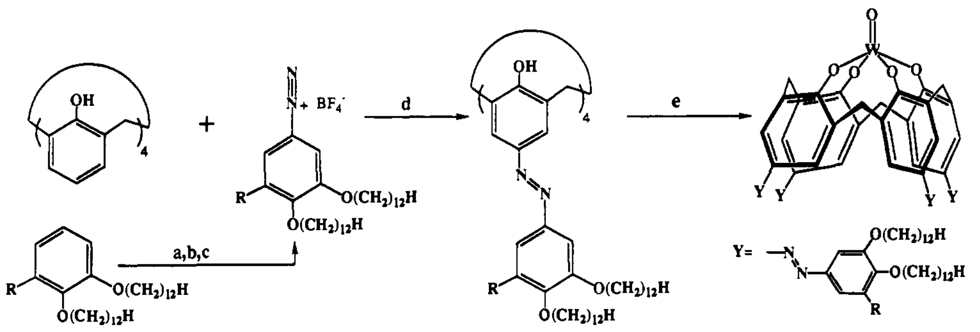
Scheme I



(a) SiO_2:HNO_3, (b) N_2H_4, graphite, (c) $NaNO_2$, HCl, $NaBF_4$ (d) Pyridine, (e) $WOCl_4$, Toluene

2a R=H
2b R=$O(CH_2)_{12}H$

1a R=H
1b R=$O(CH_2)_{12}H$

Scheme II

$$\mathbf{1a}\qquad K \xleftrightarrow[19.8\ \text{kcal/mol}]{136\,^{\circ}\text{C}} M \xleftrightarrow[1.5\ \text{kcal/mol}]{320\,^{\circ}\text{C}} I_d$$

$$\mathbf{1b}\qquad K \xleftrightarrow[7.9\ \text{kcal/mol}]{54\,^{\circ}\text{C}} M_1 \xleftrightarrow[0.6\ \text{kcal/mol}]{77\,^{\circ}\text{C}} M_2 \xleftrightarrow[5.1\ \text{kcal/mol}]{267\,^{\circ}\text{C}} I$$

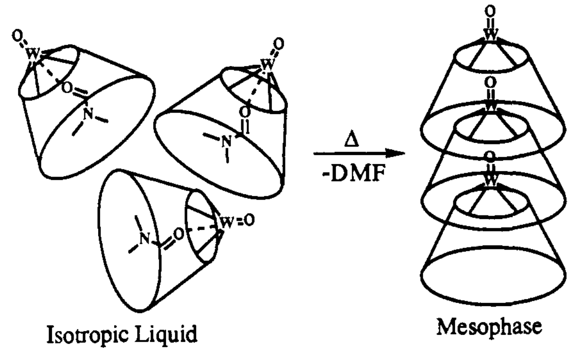
of the host–guest complex in CH_2Cl_2 solution occurs at 1645 cm^{-1}, which is 25 cm^{-1} lower in energy than uncomplexed DMF in CH_2Cl_2. The host–guest complexes exhibit more complicated ^1H NMR spectra with signals assigned to DMF at δ = 7.81, 1.55, 1.36, and 0.58 ($CDCl_3$) and at δ = 8.05, 0.41, 0.29, and 0.28 (C_6D_6).[11] The relative intensities of these signals were variable, depending on sample history; however, treatment of host–guest complexes with pyridine or diisopropylamine results in quantitative liberation of 1 equiv of DMF with signals at δ = 7.99, 2.93, and 2.86 ($CDCl_3$). In addition, we found one of the calix[4]arene methylene resonances to be sensitive to the guests and shift from δ = 4.80 to 4.85 ($CDCl_3$) with a DMF guest. The large upfield shift of the DMF protons is consistent with the shielding environment provided by the cavity.

The most dramatic consequence of host–guest complexation is its effect upon the mesomorphic behavior of the complexes. While **1a** exhibits a discotic mesophase from 135 to 330 °C, the DMF-**1a** host–guest complex melts directly to an isotropic phase at 115 °C (ΔH = 12.3 kcal/mol). Likewise, the **1b**–DMF host–guest complex melts to form an isotropic phase at 84 °C. Complexes with pyridine guests were found to exhibit isotropic transition at the same temperatures, indicating that a filled cavity is more important than the nature of the guest. With further heating (200–250 °C), the DMF slowly dissociates to form the discotic phase.

The depression of the isotropic phase through host–guest complexation allows the formation of a texture for **1a** consisting of well-defined cyclides with a tangential orientation of the optic axis characteristic of columnar phases.[8] Observation of these textures with a λ plate shows **1a** to exhibit unusual temperature dependent changes in the optic sign. These results are similar to those observed by others for bowlic liquid crystals.[3f] The K phase is optically positive; however, in heating cycles at temperatures just above K–M transition (<20 °C), the optic sign becomes negative. At higher temperatures the M phase becomes optically positive again. This transition to an optically negative material is not observed on cooling and is most likely due to conformational preferences of the azophenyl groups.

The deleterious effect of a DMF or pyridine guest on the mesophase stability indicates that occupation of the cavity is critical. This fact combined with the columnar structure of the mesophases suggests that the bowlic cores exhibit a head-to-tail arrangement whereby tungsten–oxo groups protrude into the cavity of the neighboring mesogen (Scheme III). The bowlic D_{ho} phase of **1b** is important since triangular symmetry dictates that the polar head-to-tail organization in the column must result in ferroelectric intercolumnar order or in a frustrated phase with random polarization. Additionally, the depression of the isotropic point through host–guest complexation provides a route to a low-viscosity state at reduced temperatures. This feature will allow for poling of very high melting bowlic complexes to produce noncentrosymmetric materials with high temporal stability.

Scheme III



Acknowledgment. We are grateful for financial support from the Office of Naval Research and to Dr. A. P. Sattelberger for a gift of $^{18}OH_2$.

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[11] Further investigations of host–guest complexation in these complexes is ongoing in our laboratory.

Synthesis, Crystal Structure, and Olefin Polymerization Activity of a Zwitterionic η^6-Arene Zirconium Tris(hydrocarbyl)

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The recent development of the chemistry of cationic dicyclopentadienyl group 4 metal–alkyl complexes,[1] $[Cp_2MR]^+$, functioning as single-component olefin polymerization catalysts has provided a key contribution to the understanding of the true active species involved in the metallocene-based Ziegler–Natta catalysts.[2]

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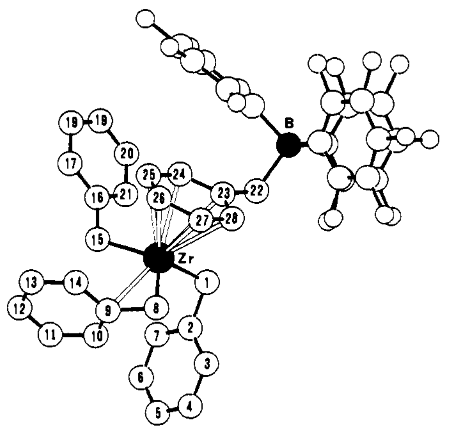

Figure 1. Structure of $[Zr(CH_2Ph)_3]^+[B(CH_2Ph)(C_6F_5)_3]^-$ (**1**). Important bond distances (Å, average standard error 0.02 Å) and bond angles (deg, average standard error 0.8°) are as follows: Zr–C(1), 2.23; Zr–C(8), 2.28; Zr–C(9), 2.57; Zr–C(15), 2.27; Zr–C(23), 2.76; Zr–C(24), 2.67; Zr–C(25), 2.65; Zr–C(26), 2.65; Zr–C(27), 2.65; Zr–C(28), 2.69; C(1)–Zr–C(15), 103.5; C(1)–Zr–C(8), 108.1; C(1)–Zr–CNT, 105.1; C(8)–Zr–C(15), 123.0; C(8)–Zr–CNT, 104.4; C(15)–Zr–CNT, 109.6; Zr–C(1)–C(2), 103.9; Zr–C(8)–C(9), 82.7; Zr–C(15)–C(16), 121.1 (CNT = center of gravity of the C(23)–C(28) ring).

Although less investigated, non-metallocene organometallic compounds, such as monocyclopentadienyl (Cp) derivatives or Cp-free hydrocarbyls, also play a role in some homogeneous Ziegler–Natta catalysts promoting polymerization of α-olefins,[3] styrene,[4] and conjugated diolefins.[5] We have recently described[6] olefin polymerization catalysts based on group 4 homoleptic hydrocarbyls or monocyclopentadienyl derivatives activated by in situ reaction with either $[C_6H_5(CH_3)_2NH]^+[B(C_6F_5)_4]^-$ or $B(C_6F_5)_3$, and we suggested that the resulting active species are cationic complexes analogous to those involved in metallocene-based catalytic systems. In our continuing efforts to isolate and characterize such highly unsaturated species, we report here the synthesis, the crystal, and the solution structure of $[Zr(CH_2Ph)_3]^+[B(CH_2Ph)(C_6F_5)_3]^-$, the first non-metallocene ionic complex functioning as single-component olefin polymerization catalyst.[7]

Reaction of $Zr(CH_2Ph)_4$ with 1 equiv of $B(C_6F_5)_3$ in toluene at room temperature yields, after addition of heptane, a red-orange crystalline solid, identified by ^1H and ^{13}C NMR analysis[8] as the ionic complex $[Zr(CH_2Ph)_3]^+[B(CH_2Ph)(C_6F_5)_3]^-$ (**1**). Noteworthy in the ^1H NMR spectrum (250 MHz, toluene-d_8, 25 °C) are the high-field resonance for the ortho hydrogens of the Zr-bound benzyl ligands (δ 6.03), indicating some interaction of the phenyl π-system with the metal,[9] and a doublet–triplet–triplet pattern of resonances at δ 6.39, 5.66, and 5.40 for the ortho, meta, and para hydrogens, respectively, of the B-bound benzyl group, suggesting the coordination of the phenyl ring to Zr.[10] Similar NMR patterns were found at low temperature for $[Zr(CH_2Ph)_3][BPh_4]$ (an η^6-coordination of a B-bound phenyl group was suggested[10a]) and for $[Cp'_2Zr(CH_3)]^+[BPh_4]^-$ (Cp′ = bridged η^5-cyclopentadienyl or indenyl) (an η^3(*m,p*)- or a fluxional η^2-(*m,p*)-coordination was proposed[10b]). Variable-temperature ^1H NMR experiments performed either in toluene-d_8 or in dichloromethane-d_2 show that the structure of **1** in aromatic solvent remains substantially unchanged between −60 and 100 °C, while some temperature-dependent fluxionality of the coordinated aromatic ring is observed in dichloromethane-d_2,[8,10] indicating a weaker metal–arene bonding, probably due to lower cation–anion electrostatic interactions in polar solvents with respect to toluene.

The crystal structure of **1**[11] (Figure 1) consists of $[Zr(CH_2Ph)_3]^+$ cations coordinated to $[B(CH_2Ph)(C_6F_5)_3]^-$ anions through metal–arene η^6 bonding of the B-benzyl. Solvated toluene molecules (1 per Zr atom) are also present. Four ligands are tetrahedrally coordinated to Zr: two benzyl groups behave as η^1 ligands, while the third one is bound in an η^2 fashion with both the methylene and the ipso phenyl carbons;[9,12] the unusual η^6-phenyl coordination to Zr is unprecedented for structurally characterized Zr(IV) derivatives.[13] The six Zr–C metal–arene distances are slightly different, with the *m,p*-carbons being found closest to Zr.

Addition of THF to a toluene solution of **1** results in the immediate precipitation of a yellow oil, identified by NMR as the 6-coordinate adduct $[Zr(CH_2Ph)_3(THF)_3]^+[B(CH_2Ph)(C_6F_5)_3]^-$ (**2**).[14]

1 promotes polymerization of ethylene and propene, performing similarly to the corresponding catalytic system prepared in situ.[6b] Typical activities are, respectively, $\sim 2.5 \times 10^4$ g of polyethylene (mol of Zr)$^{-1}$ h^{-1} atm^{-1} and $\sim 1.5 \times 10^3$ g of polypropylene (mol of Zr)$^{-1}$ h^{-1} atm^{-1}, in toluene solution at 50 °C and 5 atm of monomer pressure with catalyst concentrations of 0.5–1.0 mM and polymerization times of 0.25–1.0 h. Monitoring the polymerization of ethylene by ^1H NMR either in toluene-d_8 or in tetrachloroethane-d_2 at 25 °C shows a faster monomer consumption in the latter solvent, in which most of **1** appears to be catalytically active, consistent with the observed weaker coordination of the counterion to Zr in polar solvents with respect to toluene.[15]

These results indicate that the role of cationic complexes in the catalytic polymerization of olefins is not limited to metallocene systems. We are currently studying the chemistry of **1** and of

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(5) (a) Zambelli, A.; Ammendola, P.; Proto, A. *Macromolecules* **1989**, *22*, 2126–2128. (b) Oliva, L.; Longo, P.; Grassi, A.; Ammendola, P.; Pellecchia, C. *Makromol. Chem., Rapid Commun.* **1990**, *11*, 519–524. It is worth noting that metallocene-based catalysts are inactive in the polymerization of styrene and conjugated diolefins.[2b]

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(7) Some preliminary results have been presented at the A.O.D.C.P. International Symposium, Lyon, France, April 1992 (PII-10).

(8) See the supplementary material for detailed NMR data.

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(11) The present structure analysis is based on a limited number of measurements (reflections having $d \geq 1.3$ Å) taken from a rather large crystal (0.6 × 0.5 × 0.4 mm), since **1** is unstable under X-ray radiation. Crystallographic data for **1**·$PhCH_3$ are as follows: $a = 10.78$ (1) Å, $b = 13.41$ (1) Å, $c = 16.84$ (1) Å, $\alpha = 75.46$ (3)°, $\beta = 89.79$ (5)°, $\gamma = 77.22$ (6)°, $V = 2293$ Å^3, $Z = 2$ for space group $P\bar{1}$. $R = 0.11$ for 3357 reflections collected (Mo Kα radiation) with $F \geq 3\sigma(F)$. The structure was refined using isotropic thermal parameters for all atoms excepting Zr. Strong indications of anisotropy are evident (especially for $B(C_6F_5)_3$), but trials using anisotropic parameters for light atoms failed. A new, more complete low-temperature X-ray analysis will be performed.

(12) In spite of the limited accuracy of the present analysis, a significant lack of planarity for the η^2-bound benzyl group is observed: the root mean square deviation from the average plane for the C(9)–C(14) ring is 0.013 Å, while C(8) is displaced 0.132 Å away.

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(14) ^1H and ^{13}C NMR resonances for coordinated THF and for the "free" anion are detected: see the supplementary material.

(15) Temperature- and solvent-dependent cation–anion interactions have been suggested to play a role in both the activity and the stereospecificity of metallocene-based catalysts: (a) Reference 1g. (b) Eisch, J. J.; Caldwell, K. R.; Werner, S.; Kruger, C. *Organometallics* **1991**, *10*, 3417–3419. (c) Bochmann, M.; Jaggar, A. J. *J. Organomet. Chem.* **1992**, *424*, C5–C7. (d) Herfert, N.; Fink, G. *Makromol. Chem.* **1992**, *193*, 773–778.

related complexes, which could reveal novel, interesting features due to their increased electrophilicity and coordinative unsaturation with respect to the cationic metallocenes.

Acknowledgment. This work has been supported by MURST and by CNR (Italy).

Supplementary Material Available: Listings of ^{1}H and ^{13}C NMR data for **1** and **2**, a summary of X-ray data, tables of positional and thermal parameters and bond distances and angles, and a crystal packing diagram for **1**·PhCH$_3$ (7 pages). Ordering information is given on any current masthead page.

Ellagitannin Chemistry. The First Example of Biomimetic Diastereoselective Oxidative Coupling of a Glucose-Derived Digalloyl Substrate

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Ellagitannin natural products comprise an extensive group of structurally characterized secondary plant metabolites whose myriad subclasses result from expression of almost all the possible modes of oxidative coupling of appropriately juxtaposed galloyl groups attached to a glucose core. Thus, the monomeric ellagitannin tellimagrandin II (**1**)[2] contains a 4,6 coupled (*S*)-hexahydroxydiphenyl (HHPD) unit, while the dimeric congener oenothein B (**2**)[3] not only has this same HHDP moiety but also displays a second C–O coupling within the valoneoyl ester linking bridge. In addition, other members of this class of natural products feature C–C coupling between galloyl groups at the 2,4,[4] 3,6,[4a–c,5] and 1,6[4c] positions of the glucose ring, as well as C–O coupling (in dimeric ellagitannins) between 1,1′, 1,2′, 1,6′, 2,4′, 2,6′, and 3,4′ galloyl residues.[1c] Schmidt[1a] and later Haslam[1b,d] have postulated that the stereochemical and regiochemical outcome of the oxidative coupling processes of glucose-bound galloyl units is a direct and predictable consequence of the conformational preferences of the precursor molecules. In the results described below, we report the *first examples of high-yielding, completely diastereoselective* (*atrop-selective*) *coupling of galloyl species at the 4 and 6 positions of a glucose derivative using Pb(OAc)$_4$ as the oxidant.*[6] These experiments provide the *first support for the Haslam/Schmidt hypothesis*, as the conformational preferences of the digallate substrate (vide infra) are translated faithfully to the naturally occurring ellagitannin stereochemistry upon oxidation.

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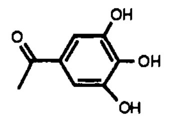

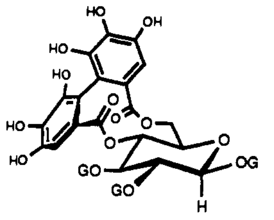

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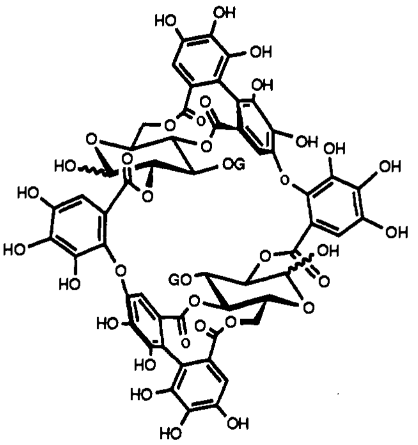

2

Historically, the defining attribute of gallotannins that has prompted much experimental inquiry is their ability to recognize and bind to almost all proteins examined.[1b] Manifestations of this phenomenon can be found, inter alia, in leather tanning, wine aging, and the salubrious properties of herbal teas. In addition to the generalized surface interactions between proteins and gallotannins which presumably underlie these applications,[1b] recent studies have documented that several of the more structurally complex ellagitannins engage in what is likely a very specific interaction with various disease-associated target enzymes, thus highlighting their therapeutic potential.[7] Despite their appealing structural complexity and potential as tools to study protein/ligand interactions, no investigations into the synthesis of the ellagitannins have been reported. Furthermore, even simple galloyl esters (and gallic acid) have continually defied attempts at oxidative phenolic coupling, instead providing modest yields of ellagic acid in most cases.[8]

We have initiated a program of synthesis whose goals include the development of diastereoselective oxidative phenolic coupling protocols to provide the various (*S*)- or (*R*)-HHDP units mentioned above. Toward this end, we have screened several di-, tetra-, and hexamethylated analogs of the digalloyl glucose-derived substrate **3** with a range of reagents reputed to be useful for oxidative phenolic coupling. One successful combination featured

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