

Preparation of CpMoX₃ (Cp = η-C₅H₅; X = Cl, Br, I) by Thermal Decarbonylation of CpMoX₃(CO)₂, a Previously Overlooked Phenomenon

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Introduction

We have been recently interested in the synthesis of new molybdenum compounds that contain only halides and one cyclopentadienyl ring, CpMoX_n, as these are useful synthons for the preparation of a variety of other organometallic products.¹ Although the CpMoX₄ (X = Cl, Br) materials have been known for a long time² and a variety of (ring)MoX₂ compounds (ring = Cp³ or other alkyl-substituted cyclopentadienyl rings⁴) have been more recently developed, monocyclopentadienyl halide derivatives of molybdenum(IV) were unknown until we recently reported the preparation of CpMoCl₃ by either oxidation of CpMoCl₂ by PhICl₂, reduction of CpMoCl₄ by TiCl₃, or conproportionation of CpMoCl₂ and CpMoCl₄.⁵

We wondered whether the decarbonylation of CpMoCl₃(CO)₂ might represent a more direct route to CpMoCl₃ and whether this synthetic procedure could be extended to the heretofore unknown bromide and iodide analogues. CpMoX₃(CO)₂ complexes (X = Cl, Br, I) were reported over 25 years ago by two research groups^{6,7} and were described as unstable toward loss of CO, but the nature of the decomposition products was not investigated. We report here that the decarbonylation strategy is indeed a most convenient one for the preparation of CpMoX₃ (X = Cl, Br, I) materials.

Experimental Section

General Procedures. All operations were carried out under an atmosphere of dinitrogen with standard glove-box and Schlenk-line techniques. Solvents were purified by conventional methods and distilled under dinitrogen prior to use. Low-energy FT-IR spectra were recorded on a Perkin-Elmer 1800 spectrophotometer. NMR spectra were obtained with a Bruker WP200 spectrometer; the peak positions are reported upfield of TMS as calculated from the residual solvent peaks (¹H and ¹³C) or external 85% H₃PO₄ (³¹P). EPR spectra were recorded with a Bruker ER200 spectrometer operating with an X-band radio-frequency generator. Room-temperature magnetic susceptibility measurements were carried out by a modified Gouy method with a Johnson Matthey balance, and low-temperature data were obtained with a computer-controlled Faraday system which consists of a Cahn 2000 microbalance, Applied Magnetics electromagnet, Lake Shore Cryotronics temperature controller, platinum resistance thermometer, and Abess instrument cryostat. The magnetic susceptibility data were corrected for the diamagnetism of the ligands by using Pascal's constants before conversion to magnetic moments. The elemental analyses were performed by M-H-W, Phoenix, AZ, or Galbraith Laboratories, Inc., Knoxville, TN.

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Thermal Decarbonylation of CpMoX₃(CO)₂. Preparation of CpMoX₃ (X = Cl, Br, I). The starting materials CpMoX₃(CO)₂ were prepared *in situ* by a slightly modified procedure with respect to those reported in the literature.^{6,7} In each case, [CpMo(CO)₃]₂ was treated in CH₂Cl₂ with 3 equiv of the oxidizing agent (PhICl₂, Br₂, and I₂, respectively). The reactions were followed by IR, showing the immediate formation of CpMoX(CO)₃, followed by the slower conversion to CpMoX₃(CO)₂, which was only sparingly soluble in the CH₂Cl₂ solvent (overnight stirring at room temperature was necessary in each case to complete this step). For each halogen system, the nature of the precipitate obtained by filtration at this point in a separate control experiment was confirmed as the desired CpMoX₃(CO)₂ complex by its IR properties (Nujol mull and CH₂Cl₂ solution) with those reported in the literature.^{6,7} We did not observe decomposition of these compounds by loss of CO upon standing in solution at room temperature. Overnight reflux of the resulting mixtures yielded insoluble materials (Cl, brown; Br, dark green; I, black), which were filtered off, washed with CH₂Cl₂, and dried under vacuum. Yields: 173 mg, 69% (Cl); 400 mg, 52% (Br); 669 mg, 61% (I).

The CpMoCl₃ material had an IR spectrum (4000–200 cm⁻¹) identical to that reported previously for the same compound obtained by a different method,⁵ and a reaction with 2 equiv of P(OCH₂)₃CET proceeded to produce CpMoCl₃[P(OCH₂)₃CET]₂, as expected on the basis of the previous report.⁵ μ_{eff} = 1.25 μ_B at room temperature, decreasing to 1.12 μ_B at 80 K (see supplemental figure). Anal. Calcd. for CpMoBr₃, C₅H₅Br₃Mo: C, 14.99; H, 1.26. Found: C, 14.9; H, 1.1. Low-energy IR (400–200 cm⁻¹; Nujol mull): 245 vs, 222 s. μ_{eff} = 1.66 μ_B. Anal. Calcd for CpMoI₃, C₅H₅I₃Mo: C, 11.09; H, 0.93. Found: C, 11.7; H, 0.8. Low-energy IR (400–200 cm⁻¹; Nujol mull): 324 w, 206 w, 188 s with sh at ca. 180. μ_{eff} = 1.84 μ_B.

For the purpose of measuring the variable-temperature magnetic moment of CpMoCl₃, a sample was obtained by refluxing *isolated* CpMoCl₃(CO)₂ in order to avoid any possible contamination with trace amounts of CpMoCl₂ or CpMoCl₄, which may form if a nonstoichiometric amount of PhICl₂ is used.

Preparation of CpMoCl₄ from [CpMo(CO)₃]₂ and PhICl₂. [CpMo(CO)₃]₂ (1.187 g, 2.42 mmol) was placed in a Schlenk tube and slurried in 20 mL of CH₂Cl₂. To the resulting suspension was added via cannula a solution of PhICl₂ (4.0 g, 14.5 mmol) in 30 mL of CH₂Cl₂ at room temperature with stirring. During the addition, vigorous gas evolution was observed, accompanied by the formation of a brown precipitate, presumably CpMoCl₃(CO)₂. When all the PhICl₂ solution was added, stirring was continued for a few minutes at room temperature, during which time the color of the precipitate changed to the characteristic purple color of CpMoCl₄. The precipitate was filtered off, washed with *n*-heptane, and dried under vacuum. Yield: 1.281 g (87%). The color and EPR properties of this compound are identical with those of an authentic sample of CpMoCl₄.⁸

Preparation of CpMoBr₄ from [CpMo(CO)₃]₂ and Br₂. [CpMo(CO)₃]₂ (0.109 g, 0.22 mmol) was dissolved in 15 mL of CH₂Cl₂, and to the solution was added Br₂ (50 μL, 0.16 g, 0.97 mmol), resulting in gas evolution and formation of a black solid. After the mixture was stirred at room temperature overnight, the solid was separated from the mother liquor, washed with heptane, and dried under vacuum. Yield: 96 mg (45%). The color of this compound is identical with that reported for a sample of CpMoBr₄ prepared from CpMoOBr₂ and HBr.^{2b}

Preparation of CpMoBr₄ from CpMo(CO)₃Me and PBr₅. CpMo(CO)₃-Me was prepared *in situ* from Mo(CO)₆ (1.914 g, 7.25 mmol) and NaCp as reported in the literature.⁸ After complete removal of the THF solvent, extraction of the residue with CH₂Cl₂ (50 mL), and filtration, the resulting solution was slowly transferred with a cannula into a suspension of PBr₅ (7.8 g, 18.1 mmol) in 50 mL of CH₂Cl₂. Gas evolution and formation of a black precipitate were observed during the transfer. The mixture was subsequently refluxed for 1.5 h. The product was recovered by filtration and washed with CH₂Cl₂. Yield: 2.645 g (76%). This compound has the same color and properties as that obtained from [CpMo(CO)₃]₂ and Br₂; see previous paragraph.

Reaction of [CpMo(CO)₃]₂ and I₂ in a 1:3.5 Ratio. Preparation of CpMoI_{3.5}. [CpMo(CO)₃]₂ (305 mg, 0.622 mmol) was dissolved in 20 mL of CH₂Cl₂, and to the resulting solution was added I₂ (556 mg, 2.19 mmol). Gas evolution and the formation of a dark brown suspension were observed. The mixture was refluxed for 2 days, resulting in the formation of a black solid. The mother solution of this solid had a pale blue-purple color. The solid was filtered off, washed with heptane, and

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dried. Yield: 560 mg (74%). Anal. Calcd for $C_5H_5I_3.5Mo$: C, 9.92; H, 0.83. Found: 9.7; H, 0.7. Low-energy IR (400–200 cm^{-1} ; Nujol mull): 333 w with sh at ca. 328, 193 w. $\mu_{eff} = 1.01 \mu_B$ per $[Cp_2Mo_2I_7]$ unit. EPR (X-band, CH_2Cl_2 , room temperature): $g = 2.054$ (broad; peak-to-peak separation = 140 G).

Reactions of $CpMoX_3$ ($X = Br, I$) with Phosphines and Phosphites. (a) Preparation of $CpMoBr_3(dppe)$. $CpMoBr_3$ (249 mg, 0.62 mmol) and $dppe$ (255 mg, 0.64 mmol) were introduced into a Schlenk tube and slurried in 20 mL of CH_2Cl_2 . The immediate formation of a red-purple solution was observed, followed by the slow precipitation of a red-brown solid. More precipitate formed after 20 mL of *n*-heptane was added to the mixture. The solid was filtered off, washed with *n*-heptane (5 mL), and dried under vacuum. Yield: 64.3%. Anal. Calcd for $C_{31}H_{29}Cl_3MoP_2$: C, 46.59; H, 3.66. Found: C, 46.8; H, 4.1. 1H -NMR ($CDCl_3$): δ 8.13, 7.61, 7.24 (m, 20H, C_5H_5), 5.24 (d, $J_{PH} = 2.6$ Hz, 5H, C_5H_5), 3.45, 3.33, 3.07, 2.93 (br, 4H, CH_2). $^{31}P\{^1H\}$ -NMR ($CDCl_3$): δ 57.1 (d, $J_{PP} = 115$ Hz, 1P), 14.5 (d, 1P).

(b) Preparation of $CpMoBr_3[P(OCH_2)_3CtEt]_2$.

 $CpMoBr_3$ (420 mg, 1.05 mmol) and $P(OCH_2)_3CtEt$ (343 mg, 2.12 mmol) were introduced into a Schlenk tube and slurried with 20 mL of CH_2Cl_2 . The solution immediately turned brown. A minor amount of insoluble solid was eliminated by filtration, and from the solution, the product was crystallized by diffusion of a heptane layer (15 mL). The resulting crystals were filtered off, washed with heptane, and dried under vacuum. Yield: 503 mg, 70.7%. Anal. Calcd for $C_{17}H_{27}Br_3MoP_2O_2$: C, 28.16; H, 3.75. Found: C, 27.7; H, 4.0. 1H -NMR (CD_2Cl_2): δ 5.75 (t, $J_{PH} = 3.8$ Hz, 5H, C_5H_5), 4.33 (vt, $J_{PH} = 2.1$ Hz, 12H, OCH_2), 1.25 (q, $J_{HH} = 7.4$ Hz, 4H, CH_2CH_3), 0.83 (t, $J_{HH} = 7.5$ Hz, 6H, CH_2CH_3). $^{31}P\{^1H\}$ -NMR ($CDCl_3$): δ 128.1.

(c) Reaction of $CpMoBr_3$ with PMe_3 .

 $CpMoBr_3$ (172 mg, 0.64 mmol) was suspended in THF (10 mL). The suspension was cooled to $-78^\circ C$ in a dry ice/acetone bath, and then PMe_3 (135 μL , 1.28 mmol) was added. The mixture allowed to slowly warm to room temperature under magnetic stirring. At $-5^\circ C$ the formation of a pale pink-red solution was noted. Further warming deepened the color of the solution to purple while a cream-white precipitate had replaced the initial insoluble $CpMoBr_3$. 1H -NMR inspection of an aliquot of the solution showed no evidence for a diamagnetic $CpMoBr_3(PMe_3)_2$ product and revealed the presence of several paramagnetically shifted resonances in the δ -2 to -50 region, two prominent resonances in an approximate 1:1 ratio being observed at δ -38 and -40, but none of these could be assigned to a paramagnetic $CpMoBr_3(PMe_3)$ adduct.

(d) Reaction of $CpMoI_3$ with $Dppe$.

 $CpMoI_3$ (536 mg, 0.989 mmol) and $dppe$ (399 mg, 1.00 mmol) were introduced into a Schlenk tube and slurried with 10 mL of CH_2Cl_2 . A deep red-brown solution immediately formed, and large amounts of a brown solid were also noted. After being stirred overnight at room temperature, the mixture had not changed in appearance. After the solid was filtered off, concentration of the mother solution, addition of toluene, and cooling to $-80^\circ C$ produced crystalline $CpMoI_2(dppe)$, which was separated from the solution, washed with *n*-heptane, and dried under vacuum. Yield: 218 mg (40.7% based on the stoichiometry of eq 4; see Results and Discussion). Anal. Calcd for $C_{31}H_{29}I_2MoP_2$: C, 45.78; H, 3.59. Found: C, 45.4; H, 3.8. This solid had an EPR spectrum identical to that reported previously for $CpMoI_2(dppe)$.⁸

(e) Reaction of $CpMoI_3$ with PMe_3 and Then with I_2 .

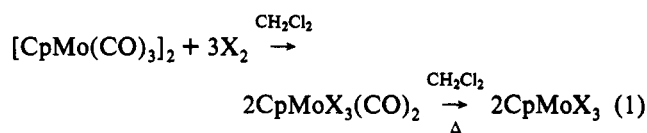
 $CpMoI_3$ (160 mg, 0.295 mmol) was slurried in a Schlenk tube with 10 mL of CH_2Cl_2 . PMe_3 (61 mL, 0.589 mmol) was added with a microsyringe while the suspension was kept at room temperature with magnetic stirring. An immediate reaction took place with formation of a green solution and a gray-green solid. An EPR spectrum of an aliquot of the supernatant liquid indicated the presence of $CpMoI_2(PMe_3)_2$ by comparison with the literature spectrum.⁹ To the reaction mixture was added I_2 (112 mg, 0.441 mmol), and the resultant mixture was stirred at room temperature. Once all the I_2 dissolved, the solution became noticeably darker green, while a green precipitate remained. At this point, an aliquot of the solution showed no residual EPR signal, while 1H -NMR spectroscopy after evaporation to dryness and redissolution in CD_2Cl_2 confirmed the presence of the $[CpMoI_2(PMe_3)]^+$ ion.¹⁰

Reaction of $CpMoX_3$ with CO. For each halide system, a small amount (ca. 50 mg) of the compound was suspended in CH_2Cl_2 (5–10 mL), and the suspension was stirred at room temperature under an atmosphere of

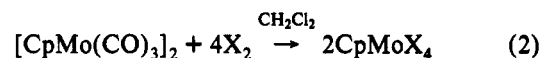
CO. The solid rapidly reacted to afford a solution of sparingly soluble $CpMoX_3(CO)_2$ (IR (cm^{-1}): $CpMoBr_3(CO)_2$, 2095 s, 2061 vs; $CpMoI_3(CO)_2$, 2071 s, 2039 vs). The chloride analogue was not sufficiently soluble for an IR investigation in CH_2Cl_2 , and it was identified from its Nujol IR spectrum (2116 s, 2076 vs cm^{-1}). Continued stirring at room temperature under CO with IR monitoring indicated no further change for $CpMoCl_3(CO)_2$ and $CpMoBr_3(CO)_2$, whereas $CpMoI_3(CO)_2$ was totally transformed to $CpMoI(CO)_3$ within 3 days (IR: 2042 s, 1966 vs, $br\ cm^{-1}$).

Results and Discussion

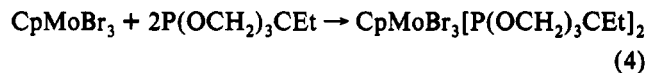
The Mo(IV) carbonyl compounds $CpMoX_3(CO)_2$ ($X = Cl, Br, I$) appear to be stable toward CO loss at room temperature, but they can be completely decarbonylated in refluxing CH_2Cl_2 to afford insoluble materials that analyze correctly for $CpMoX_3$. The $CpMoCl_3$ material obtained by this route shows physical and chemical properties identical with those of the same compound obtained by other routes.⁵ Considering that $[CpMo(CO)_3]_2$ is commercially available or can easily be prepared from $Mo(CO)_6$, the procedure reported here represents by far the most convenient method for the preparation of $CpMoCl_3$ (eq 1) and also represents a convenient route to the previously unknown $CpMoBr_3$ and $CpMoI_3$ materials. For the chloride system, the stoichiometric reagent $PhICl_2$ was used rather than gaseous Cl_2 .



Use of an excess of $PhICl_2$ or Br_2 affords a straightforward synthesis of the previously reported $CpMoCl_4$ and $CpMoBr_4$,² respectively (see eq 2), and is comparable in simplicity to the recently reported PX_5 ($X = Cl, Br$) method.^{8,11}



The identity of the bromide compound is verified by derivatization reactions analogous to those previously run on the chloride system (eqs 3 and 4).



The $CpMoBr_3(dppe)$ compound had been previously described,¹² but only a 1H -NMR characterization was given, which was not sufficient for a complete structural assignment. We find by 1H - and ^{31}P -NMR that freshly prepared solutions of this compound contain isomer I as the major species but that, upon standing at room temperature, the samples exhibit other peaks in the ^{31}P -NMR spectrum, possibly indicating equilibration with other isomers, as was also suggested for the analogous $CpMoCl_3(dppe)$ and $CpMoClBr_2(dppe)$ compounds.¹³ In particular, a ^{31}P -NMR singlet resonance at δ 39.3 is assigned to structure II [compare with δ 38.6 for $CpMoCl_3(dppe)$ and δ 39.1 for $CpMoClBr_2(dppe)$].¹³ Structure I was found in the solid state for both $CpMoCl_3(dppe)$ ¹³ and $CpMoCl_3(dmpe)$.¹⁴

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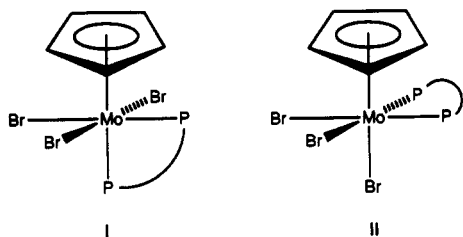
(13) Stärker, K.; Curtis, M. D. *Inorg. Chem.* 1985, 24, 3006.

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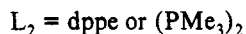
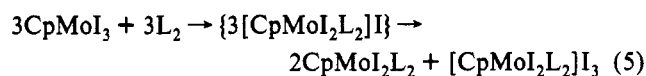
(9) Krueger, S. T.; Poli, R.; Rheingold, A. L.; Staley, D. L. *Inorg. Chem.* 1989, 28, 4599.

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$\text{CpMoBr}_3[\text{P}(\text{OCH}_2)_3\text{CEt}]_2$ has NMR properties that perfectly match these previously reported for the trichloride analogue.⁵

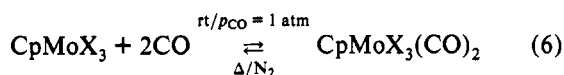


The reaction between CpMoBr_3 and PMe_3 did not produce the expected $\text{CpMoBr}_3(\text{PMe}_3)_2$ or the corresponding paramagnetic $\text{CpMoBr}_3(\text{PMe}_3)$ adduct but rather produced a mixture of other paramagnetic products, as shown by $^1\text{H-NMR}$, and thus takes a course rather similar to that of the previously investigated $\text{CpMoCl}_3/\text{PMe}_3$ reaction.⁵ CpMoI_3 rapidly reacted with *dpe* and PMe_3 , but 18-electron adducts were not obtained in these cases. Rather, the previously reported⁹ $\text{CpMoI}_2(\text{dpe})$ complex was isolated in the *dpe* reaction and the known¹⁰ $\text{CpMoI}_2(\text{PMe}_3)_2$ and $[\text{CpMoI}_2(\text{PMe}_3)_2]^+\text{I}_3^-$ complexes were identified by EPR in the PMe_3 reaction. These reduction reactions probably occur as illustrated in eq 5, since the proposed intermediate ($L = \text{PMe}_3$)



was obtained earlier by interaction of $\text{CpMoI}_2(\text{PMe}_3)_2$ and $1/2$ equiv of I_2 and shown to rapidly rearrange to the products of eq 5.^{10,15} Two possible reasons for the different reactivities of the tribromide and triiodide compounds are the greater steric requirements of the three iodide ligands and the greater reducing power of I^- versus the corresponding Br^- ion.

All three CpMoX_3 compounds promptly react with CO at room temperature to regenerate the 18-electron precursors, $\text{CpMoX}_3(\text{CO})_2$, showing the reversibility of the decarbonylation process (eq 6). Whereas the chloride and bromide $\text{CpMoX}_3(\text{CO})_2$



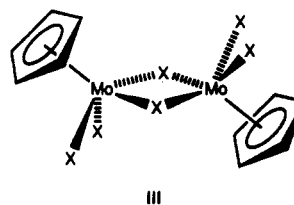
systems are stable under CO, the iodide analogue reacts further to *quantitatively* (by IR) produce the monoiodide tricarbonyl complex (eq 7), demonstrating that this step is also completely



reversible. Under CO (1 atm), equilibrium 7 is completely shifted toward the right whereas, under N_2 , the triiodide derivative can be made from the monoiodide complex in good yields. The lower solubility of the triiodide compound presumably contributes to shift this equilibrium toward the left under low CO pressure conditions.

A point of interest concerns the structure of the CpMoX_3 compounds. As these are all highly insoluble materials, solution spectroscopic methods cannot be applied. The sparing solubility also prevented recrystallization in the form of single crystals for an X-ray analysis. On the basis of the reactivity of these materials (see above) and parallels with the corresponding Cp^*Mo analogues,¹⁶ it seems most likely that the CpMoX_3 species are dinuclear or oligonuclear covalent species. Physical studies are also in accord

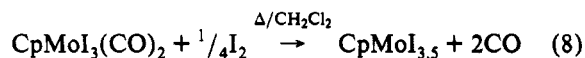
with this view and point more directly to a dinuclear structure as found for Cp^*MoX_3 ($\text{Cp}^* = \eta\text{-C}_5\text{Me}_5$; $\text{X} = \text{Cl}, \text{Br}$), e.g. III.



The low-energy IR spectra of CpMoCl_3 and Cp^*MoCl_3 are similar,¹⁶ and the two compounds also have a similar temperature dependences of their magnetic moments, which has been attributed to antiferromagnetic coupling.¹⁶ The room-temperature magnetic moment for CpMoX_3 increases in the order $\text{Cl} > \text{Br} < \text{I}$, with the moment for the iodide complex ($1.84 \mu_{\text{B}}/\text{Mo}$) being still substantially smaller than the value found for magnetically diluted $d^2 \text{Mo(IV)}$, for instance $2.63 \mu_{\text{B}}$ (temperature independent) for $\text{Cp}^*\text{MoCl}_3(\text{PMe}_3)$.¹⁶ This trend indicates stronger antiferromagnetic coupling for the lighter halide system, in accord with the presumed smaller separation of the metals in the halide-bridged dinuclear structure.

It is interesting to observe that a compound of empirical formula " $\text{Cp}^*\text{MoI}_{3.5}$ " exists but it in fact consists of a $\text{Mo}^{\text{III}}\text{Mo}^{\text{IV}}$ system, e.g. $[\text{Cp}^*\text{Mo}_2\text{I}_4]^+\text{I}_3^-$. Electrochemical work has established that further addition of I_2 would not produce a complex of the metal in a higher oxidation state.¹⁷ Consequently, a question arises as to whether it is legitimate to describe CpMoI_3 as a neutral Mo(IV) compound. It is worth mentioning here that CpVI_3 has also been shown to adopt a solid-state structure consisting of a molecular triiodo material of the metal in the +IV oxidation state whereas considerations of oxidation potentials as a function of halide for the CpVX_3 ($\text{X} = \text{Cl}, \text{Br}, \text{I}$) series would lead to the prediction of an internal redox process with production of polyiodides.¹⁸ The related Cp^*CrI_3 and $\text{Cp}^*\text{CrI}_{5.5}$ compounds, on the other hand, have been shown to be polyiodide complexes of Cr(III) .¹⁹

Support for the assignment of a neutral structure to CpMoI_3 is its rapid reaction with CO, which is analogous to those of CpMoX_3 ($\text{X} = \text{Cl}, \text{Br}$) (vide supra) and to those of the structurally characterized (see III) $[\text{Cp}^*\text{MoX}_3]_2$ ($\text{X} = \text{Cl}, \text{Br}$).^{16,20} " $\text{Cp}^*\text{MoI}_{3.5}$ ", on the other hand, does not react with CO.²¹ As further support for this argument, we have prepared a compound of stoichiometry $\text{CpMoI}_{3.5}$ by the reaction between $[\text{Cp}^*\text{Mo}(\text{CO})_3]_3$ and I_2 in a 1:3.5 ratio, followed by reflux of the resulting mixture that presumably contains $\text{Cp}^*\text{MoI}_3(\text{CO})_2$ and unreacted I_2 in a 4:1 molar ratio (eq 8). This product has



properties substantially different from those of CpMoI_3 . For instance, the two materials have different low-energy IR spectra, and while CpMoI_3 is completely insoluble in organic solvents, $\text{CpMoI}_{3.5}$ is sparingly soluble in CH_2Cl_2 , generating deep violet solutions. While CpMoI_3 rapidly reacts with CO to regenerate $\text{CpMoI}_3(\text{CO})_2$, $\text{CpMoI}_{3.5}$ does not react with CO over several days at room temperature and therefore behaves identically to " $\text{Cp}^*\text{MoI}_{3.5}$ ".

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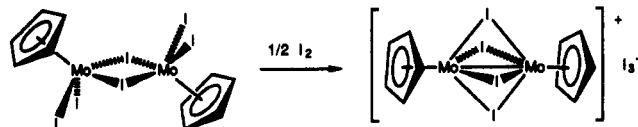
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Scheme I



We propose therefore that the structure of CpMoI_{3.5} is analogous to that of its more soluble Cp* analogue, i.e. [Cp₂Mo₂I₄]⁺I₃⁻, and that the affinity of I₂ for I⁻ induces this curious rearrangement with formal reduction of the metal by action of the oxidizing agent I₂ (see Scheme I). For the proposed structure of CpMoI_{3.5}, a single unpaired electron per dimeric

unit is expected since theoretical calculations on this system²² suggest a $\sigma^2\delta^*2\delta^1$ configuration. Consistent with this view, CpMoI_{3.5} shows a broad EPR resonance at room temperature at $g = 2.054$.

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Supplementary Material Available: A figure showing the variable-temperature magnetic moment of CpMoCl₃ (1 page). Ordering information is given on any current masthead page.

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