green acicular crystals. Yield: 437 mg, 64%. Anal. Calcd (found) for $V_2Cl_4P_2O_4C_{36}H_{44}$: C, 51.06 (51.88); H, 5.20 (5.43); P, 7.32 (7.49). IR (Nujol cm^{-1}): 2995 (s), 2852 (s), 1616 (m), 1457 (s), 1376 (s), 1260 (m), 1118 (s), 998 (s) (probably $\nu(V{=}O)$), 906 (m), 736 (s), 680 (br), 491, 461.

Structure determination of 2. A blue-green crystal of dimensions 0.20 × 0.10 × 0.10 mm was mounted on an Enraf-Nonius CAD4 diffractometer, and accurate cell parameters were derived from 25 independent reflections. The crystal data parameters and other pertinent details are presented in Table I.

Intensities were measured with graphite-monochromated Mo Kα radiation. A correction based on the decay of standard reflections was applied to the data. Lorentz and polarization corrections were applied as was an empirical absorption correction based on a series of Ψ-scans.

The centrosymmetric space group $P\bar{1}$ was indicated by intensity statistics. The positions of the V, P, Cl, and O atoms were taken from the direct-methods solution.[15] All remaining non-hydrogen atoms were found in one difference Fourier map. All hydrogen atoms were placed at ideal positions 0.95 Å from the bonded carbon atom. Refinement calculations were performed on a Digital Equipment Corp. MicroVAX II computer using the CAD4-SDP programs.[16]

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Supplementary Material Available: Tables of crystallographic data, anisotropic thermal parameters, bond lengths and bond angles, and general displacement parameters (5 pages); a table of calculated and observed structure factors (15 pages). Ordering information is given on any current masthead page.

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Simple, High-Yield Syntheses of Molybdenum(VI) Bis(imido) Complexes of the Type $Mo(NR)_2Cl_2$(1,2-dimethoxyethane)

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Introduction

Imido ligands have been widely used to stabilize high oxidation state metal centers and to provide steric protection about the metal center;[1] the latter is an important property of imido complexes that is not true of analogous oxo complexes. We have employed bis((2,6-diisopropylphenyl)imido) complexes of molybdenum[2,3] and tungsten[4,5] as part of a scheme in which metathetically active complexes of the type $M(CH\text{-}t\text{-}Bu)(N\text{-}2,6\text{-}C_6H_3\text{-}i\text{-}Pr_2)(OR)_2$ are prepared. We also have prepared a variety of rhenium,[6–13] osmium,[14,15] and ruthenium[16] (2,6-diisopropylphenyl)imido complexes. Several 2,6-disubstituted arylimido complexes have been reported from other laboratories recently.[17,18] *tert*-Butylimido ligands have been used much more commonly than 2,6-disubstituted arylimido ligands to stabilize high oxidation state transition metal complexes,[1] but in at least two cases species that contain 2,6-disubstituted arylimido ligands have been synthesized that have no *tert*-butylimido analogues.[13,15]

Certain imido complexes can be prepared most simply by adding a primary amine to a metal–oxo complex, water being the byproduct. For example, addition of *tert*-butylamine to a solution of osmium tetraoxide affords $OsO_3(N\text{-}t\text{-}Bu)$, the first reported imido complex, in up to 90% yield.[19–21] Imido complexes also can be prepared by treating an oxo complex with an isocyanate[5,6,18,22,23] or by adding a (trimethylsilyl)amine to a metal oxide or chloride.[24] A method that combines several of these approaches was perfected in order to synthesize $Mo(NAr)_2Cl_2$(DME) (Ar = 2,6-diisopropylphenyl, DME = 1,2-dimethoxyethane), i.e., adding $ArNH_2$, Me_3SiCl, and 2,6-lutidine to a solution of $MoO_2Cl_2(THF)_2$.[2] 2,6-Lutidine and Me_3SiCl can be viewed most simply as agents that remove HCl and water generated during the reaction, although they may also have more complex roles. The analogous tungsten complex has been prepared in a similar manner from $WOCl_4$.[5] Molybdenum(VI) dioxide dichloride also has been used by other researchers to prepare bis(imido) complexes from isocyanates.[18,23] We report here new syntheses of molybdenum(VI) bis(imido) complexes that employ inexpensive, air-stable $(NH_4)_2Mo_2O_7$, that are easily carried out, and that are quantitative in several cases.

Results and Discussion

Treatment of $(NH_4)_2Mo_2O_7$ with 8 equiv of triethylamine, an excess of chlorotrimethylsilane (~17 equiv), and 4 equiv of a substituted aniline (eq 1) at ~65 °C for 6–10 h in DME under

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$$(NH_4)_2Mo_2O_7 + 4RNH_2 + 8NEt_3 + 17Me_3SiCl \xrightarrow[\text{DME}]{\sim 65\ °\text{C},\ 6\text{–}10\ \text{h}} \underset{\mathbf{1}}{2Mo(NR)_2Cl_2(DME)} \quad (1)$$

	R	% yield
1a	2,6-diisopropylphenyl	98
1b	2,6-dimethylphenyl	99
1c	2-*tert*-butylphenyl	78
1d	pentafluorophenyl	74

$$(NH_4)_2Mo_2O_7 + 4PhNH_2 + 8NEt_3 + 17Me_3SiCl \xrightarrow[\text{DME}]{25\ °\text{C},\ 20\ \text{h}} \underset{\mathbf{1e}\ (97\%)}{2Mo(NPh)_2Cl_2(DME)} \quad (2)$$

$$(NH_4)_2Mo_2O_7 + 16t\text{-}BuNH_2 + 28Me_3SiCl \xrightarrow[\text{DME}]{65\ °\text{C},\ 5\ \text{h}} \underset{\mathbf{1f}\ (58\%)}{2Mo(N\text{-}t\text{-}Bu)_2Cl_2(DME)} \quad (3)$$

a nitrogen atmosphere yields $Mo(NR)_2Cl_2(DME)$ in high to essentially quantitative yields. The reaction that yields the phenylimido complex (**1e**, eq 2) did not require heating. $Mo(N\text{-}t\text{-}Bu)_2Cl_2(DME)$ (**1f**) can be prepared similarly from $(NH_4)_2Mo_2O_7$ and excess *tert*-butylamine and chlorotrimethylsilane (eq 3). (The solvent-free version of **1f** has been reported in the literature.[24,25]) Chlorotrimethylsilane is consumed to form (ultimately) hexamethyldisiloxane, and HCl is removed as the hydrochloride salt by triethylamine (or *tert*-butylamine in the reaction shown in eq 3). The reaction is easily worked up by filtering off the ammonium salts under an inert atmosphere and removing the solvents and volatile products from the filtrate in vacuo. In cases where the yield is very high, no further purification usually is necessary. The (2,6-diisopropylphenyl)imido complex (**1a**) has been prepared on a 70-g scale starting from 20 g of $(NH_4)_2Mo_2O_7$. The (2,6-diisopropylphenyl)imido derivative is the most valuable to us at this point, as it is the first intermediate in the three-step synthesis of $Mo(CH\text{-}t\text{-}Bu)(NAr)(triflate)_2(DME)$, a universal precursor to a variety of complexes of the type $Mo(CH\text{-}t\text{-}Bu)(NAr)(OR)_2$ that are active for the metathesis of olefins and are especially useful for the controlled ring opening metathesis polymerization of cyclic olefins.[26,27] NMR data for all of the complexes prepared in this work are consistent with an octahedral structure containing cis imido ligands and trans chlorides, although exchange of DME on the NMR time scale is often observed. Ready loss of DME appears to thwart elemental analysis in the case of **1e**. Treating **1e** with $AgOSO_2CF_3$ yields $Mo(NPh)_2(OSO_2CF_3)_2(DME)$, a compound in which DME exchange is slow on the NMR time scale, which analyzes reproducibly.

The syntheses reported here are analogous to those of bis- or tris(imido)rhenium(VII) compounds such as $Re(NAr)_2Cl_3(py)$ (Ar = 2,6-diisopropylphenyl or 2,6-dimethylphenyl),[8] $Re(N\text{-}t\text{-}Bu)_2Cl_3$,[8] $Re(N\text{-}t\text{-}Bu)_3(OSiMe_3)$,[8] $Re(N\text{-}2,6\text{-}C_6H_3\text{-}i\text{-}Pr_2)_3Cl$,[28] and $Re(N\text{-}2,6\text{-}C_6H_3Me_2)_3(OSiMe_3)$,[28] which may be obtained in one-step, high-yield reactions from Re_2O_7 or $(NH_4)ReO_4$. Attempts to extend this approach to tungsten by employing a variety of tungstate starting materials so far have failed, perhaps because of the relatively slow rate of reactions involving tungsten (relative to Mo) and decomposition of $W(NR)_2Cl_2(DME)$ complexes at temperatures where rates are practical.

Experimental Section

General Details. All experiments were performed under a nitrogen atmosphere in a Vacuum Atmospheres drybox or by standard Schlenk techniques unless otherwise specified. Pentane was washed with sulfuric/nitric acid (95/5 v/v), sodium bicarbonate, and then water, stored over calcium chloride, and then distilled from sodium benzophenone ketyl under nitrogen. Reagent grade diethyl ether, tetrahydrofuran, toluene, benzene, and 1,2-dimethoxyethane were distilled from sodium benzophenone ketyl under nitrogen. Reagent grade dichloromethane was distilled from calcium hydride under nitrogen. All deuterated NMR solvents were passed through a column of activated alumina prior to use. $(NH_4)_2Mo_2O_7$ was purchased commercially and used as received.

NMR data are listed in parts per million downfield from tetramethylsilane for proton and carbon and downfield from $CFCl_3$ for fluorine. Coupling constants are listed in hertz. Spectra were obtained in benzene-d_6 at 25 °C unless otherwise noted. Elemental analyses (C, H, N) were performed on a Perkin-Elmer 2400 CHN analyzer.

$Mo(N\text{-}2,6\text{-}C_6H_3\text{-}i\text{-}Pr_2)_2Cl_2(DME)$ (1a). $(NH_4)_2Mo_2O_7$ (10.00 g, 29.4 mmol) was suspended in DME (150 mL) at room temperature, and triethylamine (23.80 g, 235 mmol) in 10 mL of DME was slowly added to the stirred suspension over a period of 5 min. Chlorotrimethylsilane (54.20 g, 500 mmol) in DME (20 mL) was then added over a period of 5 min. The solution became opaque. Finally, 2,6-diisopropylaniline (20.80 g, 118 mmol) in DME (15 mL) was added over a period of 5 min. The solution turned yellow and then deep red as additional white precipitate formed. The mixture was then heated to 65 °C for 6 h. The reaction mixture was cooled to room temperature and filtered to remove the ammonium salts. The salts were washed with DME until the DME ran through colorless. The volatile components were removed from the filtrate in vacuo to yield 35 g (98%, 57.6 mmol) of the brick-red product, which is pure by NMR and can be used directly in subsequent reactions. The NMR spectra and physical properties of the compound are identical to those reported previously.[2] The solid can be purified further by washing with cold pentane or recrystallization from DME.

$Mo(N\text{-}2,6\text{-}C_6H_3Me_2)_2Cl_2(DME)$ (1b). Ammonium dimolybdate (5.00 g, 14.7 mmol) was suspended in 70 mL of DME at room temperature. Triethylamine (11.90 g, 118 mmol) in 5 mL of DME, chlorotrimethylsilane (27.10 g, 250 mmol) in 20 mL of DME, and 2,6-dimethylaniline (7.13 g, 59 mmol) in 15 mL of DME were added sequentially, each over a period of about 5 min. The mixture was heated to 60 °C for 8 h and the product isolated as described for the synthesis of **1a**; yield 14.5 g (99%, 29.2 mmol) of red-purple product. ^1H NMR: δ 6.76 (d, 4, H_m), 6.66 (m, 2, H_p), 3.44 (s, 6, *Me*O), 3.18 (s, 4, CH_2O), 2.65 (s, 12, $NC_6H_3Me_2$). ^{13}C NMR (CD_2Cl_2): δ 156.1 (s, C_i), 133.8 (s, C_o), 127.4 (d, J_{CH} = 160, C_m), 126.4 (d, J_{CH} = 160, C_p), 71.1 (t, J_{CH} = 145, OCH_2), 63.1 (q, J_{CH} = 145, *Me*O), 18.0 (q, J_{CH} = 127, $C_6H_3Me_2$). Anal. Calcd for $MoC_{20}H_{28}N_2O_2Cl_2$: C, 48.50; H, 5.70; N, 5.66. Found: C, 48.41; H, 5.85; N, 5.74.

$Mo(N\text{-}2\text{-}C_6H_4\text{-}t\text{-}Bu)_2Cl_2(DME)$ (1c). Triethylamine (0.476 g, 4.70 mmol), chlorotrimethylsilane (1.15 g, 10.6 mmol), and 2-*tert*-butylaniline (0.351 g, 2.34 mmol) were added sequentially to a stirred solution of $(NH_4)_2Mo_2O_7$ (0.200 g, 0.588 mmol) in 10 mL of DME, and the mixture was heated to 65 °C for 10 h, then cooled to room temperature, and filtered. The volatile components were removed from the filtrate in vacuo, and the solid residue was recrystallized from diethyl ether to yield 0.507 g of red needles (78%, 0.919 mmol). ^1H NMR (C_6D_6): δ 8.18 (d, 2, H_o), 7.15 (d, 2, H_m), 6.95 (t, 2, H_m), 6.75 (t, 2, H_p), 3.39 (s, 6, *Me*O), 3.17 (s, 4, CH_2O), 1.54 (s, 18, CMe_3). ^{13}C NMR (C_6D_6): δ 156.1 (s, C_i), 140.8 (s, C_o), 128.8–125.6 (m, C_o, C_m, and C_p), 71.1 (t, J_{CH} = 146, CH_2O), 62.6 (q, J_{CH} = 145, *Me*O), 35.6 (s, CMe_3), 30.9 (q, J_{CH} = 126, CMe_3). Anal. Calcd for $MoC_{28}H_{42}N_2O_2Cl_2$: C, 52.28; H, 6.58; N, 5.08. Found: C, 52.23; H, 6.90; N, 4.94.

$Mo(NC_6F_5)_2Cl_2(DME)$ (1d). To $(NH_4)_2Mo_2O_7$ (1.00 g, 2.94 mmol) in 20 mL of DME were added NEt_3 (2.38 g, 23.52 mmol) in 1 mL of DME, Me_3SiCl (5.43 g, 49.98 mmol) in 1 mL of DME, and finally $C_6F_5NH_2$ (2.15 g, 11.74 mmol) in DME (5 mL). The mixture was stirred at room temperature for 12 h and at 70 °C for another 6 h. The red reaction mixture was allowed to cool to room temperature and was filtered. The filter cake was rinsed with DME. The volatile components were removed from the filtrate in vacuo, and the waxy red solid that remained was washed with pentane (3 × 20 mL) and dried under vacuum; yield 2.70 g (74%). Recrystallization from a mixture of DME and pentane at −40 °C gave analytically pure red crystals. ^1H NMR ($CDCl_3$): δ 4.05 (s, 4, CH_2O) and 4.00 (s, 6, *Me*O). ^{19}F NMR ($CDCl_3$): δ −146.0 (d, 4, F_o), −152.9 (t, 2, F_p), −162.5 (t, 4, F_m). ^{13}C NMR ($CDCl_3$): δ 143.0 (d of d, C_o), 141.2 (d of t, C_p), 137.2 (d of t, C_m), 131.6 (t, C_i), 71.7 (s, CH_2O), 64.6 (s, *Me*O). Anal. Calcd for $MoC_{16}H_{10}Cl_2F_{10}N_2O_2$: C, 31.04; H, 1.63; N, 4.52. Found: C, 30.85; H, 1.67; N, 4.51.

$Mo(NPh)_2Cl_2(DME)$ (1e). Ammonium dimolybdate (0.200 g, 0.588 mmol) was suspended in 10 mL of DME, and triethylamine (0.476 g, 4.70 mmol), chlorotrimethylsilane (1.150 g, 10.6 mmol), and aniline (0.219 g, 2.34 mmol) were added. The reaction mixture was stirred at room temperature for 20 h. The solution was filtered, and the DME was removed from the filtrate in vacuo to afford 0.493 g of dark red product (97%, 1.12 mmol). ^1H NMR (CD_2Cl_2, 40 °C): δ 7.55–7.10 (m, 10, H_{aryl}), 3.96 (br s, 4, CH_2O), 3.89 (br s, 6, *Me*O). ^{13}C NMR (CD_2Cl_2):

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δ 156.3 (s, C_i), 128.7 (dd, J_{CH} = 159, C_o), 127.2 (br d, J_{CH} = 164, C_m), 123.5 (br d, J_{CH} = 162, C_p), 71.5 (t, J_{CH} = 145, *C*H$_2$O), 63.5 (br q, J_{CH} = 139, *Me*O). The elemental analyses of **1e** were variable, we presume, because DME is labile. The analogous triflate derivative (see below) analyzed consistently.

$Mo(NPh)_2(OSO_2CF_3)_2(DME)$. $Mo(NPh)_2Cl_2(DME)$ (1.00 g, 2.3 mmol) was dissolved in 40 mL of diethyl ether and the solution was chilled to −20 °C. Silver trifluoromethanesulfonate (1.17 g, 4.6 mmol) was added in small solid portions, and a precipitate of silver chloride formed. The reaction was allowed to warm to room temperature and was stirred overnight. The silver chloride was removed by filtration and washed with DME. The solvents were removed from the filtrate in vacuo, and the residue was recrystallized from dichloromethane layered with pentane; yield (1.27 g, 84%). This reaction may also be run in DME as the solvent. ^{1}H NMR (CD_2Cl_2): δ 7.45–7.20 (m, 10, H_{aryl}), 4.08 (s, 6, *Me*O), 4.04 (s, 4, C*H*$_2$O). ^{13}C NMR (CD_2Cl_2): δ 156.9 (s, C_i), 130.2 (d, J_{CH} = 139, C_o), 128.9 (d, J_{CH} = 144, C_m), 125.9 (d, J_{CH} = 165, C_p), 72.2 (t, J_{CH} = 148, C*H*$_2$O), 65.4 (q, J_{CH} = 144, *Me*O). ^{19}F NMR (CD_2Cl_2): δ 29.5 (s). Anal. Calcd for $MoC_{18}H_{20}F_6Cl_2N_2O_8S_2$: C, 32.44; H, 3.02; N, 4.20. Found: C, 32.31; H, 3.01; N, 4.13.

$Mo(N\text{-}t\text{-}Bu)_2Cl_2(DME)$ (1f). Chlorotrimethylsilane (8.91 g, 0.082 mol, 28 equiv) was added dropwise to a solution of *tert*-butylamine (3.43 g, 0.047 mol, 16 equiv) in DME (60 mL). A white precipitate formed. To this slurry was added ammonium dimolybdate (1 g, 2.94 mmol) all at once as a solid. The mixture was heated to 65 °C for 5 h. The mixture was then filtered. The solvent was removed from the orange-brown filtrate to give a brown residue. The brown residue was extracted with pentane to give a yellow solution, from which the solvent was removed in vacuo to give $Mo(N\text{-}t\text{-}Bu)_2Cl_2(DME)$ as a yellow powder which is >95% pure by ^{1}H NMR; yield 1.35 g (58%). Recrystallization from pentane yields analytically pure yellow needles. ^{1}H NMR: δ 3.46 (s, 6, *Me*O), 3.24 (s, 4, C*H*$_2$O), 1.40 (s, 18, C*Me*$_3$). ^{13}C NMR: δ 71.7 (s, *C*Me$_3$), 70.7 (q, *C*H$_3$O), 62.4 (d, *C*H$_2$O), 30.1 (q, C*Me*$_3$). Anal. Calcd for $MoC_{12}H_{28}N_2Cl_2O_2$: C, 36.10; H, 7.07; N, 7.02. Found: C, 35.87; H, 7.13; N, 6.76.

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Crystal Structure and Infrared and NMR Spectra of $\{[(C_5H_5N)_4Na]_2[Fe_2(CO)_8]\}_\infty$: A Two-Dimensional Extended Structure Due to Sodium–Isocarbonyl Interactions

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Introduction

Transition-metal carbonylate anions are of great importance in organometallic chemistry.[1] Solid-state structures of metal carbonylate anions are usually determined by X-ray analyses with large counterions (e.g., $[PPN]^+$, $[PPh_4]^+$), the anions and cations being well separated in crystal lattices. With alkali-metal, alkaline-earth-metal aluminum,[2] and lanthanide cations,[3] interactions between these cations and the carbonyl oxygen atoms of the metal carbonylates (i.e. isocarbonyl linkages) have been observed in solution as well as in the solid state, giving rise to contact ion pairs. Ion-pairing phenomena are most conveniently studied in solution by infrared spectroscopy and in the solid state by single-crystal X-ray analyses.[2]

Table I. Crystallographic Data for $\{[(C_5H_5N)_4Na]_2[Fe_2(CO)_8]\}_\infty$

chem formula	$C_{48}H_{40}Fe_2N_8$-Na_2O_8	*Z*	4
		d(calc), g cm^{-3}	1.346
fw	1014.58	temp, °C	−60
space group	*Pbca* (No. 61)	radiation (λ, Å)	Mo Kα (0.710 73)
a, Å	15.796 (4)	μ, cm^{-1}	6.512
b, Å	17.203 (6)	$R(F_o)$	0.057
c, Å	18.428 (4)	$R_w(F_o)$	0.045
V, Å^3	5007.3		

The solid-state structures of the sodium and potassium salts of the typical mononuclear carbonylate anions $[Fe(CO)_4]^{2-}$ have been determined.[4] They reveal interesting interactions between $[Fe(CO)_4]^{2-}$ and Na^+ or K^+. The structure of the typical dinuclear dianion $[Fe_2(CO)_8]^{2-}$ has previously been determined as its $[PPN]^+$ and $[PPh_4]^+$ salts.[5] In view of the interesting structural aspects of the ion-pairing phenomena in the solid state,[2] we set out to determine the structure of $Na_2[Fe_2(CO)_8]$. $Na_2[Fe_2(CO)_8]$ crystallizes in saturated pyridine solutions as the title compound. Reported herein is the novel two-dimensional extended sheetlike structure of $\{[(C_5H_5N)_4Na]_2[Fe_2(CO)_8]\}_\infty$ as determined by X-ray analysis and related NMR and infrared studies. The crystal structure of $\{[(C_5H_5N)_4Na]_2[Fe_2(CO)_8]\}_\infty$ is to our knowledge the first example of a dinuclear transition-metal carbonylate dianion involved in an extended structure via isocarbonyl linkages.[6]

Experimental Section

All manipulations were carried out on a standard high-vacuum line or in a drybox under an atmosphere of dry, pure N_2. Tetrahydrofuran (THF) was dried by distillation from sodium–benzophenone ketyl into storage bulbs equipped with Teflon stopcocks and containing sodium–benzophenone ketyl. CH_3CN was dried over P_4O_{10} with continuous stirring for 2–3 days followed by distillation into storage bulbs. Pyridine was dried over sodium and distilled prior to use. $Na_2[Fe_2(CO)_8]$ and $[PPh_4]_2[Fe_2(CO)_8]$ were prepared by procedures reported in the literature.[5b]

All IR spectra were recorded with 2-cm^{-1} resolution using a Mattson-Polaris FT-IR spectrometer. Solution spectra were obtained in Perkin-Elmer liquid cells with 0.1-mm Teflon spacers and KBr windows. Spectra in Nujol mull were obtained with KBr plates. Proton NMR (δ(TMS) = 0.00 ppm) and ^{13}C NMR (δ(TMS) = 0.00 ppm) spectra were obtained on a Bruker AM-250 NMR spectrometer operating at 250.14 and 101.25 MHz, respectively.

Preparation of $\{[(C_5H_5N)_4Na]_2[Fe_2(CO)_8]\}_\infty$. In the drybox 500 mg of $Na_2[Fe_2(CO)_8]$ was dissolved in a minimal amount of dry pyridine and the resulting solution was filtered and stored at −40 °C for 1 week, during which time red crystals of $\{[(C_5H_5N)_4Na]_2[Fe_2(CO)_8]\}_\infty$ bigger than 2 × 2 × 2 mm^3 were obtained.

Infrared Spectra. For $\{[(C_5H_5N)_4Na]_2[Fe_2(CO)_8]\}_\infty$ (ν_{CO}, cm^{-1}): in Nujol mull 1927 (m), 1855 (m, sh), 1830 (s); in THF 1986 (w), 1970 (vw), 1922 (s), 1875 (s), 1829 (m); in CH_3CN 1977 (w), 1939 (w), 1914 (s), 1863 (s), 1840 (w, sh); in pyridine 1973 (w), 1942 (w, sh), 1914 (m), 1863 (s), 1940 (w, sh). For $[PPh_4]_2[Fe_2(CO)_8]$ (ν_{CO}, cm^{-1}): in Nujol mull 1909 (m), 1847 (s, br); in CH_3CN[5b] 1992 (vw), 1953 (w, sh), 1914 (m), 1865 (s).

^{13}C{^{1}H} NMR Spectra. For $\{[(C_5H_5N)_4Na]_2[Fe_2(CO)_8]\}_\infty$ (THF-d_8, 303 K, ppm): δ 227.84 (s, carbonyls); δ 150.74 (s), δ 136.24 (s), δ 124.29 (s, pyridines).

X-ray Crystal Structure Determination. A crystal of the size 0.45 × 0.50 × 0.60 mm^3 was cut from a bigger crystal, coated with epoxy resin, and then mounted on the tip of a glass fiber under N_2. All crystallographic data were collected at −60 °C on an Enraf-Nonius CAD4 diffractometer with graphite-monochromated Mo Kα radiation. Unit cell parameters were obtained by a least-squares refinement of the angular

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(6) For other examples of extended structures due to Na^+–isocarbonyl linkages, see ref 13a,e,g.

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