	16.73%	16.30%	16.00%
0.80469	12.76%	12.33%	11.95%	11.65%
0.90245	7.06%	6.80%	6.55%	6.34%
0.95177	3.66%	3.52%	3.38%	3.25%

Table- 4 Average absolute % deviation and R^2 values at different temperatures

T/K	AAPD				R^2 values			
	U_{CFT}	U_{LFT}	U_{NOM}	U_{VAN}	$R^2(U_{CFT})$	$R^2(U_{LFT})$	$R^2(U_{NOM})$	$R^2(U_{VAN})$
288.15	0.828	0.976	1.753	6.106	0.9999	0.9999	0.9995	0.9942
298.15	0.632	1.101	1.361	5.989	0.9999	0.9998	0.9997	0.9944
308.15	0.453	1.232	0.959	5.591	1.0000	0.9998	0.9998	0.9946
318.15	1.357	1.346	0.623	5.534	0.9996	0.9997	0.9999	0.9947

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