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REACTION OF *trans*-$Mo(N_2)_2(Ph_2PCH_2CH_2PPh_2)_2$ WITH ALDEHYDES AND ALDEHYDIC COMPOUNDS, ALCOHOLS, AND HYDROAROMATIC COMPOUNDS: STOICHIOMETRIC DECARBONYLATION AND DEHYDROGENATION

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Summary

Reactions of formamides ($HCONR^1R^2$), formate esters ($HCOOR^3$), and aldehydes (R^4CHO) with *trans*-$Mo(N_2)_2(Ph_2PCH_2CH_2PPh_2)_2$ yield their respective decarbonylation products [amine (R^1R^2NH), alcohol (R^3OH), and alkane (R^4H)] and molybdenum carbonyl complexes. The formation of the products is accounted for by assuming oxidative addition of the compounds to molybdenum, involving the cleavage of the formyl C—H bond followed by decarbonylation. Reactions of primary alcohols (RCH_2OH) yield alkane (RH) and a mixture of molybdenum carbonyl complexes and molybdenum hydride. Reactions of secondary alcohols give the corresponding ketones and molybdenum hydride. Similar reactions of hydroaromatics have also been investigated.

Introduction

It has long been known that on treatment with alcohols some transition metal complexes give carbonyls or hydridocarbonyls [1,2]. The decarbonylation of aldehydes and acyl halides with transition metal complexes has been established as a useful synthetic reaction [2]. However, little is known concerning the decarbonylation of other carbonyl compounds. Very recently, Felkin et al. reported decarbonylation of ethyl formate by a dinitrogen complex of $Fe^{(0)}$ to yield a corresponding carbonyl complex and ethanol [3]. Rusina and Vlček reported the formation of $RhCl(CO)(PPh_3)_2$ from $RhCl_3 \cdot 3\ H_2O$ using dimethyl formamide (DMF), cyclohexanone, acetophenone, tetrahydrofuran (THF), and

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dioxane as CO sources [4]. The transition metals involved in these reactions were, to our best knowledge, limited to Group VIII metals.

We now report details of the decarbonylation of formamides, formate esters, aldehydes, and primary alcohols, and dehydrogenations of alcohols and hydroaromatic compounds promoted by *trans*-$Mo(N_2)_2(dpe)_2$ (dpe = $Ph_2PCH_2CH_2PPh_2$). A preliminary report has been published [5]. In these cases the molybdenum carbonyl complexes formed are too stable, and CO is not liberated from them; the reaction is stoichiometric with respect to the complex. Dehydrogenation also does not proceed catalytically, whereas catalytic hydrogen transfer occurs in the presence of appropriate acceptors [6].

Results and discussion

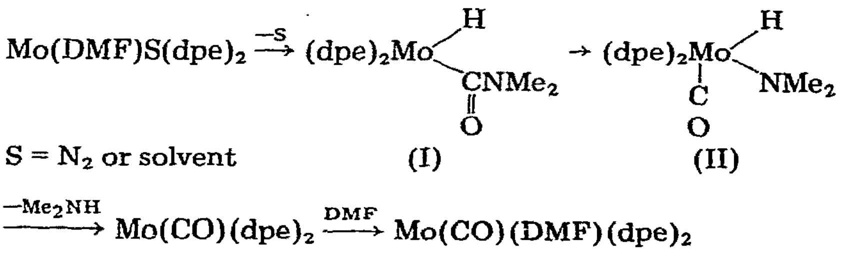
Reaction of formamides

Treatment of *trans*-$Mo(N_2)_2(dpe)_2$ with a large excess of DMF in benzene at reflux gave $Mo(CO)(DMF)(dpe)_2$. The only other reported example of the formation of a carbonyl complex from amides was found with a rhodium complex, but in this case the decarbonylation product was not determined [4]. In our case Me_2NH was obtained and the molar ratio of Me_2NH and $Mo(CO)(DMF)(dpe)_2$ was almost unity as shown in Table 1. The reaction may be expressed as follows:

$$trans\text{-}Mo(N_2)_2(dpe)_2 + 2\ HCONMe_2 \rightarrow Mo(CO)(DMF)(dpe)_2 + Me_2NH + 2\ N_2 \quad (1)$$

When diethyl formamide was employed, decarbonylation occurred similarly to give $Mo(CO)(HCONEt_2)(dpe)_2$ and Et_2NH. The decarbonylation of formamides can be expressed exactly by the reverse process of the carbonylation of amines by noble metal catalysts [7]. In contrast to formamides, acetamides such as $MeCONMe_2$ scarcely undergo reaction with *trans*-$Mo(N_2)_2(dpe)_2$, suggesting that the reaction starts with the initial cleavage of the carbon—hydrogen bond of the formyl group to give molybdenum carbonyl complexes followed by extrusion as shown in Scheme 1.

SCHEME 1



The oxidative addition of formamides to yield the hydrido-(carbamoyl)-molybdenum complex I is analogous to that of an aldehyde to form a hydrido-(acyl) complex, which are intermediates in the decarbonylation of aldehydes [8]. The ensuing extrusion of CO of I is regarded as the reverse reaction of carbamoyl complex formation by the reaction of carbonyls with R_2N^- [9]. The hydrido(amido)molybdenum complex II thus formed is assumed to undergo reductive elimination to produce Me_2NH. The presence of the Mo—H species is

TABLE 1

PRODUCTS OF THE REACTIONS OF ALDEHYDIC COMPOUNDS HCOR WITH *trans*-$Mo(N_2)_2(dpe)_2$ [a]

Compound (L) R =	L/Mo	Time (min)	Products (% yield/*trans*-$Mo(N_2)_2(dpe)_2$)			
			RH	H_2 [b]	Mo complex	Others
NMe_2	123	20	80	0.8	$Mo(CO)L(dpe)_2$ (72)	
NMe_2	10	60	72	0.6	$Mo(CO)L(dpe)_2$ (40) *cis*-$Mo(CO)_2(dpe)_2$ (17)	
NEt_2	100	30	82	0.6	$Mo(CO)L(dpe)_2$ (78)	
OMe	80	10	151	t	*trans*-$Mo(CO)(N_2)(dpe)_2 \cdot 1/2\ C_6H_6$ (45) *cis*-$Mo(CO)_2(dpe)_2$ (30)	
OMe	80	30	163	t	*cis*-$Mo(CO)_2(dpe)_2$ (76)	
OEt	80	10	140	t	*trans*-$Mo(CO)(N_2)(dpe)_2 \cdot 1/2\ C_6H_6$ (52) *cis*-$Mo(CO)_2(dpe)_2$ (20)	CH_4 (t)
$OCHMe_2$	10	30	81	2	*trans*-$Mo(CO)(N_2)(dpe)_2 \cdot 1/2\ C_6H_6$ (55) *cis*-$Mo(CO)_2(dpe)_2$ (7)	CO_2 (5) C_3H_6 (6)
C_6H_{13}	30	20	6	59	*cis*-$Mo(CO)_2(dpe)_2$ (15) [c]	1-C_6H_{12} (t)
C_6H_{13}	1.2	20	63	0.5	*trans*-$Mo(CO)(N_2)(dpe)_2 \cdot 1/2\ C_6H_6$ (55) *cis*-$Mo(CO)_2(dpe)_2$ (20)	1-$C_7H_{15}OH$ (4) 1-C_6H_{12} (4)
Ph [d]	1.0	150	51	14	*trans*-$Mo(CO)(N_2)(dpe)_2 \cdot 1/2\ C_6H_6$ (15) *cis*-$Mo(CO)_2(dpe)_2$ (60)	PhCOPh (5)
$CH_2{=}CH$ [e]	1.5	10	64 [b]	0	$Mo(CO)(C_2H_4)(dpe)_2$ (18) [f] *cis*-$Mo(CO)_2(dpe)_2$ (52)	

[a] *trans*-$Mo(N_2)_2(dpe)_2$ = 0.07 mmol, solvent; benzene = 3 ml, t; trace. Refluxed under nitrogen. When mixtures of molybdenum complexes were obtained, the respective yields were estimated from the IR spectrum of the mixture in similar manner as the reaction with ethanol (see Experimental). [b] Checked from the gas phase. [c] Assumed diacyl complex (31 mg) also was obtained. [d] Solvent; toluene = 3 ml. [e] Refluxed under argon. [f] Confirmed by IR and NMR spectroscopy [17].

supported by the formation of a small amount of H_2.

As described previously, Mo(CO)(DMF)(dpe)$_2$ can be easily converted to *trans*-Mo(CO)(N_2)(dpe)$_2$ · 1/2C_6H_6 [10]. Therefore, this indirect route is useful for the preparation of the monodinitrogen complex. The monodinitrogen complex is not too stable in solution at higher temperatures and gives *cis*-Mo(CO)$_2$(dpe)$_2$ slowly in the absence of CO. In the presence of an excess of DMF, another molecule of DMF blocks the vacant octahedral site of Mo(CO)(dpe)$_2$ [10] and stabilizes the monocarbonyl species. Further oxidative addition of the C—H bond of DMF to Mo(CO)(dpe)$_2$ did not occur, since the electron density of the metal is decreased by the strong π-acceptor CO ligand.

Reaction of formate esters

Formate esters also can be considered as containing the formyl group, and thus decarbonylation by *trans*-Mo(N_2)$_2$(dpe)$_2$ is an expected reaction. Treatment of *trans*-Mo(N_2)$_2$(dpe)$_2$ with HCOOMe in benzene at reflux afforded a dark brown solution, which became reddish orange on cooling under nitrogen. From the resulting solution *trans*-Mo(CO)(N_2)(dpe)$_2$ · 1/2C_6H_6, *cis*-Mo(CO)$_2$(dpe)$_2$, and methanol were obtained. The dark brown color at reflux seems to be due to the five-coordinate Mo(CO)(dpe)$_2$ [10].

Table 1 shows that in the decarbonylation of formate esters the products are mostly alcohols. The reaction, however, is dependent on the structure of the formate. Thus propylene, CO_2, and H_2 are formed as minor products from isopropyl formate. Alkyl acetates such as MeCOOMe and MeCOOCHMe$_2$ did not react with *trans*-Mo(N_2)$_2$(dpe)$_2$ under similar conditions. From these results we assumed that the oxidative addition of the esters to yield the hydrido(alkoxycarbonyl)molybdenum complex followed by extrusion would proceed analogously to the reaction of formamides. The oxidative addition of HCOOEt to give a dialkoxycarbonyl complex is known for Pt(PPh$_3$)$_4$ [11]. In the case of R = CHMe$_2$ in eq. 2, the alkoxycarbonyl to some extent behaved similarly to

$$\begin{tikzcd} & \text{Mo—CO} + \text{ROH} \\ \text{M—Mo—COOR} \arrow[ur] \arrow[dr, "-CO_2"'] & \\ & \text{H—Mo—R} \rightarrow \text{R(—H)} + H_2 + \text{Mo} \end{tikzcd} \qquad (2)$$

carboxylate complexes, MCO$_2$H, which have been reported to decompose to the corresponding hydride with loss of CO_2 [1a].

When heated in refluxing benzene for 30 min, *trans*-Mo(N_2)$_2$(dpe)$_2$ reacted with HCOOMe to give *cis*-Mo(CO)$_2$(dpe)$_2$ exclusively, presumably through further oxidative addition of the ester to Mo(CO)(dpe)$_2$ (eq. 3), but the latter complex showed no oxidative addition reactivity toward formamides.

$$\begin{tikzcd} \textit{trans}\text{-Mo}(N_2)_2(\text{dpe})_2 \arrow[r, "\text{HCOOMe}"] & \text{Mo(CO)(dpe)}_2 + \text{MeOH} + 2\,N_2 \arrow[d, "N_2"'] \arrow[dr, "\text{HCOOMe}"] & \\ & \textit{trans}\text{-Mo(CO)}(N_2)(\text{dpe})_2 & \textit{cis}\text{-Mo(CO)}_2(\text{dpe})_2 \end{tikzcd} \qquad (3)$$

Reaction of aldehydes

We considered that aldehydes also are promising candidates for decarbonylation by *trans*-Mo(N_2)$_2$(dpe)$_2$. However, the main product of the reaction with a large excess of aldehyde was H_2 and a complex containing acyl ligands (Table 1). The complex exhibited two strong bands in the IR spectrum due to the C=O stretching frequencies of the diacyl group. But we could not determine the structure of the complex since it proved difficult to purify; no satisfactory analyses were obtained. Harvie and Kemmitt reported that aldehydes react with $Pt(PPh_3)_4$ to yield the diacyl complex $Pt(COR)_2(PPh_3)_2$ (R = Et or Ph), probably with the evolution of H_2 [11].

In contrast, when an almost equimolar amount of an aldehyde (RCHO) was allowed to react with *trans*-$Mo(N_2)_2(dpe)_2$, alkane (RH) and carbonyl complexes were observed as major products. For example, heptanal was decarbonylated to n-hexane. Hydrogen, 1-hexene, and 1-heptanol were obtained as minor products. Again, the decarbonylation would proceed through oxidative addition of the C—H bond of aldehydes and subsequent acyl—alkyl rearrangement followed by β-elimination to give alkene or reductive elimination to produce alkane. When benzaldehyde was employed, benzophenone was formed as a minor product (eq. 4). This may be due to a coupling reaction between the benzoyl complex and the phenyl complex. Acrolein was selectivity decarbonylated to ethylene.

$$\mathrm{HCOPh}\begin{array}{l}\nearrow^{\text{minor}}\ \frac{1}{2}\,\mathrm{H_2} + \frac{1}{2}\,\mathrm{PhCOPh} + \frac{1}{2}\,\mathrm{CO}\\ \searrow_{\text{major}}\ \mathrm{PhH} + \mathrm{CO}\end{array} \qquad (4)$$

These results suggest that in the presence of a large excess of aldehyde, further oxidative addition to the hydrido(acyl)molybdenum intermediate may occur before extrusion of CO followed by β-elimination or reductive elimination. Another possible explanation is that high concentrations of the hydrido(acyl) species result in its disproportionation giving diacyl and dihydride complexes of molybdenum.

Ketones such as acetone, cyclohexanone, and acetophenone were unreactive towards *trans*-$Mo(N_2)_2(dpe)_2$.

Reaction of alcohols

Treatment of *trans*-$Mo(N_2)_2(dpe)_2$ with primary alcohols gave a mixture of *trans*-$Mo(CO)(N_2)(dpe)_2$, *cis*-$Mo(CO)_2(dpe)_2$, and $MoH_4(dpe)_2$, accompanied by formation of alkanes containing one carbon less than the starting alcohols (Table 2). We assume that the primary alcohols are dehydrogenated to aldehydes and then decarbonylated to alkanes. Since the aldehyde is formed gradually and its concentration is very low, no side reaction leading to the diacyl complex took place. Thus the removal of the -CH_2OH group of the alcohols was realized. Such a reaction is known for metallic palladium or nickel catalysts [2]. In analogy with aldehydes, alkenes also were obtained in low yields. The postulated reaction pathway may be supported by the fact that the corresponding aldehydes were obtained as by-products.

The reaction of *trans*-$Mo(N_2)_2(dpe)_2$ with 2-propanol afforded $MoH_4(dpe)_2$ in high yield. During the reaction H_2 was confirmed by GLC analysis of the gas phase but not determined quantitatively. As shown in Table 3, secondary alcohols such as 2-butanol and 1-phenylethanol were effective hydrogen

TABLE 2

PRODUCTS OF THE REACTIONS OF PRIMARY ALCOHOLS RCH_2OH WITH *trans*-$Mo(N_2)_2(dpe)_2$ [a]

Compound (L) R =	Time (min)	Products (% yield/*trans*-$Mo(N_2)_2(dpe)_2$)						
		RH	R(—H)	H_2 [b]	RCHO	Mo(CO)(N_2)	$Mo(CO)_2$	MoH_4
H	90	—	—	72	t	46	t	25
CH_3	90	43 [b]	—	33	16	24	t	51
C_2H_5	150	29 [b]	2 [b]	67	t	21	t	55
n-C_6H_{13}	180	33	2	78	29	11	18	34
Ph [c]	25	80	—	85	1	30	25	12

[a] *trans*-$Mo(N_2)_2(dpe)_2$ = 0.07 mmol, alcohol = 0.7 mmol, solvent; benzene = 3 ml, t = trace, Mo(CO)(N_2); *trans*-Mo(CO)(N_2)$(dpe)_2$ · 1/2 C_6H_6, $Mo(CO)_2$; *cis*-$Mo(CO)_2(dpe)_2$, MoH_4; $MoH_4(dpe)_2$. Refluxed under nitrogen. The yields of the complexes were estimated from the IR spectrum of the mixture (see Experimental). [b] Checked from the gas phase. [c] Solvent; toluene = 3 ml.

TABLE 3

PRODUCTS OF REACTIONS OF HYDROGEN DONORS WITH *trans*-$Mo(N_2)_2(dpe)_2$ [a]

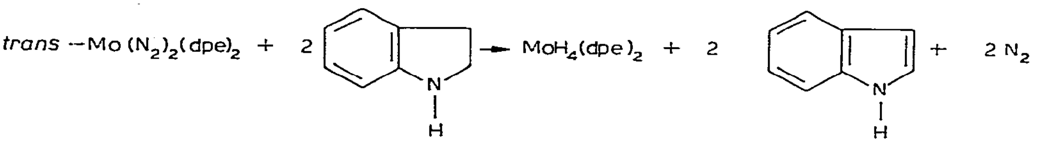
Donor (L)	L/Mo	Time (min)	Products (% yield/*trans*-$Mo(N_2)_2(dpe)_2$)		
			Dehydrogenation product	$MoH_4(dpe)_2$	Others
2-Propanol	10	15	Acetone (160)	83	
2-Butanol	10	15	MeCOEt (162)	80	
1-Phenylethanol	10	15	MeCOPh (180)	85	
Cyclohexanol	10	15	Cyclohexanone (45)	17	
Indoline	20	15	Indole (26)	11	
Pyrrolidine [b]	450	30	—	75	
Dioxane [b]	450	30	—	15	
THF [b]	450	60	C_2H_6 (14) [c], C_2H_4 (1) [c]	17	*trans*-$Mo(CO)(N_2)(dpe)$ (40) *cis*-$Mo(CO)_2(dpe)_2$ (15)
THF [b, d]	450	30	C_2H_6 (5) [c]	40	*trans*-$Mo(CO)(N_2)(dpe)_2$ (10) *cis*-$Mo(CO)_2(dpe)_2$ (13)
THF [b]	450	1080	—	42	*trans*-$Mo(CO)(N_2)(dpe)_2$ (7) *cis*-$Mo(CO)_2(dpe)_2$ (8)

[a] Footnote *a* in Table 1 applies. [b] With no solvent. [c] Checked from the gas phase. [d] Refluxed under argon.

donors. As the dehydrogenation product, the corresponding ketones were obtained. Cyclohexanol, the dehydrogenation product of which is slightly hindered, was less effective. Tertiary alcohols such as t-butanol were unreactive to *trans*-$Mo(N_2)_2(dpe)_2$.

Reaction of hydroaromatic compounds

The reaction of *trans*-$Mo(N_2)_2(dpe)_2$ with several hydroaromatics was investigated (Table 3). The complex reacted with indoline to give $MoH_4(dpe)_2$ and indole.



(5)

When pyrrolidine was used, $MoH_4(dpe)_2$ was obtained in good yield. Howeever, no low-boiling dehydrogenation products such as pyrrole could be detected. Since 1-pyrroline is reported to be so unstable as to trimerize or add to amines [12], it is presumed that the dehydrogenation intermediate might have reacted to give products of higher molecular weight which were not detectable by GLC analysis. Dehydrogenation of hydroaromatic systems may be facilitated by the resonance energy derived in forming an aromatic system; diethylamine was unreactive under similar conditions.

The reaction of *trans*-$Mo(N_2)_2(dpe)_2$ with THF was rather complicated. The reaction under argon at reflux yielded $MoH_4(dpe)_2$ as the major product. Small amounts of *trans*-$Mo(CO)(N_2)(dpe)_2$ and *cis*-$Mo(CO)_2(dpe)_2$ also were obtained. On the other hand, the reaction under nitrogen afforded principally *trans*-$Mo(CO)(N_2)(dpe)_2$. The bis-carbonyl complex *cis*-$Mo(CO)_2(dpe)_2$ and MoH_4-

$(dpe)_2$ also were obtained. At longer reaction times mainly *cis*-$Mo(CO)_2(dpe)_2$ and $MoH_4(dpe)_2$ were obtained. These mixtures were difficult to separate and thus THF was not suitable for the clean synthesis of the carbonyl or hydride complex. GLC analysis showed that there were no detectable amounts of the simply expected dehydrogenation and decarbonylation products of THF, i.e., dihydrofuran, furan, propylene, and propane. The presence of hydrogen, ethylene, and ethane was confirmed. Hydrogen abstraction from THF has been suggested in the reaction of a methylruthenium complex with THF to give the hydridoruthenium complex and methane [13]. The formation of carbonyl complexes from THF as a CO source has been found only for a rhodium complex [4].

Organolithium compounds are known to react with THF to form an α-metalated THF which cycloreverts to ethylene and the enolate of acetaldehyde [14]. In analogy we can suggest such a mechanism for the formation of the molybdenum carbonyl complexes. But the amount of liberated ethylene was not large and neither acetaldehyde nor methane (the expected decarbonylation product of acetaldehyde) was obtained. Thus the mechanism of their formation could not be clarified.

The formation of the hydride from the hydroaromatic compounds would be of particular interest since the reactions apparently occur via saturated C—H bond activation. Group VIII metal catalysts have been reported to be active in the homogeneous hydrogen transfer from the donors such as THF, pyrrolidine, dioxane, and tetralin, as well as secondary alcohols, to a variety of acceptors [15], but formation of hydride complexes was not confirmed. The isolation of $MoH_4(dpe)_2$ in our reactions may be suggestive of the possibility of the intermediacy of hydride complexes in the transfer hydrogenation catalyst systems using the hydroaromatics as a hydrogen donor. The complexes *trans*-$Mo(N_2)_2(dpe)_2$ and $MoH_4(dpe)_2$ proved to be effective for the transfer hydrogenation of ketones and olefins, as expected [6].

Experimental

All reactions were carried out under prepurified nitrogen unless otherwise noted. *trans*-$Mo(N_2)_2(dpe)_2$ was prepared by a known method [16]. DMF was dried and distilled under nitrogen. Other organic compounds were purified by distillation before use. Solvents were dried and distilled by using standard techniques.

Reaction of DMF

DMF (0.63 g, 9.6 mmol) was added to a Schlenk type flask containing *trans*-$Mo(N_2)_2(dpe)_2$ (66 mg, 0.07 mmol) in benzene (3 ml) and the mixture was heated under reflux for 20 min. During the reaction, samples of the reaction gas were withdrawn by a syringe through a serum cap and analysed by GLC on a molecular sieve 5A column. After cooling, GLC analysis of the liquid phase on a 28% Pennwalt 223-4% KOH column showed formation of 0.056 mmol of dimethylamine. Then n-hexane was added and the precipitate formed was filtered, washed with ether and n-hexane, and dried in vacuo to yield dark red crystals of $Mo(CO)(DMF)(dpe)_2$ (50 mg, 72%), whose identity was confirmed by IR [17].

Other reactions of amides and formate esters were carried out in similar manner. $Mo(CO)(HCONEt_2)(dpe)_2$ was identified by IR [17].

Reaction of an excess of heptanal

Heptanal (0.24 g, 2.1 mmol) was added to a solution of *trans*-$Mo(N_2)_2(dpe)_2$ (66 mg, 0.07 mmol) in benzene (3 ml). The mixture was heated under reflux for 20 min. GLC analysis of the gas phase showed formation of 0.041 mmol of H_2. After cooling, GLC analysis of the liquid phase on a Durapak n-octane/ Porasil C column revealed formation of 0.004 mmol of n-hexane and a trace amount of 1-hexene. Addition of n-hexane gave yellow crystals of *cis*-$Mo(CO)_2(dpe)_2$ (10 mg, 15%). The filtrate was evaporated to dryness in vacuo and washed with n-hexane to yield a brown powder (31 mg) whose IR spectrum showed two strong bands at 1630 and 1560 cm^{-1}. These peaks seem to be characteristic of two acyl ligands [11], but purification by recrystallization was unsuccessful, and we could not determine the formulation of this complex.

Other reactions of aldehydes were carried out in similar manner. In the case of benzaldehyde, toluene was used as solvent instead of benzene and GLC analysis on a 10% PEG 20M/Chromosorb W column indicated formation of benzene and benzophenone. In the case of acrolein, GLC analysis was on a Porapak Q column to check C_2 hydrocarbons.

Reaction of ethanol

Ethanol (32 mg, 0.7 mmol) was added to a solution of *trans*-$Mo(N_2)_2(dpe)_2$ (66 mg, 0.07 mmol) in benzene (3 ml). The mixture was heated under reflux for 20 min. GLC analysis of the gas phase on a molecular sieve 5A column showed formation of H_2 (0.023 mmol) and methane (0.030 mmol). After cooling, GLC-MS analysis of the liquid phase using a 20% PEG 1500/Celite 545 column indicated formation of acetaldehyde. Then n-hexane was added and the precipitate formed was filtered and washed with n-hexane to yield a mixture of *trans*-$Mo(CO)(N_2)(dpe)_2 \cdot 1/2\ C_6H_6$ (orange crystals), *cis*-$Mo(CO)_2(dpe)_2$ (yellow microcrystals), and $MoH_4(dpe)_2$ (yellow crystals). The respective IR spectrum was almost identical with that of the known compound. However, the yields could not be determined directly owing to the difficulties in separating the complexes completely from the mixture. Therefore the mixture was weighed as such and the content of each complex was estimated from the IR spectrum of the mixture by using the molar extinction coefficient of $\nu(C\equiv O)$ or $\nu(Mo—H)$ of an independently synthesized authentic sample.

Other reactions of primary alcohols were carried out in similar manner.

Reaction of indoline

Indoline (167 mg, 1.4 mmol) was added to a solution of *trans*-$Mo(N_2)_2(dpe)_2$ (66 mg, 0.07 mmol) in benzene (3 ml). The mixture was refluxed for 15 min. GLC analysis of the liquid phase on a 10% Silicone SE-30/Chromosorb W column indicated formation of 0.018 mmol of indole. Addition of n-hexane deposited yellow crystals of $MoH_4(dpe)_2$, which were filtered and washed with n-hexane (7 mg, 11%).

Other reactions of the hydrogen donors were carried out in similar manner. In the case of pyrrolidine, dioxane, and THF, no solvent was used.

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