

Complexation of Ceftazidime Antibiotic with Copper Chloride Salt in Mixed (MeOH+H₂O) Solvents

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The complexation reaction between Ceftazidime antibiotic (CFZ) and copper chloride salt (CuCl₂.2H₂O) was studied in mixed (MeOH+H₂O) using conductometric measurements. The association parameters of CuCl₂.2H₂O salt in mixed (MeOH+H₂O) with the presence of CFZ were easily calculated using Fuoss-Shedlovsky equation at different temperatures 293.15, 298.15, 303.15 and 308.15K. It was observed that, the limiting molar conductances decrease by increasing temperature due to more association. Also, the thermodynamic parameters of complexation between CuCl₂.2H₂O salt and CFZ were calculated to found the formation of (1:2) and (1:1) M/L complexes. The (1:1) M/L complex has greater formation thermodynamic values for CuCl₂.2H₂O by its interaction with CFZ at all the temperatures used.

Introduction

The studies of the conductivity behavior of substances in aqueous and nonaqueous media have received considerable importance in the recent years [1-3] due to its varied applications in various electrochemical investigations. Conductivity study is one of the most important and simplest tools to understand the transport behavior in general and solvation behavior in specific [4].

The electrical conductivity is one of the most important properties of electrolyte solution [5, 6]. From the results of the conductivity experiment a series of fundamental parameters of electrolyte solutions such as ion association constants, limiting conductivities and solvation characteristics can be obtained. Despite of more than a century the history of the measurements of the conductivity, studying of the solution of unsymmetrical electrolytes in both non-aqueous and mixed solvents has not been developed intensively. Partly, this situation can be explained by the complexity of interpretation of the experimental conductometric data for unsymmetrical electrolytes taking into account ion association [7].

Inflammatory diseases including different types of rheumatic diseases are very common throughout the world [8]. Although the rheumatism is one of the oldest known diseases of mankind and affects a large population of the world and no substantial progress has been made in achieving a permanent cure. The treatment includes intake of non steroidal anti-inflammatory drugs (NSAIDs) of the oxicam family. Ceftazidime (CFZ) [9-18] is a broad-spectrum antibiotic that kills a wide variety of bacteria.

Experimental

Chemicals and Reagents

Copper chloride salt (CuCl₂.2H₂O) was Merck pure while Ceftazidime antibiotic (CFZ) was from "GlaxoSmithKline" and used as purchased.



Ceftazidime antibiotic (CFZ)

Formula: C₂₂H₂₂N₆O₇S₂· 5H₂O

Mol. Wt.: 636.65 g/mol

Methanol solvent (MeOH) was provided from El Nasr pharmaceutical chemicals co and used directly without purification.

Experimental Method

Conductometric Measurement

A solution of CuCl_2 (10⁻⁴ M) was placed in a titration cell, thermostated at a given temperature and the conductance of the solution was measured. The ligand (10⁻³M) (Ceftazidime) was transferred step by step to the titration cell using a precaliberated micropipette and the conductance of the solution was measured after each transfer. The addition of the ligand solution was continued until the total concentration of the ligand was approximately four times higher than that of the metal ions. The conductance of the solution was measured after each addition. The complex formation constant K_f, and the molar conductance of the complex ML, were evaluated by computer fitting to the molar conductance mole ratio data. The temperatures used are (298.15, 303.15, 308.15 and 313.15K). The specific conductivity K_s was achieved by using a conductometer of the type (OAKTON WD-35607-10,-20,-30) with platinum electrode and a cell constant equal (1). The conductometer was connected with ultra-thermostat of the type (Kottermann 4130).

Results and Discussion

Conductometric measurements

The stability of a transition metal complex with a polydentate chelate ligand depends on a range of factors including the number and the type of the donor atoms presented, the number and the size of cheated rings formed on the complexation [19]. In addition, the stability and the selectivity of the complexities are strongly depend on the donor ability and the dielectric constant of the solvent [20], the shape, the size of the solvent molecules [21].

Fortum contains the active ingredient Ceftazidime, which is a type of antibiotic called a cephalosporin. This antibiotic is related to penicillin and it is a broad-spectrum antibiotic that kills a wide variety of bacteria. It is a polydentate ligand which tends to be completely coordinated to a metal ion. This ligand is soluble in water and in most organic solvents.

Calculation of association parameters for $CuCl_2.2H_2O$ in mixed (MeOH+H₂O) with presence of ligand (Ceftazidime, CFZ).

The values of specific conductance (K_s) for different concentrations of CuCl₂.2H₂O in mixed (MeOH+H₂O) were measured experimentally with presence of ligand (CFZ) at different temperatures 293.15, 298.15, 303.15 and 308.15 K. Also, the values of molar conductance (Λ_m) can be calculated [22, 23] using equation (1):

$$\Lambda_m = \frac{(K_s - K_{solv})K_{cell} \times 1000}{C} \tag{1}$$

Where; K_s and K_{solv} are the specific conductance of the solution and the solvent, respectively; K_{cell} is the cell constant and C is the molar concentration of CuCl₂.2H₂O solutions.

The limiting molar conductances (Λ_0) at infinite dilutions were estimated for CuCl₂.2H₂O in mixed (MeOH+H₂O) with presence of ligand (CFZ) at different temperatures by extrapolating the relation between Λ_m and $C_m^{\frac{1}{2}}$ to zero concentration as shown in Fig. 1.



Fig. 1. Variation of the molar conductance $\Lambda_m (\Omega^{-1}.cm^2.mol^{-1})$ with $Cm^{\frac{1}{2}}$ for $CuCl_2.2H_2O$ with CFZ in (a) dist. H_2O .

The experimental data for conductance measurements were analyzed using Fuoss-Shedlovsky [24] extrapolation techniques which follow equations (2-12): -

$$\frac{1}{\mathbf{\Lambda}\mathbf{S}_{(z)}} = \frac{1}{\mathbf{\Lambda}_{o}} + \left(\frac{K_{A}}{\mathbf{\Lambda}_{o}^{2}}\right) \left(C\mathbf{\Lambda}\boldsymbol{\gamma}_{\pm}^{2}\boldsymbol{S}_{(z)}\right)$$
(2)

Where, $S_{(Z)} = 1 + Z + Z^2/2 + Z^3/8 + \dots$ etc.

and

$$Z = \frac{S(AC)^{1/2}}{A_{o}^{3/2}}$$
(3)

The Onsager s[lope

$$S = a\Lambda_0 + b \tag{4}$$

Where,

$$a = 8.2 \times 10^5 / (\varepsilon T)^{3/2}$$
(5)

$$b = 82.4/\eta ((\epsilon T)^{1/2}$$
 (6)

Where (ϵ) is the dielectric constant of the solvent, (η) is the viscosity of the solvent.

The degree of dissociation

$$\boldsymbol{\alpha} = \Lambda \, \mathbf{S}_{(Z)} \,/\, \Lambda_{\mathrm{o}} \tag{7}$$

The mean activity coefficients

$$\log \gamma_{\pm} = -z_{-} \cdot z_{+} A(\alpha C)^{1/2} / [1 + B r^{o}(\alpha C)^{1/2})]$$
(8)

Where (z_{-}, z_{+}) are the charges of ions in solutions A, B are the Debye-Hückel constant.

A = 1.824 X 10⁶ (ϵT)^{-3/2} ; B = 50.29 X 10⁸ (ϵT)^{-1/2} and (r^o) is the solvated radius.

The association constant

$$\mathbf{K}_{\mathbf{A}} = \frac{C_{[MX_{a}]} \cdot \boldsymbol{\gamma}_{[MX_{a}]}}{C_{M^{n+1}} \cdot \boldsymbol{\gamma}_{M^{n+1}} \cdot C_{x^{-1}}^{n} \cdot \boldsymbol{\gamma}_{x^{-1}}}$$
(9)

The dissociation constant

 $K_{\rm D} = 1/K_{\rm A} \tag{10}$

The values of the triple ion association constant (K_3) were calculated [26] by using the equation (11):

$$\frac{\boldsymbol{\Lambda}C^{1/2}}{(1-\frac{\boldsymbol{\Lambda}}{\boldsymbol{A}_{o}})^{1/2}} = \frac{\boldsymbol{\Lambda}_{o}}{(K_{A})^{1/2}} + \frac{\lambda_{3}^{o}C}{K_{3}(K_{A})^{1/2}}(1-\frac{\boldsymbol{\Lambda}}{\boldsymbol{A}_{o}})$$
(11)

Equation (11) was derived by Fuoss [25, 26] and using Walden approximation ($\Lambda_0 = 3\lambda_0$).

The values of Gibbs free energy change of association (ΔG_A) for CuCl₂.2H₂O in mixed (MeOH+H₂O) at different temperatures (293.15, 298.15, 303.15 and 308.15 K) were calculated [27, 28] by using equation (12).

$$\Delta G_A = -2.303 \text{ RT} \log K_A \tag{12}$$

Where R is the gas constant (8.314 J.mol⁻¹ degree⁻¹). The calculated values of η_0 , Λ_0 , Λ , C, S, Z, $S_{(Z)}$, γ_{\pm} , K_A , K_D , α , K_3 and Gibbs free energies change for the solutions of CuCl₂.2H₂O in mixed (MeOH+H₂O) at different temperatures 293.15, 298.15, 303.15 and 308.15 K were calculated and reported in Table.1(a, b, c).

Table 1(a). The values of viscosity (η_0) , limiting molar conductance (Λ_m) , molar conductance (Λ) , Fuoss-Shedlovsky parameters $(S, Z \text{ and } S_{(z)})$, activity coefficient (Y_{z}) , association constant (log K_A), dissociation constant (K_D) , degree of dissociation (α) , triple ion association constant (K_3) , Gibbs free energy change of association (ΔG_A) for CuCl₂.2H₂O with CFZ in dist.H₂O at different temperatures.

Temp. (K)	10² η₀ (poise)	$\Lambda_{\rm m}$	L	S	Z	S _(z)	γ_{\pm}	α	10 ³ K _D	10 ⁵ K ₃	log K _A	D GA
293.15	1.001	1827.2	213.9	470.1	0.00145	1.00145	0.98688	0.1172	0.0041	18.03	5.383	-30.21
298.15	0.892	1788.1	202.0	471.5	0.00146	1.00146	0.98700	0.1131	0.0038	18.53	5.416	-30.91
303.15	0.800	1680.9	182.6	459.4	0.00148	1.00148	0.98708	0.1087	0.0035	19.09	5.452	-31.64
308.15	0.722	1556.7	161.9	443.8	0.00151	1.00152	0.98715	0.1041	0.0032	19.71	5.492	-32.40

 Λ_{\circ} in (Ω^{-1} .cm².mol⁻¹), Λ in (Ω^{-1} .cm².mol⁻¹) and Δ G_A in (kJ mol⁻¹).

Table 1(b). The values of viscosity (η_0), limiting molar conductance (Λ_m), molar conductance (Λ), Fuoss-Shedlovsky parameters (S, Z and S₍₂₎), activity coefficient (Υ_{+}), association constant (log K_A), dissociation constant (K_D), degree of dissociation (α), triple ion association constant (K₃), Gibbs free energy change of association (ΔG_A) for CuCl₂.2H₂O with CFZ in (40% MeOH+60% H₂O) at different temperatures.

Temp. (K)	10² η₀ (poise)	$\Lambda_{\rm m}$	L	S	Z	S _(z)	${\gamma}_{\pm}$	α	10 ³ K _D	10 ⁵ K ₃	log K _A	D G _A
293.15	1.83	957.2	114.8	302.1	0.00180	1.00180	0.9835	0.1201	0.0043	17.52	5.363	-30.10
298.15	1.62	945.9	109.6	305.1	0.00181	1.00181	0.9837	0.1161	0.0040	18.00	5.395	-30.79
303.15	1.43	869.7	96.68	292.4	0.00185	1.00185	0.9837	0.1113	0.0036	18.58	5.433	-31.53
308.15	1.24	823.5	87.08	289.3	0.00188	1.00188	0.9839	0.1059	0.0033	19.29	5.479	-32.33

 Λ_{\circ} in (Ω^{-1} .cm².mol⁻¹), Λ in (Ω^{-1} .cm².mol⁻¹) and ΔG_{A} in (kJ mol⁻¹).

Table 1(c). The values of viscosity (η_0) , limiting molar conductance (Λ_m) , molar conductance (Λ) , Fuoss-Shedlovsky parameters $(S, Z \text{ and } S_{(z)})$, activity coefficient (Υ_*) , association constant (log K_A), dissociation constant (K_D) , degree of dissociation (α) , triple ion association constant (K_3) , Gibbs free energy change of association (ΔG_A) for CuCl₂.2H₂O with CFZ in (60% MeOH+40% H₂O) at different temperatures.

Temp. (K)	10² η ₀ (poise)	$\Lambda_{\rm m}$	L	S	Z	S _(z)	${\gamma}_{\pm}$	α	10 ³ K _D	10 ⁵ K ₃	log K _A	$D G_A$
293.15	1.72	780.94	95.81	301.2	0.00223	1.00223	0.9800	0.1229	0.00451	17.06	5.345	-30.00
298.15	1.54	750.70	90.18	296.8	0.00226	1.00226	0.9801	0.1204	0.00431	17.34	5.364	-30.62
303.15	1.36	681.78	78.02	283.1	0.00232	1.00232	0.9802	0.1147	0.00389	18.00	5.409	-31.39
308.15	1.21	616.89	66.58	269.9	0.00237	1.00237	0.9804	0.1081	0.00344	18.81	5.463	-32.23

 Λ_{\circ} in (Ω^{-1} .cm².mol⁻¹), Λ in (Ω^{-1} .cm².mol⁻¹) and ΔG_{Λ} in (kJ mol⁻¹).

The enthalpy change (ΔH_A) for CuCl₂.2H₂O in mixed (MeOH+H₂O) at different temperatures were calculated by using Van't Hoff equation (13) [29, 30]:

$$\log K = -\frac{\Delta H}{2.303R} \left(\frac{1}{T}\right) + \text{ constant}$$
(13)

The calculated values of (ΔH_A) and (ΔS_A) for CuCl₂.2H₂O are presented in Table 2.

Table 2. Gibbs free energy change of association (ΔG_A), enthalpy change (ΔH_A) and entropy change (ΔS_A) for CuCl₂.2H₂O with CFZ in dist.H₂O, (40% MeOH+ 60% H₂O) and (60% MeOH+ 40% H₂O) at different temperatures.

	$\Delta G_A (kJ mol^{-1})$			$\Delta H_A (kJ mol^{-1})$			$\Delta S_A (J mol^{-1}K^{-1})$		
Temp(K)	Dist.H ₂ O	(40%MeOH + 60% H2O)	(60%MeOH + 40% H2O)	Dist.H ₂ O	(40% MeOH + 60% H ₂ O)	(60% MeOH + 40% H ₂ O)	Dist.H ₂ O	(40% MeOH + 60% H ₂ O)	(60% MeOH + 40% H2O)
293.15	-30.21	-30.1	-30		13.378	13.759	0.1458	0.1483	0.1492
298.15	-30.91	-30.79	-30.62	10 521			0.1457	0.1481	0.1488
303.15	-31.64	-31.53	-31.39	12.531			0.1457	0.1481	0.1489
308.15	-32.4	-32.33	-32.23				0.1458	0.1483	0.1492

By drawing the relation between log K_A and 1/T, Δ H_A can be calculated from the slope of each line which equal (- Δ H_A/2.303R) as shown in Fig. 2. The entropy change (Δ S_A) for CuCl₂.2H₂O was calculated by using equation (14):

$$\Delta G_A = \Delta H_A - T \Delta S_A \tag{14}$$

Where, (S) is the entropy change of the system.



Fig. 2. Variation of log K_A with l/T (K^{-1}) of CuCl₂₋₂H₂O with CFZ in dist.H₂O, (40 % MeOH+ 60% H₂O) and (60% MeOH+ 40% H₂O).

Calculation of formation parameters for $CuCl_2.2H_2O$ in mixed (MeOH+H₂O) with presence of ligand (Ceftazidime, CFZ).

The values of specific conductance (K_s) for different concentrations of CuCl₂.2H₂O in mixed (MeOH+H₂O) were measured experimentally in the presence of ligand (CFZ) at different temperatures 293.15, 298.15, 303.15 and 308.15 K. Also, the values of molar conductance (Λ_m) can be calculated [22, 23] using equation (1).

By drawing the relation between molar conductance (Λ_m) for CuCl₂.2H₂O with presence of ligand (CFZ) at different temperatures and the molar ratio of metal to ligand [M]/[L] concentrations, Fig. 3 different lines were obtained with breaks indicating the formation of 1:2 and 1:1 (M:L) stoichiometric complexes.



Fig. 3. Variation of the molar conductance $A_m (\Omega^{-1}.cm^2.mol^{-1})$ with [M]/[L] for CuCl₂.2H₂O with CFZ in (a) dist.H₂O (b) (40% MeOH+60% H₂O) (c) (60% MeOH+ 40% H₂O) at (293.15, 298.15, 303.15 and 308.15 K).

The formation constants (K_f) for CuCl₂.2H₂O complexes were calculated for each type of complexes (1:2) and (1:1) (M:L) by using equation (15) [31-49]:

$$K_{f} = \frac{[ML]}{[M][L]} = \frac{\Lambda_{M-}\Lambda_{obs}}{(\Lambda_{obs-}\Lambda_{ML})[L]} \text{ and } [L] = ([L]t - [M]t)\frac{(\Lambda M - \Lambda Obs)}{(\Lambda Obs - \Lambda ML)}$$
(15)

Where, Λ_M is the limiting molar conductance of CuCl₂.2H₂O alone, Λ_{obs} is the molar conductance of solution during titration, Λ_{ML} is the molar conductance of the complex and [L] is the ligand concentration.

The Gibbs free energies change of formation for each stoichiometric complex (ΔG_f) was calculated by using the equation (16):

$$\Delta G_{\rm f} = -2.303 \,\mathrm{RT} \log \mathrm{K}_{\rm f} \tag{16}$$

The obtained values (K_f) for CuCl₂.2H₂O stoichiometric complexes and their calculated ΔG_f values were presented in Table 3 (a, b, c).

Table 3(a). Formation constants (log K_{f}) and Gibbs free energies change of formation (ΔG_{f}) for 1:2 and 1:1 (M/L) of CuCl₂.2H₂O with CFZ in dist.H₂O at (293.15, 298.15, 303.15 and 308.15 K).

Complex ratio (M:L)	Temp. (K)	log K _f	ΔG_{f}
	293.15	2.979	-16.72
(1.2)	298.15	3.012	-17.19
(1.2)	303.15	3.055	-17.73
	308.15	3.124	-18.43
	293.15	3.471	-19.48
(1.1)	298.15	3.527	-20.13
(1.1)	303.15	3.65	-21.19
	308.15	3.679	-21.7

Table 3(b). Formation constants (log K_{f}) and Gibbs free energies change of formation (ΔG_{f}) for 1:2 and 1:1 (M/L) of CuCl₂.2H₂O with CFZ in (40%MeOH+60% H₂O) at (293.15, 298.15, 303.15 and 308.15 K).

Complex ratio (M:L)	Temp. (K)	log K _f	ΔG_{f}
	293.15	2.956	-16.59
(1.2)	298.15	2.995	-17.1
(1.2)	303.15	3.035	-17.61
	308.15	3.1	-18.29
	293.15	3.452	-19.37
(1.1)	298.15	3.496	-19.96
(1.1)	303.15	3.577	-20.76
	308.15	3.631	-21.42

Table 3(c). Formation constants (log K_{f}) and Gibbs free energies change of formation (ΔG_{f}) for 1:2 and 1:1 (M/L) of CuCl₂.2H₂O with CFZ in (60% MeOH+40% H₂O) at (293.15, 298.15, 303.15 and 308.15 K).

Complex ratio (M:L)	Temp. (K)	log K _f	ΔG_{f}
	293.15	2.932	-16.45
(1.2)	298.15	2.979	-17.00
(1.2)	303.15	3.027	-17.57
	308.15	3.062	-18.07
	293.15	3.399	-19.08
(1.1)	298.15	3.459	-19.74
(1.1)	303.15	3.522	-20.44
	308.15	3.568	-21.05

By drawing the relation between log K_f and 1/T, different lines were obtained indicating the formation of 1:2 and 1:1 (M:L) stoichiometric complexes Fig. 4. From the relation between log K_f and 1/T, Δ H_f can be calculated for each type of complexes, from the slope of each line which equal (- Δ H_f/2.303R). The entropy change (Δ S_f) for CuCl₂.2H₂O stoichiometric complexes was calculated [35] for each type of complexes (1:2) and (1:1) (M:L) by using equation (17):

$$\Delta G_{\rm f} = \Delta H_{\rm f} - T \Delta S_{\rm f} \tag{17}$$



Fig. 4. Variation of log K_f with $l/T(K^{-1})$ of CuCl₂ with CFZ in dist. H_2O , (40% MeOH+60% H_2O) and (60% MeOH+40% H_2O).

The calculated values of (ΔH_f) and (ΔS_f) for CuCl₂.2H₂O stoichiometric complexes were presented in Table 4 (a, b, c).

Table 4(a). Gibbs free energy change of formation (ΔG_{θ}), enthalpy change (ΔH_{θ}) and entropy change (ΔS_{θ}) for CuCl₂.2H₂O with CFZ in dist.H₂O at different temperatures.

Temp. (K)	$\Delta G_{f}(kJ mol^{-1})$		ΔH_{f} (kJ mol ⁻¹)		$\Delta S_{f} (J \text{ mol}^{-1} \text{K}^{-1})$	$\Delta S_{f} (J \text{ mol}^{-1} \text{K}^{-1})$		
	(1:2) Complex	(1:1) Complex	(1:2) Complex	(1:1) Complex	(1:2) Complex	(1:1) Complex		
293.15	-16.72	-19.48			0.1156	0.1583		
298.15	-17.19	-20.13	17 10	26.04	0.1153	0.1579		
303.15	-17.73	-21.19	17.18	20.94	0.1152	0.1587		
308.15	-18.43	-21.70			0.1155	0.1578		

Table 4(b). Gibbs free energy change of formation (ΔG_{d}), enthalpy change (ΔH_{d}) and entropy change (ΔS_{d}) for CuCl₂.2H₂O with Ceftazidime in (40% MeOH+60% H₂O) at different temperatures.

Temp. (K)	$\Delta G_{f}(kJ mol^{-1})$		ΔH_{f} (kJ mol ⁻¹)		$\Delta S_{f} (J \text{ mol}^{-1} \text{K}^{-1})$	$\Delta S_{f} (J \text{ mol}^{-1} \text{K}^{-1})$		
	(1:2) Complex	(1:1) Complex	(1:2) Complex	(1:1) Complex	(1:2) Complex	(1:1) Complex		
293.15	-16.59	-19.37			0.1142	0.1418		
298.15	-17.10	-19.96	16.00	22.21	0.1139	0.1414		
303.15	-17.61	-20.76	10.88	22.21	0.1138	0.1417		
308.15	-18.29	-21.42			0.1141	0.1416		

Table 4(c). Gibbs free energy change of formation (ΔG_{d}), enthalpy change (ΔH_{d}) and entropy change (ΔS_{d}) for CuCl₂.2H₂O with CFZ in (60% MeOH+ 40% H₂O) at different temperatures.

Temp. (K)	$\Delta G_{\rm f}({\rm kJ~mol^{-1}})$		ΔH _f (kJ mol ⁻¹)		$\Delta S_{f} (J \text{ mol}^{-1} \text{K}^{-1})$		
	(1:2) Complex	(1:1) Complex	(1:2) Complex	(1:1) Complex	(1:2) Complex	(1:1) Complex	
293.15	-16.45	-19.08			0.1103	0.1351	
298.15	-17.00	-19.74	15.90	20.54	0.1103	0.1351	
303.15	-17.57	-20.44	15.89	20.34	0.1104	0.1352	
308.15	-18.07	-21.05			0.1102	0.1349	

Conclusion

From Conductometric measurements it was observed that, the limiting molar conductances decrease by increasing temperature due to more association. Also, the degree of dissociation $((\alpha))$ increases by increasing temperature due to more solvation. The association parameters do not change clearly by increasing in the percentage of (MeOH+H₂O) mixtures for CuCl₂.2H₂O with CFZ indicating independent solvent behavior in this case. Little effect of different (MeOH+H₂O) mixtures on the complexation thermodynamic parameters for CuCl₂.2H₂O salt with CFZ. Inflections at (0.5, 1) molar ratio was observed indicating the formation of (1:2) and (1:1) M/L complexes. The (1:1) M/L complex has greater formation values for CuCl₂.2H₂O by its interaction with CFZ at all the temperatures used. The negative free energies change, positive enthalpies change and positive entropies change indicate the spontaneous character of the interaction in different percentage of the used mixed solvents.



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