refined by the block-diagonal least-squares method, using anisotropic temperature factors for all the nonhydrogen atoms. The weighting scheme $w = [\sigma^2(F) + 0.000625F^2]^{-1}$ was employed, and the final R value was 0.062 for 2257 independent reflections [$\sin\theta/\lambda \leq 0.76$, $|F| \geq 3\sigma$, without absorption correction].

The polyhedral model of the heteropolyanion $PV_{14}O_{42}^{9-}$ is shown in Figure 1. The central PO_4 tetrahedron shares its oxygen atoms with four V_3O_{13} groups, each of which is made up of three edge-sharing VO_6 octahedra, defining the well-known α-Keggin structure. There are 14 possible sites which are "pits" on a Keggin molecule for further coordination of vanadium atoms (six A sites, four B sites, and four C sites as shown in Figure 1). The last two VO units occupy trans-located A sites, forming trigonal bipyramidal caps. The geometry of the anion is shown in Figure 2, which includes some bond parameters.

In the region of pH 1.3–4.0, each of the 40.5-MHz ^{31}P NMR spectra of P/V 1:4 solutions showed only a singlet peak at −0.2 to +0.7 ppm (with respect to 85% H_3PO_4 as external standard) accompanied by a phosphate ion peak.

Such a Keggin structure with the capping 5-coordinated metal atoms has not been previously observed in the structures of polyanions. Compared with the polyanions of Mo(VI) and W(VI), a high negative charge will prevent the formation of Keggin-type anion $PV_{12}O_{40}^{15-}$ whereas bicapped Keggin anion $PV_{14}O_{42}^{9-}$ is stabilized by the two capping VO^{3+} units. The results of this work suggest the possibility of a capped Keggin ion series $PV_{12+n}O_{40+n}^{-(15-3n)}$ (n = 1–6).[5]

Acknowledgment. We are grateful to Dr. K. Sato for measuring ^{31}P NMR spectra.

Supplementary Material Available: Three tables listing atomic positions, thermal parameters, and structure factors (18 pages). Ordering information is given on any current masthead page.

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(5) Very recently we have obtained a crystal of heteropolyvanadate containing arsenic as a heteroatom whose structure seems to be a capped Keggin type (n = 14 or 15?). A crystallographic analysis is in progress.

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Received May 27, 1980

Olefin Metathesis Reaction: Characterization of an Active Catalyst Precursor, $CH_3WOCl_3\cdot O(C_2H_5)_2$, from the $WOCl_4$–$(CH_3)_2Mg$ Reaction

Sir:

Earlier[1] we had shown that $WOCl_4$ in combination with main group metal alkyls provided a consistent source of an exceedingly active catalyst precursor for the olefin metathesis reaction.[2] With the report of the synthesis, isolation, and characterization of CH_3WOCl_3,[4] we set out to identify the precursor and the intermediate(s) in the $WOCl_4$-based olefin metathesis reaction. We describe here the character of the $(CH_3)_2Mg$–$WOCl_4$ reaction, the spectroscopic and complex chemical properties of methyltungsten oxytrichloride–diethyl etherate complexes, and the catalytic properties of these methyltungsten complexes.

Riess and Santini[4] described the isolation of unsolvated CH_3WOCl_3 from the reaction of $(CH_3)_2Mg$ and $WOCl_4$ in a pentane–diethyl ether medium.[5,6] After an intensive examination of this reaction under the precise Riess and Santini conditions, and also widely varying conditions, we report the following. Reaction between $(CH_3)_2Mg$ and $WOCl_4$ did not proceed until temperatures of −30 to −20 °C were attained.[7] At minimal reaction temperatures, methane and methyl chloride were formed in addition to small amounts of ethane, ethylene, and propylene, with no detectable CH_3OH or H_2.[8] A major[10] coproduct was a green insoluble solid that had no hydrolyzable W–CH_3 or W–OCH_3 function, contained W, Cl, and Mg, and was catalytically inactive.[11a] The soluble fraction was isolated as red, apparently[11e] single crystals at low temperatures (<−30 °C).[12] These crystals contained tungsten and chlorine in the ratio of 1:2.93 and diethyl ether, but no magnesium; maximal yields of the isolated crystals were 45%. The extraordinary thermal reactivity precluded a normal high-precision analysis of all elements.[12] Nevertheless, the definition of the W/Cl ratio and the following spectroscopic studies establish these crystals to be an etherate of CH_3WOCl_3.[11f]

Red crystals of $CH_3WOCl_3\cdot O(C_2H_5)_2$ derived from a $WOCl_4$–$Mg(^{13}CH_3)_2$[11d] reaction were examined by ^{13}C and ^{1}H NMR as a function of temperature.[12] At low temperatures, there were

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(1) M. T. Mocella, R. Rovner, and E. L. Muetterties, *J. Am. Chem. Soc.*, **98**, 4689 (1976).

(2) Especially active was the product from alkylaluminum chlorides.[1] Products from WCl_6 and main group metal alkyls were shown to be inactive except under conditions wherein small amounts of air were admitted to the reaction system.[1] The reaction system of WCl_6, $[(C_2H_5)_xAlCl_{3-x}]_2$, and ethanol is an alternative source of an active olefin metathesis catalyst.[3]

(3) N. Calderon, E. A. Ofstead, J. P. Ward, W. A. Judy, and K. W. Scott, *J. Am. Chem. Soc.*, **90**, 4133 (1968).

(4) C. C. Santini-Scampucci and J. G. Reiss, *J. Organomet. Chem.*, **73**, C13 (1974); *J. Chem. Soc., Dalton Trans.*, 196 (1976).

(5) C. Santini, Ph.D. Thesis, Institut de Mathematiques and Sciences Physiques, Nice, France (1973).

(6) We are indebted to Professor Riess and to Dr. Santini for detailed information. Unfortunately, the original NMR spectral data for unsolvated CH_3WOCl_3 could not be obtained for precise comparison. They reported J_{H-W} = 3 Hz for CH_3WOCl_3.

(7) Variations of temperature, ratio of pentane to diethyl ether solvent, reactant ratios, and modes of addition were explored in a glass high-vacuum system wherein all operations of filtration, gas-phase analysis, and recrystallization could be performed quantitatively as a function of temperature.

(8) With $(CH_3)_2Mg$ to $WOCl_4$ reactant ratios of 0.5:1.0, the percentages of methyl equivalents in the hydrocarbons formed at reaction temperatures were CH_4 (3.2%), CH_3Cl (10.9%), C_2H_6 (2.7%), C_2H_4 (0.2%), and C_3H_6 (~0.1%). These hydrocarbons when generated from the $(CD_3)_2Mg$ reaction were fully deuterated with the exception of methane, which contained ~30% CD_3H. CD_3H formation may have been due to protons present on the surface on the Pyrex vessel.[9]

(9) Hydrogen incorporation was observed in the alkanes produced from the decomposition of tantalum perdeuterioalkyls when Pyrex vessels (flamed out under vacuum) were used but not when Vycor vessels (flamed out under vacuum) were employed: R. R. Schrock and J. D. Fellmann, *J. Am. Chem. Soc.*, **100**, 3359 (1978).

(10) 90% of the tungsten was present in this material with reactant ratios of $(CH_3)_2Mg$ to $WOCl_4$ of 1:1. This yield dropped to the 55–65% level at reactant ratios of $(CH_3)_2Mg$ to $WOCl_4$ of 0.5:1.0. The other product was crystalline CH_3WOCl_3 etherate.

(11) (a) This green solid contained W, Cl, and Mg in the ratios of 1:3.46:0.65 (typical analysis). (b) Qualitatively, some of our observations did not coincide with those of Santini and Riess. Their insoluble product was described as brown and CH_3WOCl_3 as yellow; the only yellow species we observed was $WOCl_4[O(C_2H_5)_2]$. Our soluble tungsten products invariably contained diethyl ether;[11c] Riess and Santini pointedly described their CH_3WOCl_3 as ether free. Their reported optimal conditions of stoichiometry diverged from our findings: Our optimal yields of methyltungsten complexes were realized at $WOCl_4/(CH_3)_2Mg$ ratios of 2:1. Extensive variations of these procedures were examined over a 3-year period, but we were unable to achieve the Riess–Santini accomplishments. Reaction of the methyl–tungsten complexes produced in our research with strong donor molecules led to formation of green, reduced tungsten species. (c) Quantitative gravimetric studies with $WOCl_4$ established that this oxychloride formed a 1:1 etherate isolable as a crystalline solid, but high-vacuum pumping converted the crystals to an ether-free form at 20 °C in about 2 h. At −30 °C, evacuation removed only about 2% of the ether from the solid etherate at −30 °C in ~1 h. Because of the thermal reactivity of $CH_3WOCl_3\cdot O(C_2H_5)_2$[12], ether removal by evacuation was not feasible. (d) The ^{13}C content was 18%. (e) The CH_3WOCl_3 etherate was purified by low-temperature crystallization to give large red needles. Visually, the crystals appeared to be single crystals. Attempts to obtain X-ray diffraction data at low temperatures failed in the processes of mounting the crystals in capillaries, where, because of the extreme thermal reactivity of the crystals, inadvertant temperature rises led to loss of crystallinity.[12] (f) The ratio of ether to tungsten in the complex was not established but was assumed to be 1:1 by analogy to $WOCl_4[O(C_2H_5)]$.

(12) These crystals decomposed rapidly at 0–20 °C (s), slowly at −30 °C (min), detectably at −78 °C (h), and undetectably at −197 °C. In solution, the decomposition was slow at 20 °C.

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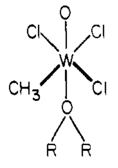
three $^{13}C\{^1H\}$ resonances at 101.5, 101.1, and 86.6 ppm [referenced to $(CH_3)_4Si$] of respective relative intensities 1:0.2:1.5, with the two intense resonances showing tungsten satellites, $J_{^{13}C-^{183}W} = 48$ Hz. In the ^{13}C spectrum, the two intense resonances were quartets, $J_{^{13}C-H} = 132$ Hz, clearly establishing the presence of W–CH_3 groups. Also detectable in the ^{13}C spectrum were two broad resonances at 65.6 and 12.4 ppm assignable to the CH_2 and CH_3 groups of diethyl ether. The ^{13}C spectra were temperature dependent. As the temperature was raised, the 101.5-ppm resonance broadened, and a new resonance at 103.7 ppm grew in. At −30 °C, the resonances at 101.5 and 103.7 ppm were of equal intensity, and both were binomial quartets in the ^{13}C spectrum. With further temperature increase, the 103.7-ppm resonance sharpened so that tungsten satellites became clearly evident, $J_{^{13}C-^{183}W} = 39$ Hz, and the other resonances decreased in intensity and then disappeared; these changes were reversible with temperature decrease.[12] The ^1H NMR spectrum of the methyltungsten complexes derived from $Mg(^{13}CH_3)_2$ was complex because the W–CH_3 resonances overlapped the CH_2 resonance of the complexed diethyl ether. At low temperatures, there were two readily distinguishable W–CH_3 singlets at 3.25 and 1.94 ppm, each with ^{13}C and ^{183}W satellites, $J_{H-^{13}C} = 132$ Hz and $J_{H-^{183}W} = 5$ Hz. The spectra were also temperature dependent, and correlations with the ^{13}C spectra could be made. No NMR evidence for W–CH_2 or W–H species was obtained.

The multiplicity of ^{13}C NMR resonances for the $^{13}CH_3WOCl_3$–ether complex may be explained in terms of isomeric solution species at low temperatures. Of the five possible isomers, the predominant one should be **1** by analogy to stereochemistry **2** established for the associated state of crystalline $WOCl_4$. The temperature dependence of the ^{13}C spectra is ascribed to a shift in equilibrium (eq 1), with the rates of ether dissociation and association low with respect to the NMR time scale. This interpretation is fully consistent with the observed increase in free ether (methylene) resonance intensities with temperature increase.



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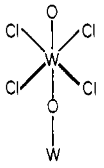


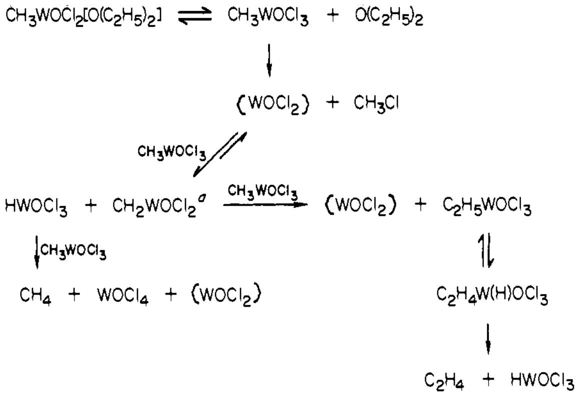
2

$$CH_3WOCl_3{\cdot}O(C_2H_5)_2 \rightleftharpoons CH_3WOCl_3 + O(C_2H_5)_2 \quad (1)$$

Thermal decomposition of crystalline CH_3WOCl_3 etherate at 20 °C primarily produced methyl chloride, methane, polymethylene, and a reduced tungsten oxychloride; minor products were ethane, ethylene, and propylene. All hydrocarbons with the exception of polymethylene were analyzed by gas chromatography (GC) and GC/MS (mass spectrometry); the polymethylene was characterized by ^{13}C NMR, following the products produced from $^{13}CH_3WOCl_3$. The relative proportions of CH_4 and CH_3Cl were dependent upon the state of the methyl–tungsten complex: the ratio of CH_3Cl to CH_4 was ~1.5:1.0 and 0.2:1.0 for the solution and solid states, respectively. In the solution-state decomposition of CD_3WOCl_3, methyl chloride and all the hydrocarbon products were fully deuterated, with the exception of methane which contained ~10% CD_3H.[9]

Decomposition of a mixture of CH_3WOCl_3 and CD_3WOCl_3, each prepared and isolated separately as red crystals, in solution at 20 °C produced a surprising pattern of deuterated hydrocarbons.[13] Methyl chloride consisted only of CH_3Cl and CD_3Cl whereas methane contained comparable amounts of CH_4, CH_3D, CD_3H, and CD_4, as well as much smaller quantities of CH_2D_2. Ethylene was comprised of a nearly *statistical* mixture of all isotopic species from d_0 to d_4 and thus must be generated in a reaction pathway separate from the methane and methyl chloride pathways. Ethane consisted solely of C_2H_6 and C_2D_6. This ethane, a trace product, must be produced in a reaction uncoupled from those producing methane and ethylene, probably from trace amounts of $(CH_3)_2WOCl_2$ and $(CD_3)_2WOCl_2$.[14] All methyl chloride elimination must follow a reaction sequence separate from the methane formation reaction. We suggest that methyl chloride formation is the initiating step of a chain reaction (Scheme I) involving $WOCl_2$ as the chain-propagating species and CH_2WOCl_2 as the metathesis intermediate (vide infra).

Scheme I



[a] Metathesis intermediate.

Thermal decomposition of benzene solutions of $CH_3WOCl_3{\cdot}O(C_2H_5)_2$ in the presence of an olefin initiated catalytic metathesis of the olefin. Thus, 2-pentene was converted to 2-butene and 3-hexene in equimolar amounts. In addition, propylene and 1-butene were formed in a 1:1 molar ratio and in molar amounts comparable to the moles of CH_3WOCl_3 used. These two olefins consisted solely of the $C_3H_4D_2$ and $C_4H_6D_2$ isotopic species when the methyltungsten complex was $CD_3WOCl_3{\cdot}O(C_2H_5)_2$.[16] It is important to note that formation of ethylene from the thermal decomposition of CH_3WOCl_3 was completely suppressed by the presence of an olefin.

Our studies unambiguously establish that $CH_3WOCl_3{\cdot}O(C_2H_5)_2$ is an olefin metathesis catalyst precursor and that a CH_2 fragment of the CH_3–W complex is incorporated into the olefin metathesis products, the first demonstration of the incorporation of a CH_2 fragment, from a characterized methyl–transition-metal complex, in olefin metathesis reaction products. The metathesis intermediate is proposed to be CH_2WOCl_2 (Scheme I).[17,18] Decomposition of CH_3WOCl_3 is unquestionably complex, with three major "reductive elimination" products, methyl chloride, polymethylene, and methane.

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(13) We accounted for about 50% of the CH_3 equivalents derived from CH_3WOCl_3, exclusive of the polymethylene product. A quantitative measure of this latter product was not obtained.

(14) Methyltungsten complexes, detected by NMR, equilibrated CH_3 groups at a rate fast with respect to time of mixing (20 °C). None of these resonances, therefore, can be representative of a $(CH_3)_2W$ species.

(15) Decomposition of CH_3WOCl_3 is atypical of methyl–transition-metal complexes in that CH_3Cl is a major product although the remaining products of CH_4 and small amounts of ethane, ethylene, and propylene are typical of CH_3–M complex decompositions. Polymethylene formation may well have been missed in some studies since these studies have never accounted for more than 50% of the CH–M carbon atoms. Observations of polymethylene formation from methyl–transition compound decomposition are CH_3TiCl_3 [Hn. DeVries, *Recl. Trav. Chim. Pays-Bas*, **80**, 866 (1961)] and $W(CH_3)_6$ [A. J. Shortland and G. Wilkinson, *J. Chem. Soc., Dalton Trans.*, 872 (1973)].

(16) R. H. Grubbs and C. R. Hoppin [*J. Chem. Soc., Chem. Commun.*, 634 (1977)] have demonstrated that WCl_6 and $(CD_3)_4Sn$ initiate olefin metathesis with early production of a CD_2-labeled terminal olefin; however, no CD_3W intermediate was characterized. However, we[1] have shown definitively that WCl_6 is an inactive reactant for a metathesis precursor unless oxygen is present.

(17) R. Schrock, S. Rocklage, J. Wengrovius, G. Rupprecht, and J. Fellman, *J. Mol. Catal.*, **8**, 73–83 (1980). Phosphine complexes of CH_2WOCl_2 have been isolated and shown to be slightly active metathesis catalysts.

(18) Since our work was completed, Schrock et al. have described $(CH_3)_3CCHW(O)Cl_2[P(C_2H_5)_3]$ as an active olefin metathesis catalyst although rates were extremely low: J. H. Wengrovius, R. R. Schrock, M. R. Churchill, J. R. Misset, and W. J. Youngs, *J. Am. Chem. Soc.*, **102**, 4515 (1980).

J. Am. Chem. Soc. **1980**, *102*, 6574–6575

Acknowledgment. This research was supported by the National Science Foundation. We especially thank Professor J. Riess and Dr. Santini-Scampucci for experimental information and Gregory F. Schmidt for constructive comment on mechanistic features of the decomposition reaction.

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Received May 19, 1980

Positive Halogen Cryptate: Complexation of Iodine with [2.2.2]Cryptand

Sir:

Many cationic species (e.g., metal cations and ammonium, guanidinium, imidazolidium, and diazonium cations) are known to form inclusion complexes with macrocyclic ligands.[1] We report in this communication *the first example of a positive halogen ion cryptation*, formed by complexation of molecular iodine with the [2.2.2]cryptand ([2.2.2]) (Figure 1).[2]

The formation of charge-transfer complexes between diatomic halogens and amines or ethers has been extensively investigated.[3] In these complexes, a linear orientation of the halogen molecule with the donor atom is generally observed in the solid state.[4] Recently, Schori[5] and Pannell[6] have investigated the interaction between bromine and several crown ethers. They have concluded that the binding of the bromine to the polyether does not involve the cavity of the crown but rather that the bonding is essentially the same as in tetrahydrofuran or dioxan complexes, which involves a single oxygen–bromine interaction. The same conclusion was reached by Hopkins[7] for crown ether–iodine complexes.

The NMR spectrum of a solution of [2.2.2] in $CDCl_3$ shows a cleanly resolved triplet at 2.65 ppm for the CH_2N protons, with no significant alteration between −40 and +50 °C. When 1 equiv of I_2 is added to this solution, a new triplet appears at 3.22 ppm for the CH_2N protons, and the signal at 2.65 ppm nearly disappears.[5] The same spectra are obtained from +24 to −40 °C. This new spectrum must be due to the formation of a 1:1 complex between [2.2.2] and iodine. On the other hand, when 0.5 equiv of I_2 is added to the solution at 24 °C, two broad singlets of approximately equal intensity appear at 2.65 and 3.23 ppm. When the solution is cooled to −40 °C, triplets at 2.65 and 3.23 ppm are obtained which correspond to the free [2.2.2] and the [2.2.2]–iodine complex, respectively. When the solution is heated to 50 °C, a singlet is obtained at 2.97 ppm. At the coalescence temperature (T_c 30 °C), the corresponding exchange rate (k_c) and free energy of activation can be calculated in the usual way from the spectral parameters by using the Eyring rate equation ($k_c = 129\ s^{-1}$, $\Delta G_c^{\ddagger} = 14.9\ kcal{\cdot}mol^{-1}$) (Figure 2).[8]

Figure 1. [2.2.2]Cryptand

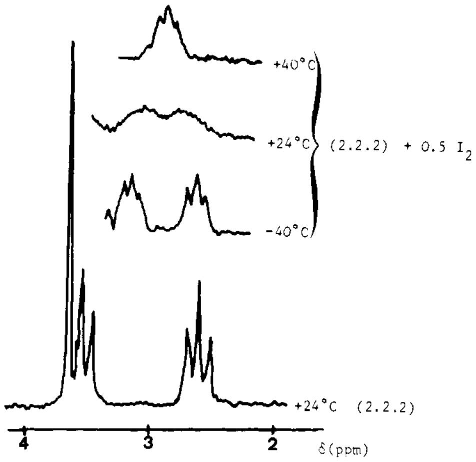

Figure 2. 100-MHz NMR spectra of $CDCl_3$ solutions.

Similar experiments were performed with triethylamine instead of [2.2.2]. The methylene protons of free triethylamine and triethylamine–iodine complex are observed at 2.55 and 3.55 ppm, respectively. When 0.5 equiv of I_2 is added, the quartet is displaced to 3.07 ppm, corresponding to a fast exchange rate between the complexed and noncomplexed species; at −40 °C, the exchange remains rapid, normally indicating a less stable complex.

In the present case, NMR spectroscopy is not a convenient method for estimating the equilibrium constant (K_s) of the complex formation since the Benesi–Hildebrand method requires a large excess of I_2.[9]

When iodine is added to chloroform solutions of [2.2.2], conductance measurements reveal an *ionic character* for the resulting complex: the conductance of a 2×10^{-2} M solution of [2.2.2] and I_2 (1:1 stoichiometry) was found to be $6 \times 10^{-2}\ \Omega^{-1}{\cdot}M^{-1}{\cdot}cm^{-2}$ ([2.2.2]cryptand or I_2 alone is not conducting). This value is similar to that obtained for a 2×10^{-2} M solution of N^+Bu_4, I^- ($3.45 \times 10^{-3}\ \Omega^{-1}{\cdot}M^{-1}{\cdot}cm^{-2}$).

Voltamperometric measurements in $CHCl_3$ (2.9×10^{-4} M) unambiguously confirm the ionic character of the complex. I_2, I^- and I_3^- are the only species detected (I_3^- is also detected by UV analysis). The quantities present of these species add up to less than the initial I_2 concentration; thus, from the law of conservation of both charge and mass, one can state that there is necessarily present additional, positively charged iodine species. I_3^- is formed in the well-known equilibrium (eq 2). The results favor a mechanism corresponding to eq 1 and 2.

$$[2.2.2] + I_2 \underset{k_{-1}}{\overset{k_1}{\rightleftarrows}} ([2.2.2]\text{–}I^+),\ I^-\quad K_s = \frac{k_1}{k_{-1}} = \frac{[2.2.2\text{–}I]^+[I^-]}{[2.2.2][I_2]} \tag{1}$$

$$I^- + I_2 \rightleftarrows I_3^-\quad K_2 = \frac{[I_3^-]}{[I^-][I_2]} \tag{2}$$

The equilibrium constant K_s is estimated to be $\geq 10^7$ (for the $Et_3N\text{–}I_2$ complex, the K_s value is only 4.6×10^3 M). K_2 is obtained (3.12×10^6 M) in a manner similar to that used by Guidelli and Piccardi[10] for CH_3CN solutions. The relative values of K_s and K_2 are in agreement with the stoichiometry determined from NMR measurements (vide supra).

As the complex is more stable than the dissociated state ($K_s > 1$), the k_c and $\Delta G_c^{\ddagger}$ values correspond to the rate (k_{-1}) of the I^+ leaving the $[2.2.2]\text{–}I^+$ complex. Attention must be focused on the fact that the spectrum remains symmetrical at −40 °C (one

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[1] Izatt, R. M.; Christensen, J. J., Eds. "Synthetic Multidentate Macrocyclic Compounds", Academic Press: 1979.

[2] The cavity of [2.2.2] is approximately 2.8 Å. The diameter of I^+ is approximately 1.6 Å (calculated value).

[3] Foster, R. "Organic Charge-Transfer Complexes"; Academic Press: 1969.

[4] In solution, the ionic character of the amine–I_2 complexes increases with the polarity of the solvent. The formation of conducting species occurs, possibly arising from an electron transfer in which the complex $Et_3N\cdots I\text{–}I$ turns to the ion pair $Et_3N^+\text{–}I$, I^-.

[5] Schori, E.; Jagur-Grodzinsky, J. *Isr. J. Chem.* **1972**, *10*, 935.

[6] Pannell, K. H.; Mayr, A. *J. Chem. Soc., Chem. Commun.* **1979**, 132.

[7] Hopkins, H. P.; Jahagirdal, D. V.; Windler, F. J. *J. Phys. Chem.* **1978**, *82*, 1254.

[8] The spectrum of [2.2.2] shows a triplet at 3.60 ppm (CH_2O protons) and a singlet at 3.70 ppm (OCH_2CH_2O protons). Modifications of these resonances upon complexation with iodine (3.70 ppm for the triplet and 3.65 ppm for the singlet) have not been considered since the CH_2N proton shift is more significant.

[9] Near 1.3:1 stoichiometry, the solutions became turbid; more dilute solutions (2×10^{-4} M) remain homogeneous.

[10] Guidelli, R.; Piccardi, G. *Electrochim. Acta* **1967**, *12*, 1085.

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