Front. Chem. China 2011, 6(2): 113–119
DOI 10.1007/s11458-011-0234-9

RESEARCH ARTCILE

Effects of $CuCl_2 \cdot 6H_2O$ and $ZnCl_2 \cdot 6H_2O$ on the viscosity of aqueous ethanol mixtures

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The effects of $CuCl_2$ and $ZnCl_2$ on the viscosity in aqueous ethanol mixtures (10%–50% v/v) were studied in the concentration range 1.0×10^{-2}–8.0×10^{-2} mol·dm^{-3} at different temperatures. It was found that the viscosities increased with an increase in the concentration of the salts and percent composition of ethanol content, whereas it decreased with an increase in temperature. Ion-ion and ion-solvent interactions are determined with the help of *A*- and *B*-coefficients of Jones-Dole equation. The values of *A*- and *B*-coefficients are irregular and increase with a rise in temperature and also with an increase in ethanol contents for both salts. Negative values of *B*-coefficients show that ion solvent interactions is comparatively small and suggest that $CuCl_2$ and $ZnCl_2$ behave as structure breakers in aqueous ethanol mixtures. Thermodynamic parameters like the energy of activation (E_η) and change in entropy of activation (ΔS^*) were also evaluated which confirm the structure breaker behavior of salts in aqueous ethanol mixtures.

Keywords viscosity, Jones-Dole equation, ionic interactions, transition metal chlorides, thermodynamic parameters

1 Introduction

In our earlier publication [1] viscosity of transition metal chlorides (cobalt chloride and nickel chloride) in aqueous ethanol mixtures at different temperatures has been reported. Ion-solvent and ion-ion interactions were studied by analyzing the viscosity data with the help of Jones-Dole Eq. (2).

$$\eta_{sp}/\sqrt{C} = A + B\sqrt{C} \tag{1}$$

where η_{sp} is the specific viscosity, $\sqrt{C}$ is the square root of the concentration of salt and *A* and *B* are the coefficient viscosity which are respectively obtained from the intercept and slope of linear plots of $\eta_{sp}\sqrt{C}$ vs. $\sqrt{C}$. The values of *A*-coefficient get increased with the increase of salt, percent composition of the ethanol mixtures and temperature. Negative values of *B*-coefficient reveal that ion-solvent interaction decreases and suggest that these metal salts behave as a structure breaker in aqueous ethanol mixtures. Furthermore, the energy of activation was also determined, which decreases with an increase in the concentration of salt and increases with a rise in percent composition of ethanol mixture. The present paper deals with the effect copper chloride ($CuCl_2$) and zinc chloride ($ZnCl_2$) on the viscosity of aqueous ethanol.

2 Experimental

Analar grade copper chloride (mol.wt 134.5 g/mol) and zinc chloride (mol.wt 136 g/mol), 99.9% pure ethanol of BDH are freshly prepared in double distilled water (conductivity 0.06 μS·cm^{-1}) were used.

Viscosity measurements were accompanied according to the procedure given elsewhere [1]. Aqueous ethanol mixtures of different composition (v/v) were prepared in double distilled water. Solutions of metal chloride were prepared in different percent composition of aqueous ethanol .Viscosities of these solutions were measured with the help of Ostwald Viscometer (type Techniconominal constant 0.1 (Cs/s) capillary ASTMD 445) by taking time of flow of solvents and solutions at different temperatures, 30°C–50°C with the interval of 5°C. The temperature of the solutions were kept constant throughout the experimental work with the help of thermostatic water bath (VWP Scientific model 1120 SER9143791). Reproducibility of the observations was made sure.

3 Results and discussion

Table 1 includes the viscosities and densities of aqueous ethanol (10%, 20%, 30%, 40% and 50%) in the absence of salt. The observations are collected at 30°C. The viscosities of aqueous ethanol solvents increased with the increase in percentage concentration of aqueous ethanol, whereas the density gets decreased with the decrease in percentage concentration of aqueous ethanol. The time of flow for $CuCl_2$ and $ZnCl_2$ was measured in 10%, 20%, 30%, 40% and 50% aqueous ethanol at various temperatures (30°C–50°C)± 0.1°C with a difference of 5°C. The results are summarized in Tables 2 and 3 which indicate that the viscosity values

Received February 21, 2011; accepted March 4, 2011
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increased with the increase in concentration of salts whereas it decreased with the rise in temperature. Similar behavior was also noted in the case of $CoCl_2$ and $NiCl_2$ [1] and electrolyte of group I of the periodic table in aqueous ethanol system [3] and strong electrolyte like HCl, NaCl and NaOH on the viscosity values of edible oils, e.g sunflower, maize, canola and soybean in 1,4 dioxane [4–7].

The viscosities of metal chloride in aqueous ethanol are also interpreted in terms of ion-ion and ion solvent interactions using Jones-Dole Eq. (1) to characterize the behavior of ectrolytes. The Jones- Dole equation contains two coefficients A and B which are obtained graphically ($\eta_{sp}\sqrt{C}$ vs. $\sqrt{C}$). Here η_{sp} defines specific viscosity of the solutions and $\sqrt{C}$ is the square root of the concentration of metal chloride in aqueous ethanol. The slope and intercept of the plot gives the value of both B- and A-coefficient respectively. The representative plots for $CuCl_2$ and $ZnCl_2$ are shown in Figs. 1 and 2 and the results are summarized in Tables 4 and 5. The A-coefficient of the equation indicates the ion-ion

Table 1 Viscosity of aqueous ethanol mixtures in the absence of salt at 30°C.

Aqueous ethanol/% (v/v)	Densities / (gm·mL^{-1})	Viscosities / (mPa·s)
10	0.9996	8.945
20	0.9849	10.849
30	0.9732	12.877
40	0.9568	14.711
50	0.9368	16.247

Table 2 Viscosity of copper chloride ($CuCl_2 \cdot 6H_2O$) in aqueous ethanol.

Salt / (10^2 mol·dm^{-3})	Viscosities at different temperatures / (mPa·s)				
	30°C	35°C	40°C	45°C	50°C
	10% aqueous ethanol				
1.00	9.214	7.758	7.697	7.232	6.651
2.00	9.481	7.809	7.746	7.274	6.696
5.00	9.638	7.953	7.901	7.327	6.838
7.00	9.760	8.045	8.002	7.520	6.929
9.00	9.835	8.117	8.081	7.614	7.014
	20% aqueous ethanol				
1.00	11.560	11.020	9.150	8.451	7.582
2.00	11.657	11.078	9.198	8.461	7.626
5.00	11.834	11.240	9.351	8.568	7.762
7.00	11.954	11.386	9.450	8.673	7.849
9.00	12.081	11.479	9.545	8.775	7.946
	30% aqueous ethanol				
1.00	13.273	11.715	10.016	8.789	7.807
2.00	13.378	11.812	10.066	8.839	7.838
5.00	13.650	12.035	10.216	8.985	8.036
7.00	13.786	12.150	10.298	9.083	8.137
9.00	13.978	12.304	10.384	9.182	8.232
	40% aqueous ethanol				
1.00	15.220	13.414	10.439	9.188	8.136
2.00	15.281	13.469	10.530	9.275	8.236
5.00	15.485	13.639	10.726	9.497	8.499
7.00	15.627	13.756	10.829	9.637	8.705
9.00	15.750	13.855	11.008	9.733	8.832
	50% aqueous ethanol				
1.00	16.790	15.940	13.007	11.741	9.876
2.00	16.894	16.002	13.194	11.829	9.994
5.00	16.219	16.265	13.344	12.022	10.244
7.00	16.427	16.389	13.471	12.180	10.409
9.00	16.643	16.541	13.679	12.378	10.543

Table 3 Viscosity of zinc chloride ($ZnCl_2 \cdot 6H_2O$) in aqueous ethanol.

Salt / (10^2 mol·dm^{-3})	Viscosities at different temperatures / (mPa·s)				
	30°C	35°C	40°C	45°C	50°C
			10% aqueous ethanol		
1.00	10.370	9.032	7.939	7.760	7.075
2.00	10.528	9.133	7.993	7.766	7.120
5.00	10.801	9.449	8.121	8.002	7.268
7.00	10.919	9.607	8.347	8.098	7.360
9.00	11.040	9.718	8.452	8.196	7.453
			20% aqueous ethanol		
1.00	11.181	10.864	9.312	8.433	7.576
2.00	11.239	10.966	9.492	8.502	7.621
5.00	11.387	11.275	9.660	8.649	7.760
7.00	11.437	11.392	9.790	8.732	7.852
9.00	11.493	11.506	9.898	8.821	7.945
			30% aqueous ethanol		
1.00	14.506	12.503	10.590	9.290	8.213
2.00	14.655	12.607	10.684	9.380	8.259
5.00	15.169	12.959	11.011	9.571	8.392
7.00	15.381	13.227	11.163	9.668	8.524
9.00	15.836	13.485	11.354	9.808	8.617
			40% aqueous ethanol		
1.00	15.369	13.779	11.061	9.363	8.426
2.00	15.480	13.881	11.155	9.450	8.474
5.00	15.795	14.191	11.436	9.719	8.684
7.00	16.054	14.303	11.625	9.893	8.811
9.00	16.269	14.424	11.815	9.791	8.903
			50% aqueous ethanol		
1.00	17.183	16.139	14.111	12.217	10.494
2.00	17.297	16.238	14.208	12.271	10.524
5.00	17.668	16.504	14.465	12.537	10.673
7.00	17.841	16.666	14.574	12.681	10.769
9.00	18.054	16.886	14.727	12.879	10.861

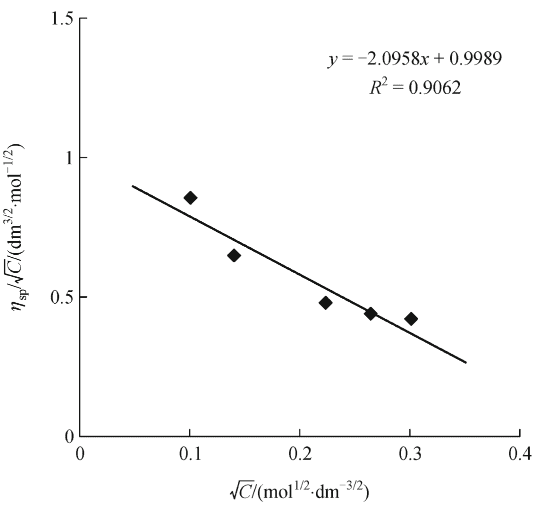

Figure 1 Plot of $\eta_{sp}\sqrt{C}$ versus $\sqrt{C}$ at 30% aqueous ethanol at 40°C for $CuCl_2 \cdot 6H_2O$.

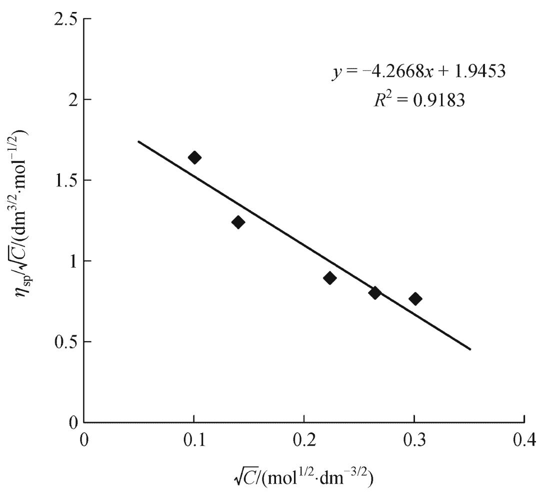

Figure 2 Plot of $\eta_{sp}\sqrt{C}$ versus $\sqrt{C}$ at 30% aqueous ethanol at 45°C for $ZnCl_2 \cdot 6H_2O$.

Table 4 The values of parameters of Jones-Dole equation at various compositions of $CuCl_2 \cdot 6H_2O$ in aqueous ethanol at different temperatures.

Temperature/°C	*A*- and *B*-coefficient of Jones-Dole equation in different aqueous ethanol solvents				
	10%	20%	30%	40%	50%
	A-coefficient/($mol^{-1/2} \cdot dm^{3/2}$)				
30	0.3717	0.5551	0.2965	0.3948	0.3141
35	0.6041	0.8315	0.6385	0.3987	0.6591
40	0.6243	0.9285	0.9520	0.4228	0.7116
45	0.6248	1.1029	0.2900	0.9415	1.1868
50	0.7044	1.2322	0.3424	0.9685	1.1790
	B-coefficient/($mol^{-1} \cdot dm^{3}$)				
30	−0.1025	−0.2188	−0.0918	−0.4443	−0.1545
35	−0.9466	−1.6829	−0.9406	−0.6022	−1.2048
40	−0.9299	−1.8134	−1.9587	−0.4272	−1.2069
45	−0.9252	−2.2944	−3.1723	−1.6696	−2.3551
50	−1.0928	−2.5271	−2.6153	−1.2847	−2.1390

Table 5 The values of parameters of Jones-Dole equation at various compositions of $ZnCl_2 \cdot 6H_2O$ in aqueous ethanol at different temperatures.

Temperature/°C	*A*- and *B*-coefficient of Jones-Dole equation in different aqueous ethanol solvents				
	10%	20%	30%	40%	50%
	A-coefficient /($mol^{-1/2} \cdot dm^{3/2}$)				
30	0.4342	0.2507	1.3620	0.6881	0.5642
35	0.7761	0.6400	1.4210	0.7504	0.6174
40	0.9103	0.9799	1.4420	0.9503	1.6015
45	1.5748	1.9060	1.8690	1.0610	1.6918
50	1.7391	1.3276	1.5500	1.5562	1.9345
	B-coefficient/($mol^{-1} \cdot dm^{3}$)				
30	−0.2605	−0.2743	−2.3260	−1.0081	−0.5640
35	−0.9114	−0.8259	−2.5810	−1.3240	−0.7880
40	−1.4454	−1.6917	−2.6551	−1.5410	−3.5167
45	−3.2804	−2.4334	−4.0600	−1.7006	−3.6070
50	−3.7223	−2.7880	−5.8600	−3.3040	−4.4250

interaction. The values get increased with the rise in temperature and also with an increase in ethanol content for both salts. This shows that ion-ion interactions increase with the temperature and percent composition of ethanol with some variations. As the concentration of the organic solvent is increased the dielectric constant of the medium decreases, and the electrostatic ion-ion interaction (*A*-coefficient) shows an increase with some expectations. A solute alters the viscosity of the solvent as the ions and water molecules exert a viscous drag on the rest of the solvent and the change in viscosity occurs in mixed solvent system. In ethanol the oxygen of alcohol has a tendency of holding water molecules, i.e, in liquid alcohol an oxygen atom carries one proton and two lone pairs of electron. This may lead to intermolecular hydrogen bonded polymer instead of formation of cluster. With the increase in percent composition of aqueous ethanol content, the viscosities of salt vary because of the formation of the complex. Variation in *A*-values is attributed to the size of the ion which differs in their degree of hydration.

In mixed solvent systems such as aqueous ethanol mixtures, H-bonding to O-H continuously changes the structure of water because of both their cluster forming and cluster distributing character [8]. Water forms strong coordinating bonds with the transition metal ring and also form strong hydrogen bond with negative ions. When the solute is dissolved into the solvent two types of interactions ion-ion and ion-solvent occur. This may be due to the inter penetration effect of cation-cation and cation-anion which brings ions together and is responsible for the increase of ion-ion interactions [9–15].

Similarly the values of *B*-coefficient of Jones Dole equation were obtained from the slope of the plot $\eta_{sp}\sqrt{C}$ vs. $\sqrt{C}$. The observations are found to be effected upon both the temperature and composition of aqueous ethanol mixtures. This *B*-coefficient describes the ion-solvent interactions. In both $CuCl_2$ and $ZnCl_2$ systems, the values are negative and the variation is irregular. The negative values of *B*-coefficient in aqueous ethanol reveal the structure breaking effect [11,12]. The variation of *B* values with a change in percent composition of the solvent represents the electrostatic ion solvent interaction in an aqueous ethanol mixture. The smaller the ion is, the stronger the electrostatic interaction, and hence the greater is the size of the solvent ion. On the other hand, the solute with the positive values of *B*-coefficient in a given solvent is expected to have the structure making effect. Furthermore, a solute with less positive or negative values of *B*-coefficient in a given solvent is considered to be a structure breaker [11]. A large solute shows a large obstructive effect meaning a bending of the stream lines around a large solute particle. In such a solvent *B*-coefficient may be always negative irrespective of how it behaves with the solvent as in the present case ethanol (C_2H_5OH) is not very bulky. Besides this the above mentioned irregularities in the values of *B*-coefficient is due to the degree of hydrolysis in different percent compositions of aqueous ethanol.

The viscosity of a liquid generally decreases with the rise in temperature. This has been explained in terms of "the hole theory of liquids" [15–20]. Intermolecular distances are relatively bigger in liquid than in solid. A liquid molecule therefore needs some activation energy to move in a hole. As the activation energy increases with the rise in temperature, a liquid can flow more easily at high temperature. The coefficient of viscosity thus falls appreciably with the rise of temperature. Arrhenius [13] pointed out the effect of temperature on viscosity by means of an exponential equation measuring the activation energy,

$$\log \eta = \log A + E_\eta / 2.303\, RT \qquad (2)$$

where η is the coefficient of viscosity; *A* is called the pre-exponential factor constant for a given liquid; E_η is the energy of activation; *R* is the gas constant and *T* is the absolute temperature. The values of energy of activation were obtained from the slopes of linear plots of log η vs. $1/T$ for the aqueous ethanol for $CuCl_2$ and $ZnCl_2$. The values of activation energy (E_η) as a function of salt concentration and solvent composition are tabulated in Tables 6 and 7. The representative plot of energy of activation is shown in Fig. 3. Here both the concentrations of salt and percent composition of solvent influence the value of energy of activation. There is an irregular variation in activation energy observed with the increment of the concentration of salt. At a $CuCl_2$ concentration of $1\times10^{-2}\ \mathrm{mol\cdot dm^{-3}}$, the activation energy is determined as $11.647\ \mathrm{kJ\cdot mol^{-1}}$ in 10% aqueous ethanol mixture, whereas in the same composition for an increased concentration of $CuCl_2$ as $2\times10^{-2}\ \mathrm{mol\cdot dm^{-3}}$, the value of activation energy increases to $12.384\ \mathrm{kJ\cdot mol^{-1}}$. Furthermore, the increase in concentration of salt, until up to a concentration of $6\times10^{-2}\ \mathrm{mol\cdot dm^{-3}}$, decreases regularly the value of activation energy. Ultimately

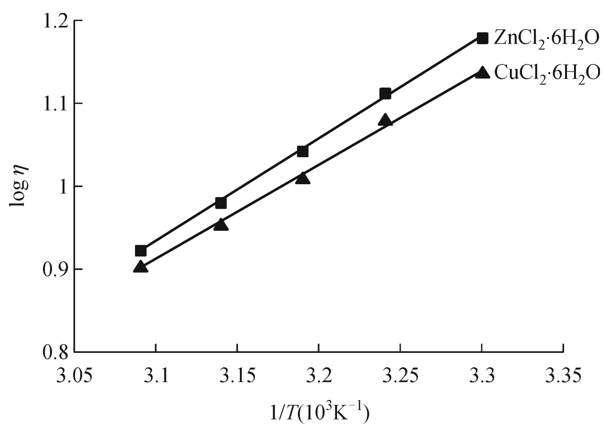

Figure 3 Plot of log η versus $1/T$ for $5\times10^{-2}\ \mathrm{mol\cdot dm^{-3}}$ $CuCl_2 \cdot 6H_2O$ and $ZnCl_2 \cdot 6H_2O$ in 30% aqueous ethanol.

Table 6 Energy of activation for $CuCl_2 \cdot 6H_2O$ in 10%–50% aqueous ethanol.

Salt/($10^2\ \mathrm{mol\cdot dm^{-3}}$)	Activation energy E_η/($\mathrm{kJ\cdot mol^{-1}}$) in aqueous ethanol solvent				
	10%	20%	30%	40%	50%
1.00	11.647	17.716	21.532	25.777	21.716
2.00	12.384	17.809	21.634	25.696	21.489
3.00	12.373	17.757	21.675	25.489	21.450
4.00	12.305	17.821	21.743	24.917	21.347
5.00	12.202	17.737	21.597	25.004	21.324
6.00	12.102	17.724	21.530	24.663	21.204
7.00	12.167	17.726	21.493	24.384	21.113
8.00	12.140	17.631	21.564	24.258	20.965
9.00	11.965	17.618	21.501	24.127	20.996

at concentrations of 7×10^{-2}, 8×10^{-2} and 9×10^{-2} $mol\cdot dm^{-3}$ the values of activation energy are obtained respectively as 12.167, 12.140, 11.965 $kJ\cdot mol^{-1}$. So the irregular behavior of concentration of salt and composition of solvent is also observed for $ZnCl_2$ as shown in Table 7. This behavior may also be seen in another composition of solvent. The increase in activation energy may be due to the fact that at higher solvent composition there is a decrease in mobility of ions, which will make it difficult to produce vacant sites in the solvent matrix resulting in the high energy of activation. As a principle the activation energy which are slightly higher than the threshold activation energy at higher temperature provides an additional activation energy which helps the free movement of the liquid and suppresses the friction of liquid in the capillary, thereby the coefficient of viscosity falls appreciable with the rise of temperature.

Entropy is an important thermodynamic parameter which can be calculated from the following expression:

$$\Delta S^* = \frac{E_\eta - \Delta G}{T} \tag{3}$$

Here ΔS^* is defined as the change in entropy, E_η represents the activation energy, T is the absolute temperature and ΔG is the change in free energy. The values of free energy of different solutions control the rate of flow in fluid process. This flow process is governed by the ability of a molecule to move into the prepared hole and the readiness with which the holes are prepared in the liquid. The changes in entropy values (ΔS^*) at different concentrations of salts, percentages composition of aqueous ethanol mixtures and different temperatures from 30°C to 50°C are summarized in Tables 8 and 9 for $CuCl_2$ and $ZnCl_2$ respectively. The results indicate that the values of change in entropy of activation are negative. There is an irregular increase or decrease with the increase of concentration of salts and with percentage composition of the aqueous ethanol mixtures from 10%–50%.

4 Conclusion

Ion-solvent and ion-ion interactions in the presence of transition metal chloride were evaluated in terms of Jones-Dole coefficient *A* and *B* respectively. The results revealed that $CuCl_2$ and $ZnCl_2$ behave as structure breakers in aqueous ethanol solvents. The irregular increase or decrease of the values of activation energy and change in entropy also support that these metal chlorides act as structure breakers.

Table 7 Energy of activation for $ZnCl_2\cdot 6H_2O$ in 10%–50% aqueous ethanol.

Salt / (10^2 $mol\cdot dm^{-3}$)	Activation energy E_η / ($kJ\cdot mol^{-1}$) in aqueous ethanol solvent				
	10%	20%	30%	40%	50%
1.00	14.739	16.379	22.936	25.328	20.250
2.00	15.177	16.363	23.057	25.348	20.226
3.00	15.235	16.372	23.490	25.229	20.254
4.00	15.248	16.351	23.595	25.124	20.388
5.00	15.391	16.356	23.770	25.111	20.387
6.00	15.446	16.240	23.895	24.946	20.386
7.00	15.417	16.290	23.870	25.020	20.388
8.00	15.349	16.014	24.116	25.305	20.576
9.00	15.341	15.894	24.547	25.412	20.663

Table 8 Entropy change of activation for $CuCl_2\cdot 6H_2O$ in aqueous ethanol at 303 K.

Salt / (10^2 $mol\cdot dm^{-3}$)	Entropy change of activation (ΔS^*) in aqueous ethanol solvent /($J\cdot K^{-1}\cdot mol^{-1}$)				
	10%	20%	30%	40%	50%
1.00	–185.223	–180.845	–185.002	–185.098	–184.995
2.00	–183.041	–178.504	–182.702	–182.834	–182.768
3.00	–183.125	–178.612	–182.790	–182.920	–182.856
4.00	–183.398	–178.862	–183.060	–183.189	–183.121
5.00	–183.781	–179.243	–183.437	–183.562	–183.496
6.00	–184.160	–179.619	–183.791	–183.930	–183.802
7.00	–183.990	–179.455	–183.653	–183.775	–183.704
8.00	–184.185	–179.594	–183.790	–183.914	–183.846
9.00	–184.708	–180.187	–184.375	–184.512	–184.432

Table 9 Entropy change of activation for $ZnCl_2 \cdot 6H_2O$ in aqueous ethanol at 30°C.

Salt / (10^2 mol·dm^{-3})	Entropy change of activation (ΔS*) in aqueous ethanol solvent /(J·K^{-1}·mol^{-1})				
	10%	20%	30%	40%	50%
1.00	–175.987	–175.639	–175.312	–175.909	–175.873
2.00	–174.654	–174.306	–174.007	–174.522	–174.567
3.00	–174.547	–174.209	–173.875	–174.492	–174.457
4.00	–174.580	–174.255	–173.913	–174.450	–174.513
5.00	–174.151	–173.878	–173.542	–174.093	–174.057
6.00	–174.031	–173.788	–173.403	–174.975	–173.0939
7.00	–174.151	–173.929	–173.288	–174.115	–174.079
8.00	–174.416	–174.190	–173.210	–174.375	–174.344
9.00	–174.486	–174.208	–173.922	–174.450	–174.414

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