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Ultrasonic, Volumetric and Isentropic Compressibility of Binary Mixtures of 1,4-Dioxane with Primary Alcohols at 303.15 K

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Abstract

Density, viscosity and sound velocity of six binary liquid mixtures of methanol, ethanol, propanol, butanol, hexanol and octanol with 1,4-dioxane have been measured over the entire range of composition at temperature 303.15K. From the experimental densities, viscosities and sound velocity, the excess molar volume (V^E), deviation in viscosity ($\Delta\eta$) and deviation in isentropic compressibility (ΔK_s) have been calculated. The results have been used to discuss the nature and strength of intermolecular interactions in these mixtures.

Keywords

Density, Viscosity, Sound Velocity, Excess Molar Volume, Viscosity Deviation, Isentropic Compressibility Binary Mixtures, Alcohols

1. Introduction

The present paper is part of our ongoing research on the thermodynamic properties of binary liquid mixtures [1] containing 1,4-dioxane with 1-alkanol at 303.15 K. Therefore their binary mixture properties are needed as a useful database in a variety of industrial applications [2] [3] [4] [5] [6]. Therefore, their interactions with different types of liquids such as 1,4-dioxane, methanol, ethanol, propanol, butanol, hexanol and octanol are important from a fundamental viewpoint. A wide range of important binary mixtures containing the above liquids have been studied by different authors [7] [8] [9] [10]. More over, to the best of our knowledge, no physical property data on the mixtures in the present study are available. This prompted us to undertake a study on the measurement of physical properties such as density (ρ), viscosity (η) and sound velocity (u) at 303.15 K. Using

these data, the excess molar volume (VE), deviations in viscosity ($\Delta\eta$) and deviation in isentropic compressibility (ΔK_s) were calculated. The results are graphically presented. The excess properties values have been interpreted in terms of the nature of intermolecular interactions between constituent molecules of mixtures [11]-[19].

2. Experimental

Materials and Method

The chemicals (AR grade) employed were supplied by Merck. Chem. Ltd. India, Their purities (in mass percent) were 1,4-dioxane 99%, methanol 99.27%, ethanol 99.2%, propanol 99.2%, butanol 99.5%, hexanol 99.3% and octanol 99%. All the chemicals were purified by a method given in the literature [20]. The purity of the liquids was also checked by measuring their densities, viscosities and sound velocities at 303.15 K and were in agreement with the literature values [21]-[29] are depicted in Table 1.

All six binary liquid mixtures Viz. 1,4-dioxane + methanol, 1,4-dioxane + ethanol, 1,4-dioxane + propanol, 1,4-dioxane + butanol, 1,4-dioxane + hexanol and 1,4-dioxane + octanol were studied. Binary mixtures were prepared by weight covering the entire mole fraction range. The components of binary mixtures were injected by means of syringe in to the glass vials of sealed with rubber stopper in order to check evaporation losses during sample preparation. The weight of the sample was measured using electrical single pan analytical balance (K-roy instruments Pvt. Ltd. Varanasi (U.P.) India. The densities of pure liquids and their binary mixtures were measured (303.15 K) using a single-capillary pycnometer, made of borosil glass, having a bulb capacity of 30 cm^3. The capillary, with graduated marks, had a uniform pore and could be closed by a well-fitted glass cap. The marks on the capillary were calibrated by using double-distilled water at 303.15 K. The pycnometer was kept for about 30 minute in an electronically controlled thermostate water bath (MSI Goyal Scientific Meerut) 303.15 ± 0.02 K and the position of the liquid level on the capillary was noted. The volume of the pycnometer at each mark was calculated by using the literature [30] value of the density of

Table 1. Physical properties of pure components at 303.15 K.

Component	ρ (g-m^{-3})		η (CP)		u (m-s^{-1})	
	Observed	Literature	Observed	Literature	Observed	Literature
1,4-Dioxane	1.0108	1.0229 [25]	1.0303	1.0690 [24]	1348.0	1322.3 [50]
Methanol	0.7840	0.7817 [21]	0.4949	0.5040 [22]	1084.0	1084.0 [29]
Ethanol	0.7720	0.7807 [8]	1.1399	1.3560 [8]	1141.0	1144.3 [22]
Propanol	0.8070	0.8003 [28]	1.5477	1.6626 [22]	1182.0	1182.6 [22]
Butanol	0.8040	0.8020 [22]	2.2045	2.2740 [23]	1196.0	1196.6 [22]
Hexanol	0.8128	0.8118 [22]	4.5642	4.5930 [26]	1298.0	1282.0 [45]
Octanol	0.8242	0.8187 [27]	7.8512	7.6630 [26]	1327.0	1330.8 [27]

pure water at 303.15 K. The volume these obtained is used to determine the density of the unknown liquid. The observed values of densities of pure 1,4-dioxane, methanol, ethanol, propanol, butanol, hexanol and octanol at 303.15 K were 1.0108, 0.7840, 0.7720, 0.8070, 0.8040, 0.8128 and 0.8242 $g \cdot m^{-3}$ which compare well with corresponding literature values of respectively. The ultrasonic velocities were measured using a multifrequency ultrasonic interferometer (Model F-80D, Mittal Enterprise, New Delhi) working at 3 MHz. The meter was calibrated with water and benzene at 303.15 K. The measured values of ultrasonic velocities of pure 1,4-dioxane, methanol, ethanol, propanol, butanol, hexanol and octanol at 303.15 K were 1348, 1084, 1141, 1182, 1196,1298 and 1327 $m \cdot s^{-1}$ respectively, which compare well with the corresponding literature values. The viscosity was measured by means of a suspended Ubbelohde type viscometer [31] calibrate was done at 303.15K with double distilled water and purified methanol. An electronic digital stop watch with readability of ±0.01 was used for the flow time measurements. The measured values of viscosities of pure 1,4-dioxane, methanol, ethanol, propanol, butanol, hexanol and octanol at 303.15 K were 1.0303, 0.4949, 1.1399, 1.5477, 2.2045, 4.5642 and 7.8512 C.P. which compare well with the corresponding literature values. The mixtures were prepared by mixing known volumes of the pure liquids in air tight stoppered bottles. The weights were taken on a single pan electronic balance (Mittal Enterprises New Patel Nagar, New Delhi, India) accurate to 0.01 mg.

3. Results and Discussion

The experimental values of density, viscosity, and sound velocity data for mixtures of 1,4-dioxane 1) and primary alcohols 2) such as methanol, ethanol, propanol, butanol, hexanol and octanol were used to calculate the excess molar volume, viscosity deviations and isentropic compressibility. The results are presented in **Table 2**.

The excess molar volume is calculated using the equation ([32] [33])

$$V^E = \left(M_1 x_1 + M_2 x_2\right) / \rho_{mix.}$$

where $\rho_{mix.}$ is the density of the mixture and M_1, M_2, x_1, x_2, ρ_1 and ρ_2 are the molecular weights, mole fraction and densities of pure components 1 and 2 respectively.

Quantitatively as per the absolute reaction rate theory [34] [35] the deviation of the viscosities ($\Delta\eta$) from the ideal mixture values are calculated as [36]

$$\Delta\eta = \eta_{mix.} - \left[x_1\eta_1 + x_2\eta_2\right]$$

where $\eta_{mix.}$ are the viscosity of the mixture and η_1, η_2 are the viscosity of pure components (1) and (2) respectively.

Isentropic compressibility (K_S) and excess isentropic compressibility (K_S^E) are calculated from the experiment density (ρ) and sound velocity (u) using the following equations [37] [38] [39] [40] [41]

$$K_S = 1/u^2\rho$$

$$\Delta K_S = K_S - x_1 K_{S1} - x_2 K_{S2}$$

where K_S, K_{S1} and K_{S2} are the isentropic compressibility of the mixture, pure component 1 and pure component 2 respectively.

We have calculated excess viscosity, excess molar volume and excess isentropic compressibility at 303.15 K for the binary mixture of 1,4-dioxane (1) with the primary alcohols (2). The variation of the excess properties over the entire range of compositions for the binary mixtures is depicted in Figures 1-3.

The value of excess molar volume was found to be negative value for 1,4-dioxane with methanol, ethanol and the positive value increase with increasing chain

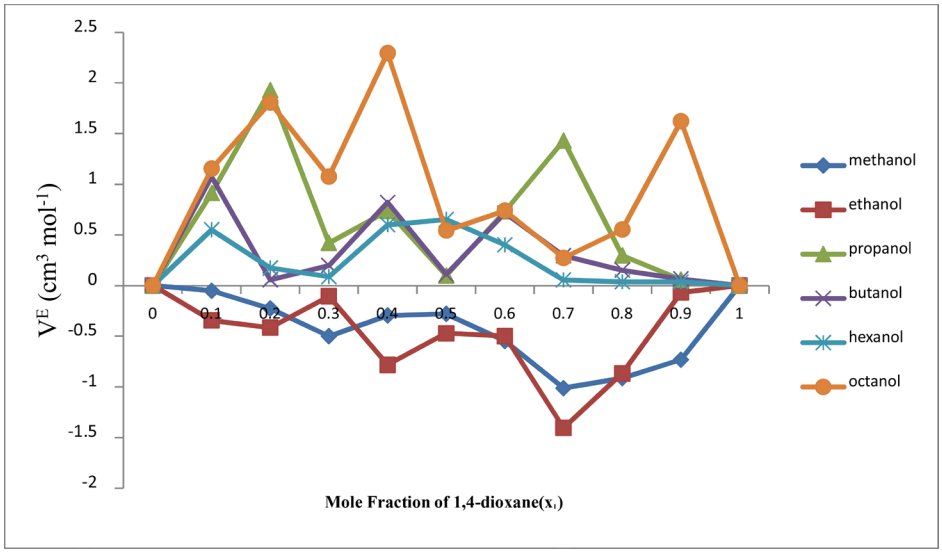

Figure 1. Plots of excess molar volume (V^E) versus mole fraction of 1,4-dioxane (x_1) at 303.15 K for binary mixtures of 1,4-dioxane with methanol, ethanol, propanol, butanol, hexanol and octanol.

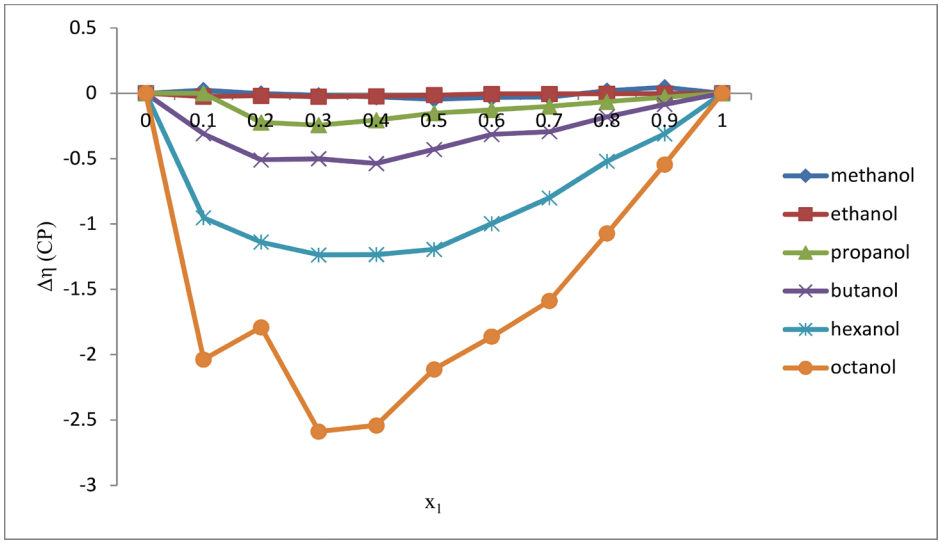

Figure 2. Plots of viscosity deviation ($\Delta\eta$) versus mole fraction of 1,4-dioxane (x_1) at 303.15 K for binary mixtures of 1,4-dioxane with methanol, ethanol, propanol, butanol, hexanol and octanol.

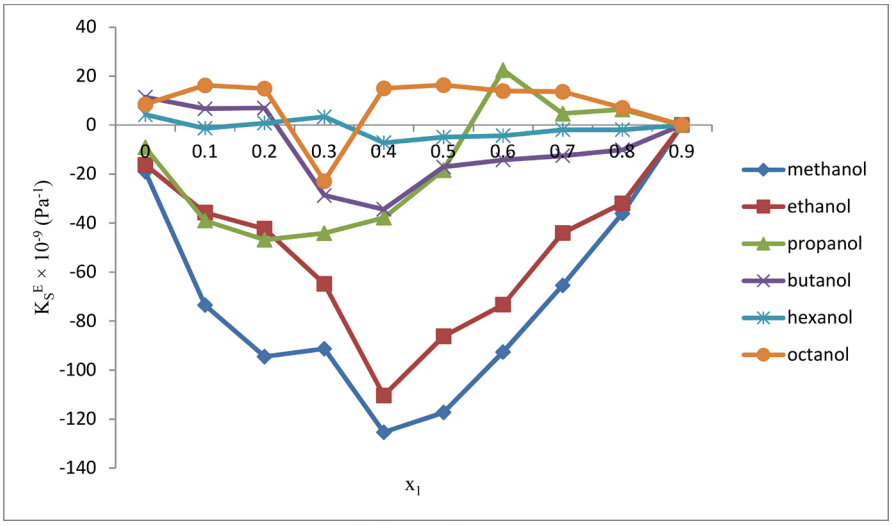

Figure 3. Plots of excess isentropic compressibility (K_S^E) versus mole fraction of 1,4-dioxane at 303.15 K for binary mixtures of 1,4-dioxane with methanol, ethanol, propanol, butanol, hexanol and octanol.

length of the alcohols in **Figure 1**.

The trend it follows is $CH_3OH < C_2H_5OH < C_3H_7OH < C_4H_9OH < C_5H_{11}OH < C_6H_{13}OH < C_7H_{15}OH < C_8H_{17}OH$.

The negative molar volume values indicate the pressure of strong molecular interactions between the components of the mixtures. So many effects may contribute to the value of V^E, such as [42] [43] [44] (I) dipolar-interaction (II) interstitial accommodation of one component in to the other (III) possible hydrogen bond interactions between unlike molecules. The observed positive trends in (V^E) values indicate that the effect due to the breaking up of self-associated structure of the components of the mixtures is dominant over the effect of H-bonding and dipole-dipole interaction between unlike molecule. The V^E values increase in the sequence butanol < hexanol < octanol which also reflects the decreasing strength of interaction between unlike molecules in the mixture. As the size of the alkyl group increases from propanol to octanol, the steric hindrance also increases. Thus the extent of positive deviation (**Figure 1**) supports our view [45].

A correlation between the sign of $\Delta\eta$ and V^E has been observed for a number of binary solvent system ([46] [47]) *i.e.* $\Delta\eta$ is negative when VE is positive and vice-versa. In general for systems where dispersion and dipolar interactions are operating, $\Delta\eta$ values are found to be negative, whereas charge transfer and hydrogen bonding interactions lead to the formation of complex species between unlike molecules.

The calculated ΔK_S values for the binary liquid mixture listed in **Table 3**. The change of this property has been shown in **Figure 3**. The ΔK_S values are negative over the entire mole fraction range and become more negative with increasing the mole fraction of second component for all binary mixtures. These results can be explained in term of molecular interactions and structured effects.

The variation of ΔK_S with volume fraction of 1,4-dioxane x_1 is represent in

Table 2. Experimental Results for the binary Liquid Mixtures of 1,4-Dioxane (1) + Primary alcohols (2) at 303.15 K.

Mole fraction Dioxane (x_1)	ρ (g·m^{-3})	η (CP)	$\Delta\eta$ (CP)	VE (cm^3·mole^{-1})
1,4-Dioxane + Methanol				
0.0000	0.7840	0.4949	0.0000	0.0000
0.0977	0.8274	0.5704	+0.0232	−0.0505
0.2004	0.8669	0.6005	−0.0015	−0.2259
0.2867	0.8969	0.6339	−0.0143	−0.4987
0.3801	0.9171	0.6703	−0.0280	−0.2947
0.4985	0.9422	0.7168	−0.0449	−0.2794
0.5919	0.9631	0.7802	−0.0315	−0.5467
0.7086	0.9877	0.8441	−0.0301	−1.0120
0.8002	0.9987	0.9426	+0.0193	−0.9140
0.9036	1.0089	1.0244	+0.0457	−0.7305
1.0000	1.0108	1.0303	0.0000	0.0000
1,4-Dioxane + Ethanol				
0.0000	0.7720	1.1399	0.0000	0.0000
0.0988	0.8094	1.1038	−0.0251	−0.3437
0.2046	0.8426	1.0986	−0.0187	−0.4155
0.2996	0.8652	1.0814	−0.0255	−0.1063
0.3974	0.8990	1.0740	−0.0222	−0.7812
0.5022	0.9201	1.0710	−0.0137	−0.4701
0.5950	0.9410	1.0697	−0.0049	−0.4986
0.6900	0.9720	1.0593	−0.0049	−1.4042
0.7993	0.9862	1.0485	−0.0037	−0.8659
0.8934	0.9935	1.0406	−0.0013	−0.0668
1.0000	1.0108	1.0303	0.0000	0.0000
1,4-Dioxane + Propanol				
0.0000	0.8070	1.5477	0.0000	0.0000
0.1000	0.8206	1.4964	+0.0005	+0.9139
0.1226	0.8407	1.2610	−0.2232	+1.9297
0.2982	0.8700	1.1498	−0.2435	+0.4208
0.4057	0.8893	1.1319	−0.2058	+0.7359
0.5043	0.9167	1.1274	−0.1515	+0.0990
0.6025	0.9290	1.1097	−0.1262	+0.7335
0.6941	0.9558	1.0881	−0.1004	+1.4319
0.7962	0.9708	1.0711	−0.0645	+0.2982
0.8992	0.9924	1.0534	−0.0290	+0.0583

Continued

1.0000	1.0108	1.0303	0.0000	0.0000
1,4-Dioxane + Butanol				
0.0000	0.8040	2.2045	0.0000	0.0000
0.0973	0.8136	1.7804	−0.3097	+1.0740
0.1975	0.8425	1.4627	−0.5096	+0.0580
0.3044	0.8626	1.3458	−0.5011	+0.1987
0.4048	0.8769	1.1934	−0.5367	+0.8202
0.4944	0.9022	1.1939	−0.4299	+0.1106
0.5976	0.9174	1.1879	−0.3147	+0.7156
0.6862	0.9402	1.1044	−0.2941	+0.2949
0.7907	0.9639	1.0953	−0.1805	+0.1522
0.8909	0.9863	1.0728	−0.0855	+0.0650
1.0000	1.0108	1.0303	0.0000	0.0000
1,4-Dioxane + Hexanol				
0.0000	0.8128	4.5642	0.0000	0.0000
0.0910	0.8219	3.2904	−0.9518	+0.5509
0.1948	0.8400	2.7369	−1.1385	+0.1738
0.2984	0.8572	2.2727	−1.2368	+0.0883
0.4043	0.8714	1.9013	−1.2337	+0.59964
0.4543	0.8799	1.7643	−1.1943	+0.6533
0.6028	0.9107	1.4365	−0.9971	+0.4033
0.6997	0.9346	1.2914	−0.7999	+0.0548
0.8018	0.9584	1.2103	−0.5202	+0.0373
0.8883	0.9800	1.1144	−0.3103	+0.0356
1.0000	1.0108	1.0303	0.0000	0.0000
1,4-Dioxane + Octanol				
0.0000	0.8242	7.8512	0.0000	0.0000
0.0978	0.8284	5.1466	−2.0374	+1.1543
0.2065	0.8370	4.6513	−1.7911	+1.8065
0.2981	0.8529	3.2294	−2.5883	+1.0755
0.4027	0.8595	2.5625	−2.5414	+2.2951
0.4922	0.8852	2.3806	−2.1126	+0.5428
0.6006	0.9030	1.8916	−1.8623	+0.7398
0.6988	0.9266	1.4950	−1.5891	+0.2733
0.7961	0.9464	1.3490	−1.0720	+0.5537
0.8974	0.9759	1.1845	−0.5448	+1.6229
1.0000	1.0108	1.0303	0.0000	0.0000

Table 3. Speeds of sound (u), Isentropic Compressibility (K_S) and Excess Isentropic Compressibility (K_S^E) of Binary Mixtures of Various Composition (Mole Fraction) at 303.15 K.

Mole fraction Dioxane (x_1)	u (m-s^{-1})	$K_S \times 10^{-9}$ (Pa^{-1})	$K_S^E \times 10^{-9}$ (Pa^{-1})
1,4-Dioxane + Methanol			
0.0000	1084.0	1085.4	0.0
0.0977	1092.0	1013.4	−19.0
0.2004	1130.0	903.3	−73.5
0.2867	1155.0	835.7	−94.5
0.3801	1176.0	788.3	−91.3
0.4985	1240.0	690.2	−125.4
0.5919	1266.0	647.8	−117.3
0.7086	1289.0	609.3	−92.6
0.8002	1306.0	587.0	−65.4
0.9036	1330.0	560.3	−36.1
1.0000	1348.0	544.4	0.0
1,4-Dioxane + Ethanol			
0.0000	1141.0	994.9	0.0
0.0988	1150.0	934.1	−16.2
0.2046	1170.0	866.9	−35.7
0.2996	1189.0	817.5	−42.3
0.3974	1217.0	751.0	−64.8
0.5022	1285.0	658.1	−110.4
0.5950	1288.0	640.5	−86.2
0.6900	1298.0	610.6	−73.3
0.7993	1310.0	590.8	−44.0
0.8934	1340.0	560.5	−31.9
1.0000	1348.0	544.4	0.0
1,4-Dioxane + Propanol			
0.0000	1182.0	886.8	0.0
0.1000	1202.0	843.4	−9.1
0.1226	1215.0	805.7	−39.0
0.2982	1248.0	737.9	−46.8
0.4057	1264.0	703.7	−44.1
0.5043	1270.0	676.3	−37.8
0.6025	1275.0	662.1	−18.4
0.6941	1284.0	671.7	+22.5
0.7962	1290.0	618.9	+4.7
0.8992	1312.0	585.3	+6.4

Continued

1.0000	1348.0	544.4	0.0
1,4-Dioxane + Butanol			
0.0000	1196.0	869.5	0.0
0.0973	1203.0	849.2	+11.4
0.1975	1209.0	812.0	+6.7
0.3044	1221.0	777.5	+7.0
0.4048	1268.0	709.2	−28.7
0.4944	1282.0	674.3	−34.4
0.5976	1287.0	658.0	−17.1
0.6862	1297.0	632.2	−14.2
0.7907	1315.0	599.9	−12.5
0.8909	1334.0	569.7	−10.1
1.0000	1348.0	544.4	0.0
1,4-Dioxane + Hexanol			
0.0000	1298.0	730.2	0.0
0.0910	1302.0	717.6	+4.3
0.1948	1311.0	692.6	−1.3
0.2984	1314.0	675.6	+0.8
0.4043	1320.0	658.5	+3.4
0.4543	1334.0	638.5	−7.2
0.6028	1338.0	613.3	−4.9
0.6997	1340.0	595.8	−4.3
0.8018	1342.0	579.3	−1.9
0.8883	1346.0	563.2	−1.9
1.0000	1348.0	544.4	0.0
1,4-Dioxane + Octanol			
0.0000	1327.0	689.0	0.0
0.0978	1329.0	683.4	+8.5
0.2065	1330.0	675.3	+16.2
0.2981	1332.0	660.8	+14.9
0.4027	1334.0	653.8	−22.9
0.4922	1336.0	632.8	+15.0
0.6006	1338.0	618.5	+16.3
0.6988	1339.0	601.9	+13.9
0.7961	1341.0	587.5	+13.6
0.8974	1345.0	566.4	+7.1
1.0000	1348.0	544.4	0.0

Figure 3. Kiyohara and Benson [48] have suggested that ΔK_S is the result of several opposing effects. Strong molecular interactions occur through charge transfer, dipole-induced dipole and dipole-dipole interaction [49], interstitial accommodation and orientation ordering all lead to a more compact structure making ΔK_S negative, whereas breakup of the alkanol structure trends to make ΔK_S positive. The magnitudes of the various contributions depend mainly on the relative molecular size of the components.

4. Conclusion

Thus paper reported the densities, viscosities and sound velocity of six pure methanol, ethanol, propanol, butanol, hexanol and octanol and mixture at 303.15 K over the entire range of mole fraction. After a thorough study of the behavior of primary alcohols and 1,4-dioxane we get a clear idea about the type and amount of molecular interactions between the components. The study of excess properties along with the speed of sound has been found to very useful in understanding the nature of the interactions within binary liquid mixtures. These excess properties obtained from the correlating equations have also provided the very important and useful information.

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Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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