

Chromium-Centered Imido Group Transfer

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Introduction

Although high-valent chromium oxides and chromyl chloride are widely used in organic oxidations,¹ examples of chromium-centered nitrogen atom transfer reactions are rare. The organoimido–Cr complexes isolated thus far are relatively inert toward imido group transfer.^{2,3,4} In fact, there are few mechanistic studies on the atom transfer reactions of organoimido complexes,⁵ in contrast to a wealth of information on oxo transfer chemistry of the isoelectronic oxometal counterparts.⁶ Metal-catalyzed aziridination of olefins by PhI=NTs (Ts = *p*-toluenesulfonyl), presumably via the unisolated (tosylimido)-metal intermediates, is well documented, however.⁷ Recently, Bruce and coworkers demonstrated that the rate of olefin epoxidation by oxochromium(V) porphyrins correlates with the Cr^{VI}–Cr^V reduction potential, suggesting that charge transfer plays a key role in the chromium-centered oxo transfer.⁸ In this regard, we believe that cationic imido–Cr complexes with high formal potentials could be more reactive toward atom transfer reactions than their neutral analogues. As our continuous effort in elucidating the factors governing metal-mediated nitrogen atom transfer reaction, we here report the synthesis, crystal structure, and imido transfer reaction of a dicationic diimido–Cr(VI) complex.

Experimental Section

All solvents were dried and distilled prior to use. NMR spectra were recorded on a JEOL EX 400 spectrometer, mass spectra on a

Table 1. Crystallographic Data for 1·0.5C₆H₁₄

chem formula	[C ₂₃ H ₂₉ N ₅ B ₂ F ₈ Cr]·0.5C ₆ H ₁₄	<i>T</i> , °C	25
<i>a</i> , Å	12.302 (7)	<i>λ</i> , Å	0.71073
<i>b</i> , Å	20.483 (3)	<i>Q</i> _{calcd.} , g cm ⁻³	1.338
<i>c</i> , Å	25.384 (8)	<i>μ</i> , cm ⁻¹	4.17
<i>V</i> , Å ³	6396 (5)	<i>R</i> ^a	0.091
<i>Z</i>	8	<i>R</i> _w ^b	0.090
fw	644.21	GOF ^c	1.180
space group	<i>Pbcn</i> (No. 60)		

^a $R = \sum |F_o| - |F_c| / \sum |F_o|$. ^b $R_w = [\sum w(|F_o| - |F_c|)^2 / \sum w|F_o|^2]^{1/2}$. ^c GOF = $[\sum w(|F_c| - |F_o|)^2 / (N_{obs} - N_{param})]^{1/2}$.

Table 2. Selected Bond Lengths (Å) and Angles (deg) for 1·0.5C₆H₁₄

Cr(1)–N(1)	2.04(1)	Cr(1)–N(2)	2.03(1)
Cr(1)–N(3)	2.05(1)	Cr(1)–N(4)	1.62(1)
Cr(1)–N(5)	1.65(1)	N(1)–C(1)	1.40(2)
N(1)–C(5)	1.35(2)	N(2)–C(10)	1.30(2)
N(3)–C(11)	1.35(2)	N(3)–C(15)	1.32(2)
N(4)–C(16)	1.47(2)	N(5)–C(2)	1.45(2)
N(1)–Cr(1)–N(2)	75.7(6)	N(1)–Cr(1)–N(3)	153.4(5)
N(1)–Cr(1)–N(4)	94.9(7)	N(1)–Cr(1)–N(5)	100.1(7)
N(2)–Cr(1)–N(3)	77.8(6)	N(2)–Cr(1)–N(4)	125.8(6)
N(2)–Cr(1)–N(5)	118.7(6)	N(3)–Cr(1)–N(4)	98.7(7)
N(3)–Cr(1)–N(5)	94.5(7)	N(4)–Cr(1)–N(5)	115.6(6)
Cr(1)–N(1)–C(5)	117(1)	C(1)–N(1)–C(5)	117(1)
Cr(1)–N(1)–C(1)	124(1)	Cr(1)–N(2)–C(6)	118(1)
Cr(1)–N(2)–C(10)	118(1)	C(6)–N(2)–C(10)	122(1)
Cr(1)–N(3)–C(11)	118(1)	Cr(1)–N(3)–C(15)	124(1)
C(11)–N(3)–C(15)	117(1)	Cr(1)–N(4)–C(16)	168(1)
Cr(1) N(5) C(20)	165(1)		

Kratos MS 80FRAQ spectrometer, and UV/vis spectra on a Milton Roy Spectronic 3000 diode array spectrophotometer. Cyclic voltammetry was performed with a Princeton Applied Research (PAR) Model 273A potentiostat. Potentials were with respect to a Ag⁺/Ag reference electrode in acetonitrile but are reported respect to the ferrocenium/ferrocene couple as measured in the same solution. Elemental analyses were done by Medac Ltd., Brunel University, U.K.

Caution! High-valent chromium compounds are potentially carcinogenic and should be handled appropriately, avoiding skin contact and inhalation. [Cr(*N-t*-Bu)₂Cl₂]^{2b} was prepared as described previously. The terpy (2,2':6',2''-terpyridine) ligand was obtained from Aldrich and used as received. NaOAr (Ar = 2,6-diisopropylphenyl) was synthesized by reaction of HOAr with 2 equiv of NaH in THF.

Preparation. [Cr(*N-t*-Bu)₂(terpy)](BF₄)₂ (1). To a solution of Cr(*N-t*-Bu)₂Cl₂ (0.3 g, 1.13 mmol) in CH₂Cl₂ (10 mL) was added 1 equiv of terpy (0.26 g, 1.13 mmol) and the mixture was stirred at room temperature for 2 h. The orange precipitate was collected and washed with ether. To the orange solid was added acetonitrile (10 mL) and AgBF₄ (0.44 g, 2.26 mmol), and the resulting mixture was stirred overnight and filtered. The filtrate was layered with ether (20 mL) and left to stand at room temperature for 2 day. The orange crystals was collected and washed with ether (Yield 75%). ¹H-NMR (CD₃CN): δ 1.55 (s, 18H, *N-t*-Bu), 7.84–8.77 (m, 11H, terpy). IR: ν (Cr=N) 1176 cm⁻¹. MS (FAB) *m/e* 427 (M⁺ – 2BF₄). UV/vis (CH₃CN): λ_{max}/nm ($\epsilon/M^{-1} cm^{-1}$): 354 (10 600), 370 (10300). Anal. Calcd for [C₂₃H₂₉N₅B₂F₈Cr]: C, 46.0; H, 4.8; N, 11.7. Found: C, 45.5; H, 4.8; N, 11.3. The triflate salt can be prepared similarly using Ag(OTf) (OTf = triflate) instead of AgBF₄.

[Cr(terpy)(PMe₃)₃](BF₄)₃ (2). To a solution of 1 (0.3 g, 0.5 mmol) in acetonitrile (10 mL) was added excess PMe₃ (3 mmol, 3 mL of a 1M solution in toluene), and the solution was stirred at room temperature for 4 h, during which the color changed from orange to yellowish-green to dark green. The solvent was pumped off, and the residue was washed with ether and extracted with CH₂Cl₂ (12 mL). The filtrate was concentrated and layered with ether for 2 days to give green crystals (yield 0.17 g, 45%). UV/vis (CH₃CN): λ_{max}/nm ($\epsilon/M^{-1} cm^{-1}$) 285 (17 100), 334 (11 200), 441 (2090), 642 (963). $\mu_{eff} = 3.3$

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Table 3. Atomic Coordinates

atom	x	y	z	atom	x	y	z
Cr(1)	0.7880(2)	0.3326(1)	0.1234(1)	C(24)	0.5971	0.5026	0.3997
F(1)	0.223(3)	0.094(2)	0.207(2)	C(25)	0.6271	0.4650	0.4481
F(2)	0.281(2)	0.178(2)	0.175(2)	C(26)	0.5673	0.4979	0.4865
F(3)	0.378(2)	0.092(2)	0.196(2)	C(27)	0.5864	0.5084	0.6552
F(4)	0.316(3)	0.154(2)	0.255(2)	C(28)	0.6031	0.5080	0.7077
F(5)	0.403(2)	0.161(3)	0.501(2)	C(29)	0.5273	0.4869	0.7296
F(6)	0.414(2)	0.100(2)	0.562(3)	B(1)	0.295(3)	0.128(2)	0.208(2)
F(7)	0.314(2)	0.184(2)	0.576(2)	B(2)	0.347(3)	0.139(2)	0.539(2)
F(8)	0.275(3)	0.113(2)	0.527(2)	H(1)	0.7412	0.2680	0.0143
N(1)	0.798(1)	0.3541(7)	0.0453(4)	H(2)	0.7474	0.2967	-0.0734
N(2)	0.8460(10)	0.4253(5)	0.1216(6)	H(3)	0.8080	0.3976	-0.0952
N(3)	0.811(1)	0.3525(7)	0.2019(5)	H(4)	0.8553	0.4782	-0.0311
N(4)	0.860(1)	0.2666(6)	0.1195(6)	H(5)	0.9073	0.5464	0.0423
N(5)	0.6556(10)	0.3214(6)	0.1291(6)	H(6)	0.9530	0.5922	0.1210
C(1)	0.766(1)	0.3118(8)	0.0049(7)	H(7)	0.9182	0.5393	0.2036
C(2)	0.770(2)	0.327(1)	-0.0456(6)	H(8)	0.8808	0.4792	0.2729
C(3)	0.800(2)	0.386(1)	-0.0571(7)	H(9)	0.8419	0.4091	0.3434
C(4)	0.833(2)	0.4344(10)	-0.0198(8)	H(10)	0.7992	0.2959	0.3233
C(5)	0.829(1)	0.4152(8)	0.0317(6)	H(11)	0.7634	0.2659	0.2315
C(6)	0.862(1)	0.4554(10)	0.0729(7)	H(12)	1.0001	0.1542	0.0621
C(7)	0.900(2)	0.520(1)	0.0743(7)	H(13)	0.9152	0.2017	0.0382
C(8)	0.922(1)	0.5481(8)	0.119(1)	H(14)	1.0200	0.2289	0.0641
C(9)	0.902(2)	0.518(1)	0.1687(8)	H(15)	0.7800	0.1584	0.1579
C(10)	0.864(1)	0.4554(9)	0.1659(8)	H(16)	0.7647	0.1557	0.0972
C(11)	0.844(1)	0.4130(9)	0.2157(7)	H(17)	0.8431	0.1072	0.1248
C(12)	0.858(2)	0.4327(9)	0.2661(7)	H(18)	1.0172	0.1546	0.1683
C(13)	0.838(2)	0.392(1)	0.3061(7)	H(19)	1.0383	0.2290	0.1631
C(14)	0.808(2)	0.328(1)	0.2946(8)	H(20)	0.9426	0.2039	0.1974
C(15)	0.791(1)	0.3108(9)	0.2404(6)	H(21)	0.4872	0.3640	0.0836
C(16)	0.905(1)	0.2001(8)	0.1176(8)	H(22)	0.5245	0.3027	0.0526
C(17)	0.967(2)	0.196(1)	0.067(1)	H(23)	0.4158	0.3016	0.0836
C(18)	0.818(1)	0.1521(7)	0.1269(9)	H(24)	0.4700	0.2121	0.1267
C(19)	0.979(2)	0.198(1)	0.164(1)	H(25)	0.5781	0.2131	0.0952
C(20)	0.547(1)	0.295(1)	0.1291(10)	H(26)	0.5803	0.2103	0.1563
C(21)	0.489(2)	0.318(2)	0.085(1)	H(27)	0.5443	0.3080	0.2094
C(22)	0.542(2)	0.229(1)	0.121(3)	H(28)	0.5044	0.3698	0.1797
C(23)	0.505(3)	0.322(2)	0.179(1)	H(29)	0.4276	0.3102	0.1862

μ_B (Evans method⁹). Anal. Calcd: C, 37.2; H, 4.9; N, 5.4. Found: C, 37.2; H, 5.0; N, 5.7.

[Cr(N-*t*-Bu)₂(OAr)₂] (3). A mixture of [Cr(N-*t*-Bu)₂Cl₂] (0.3 g, 1.13 mmol) and 2 equiv of NaOAr (0.45g, 2.26 mmol) in toluene (15 mL) was stirred at room temperature overnight. The solvent was evaporated to dryness, and the residue was extracted with hexane. Concentrating and cooling the hexane extract at -10 °C afforded air-stable dark red crystals (yield 0.46 g, 75%). ¹H-NMR (CDCl₃): δ 1.16 (s, 18H, *t*-Bu), 1.26 (d, 24H, CH(CH₃)₂), 3.54 (sept, 4H, CH(CH₃)₂), 6.87–7.07 (m, 6H, aromatic). Anal. Calcd for [C₃₂H₃₅N₂O₂Cr]: C, 67.3; H, 6.6; N, 5.3. Found: C, 66.2; H, 6.8; N, 5.4.

Kinetic Measurement. Kinetic experiments were performed spectrophotometrically under pseudo-first-order conditions in acetonitrile using a Hewlett-Packard 8452A diode array spectrophotometer. The progress of the reaction was monitored at 362 nm. The decay of **1** was found to exhibit biphasic behavior and the pseudo-first-order rate constants k_{obs1} and k_{obs2} for the first and second step respectively were obtained by a non-linear least-square fit of the absorbance (A_t) to time t according to the equation $A_t = A_1 \exp(-k_{obs1}t) + A_2 \exp(-k_{obs2}t) + A_3$.

X-ray Analysis. A summary of the crystal and data processing parameters is given in Table 1 and selected bond lengths and angles are given in Table 2. X-ray quality crystals were obtained by slow diffusion of ether to an acetonitrile solution. Three independent batches of crystals were examined, all suffering solvent loss during crystallization; the reported set is the best of these three. An orange block (0.22 × 0.24 × 0.36 mm) was used for data collection on a Rigaku RU200-AFC7R diffractometer. Intensity data were corrected for Lorentz and polarization effects. Absorption correction by ω -scan method was also applied. The structure was solved by Patterson method and refined by a full-matrix least-squares analysis. A partially occupied hexane molecule (occupancy factor 0.5) was revealed. Their contribu-

tions are included in the structure factor calculations but were not refined. All hydrogen atoms were refined isotropically and all other atoms except B were refined anisotropically. The hydrogen atoms were included as fixed atoms in idealized positions (C–H, 0.95 Å).

Results and Discussion

Syntheses. Interaction of [Cr(N-*t*-Bu)₂Cl₂] with terpy (2,2':6',2''-terpyridine) followed by treatment with 2 equiv of AgBF₄ afforded air-stable orange [Cr(N-*t*-Bu)₂(terpy)](BF₄)₂ (**1**). The structure of **1** has been established by X-ray crystallography, and Figure 1 shows a perspective view of the cation [Cr(N-*t*-Bu)₂(terpy)]²⁺; selected bond lengths and angles are given in Table 2. To our knowledge, complex **1** is the first example of structurally characterized dicationic chromium(VI) complex. The geometry around Cr in **1** is best described as trigonal bipyramidal with two imide ligands and one terpyridyl nitrogen in the equatorial plane. The Cr–N(imido) distance of ca. 1.64 Å and Cr–N–C angle of ca. 167° are typical for diimido–Cr(VI) complexes,² indicative of triple bond character of the Cr–N(imido) bond. The Cr–N(terpy) distance of ca. 2.03 Å is comparable to that for Cr(VI)–N(pyridine) in [Cr(N-*t*-Bu)₂py₂(η^1 -O₃SCF₃)](CF₃SO₃).^{2b} The IR band at 1010 cm⁻¹ is attributed to the Cr=N-*t*-Bu stretching mode. The UV/vis spectrum of **1** in acetonitrile displays a characteristic absorption at ca. 300–400 nm, which is tentatively assigned to the N(terpy) → Cr LMCT transition.

1 reacts with tertiary phosphines such as PMe₃ cleanly to give phosphinimines *t*-BuN=PR₃, identified by ³¹P NMR spectroscopy, along with a green Cr-containing species analyzed as [Cr^{III}(terpy)(PMe₃)₃](BF₄)₃ (**2**). Complex **2** was formed presumably via oxidation of the Cr(II) product [Cr^{II}(terpy)-

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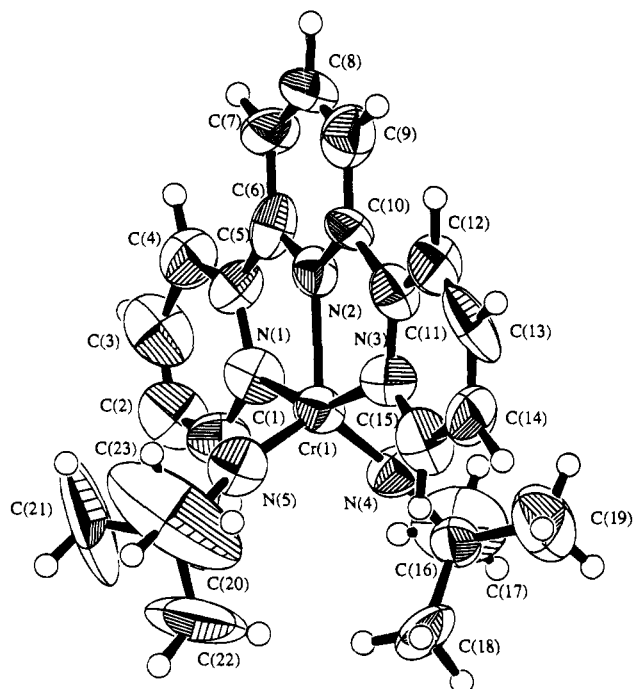


Figure 1. Perspective view of the cation $[\text{Cr}(\text{N}-t\text{-Bu})_2(\text{terpy})]^{2+}$.

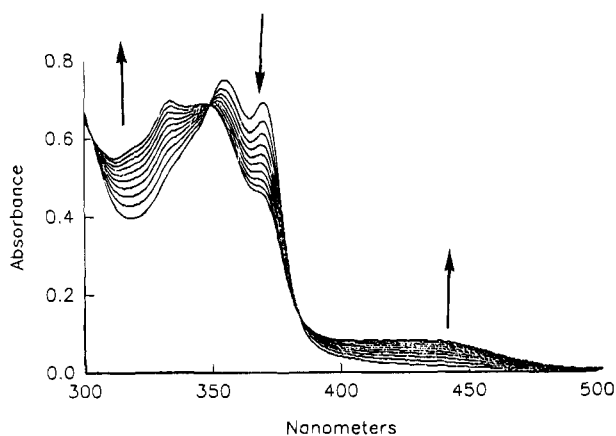


Figure 2. Optical spectral trace for the reaction between **1** and PMe_3 in CH_3CN ($[\mathbf{1}] = 0.1 \text{ mM}$, $[\text{PMe}_3] = 10 \text{ mM}$).

$(\text{PMe}_3)_3(\text{BF}_4)_2$ by adventitious air during crystallization. It should be noted that reaction of PMe_3 with neutral $[\text{Cr}(\text{N}-t\text{-Bu})_2\text{Cl}_2]$ only yields the phosphine adduct $[\text{Cr}(\text{N}-t\text{-Bu})_2\text{Cl}_2(\text{PMe}_3)]$.

Kinetics of Imido Transfer from Bis(imido)-Cr(VI). The imido transfer reactions of **1** with tertiary phosphines have been followed by UV/vis spectroscopy. Figure 2 displays a spectral trace for a reaction mixture containing ca. 0.1 mM **1** and 10 mM PMe_3 , which shows well-defined isosbestic points at 348 and 382 nm. The kinetics of reaction was followed at 362 nm with a large excess of PMe_3 : $[\mathbf{1}] = 0.1 \text{ mM}$ and $[\text{PMe}_3] = 9 \times 10^{-3}$ to $7 \times 10^{-2} \text{ M}$. Under these conditions the decay of **1** was found to follow biphasic kinetics and absorbance–time data fitted well to a biexponential model. Representative pseudo-first-order rate constants k_{obs1} and k_{obs2} at 25.0 °C for steps 1 and 2, respectively, are listed in Table 4. Plot of k_{obs1} vs $[\text{PMe}_3]$ is linear, i.e. rate = $k_2 [\text{PMe}_3]$. On the other hand, a plot of k_{obs2} vs $[\text{PMe}_3]^2$ is linear, i.e. rate = $k_3 [\text{PMe}_3]^2$. At 25.0 °C $k_2 = 0.48 \pm 0.10 \text{ M}^{-1} \text{ s}^{-1}$ and $k_3 = 1.44 \pm 0.16 \text{ M}^{-2} \text{ s}^{-1}$. These results are consistent with the following mechanism (Scheme 1). For step 2, at $K[\text{PMe}_3] \gg 1$, rate $\approx Kk'[\text{Cr}^{\text{IV}}][\text{PMe}_3]^2$, which agrees with the observed rate law with $k_3 = Kk'$. Despite many

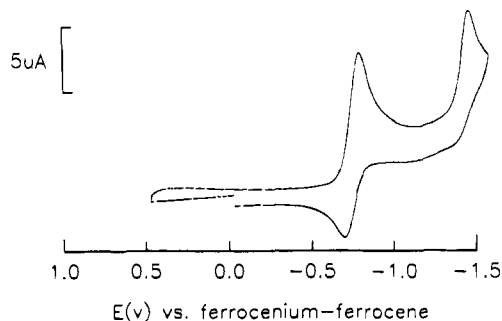


Figure 3. Cyclic voltammogram of **1** at a glassy carbon electrode with 0.1 M $[\text{n-Bu}_4\text{N}]\text{BF}_4$ in CH_3CN as supporting electrolyte, scan rate = 100 mV s^{-1} .

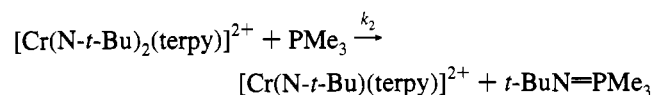
Table 4. Pseudo-First-Order Rate Constants for the Reaction of **1** with PMe_3 at 25.0 °C in Acetonitrile

$10^2[\text{PMe}_3], \text{ M}$	$10^3 k_{\text{obs1}}, \text{ s}^{-1}$	$10^4 k_{\text{obs2}}, \text{ s}^{-1}$
4.50	1.6	1.0
5.50	4.0	2.9
16.5	6.6	5.4
27.5	9.2	9.9
33.5	19	18

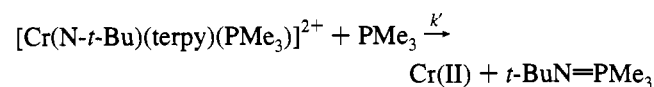
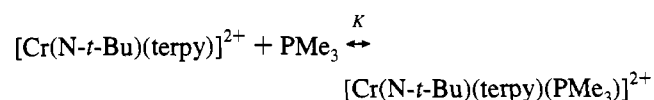
attempts we have been unable to isolate the imido-Cr(IV) intermediate.

Scheme 1

step 1



step 2



Electrochemistry. To understand the thermodynamic driving force governing the imido transfer reaction of **1**, we determined the formal potentials of diimido-Cr(VI) complexes by cyclic voltammetry. The cyclic voltammogram of **1** shown in Figure 3 consists of a reversible couple at -0.74 V and an irreversible wave at -1.45 V vs ferrocenium-ferrocene. The former couple is attributed to the metal-centered Cr(VI)/Cr(V) reduction, which as far as we are aware, is the first observation of reversible Cr(VI)/Cr(V) couple. The Cr(VI)/Cr(V) formal potential for cationic **1** is considerably less anodic than those for the neutral diimido-Cr(VI) analogs. For example, $[\text{Cr}(\text{N}-t\text{-Bu})_2\text{Cl}_2]$ is reduced irreversibly at ca. -2.0 V . The irreversibility of reduction of $[\text{Cr}(\text{N}-t\text{-Bu})_2\text{Cl}_2]$ is possibly due to instability of the reduced bis(imido)-Cr(V) species. To this end, we also prepared the sterically encumbered diaryloxo complex $[\text{Cr}(\text{N}-t\text{-Bu})_2(\text{OAr})_2]$ (**3**, Ar = 2,6-diisopropylphenyl) that should be kinetically stable with respect to reduction. **3** is an air-stable compound and shows no reactions with phosphines. The cyclic voltammogram of **3** in acetonitrile displays a reversible couple at -1.2 V , ascribable to the metal-centered Cr(VI)/Cr(V) reduction. It seems that the less negative formal potential for cationic **1** provides the driving force for its high reactivity in imido transfer with phosphines.

In summary, we have successfully isolated a dicationic bis-(imido)–Cr(VI) complex that is more reactive toward imido transfer than its neutral analogs. By judicious choice of coligands and fine-tuning the formal potential, we believe that highly oxidizing imido complexes should undergo nitrogen atom transfer reactions toward other kinds of organic substrates such as hydrocarbons.

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Supporting Information Available: Listings of crystal data (Table S1), complete bond distances (Table S2) and angles (Table S3), atomic coordinates with isotropic thermal parameters (Table S4), and anisotropic displacement parameters (Table S5) (8 pages). Ordering information is given on any current masthead page.

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