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Table 1

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## Abstract

## Full Text

## CHEMISTRY

Academician A. A. GRINBERG, M. I. GEL’ FMAN

# ON THE QUESTION OF THE STABILITY OF COMPLEX COMPOUNDS OF DIVALENT PLATINUM OF THE DIACIDODIAMMINE TYPE

In previously published works we investigated the stability of certain platinites ($^1$) and compounds of the tetrammine type ($^2$). Comparison of the obtained values of the instability constants shows that replacement of the acid residues $Cl^-$, $Br^-$, and $J^-$ in the inner sphere by molecules of ammonia or fatty amines leads to an increase in the stability of the complex. The next stage of our investigations was the determination of the overall instability constants of certain compounds of the diacidodiammine type—products of partial substitution of the acid residues $Cl^-$ and $J^-$ in the inner sphere of the complex by molecules of ammonia and methylamine.

**Table 1**

Determination of the instability constant

| Conc. of complex, M/l | $[NH_3]$, M/l | $[Cl^-]$, g-ion/l | $E = -E_{Pt} - E_{cal}$, mV | $E_{Pt}$, mV | $-\lg[Pt^{2+}]$ | pKc |
|---|---|---|---|---|---|---|
| **cis-** $[Pt(NH_3)_2Cl_2]$ | **cis-** $[Pt(NH_3)_2Cl_2]$ | **cis-** $[Pt(NH_3)_2Cl_2]$ | **cis-** $[Pt(NH_3)_2Cl_2]$ | **cis-** $[Pt(NH_3)_2Cl_2]$ | **cis-** $[Pt(NH_3)_2Cl_2]$ | **cis-** $[Pt(NH_3)_2Cl_2]$ |
| 0.0023 | $3.9 \cdot 10^{-3}$ | 1.0 | 181 | 429 | 26.6 | 28.8 |
| 0.0033 | $3.7 \cdot 10^{-3}$ | 1.0 | 163 | 411 | 27.2 | 29.5 |
| 0.0036 | $3.7 \cdot 10^{-3}$ | 0.1 | 219 | 467 | 25.3 | 29.6 |
| 0.0027 | $3.7 \cdot 10^{-4}$ | 1.0 | 228 | 467 | 25.0 | 29.2 |
| 0.0030 | $3.7 \cdot 10^{-4}$ | 0.1 | 249 | 497 | 24.2 | 30.5 |
| 0.0064 | $3.8 \cdot 10^{-5}$ | 1.0 | 261 | 509 | 23.8 | 30.4 |
| 0.0018 | $1.8 \cdot 10^{-5}$ | 1.0 | 274 | 522 | 23.4 | 30.1 |
| 0.0093 | $1.8 \cdot 10^{-5}$ | 1.0 | 340 | 588 | 21.1 | 28.7 |
| 0.0033 | $1.8 \cdot 10^{-5}$ | 0.1 | 385 | 633 | 19.5 | 28.6 |
|  |  |  |  | **Average** |  | 29.5 |
| **trans-** $[Pt(NH_3)_2Cl_2]$ | **trans-** $[Pt(NH_3)_2Cl_2]$ | **trans-** $[Pt(NH_3)_2Cl_2]$ | **trans-** $[Pt(NH_3)_2Cl_2]$ | **trans-** $[Pt(NH_3)_2Cl_2]$ | **trans-** $[Pt(NH_3)_2Cl_2]$ | **trans-** $[Pt(NH_3)_2Cl_2]$ |
| 0.0010 | $1 \cdot 10^{-3}$ | 1.0 | 247 | 495 | 24.4 | 27.4 |

| Conc. of complex, M/l | $[NH_3]$, M/l | $[Cl^-]$, g-ion/l | $E = -E_{Pt} - E_{cal}$, mV | $E_{Pt}$, mV | $-\lg[Pt^{2+}]$ | pKc |
|---|---|---|---|---|---|---|
| 0.0007 | $1.5 \cdot 10^{-3}$ | 1.0 | 228 | 476 | 25.0 | 27.5 |
| 0.0010 | $1 \cdot 10^{-3}$ | 0.1 | 284 | 532 | 23.0 | 28.0 |
| 0.0010 | $1 \cdot 10^{-4}$ | 1.0 | 247 | 495 | 24.4 | 29.4 |
| 0.0010 | $1.3 \cdot 10^{-4}$ | 1.0 | 252 | 500 | 24.2 | 29.0 |
| 0.0010 | $8 \cdot 10^{-5}$ | 0.1 | 318 | 566 | 21.9 | 29.1 |
| 0.0009 | $1.8 \cdot 10^{-5}$ | 1.0 | 325 | 593 | 21.0 | 27.6 |
| 0.0010 | $1.8 \cdot 10^{-5}$ | 0.1 | 382 | 630 | 19.7 | 28.3 |
| 0.0010 | $5 \cdot 10^{-6}$ | 1.0 | 371 | 619 | 20.0 | 27.6 |
| | | | | **Average** | | 28.4 |

There are no data on the overall instability constants of diammines in the literature. However, it was to be expected that the instability constant of a diammine should occupy some intermediate position between the values of the constants of the corresponding platinite and tetrammine. Of great interest is the question of whether geometrically isomeric platodiammines can be characterized by instability constants. As has already been shown, for the process of dissociation of a chlorine ion (or bromine ion) from the inner sphere of compounds of the type $[Pt(NH_3)_2X_2]$ this is quite possible, since the corresponding process is completely reversible with preservation of the configuration of the complex. However, detachment of ammonia or amine molecules leads to the decomposition of definite stereochemical configurations and creates the fundamental possibility of isomerization occurring. Such isomerization does not occur in aqueous solution only because of the inertness of the platinum—ammonia or platinum—amine bonds. The dissociation equilibrium of ammine complexes is established extremely slowly or is not established at all. We had previously found that in the presence of a platinized electrode equilibrium is established comparatively rapidly. This fact indicates that platinum black catalytically accelerates the dissociation of ammine complexes. It could be expected that in the presence of platinum black in solution, isomerization occurs. Therefore, in addition to potentiometric studies, we carried out experiments with the aim of detecting isomerization.

## Experimental Part

### 1. Determination of overall instability constants.

The diammines studied were synthesized by methods described in the literature. The composition of all compounds was established by chemical analysis. The concentration of divalent platinum ions in solution was determined by measuring the emf of the cells:

$$\overset{+}{\mathrm{Pt}}\,[\mathrm{PtA_2X_2}],\ \mathrm{A},\ \mathrm{X^-}\,\|\,\mathrm{KCl},\ \mathrm{Hg_2Cl_2}\,|\,\mathrm{Hg}^-$$

$c_k$ $c_A$ $c_X$ saturated

The measurement procedure did not differ from that described in previous works. All measurements were carried out at a temperature of 18° and at an ionic strength equal to unity.

A definite concentration of $A$ and $X^-$ in solution was produced by dissolving $NH_4Cl$, $CH_3NH_3Cl$, KJ, or by adding free bases. To determine the concentration of the base in solution, the pH was measured after equilibrium had been established.

From the values of the electrode potentials, the concentration of $Pt^{2+}$ ions was calculated (taking the normal potential $Pt^{2+}/Pt^0$ as equal to 1.2 V), and then the values of the overall instability constants were calculated from the formula:

$$K^c = \frac{|\mathrm{Pt}^{2+}|\,|\mathrm{A}|^2\,|\mathrm{X}^-|^2}{|\mathrm{PtA_2X_2}|}$$

The experimental data and calculations of the instability constants for various compounds are given in Tables 1-4.

**Table 2**

**Determination of the instability constant**

| Concentration of complex, mol/L | $[CH_3NH_2]$, mol/L | $[Cl^-]$, g-ion/L | $E - E_{Pt} - E_{cal}$ | $E_{Pt}$ | $-\lg[Pt^{2+}]$ | $pK^c$ |
|---|---|---|---|---|---|---|
| **cis-** $[Pt(CH_3NH_2)_2Cl_2]$ | **cis-** $[Pt(CH_3NH_2)_2Cl_2]$ | **cis-** $[Pt(CH_3NH_2)_2Cl_2]$ | **cis-** $[Pt(CH_3NH_2)_2Cl_2]$ | **cis-** $[Pt(CH_3NH_2)_2Cl_2]$ | **cis-** $[Pt(CH_3NH_2)_2Cl_2]$ | **cis-** $[Pt(CH_3NH_2)_2Cl_2]$ |
| 0.0559 | $1.6 \cdot 10^{-3}$ | 1.0 | 177 | 425 | 26.8 | 30.9 |
| 0.0026 | $2.5 \cdot 10^{-4}$ | 1.0 | 224 | 472 | 25.1 | 29.8 |
| 0.0032 | $1.2 \cdot 10^{-4}$ | 0.2 | 293 | 541 | 22.8 | 29.5 |
| 0.0029 | $3 \cdot 10^{-5}$ | 1.0 | 266 | 514 | 23.7 | 30.2 |
| 0.0039 | $3 \cdot 10^{-5}$ | 0.2 | 281 | 529 | 23.1 | 31.1 |
| 0.0040 | $5 \cdot 10^{-6}$ | 1.0 | 292 | 540 | 22.8 | 31.0 |
| 0.0029 | $5 \cdot 10^{-6}$ | 0.2 | 333 | 581 | 21.4 | 30.9 |
| 0.0095 | $1 \cdot 10^{-6}$ | 1.0 | 333 | 581 | 21.4 | 31.4 |
| 0.0015 | $0.7 \cdot 10^{-6}$ | 1.0 | 363 | 611 | 20.4 | 30.0 |
| Average | Average | Average | Average | Average | Average | 30.5 |

| Concentration of complex, mol/L | $[CH_3NH_2]$, mol/L | $[Cl^-]$, g-ion/L | $E - E_{Pt} - E_{cal}$ | $E_{Pt}$ | $-\lg[Pt^{2+}]$ | $pK^c$ |
|---|---|---|---|---|---|---|
| **trans-** $[Pt(CH_3NH_2)_2Cl_2]$ | **trans-** $[Pt(CH_3NH_2)_2Cl_2]$ | **trans-** $[Pt(CH_3NH_2)_2Cl_2]$ | **trans-** $[Pt(CH_3NH_2)_2Cl_2]$ | **trans-** $[Pt(CH_3NH_2)_2Cl_2]$ | **trans-** $[Pt(CH_3NH_2)_2Cl_2]$ | **trans-** $[Pt(CH_3NH_2)_2Cl_2]$ |
| 0.0027 | $1 \cdot 10^{-3}$ | 1.0 | 167 | 415 | 27.1 | 30.5 |
| 0.0027 | $2.5 \cdot 10^{-3}$ | 0.2 | 174 | 422 | 26.8 | 30.8 |
| 0.0025 | $0.8 \cdot 10^{-4}$ | 1.0 | 245 | 493 | 24.4 | 30.0 |
| 0.0024 | $0.8 \cdot 10^{-4}$ | 0.2 | 275 | 523 | 23.3 | 30.3 |
| 0.0036 | $1.1 \cdot 10^{-4}$ | 1.0 | 200 | 448 | 26.0 | 31.5 |
| 0.0036 | $2.5 \cdot 10^{-4}$ | 0.1 | 237 | 485 | 24.7 | 31.5 |
| 0.0024 | $3 \cdot 10^{-5}$ | 1.0 | 230 | 478 | 25.0 | 31.4 |
| 0.0025 | $3 \cdot 10^{-5}$ | 0.2 | 274 | 522 | 23.1 | 30.8 |
| 0.0010 | $1 \cdot 10^{-6}$ | 1.0 | 355 | 603 | 20.6 | 29.6 |
| 0.0053 | $0.7 \cdot 10^{-6}$ | 1.0 | 368 | 616 | 20.1 | 30.2 |
| Average | Average | Average | Average | Average | Average | 30.70 |

## 2. Isomerization of Peyrone' s chloride.

Experiments aimed at detecting the mutual conversion of isomeric diammines were carried out with Peyrone' s chloride, cis-$Pt(NH_3)_2Cl_2$, and Reiset' s second-base chloride, trans-$Pt(NH_3)_2Cl_2$. The configuration of both isomers was checked by means of the thiourea reaction of N. S. Kurnakov.

The procedure for carrying out the experiments was as follows. A definite amount of the diammine was dissolved with heating in 100 ml of water, the solution was cooled to room temperature, and a certain amount of platinum black was added to it. In individual experiments, besides the black, a platinized platinum electrode was immersed in the solution. The solution with the black was kept in a closed flask and stirred periodically. Then the solution was filtered from the platinum and evaporated on a water bath to a small

volume. The thiourea reaction was carried out with this solution. The reaction product was isolated from the solution, dried to constant weight, and analyzed for platinum content. From the platinum content in the product of the thiourea reaction it was determined which isomer was present in the solution, or what the composition of the mixture of isomers was. In addition, the reaction product was examined under a microscope.

**Table 3**

Determination of the instability constant

| Complex concentration, mol/l | $[NH_3]$, mol/l | $[J^-]$, g-ion/l | $E -$ $E_{Pt}$—$E_{\text{cal}}$, mV | $E_{Pt}$, mV | $-\lg[Pt^{2+}]$ | $pK^c$ |
|---|---|---|---|---|---|---|
| | | | **cis-** $[Pt(NH_3)_2J_2]$ | | | |
| 0.0013 | $1.05 \cdot 10^{-1}$ | 1.0 | −22 | 226 | 33.6 | 32.7 |
| 0.0017 | $1.0 \cdot 10^{-1}$ | 1.0 | 6 | 242 | 33.1 | 32.3 |
| 0.0017 | $1.05 \cdot 10^{-1}$ | 0.1 | 30 | 278 | 31.8 | 33.0 |
| 0.0012 | $1.0 \cdot 10^{-2}$ | 1.0 | 42 | 290 | 31.4 | 32.5 |
| 0.0009 | $1.3 \cdot 10^{-2}$ | 0.1 | 57 | 305 | 30.9 | 33.7 |
| 0.0018 | $1.0 \cdot 10^{-3}$ | 1.0 | 63 | 311 | 30.7 | 34.0 |
| 0.0010 | $1.0 \cdot 10^{-3}$ | 0.1 | 110 | 358 | 29.0 | 34.0 |
| 0.0013 | $1.0 \cdot 10^{-4}$ | 1.0 | 86 | 334 | 29.9 | 35.0* |
| 0.0012 | $1.0 \cdot 10^{-4}$ | 0.1 | 124 | 372 | 28.6 | 35.7* |
| 0.0013 | $1.0 \cdot 10^{-5}$ | 1.0 | 74 | 322 | 30.3 | 37.4* |
| 0.0017 | $1.0 \cdot 10^{-5}$ | 0.1 | 114 | 362 | 28.9 | 38.1* |
| | | | | | Average | 33.2 |
| | | | **trans-** $[Pt(NH_3)_2J_2]$ | | | |
| 0.0007 | $1.3 \cdot 10^{-1}$ | 1.0 | −8 | 240 | 33.1 | 31.8 |
| 0.0007 | $1.3 \cdot 10^{-1}$ | 0.1 | 34 | 282 | 31.7 | 32.3 |
| 0.0009 | $1.0 \cdot 10^{-2}$ | 1.0 | 42 | 290 | 31.4 | 32.4 |
| 0.0008 | $0.6 \cdot 10^{-2}$ | 0.1 | 87 | 335 | 29.8 | 33.1 |
| 0.0009 | $0.6 \cdot 10^{-3}$ | 1.0 | 124 | 372 | 28.6 | 32.0 |
| 0.0006 | $1.4 \cdot 10^{-3}$ | 0.1 | 148 | 396 | 27.8 | 32.4 |
| 0.0008 | $0.6 \cdot 10^{-3}$ | 0.1 | 156 | 404 | 27.5 | 32.8 |
| 0.0007 | $1.4 \cdot 10^{-4}$ | 1.0 | 129 | 377 | 28.4 | 33.0 |
| 0.0009 | $1.1 \cdot 10^{-4}$ | 1.0 | 130 | 378 | 28.4 | 33.4 |
| 0.0008 | $0.6 \cdot 10^{-4}$ | 1.0 | 132 | 380 | 28.3 | 33.6 |
| | | | | | Average | 32.7 |

* The latter *pK* values show that at low ammonia concentrations the values of the instability constant calculated for the diamine do not remain constant when the ammonia concentration is varied. This is associated with the replacement of ammonia molecules in the inner sphere of the complex by acidic residues. At the same time, intense coloration of the solution is observed. The latter *pK* values were not taken into account in calculating the average value.

The data from the study of Peyrone’ s chloride are given in Table 5. When the crystals were examined under a microscope, along with platelets characteristic of the tetrathiourea complex, needle-shaped crystals were clearly visible.

In parallel, an experiment was carried out with Peyrone's chloride in the absence of platinum black. After 2.5 months the thiourea reaction gave a product containing 34.7% Pt, which corresponds to conversion of 5%, or 0.007 g of Peyrone' s chloride.

At the same time, attempts were made to detect isomerization of Reiset' s second-base chloride. However, even after the solution had been in contact with platinum black for five months, the product of the thiourea reaction contained 43.2% platinum, which corresponds to pure trans-$[Pt(NH_3)_2Cl_2]$.

## Discussion of the results

The study of the stability of complexes of the diacidodiamine type is complicated by the fact that, in the presence of an excess of molecules $A$ and ions $X^-$, substitution reactions inevitably proceed in solution. Since this could not be taken into account in calculating the instability constants, the values obtained can be regarded only as very approximate. Nevertheless, from comparison of the $pK^c$ values certain conclusions can be drawn.

Diamines, in stability, occupy an intermediate position between the corresponding platinites and tetrammines. The stability of the iodo derivatives is higher than that of the chloro derivatives, and the methylamine complexes are stronger than the ammonia complexes. Geometrically isomeric diamines are close in stability. The study of isomerization showed that Peyrone' s chloride gradually turns into a product that gives the trans reaction according to Kurnakov. Such a product may be the ion $[Pt(NH_3)_3Cl]^+$ or Reiset' s second-base chloride. Since analysis of the conversion product corresponds to the empirical composition $Pt(NH_3)_2Cl_2$, the conversion products can only be trans-$Pt(NH_3)_2Cl_2$ or the dimer $[Pt(NH_3)_3Cl][PtNH_3Cl_3]$. In either case, of course, isomerization of the initial product takes place. Final clarification of the nature of the isomerization product is currently under investigation. Conversion of Reiset' s second-base chloride into the cis isomer does not occur under the conditions studied.

Since cis-platodiamines undergo transformations under the conditions of measurement, the values of the "instability constants" obtained for these systems are, of course, only conditional. A thermodynamically reversible process under the conditions studied takes place only in the case of the trans isomers. However, the fact that the potentials of the platinum electrode in solutions of isomeric diammines are quite close indicates an approximately identical strength of these compounds. In accordance with the thermochemical data of Chernyaev, Palkin, and Sokolov, the trans isomer $Pt(NH_3)_2Cl_2$ should be somewhat more stable than the cis isomer (the heat effect of isomerization at 70 °C is 3 kcal). The circumstance that in some systems the average value of pK for the cis forms is somewhat greater than for the trans forms is most likely explained by secondary reactions of the type:

**Table 4**

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Determination of the instability constant

| Conc. of complex, M/l | $[CH_3NH_2]$, M/l | $[J^-]$, g-ion/l | $E = -E_{Pt} - E_{cal}$, mV | $E_{Pt}$, mV | $-\lg[Pt^{2+}]$ | pKc |
|---|---|---|---|---|---|---|
| **cis-** $[Pt(CH_3NH_2)_2J_2]$ | **cis-** $[Pt(CH_3NH_2)_2J_2]$ | **cis- $[Pt(CH_3NH_2)_2J_2]$ | **cis-** $[Pt(CH_3NH_2)_2J_2]$ | **cis-** $[Pt(CH_3NH_2)_2J_2]$ | **cis-** $[Pt(CH_3NH_2)_2J_2]$ | **cis-** $[Pt(CH_3NH_2)_2J_2]$ |
| 0,0017 | $1,2 \cdot 10^{-2}$ | 1,0 | −14 | 234 | 33,4 | 34,4 |
| 0,0018 | $1,2 \cdot 10^{-2}$ | 0,1 | 42 | 290 | 31,4 | 34,5 |
| 0,0016 | $1,2 \cdot 10^{-3}$ | 1,0 | 29 | 277 | 31,8 | 35,0 |
| 0,0018 | $1,2 \cdot 10^{-3}$ | 0,1 | 74 | 322 | 30,3 | 35,4 |
| 0,0019 | $1,2 \cdot 10^{-4}$ | 1,0 | 114 | 362 | 29,0 | 34,1 |
| 0,0017 | $1,2 \cdot 10^{-4}$ | 0,1 | 166 | 414 | 27,1 | 34,2 |
| 0,0016 | $1,2 \cdot 10^{-5}$ | 1,0 | 134 | 382 | 28,2 | 35,2 |
| 0,0018 | $2,1 \cdot 10^{-5}$ | 1,0 | 163 | 411 | 27,2 | 34,0 |
| 0,0018 | $2,1 \cdot 10^{-5}$ | 0,1 | 167 | 415 | 27,1 | 35,8 |
| | | | | | Average | 34,7 |
| **trans-** $[Pt(CH_3NH_2)_2J_2]$ | **trans-** $[Pt(CH_3NH_2)_2J_2]$ | **trans-** $[Pt(CH_3NH_2)_2J_2]$ | **trans-** $[Pt(CH_3NH_2)_2J_2]$ | **trans-** $[Pt(CH_3NH_2)_2J_2]$ | **trans-** $[Pt(CH_3NH_2)_2J_2]$ | **trans-** $[Pt(CH_3NH_2)_2J_2]$ |
| 0,0012 | $1 \cdot 10^{-1}$ | 1,0 | −48 | 200 | 34,5 | 33,6 |
| 0,0010 | $1 \cdot 10^{-1}$ | 0,1 | −43 | 205 | 34,3 | 35,3 |
| 0,0010 | $1,4 \cdot 10^{-2}$ | 1,0 | −1 | 247 | 32,9 | 33,6 |
| 0,0010 | $1,4 \cdot 10^{-2}$ | 0,1 | 28 | 276 | 31,9 | 34,6 |
| 0,0014 | $8 \cdot 10^{-3}$ | 1,0 | 0 | 248 | 32,8 | 34,2 |
| 0,0016 | $8 \cdot 10^{-3}$ | 0,1 | 39 | 287 | 31,5 | 34,8 |
| 0,0010 | $1 \cdot 10^{-3}$ | 1,0 | 62 | 310 | 30,7 | 33,7 |
| 0,0011 | $1 \cdot 10^{-3}$ | 0,1 | 105 | 353 | 29,1 | 34,1 |
| 0,0016 | $1,4 \cdot 10^{-4}$ | 1,0 | 123 | 371 | 28,6 | 33,5 |
| 0,0012 | $8 \cdot 10^{-5}$ | 0,1 | 166 | 414 | 27,1 | 34,4 |
| | | | | | Average | 34,2 |

secondary reactions of the type:

$$\text{cis-}[Pt(NH_3)_2Cl_2] + NH_4Cl \rightleftarrows NH_4[PtNH_3Cl_3] + NH_3 \quad \text{and}$$

$$\text{cis-}[Pt(NH_3)_2Cl_2] + HN_3 \rightleftarrows [Pt(NH_3)_3Cl]\,Cl$$

Such reactions do indeed occur with respect to the cis forms.

**Table 5**

Isomerization of Peyrone’ s chloride

| No. | Amount of Peyrone, g | Amount of Pt black, g | Temp., °C | Duration of experiment | Pt in the thiourea reaction product, % | Peyrone isomerized, g | Peyrone isomerized, % |
|---|---|---|---|---|---|---|---|
| 1 | 0,14 | 0,1 | 18 | 18 days | 35,4 | 0,02 | 14 |
| 2 | 0,14 | 0,1 | 18 | 25 days; 30 days | 35,8 | 0,025 | 18 |
| 3 | 0,14 | 0,1 | 18 | 10 h | 36,2 | 0,032 | 23 |
| 4 | 0,14 | 0,1 | 18 | 75 days | 37,8 | 0,056 | 40 |
| 5* | 0,3 | 1,0 | 18 | 10 days | 35,2 | 0,039 | 13 |
| 6* | 0,41 | 1,0 | 18 | 20 days | 36,6 | 0,1 | 26 |

* In the presence of a Pt electrode.

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*Note: Figure translations are in progress. See original paper for figures.*

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