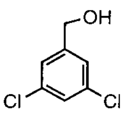

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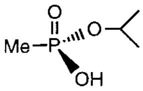

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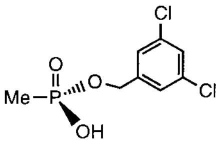

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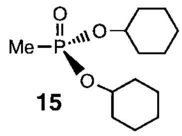

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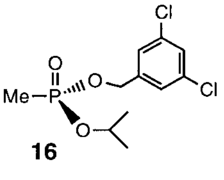

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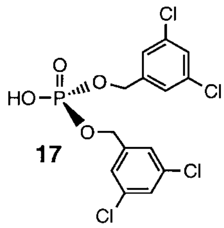

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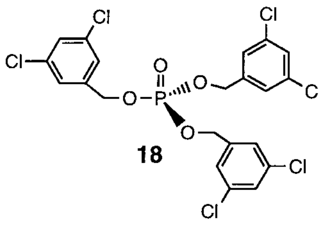

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Based on principles of antibody fine specificity and that the ligands tested were structurally related to **6**, there should be no interference from other compounds present at test sites. It was estimated that 170 ± 10 ppb of MPA analyte could be assayed as derivative **6**. A sensitivity comparable to many reported instrumental technologies that are expensive and usually necessitate specialized training. The sensitivity was also similar to other immunoassays developed for chemical warfare agents themselves.[6] Depending on the confidence limit chosen for detection, we anticipate the derivatization–ELISA procedure will accurately reflect the presence of MPA in an appropriate field sample.

Other methods routinely used for MPA analysis invoked chromatographic and/or spectrometric techniques.[7] Most of these approaches also required a prior derivatization step and furthermore were generally not amenable for the testing of crude samples. Our method is inexpensive, sensitive, convenient, robust, and would require only a minimum of sample preparation. For instance, a field sample from a suspected chemical warfare agent manufacturing, storage, or deployment site could be obtained by swipe and extracted with dioxane/water or a collected wet sample evaporated and redissolved. A simple filtration step might then be employed followed by addition of the reagent **5** and then assay. Notably, the presence of **5** in a 100-fold molar excess relative to antibody had no detrimental effects on binding. Hence, no other handling or purification steps should be necessary. While refinements are necessary to establish a field kit, we believe derivatization–ELISA can complement other MPA detection methods. These efforts and adaptation to other compounds related to chemical warfare agents are in progress.

Experimental Section

6: Reagent **5** (187 mg, 1 mmol) was added slowly to a solution of MPA (48 mg, 0.50 mmol) in 1,4-dioxane/0.5% water (5 mL). Bubbling was immediately observed upon addition. The resulting solution was stirred at room temperature for 1 h and then diluted with ethyl acetate (10 mL). After washing with brine (5 mL), the organic layer was dried with $MgSO_4$ and concentrated to give a yellow solid. The solid was purified using flash chromatography (95/5 CH_2Cl_2/EtOAc) affording a white solid (138 mg, 67%).

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Keywords: analytical methods · chemical warfare agents · immunoassays · methyl phosphonic acid · phosphorus

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$[Mn(en)]_3[Cr(CN)_6]_2 \cdot 4H_2O$: A Three-Dimensional Dimetallic Ferrimagnet ($T_c = 69$ K) with a Defective Cubane Unit[**]

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Recently, there has been increasing interest in metal assemblies of ordered networks.[1–11] One fascinating target in this research area are molecular-based magnets that exhibit

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spontaneous magnetization. Hexacyanometalate ions $[M(CN)_6]^{n-}$ are often used as building blocks for dimetallic magnetic materials. Two types of dimetallic compounds have been derived from $[M(CN)_6]^{n-}$: 1) The Prussian-Blue family constructed with a simple metal ion[6] and 2) complex-based dimetallic assemblies constructed with a complex cation such as $[Ni(L)_2]^{2+}$ (L = ethylenediamine derivative) and $[M^{III}(salen)]^+$ (M = Fe or Mn; salen = *N,N'*-ethylenebis(salicylideneaminato) dianion).[8–10] Almost all members of the Prussian-Blue family show spontaneous magnetization, and high magnetic ordering temperatures T_c are reported for some of them. However, the magnetostructural correlation for Prussian-Blue compounds is still unclear due to the lack of structural information. Complex-based dimetallic assemblies with one-dimensional (1D) chain, 1D rope ladder, two-dimensional (2D) honeycomb, and 2D square structures have been obtained, and magnetic ordering is recognized only for the 2D network dimetallic assemblies.[8] The first three-dimensional (3D) dimetallic assemblies with a cubane network, $[Ni(L)_2]_3[Fe(CN)_6]X_2$ (L = ethylenediamine (en) or trimethylenediamine, X = ClO_4^- or PF_6^-)[9] were recently reported but they showed no magnetic ordering due to the diamagnetic nature of Fe^{II}. Here we report on a dimetallic compound $[Mn(en)]_3[Cr(CN)_6]_2 \cdot 4H_2O$ that has a 3D network structure extended by the Cr^{III}-CN-Mn^{II} linkage and that shows magnetic ordering at $T_c = 69$ K.

The dimetallic assembly $[Mn(en)]_3[Cr(CN)_6]_2 \cdot 4H_2O$ was obtained as light green crystals by the reaction of $K_3[Cr(CN)_6]$, $MnCl_2 \cdot 4H_2O$, and ethylenediamine in 2:3:3 molar ratio in an aqueous solution under argon. The X-ray crystal structure analysis[12] reveals that all the cyanide groups of $[Cr(CN)_6]^{3-}$ are involved in the coordination to adjacent Mn^{2+} ions. The asymmetric unit consists of a $[Cr(CN)_6]^{3-}$ ion, three halves of $[Mn(en)]^{2+}$ ions, and two water molecules. The geometry about Mn1 and Mn2 is pseudo-octahedral with a chelating en ligand and four cyanide nitrogen atoms from adjacent $[Cr(CN)_6]^{3-}$ units (Figure 1). The Cr–C and Mn–N bond lengths range between 2.065(3) and 2.080(3) Å and between 2.208(3) and 2.287(3) Å, respectively. The Mn-N-C bond angle ranges between 145.7(3)° and 167.4(3)°. In the lattice, a 3D network structure is formed by the alternate array of $[Cr(CN)_6]^{3-}$ and $[Mn(en)]^{2+}$ ions (Figure 2). The network comprises defective cubes with three Cr atoms, three Mn1 atoms, and one Mn2 atom at the seven corners and eight Cr-CN-Mn edges. The shortest Cr⋯Mn(1), Cr⋯Mn(2), and Mn(1)⋯Mn(2) distances are 5.1553(8), 5.2842(8), and 6.659(1) Å, respectively. The lattice water molecules reside in cavities of the network and form hydrogen bonds with the cyano nitrogen atoms (O(1)–H(13)⋯N(5) and O(2)–H(15)⋯N(2)).

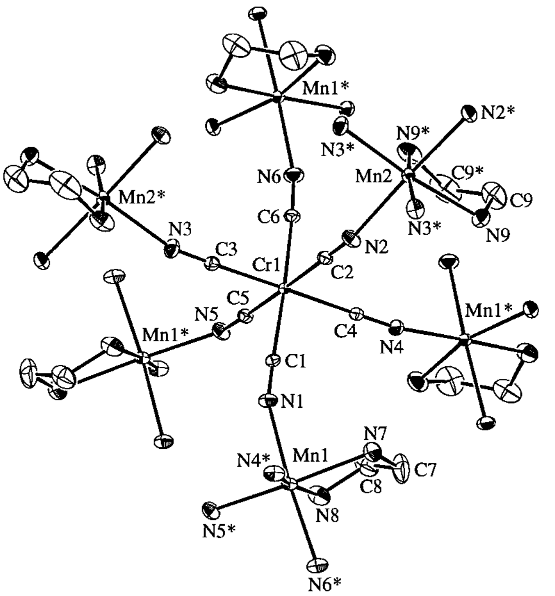

Figure 1. ORTEP drawing of the heptanuclear unit of $[Mn(en)]_3[Cr(CN)_6]_2 \cdot 4H_2O$ (H_2O are omitted).

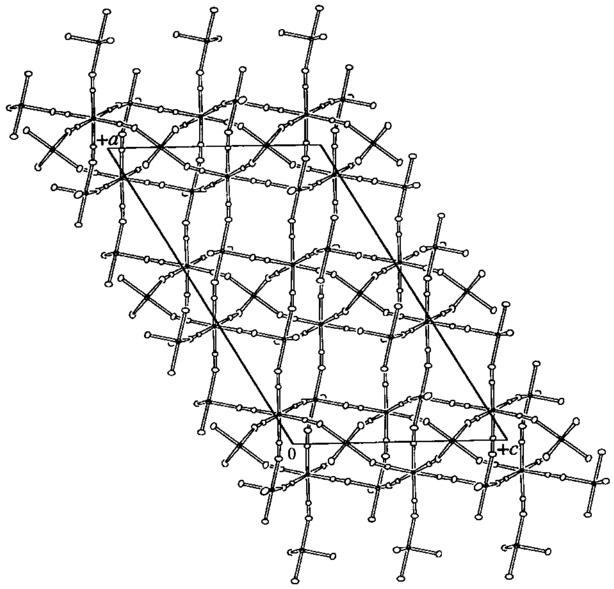

Figure 2. Projection of the 3D network structure onto the *ac* plane (en and H_2O are omitted).

The magnetic behavior[13] of the dimetallic compound is shown in Figure 3 in the form of $\chi_M T$ versus T and χ_M versus T plots. The $\chi_M T$ value is 13.08 cm³ K mol⁻¹ (10.23 μ_B per Mn_3Cr_2) at room temperature and decreases with decreasing temperature down to a minimum value of 12.04 cm³ K mol⁻¹ (9.82 μ_B) at 156 K. Upon further cooling, the $\chi_M T$ value increases up to a maximum value of 5671 cm³ K mol⁻¹ (213 μ_B)

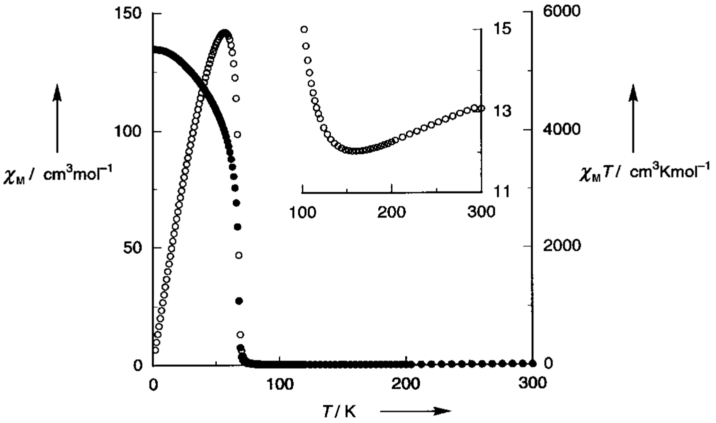

Figure 3. Plots of χ_M (●) and $\chi_M T$ (○) versus T for $[Mn(en)]_3[Cr(CN)_6]_2 \cdot 4H_2O$ per Mn_3Cr_2 unit. Insert: Expanded view of the minimum region of $\chi_M T$.

at 50 K and then decreases below this temperature. The $1/\chi_M$ versus T plot in the range from 300 to 156 K obeys the Curie–Weiss law with a Weiss constant of -38 K. The negative Weiss constant indicates an intramolecular antiferromagnetic interaction between the adjacent Cr^{III} and Mn^{II} ions through the cyano bridge. The minimum $\chi_M T$ value agrees with the spin-only value of 12.38 $cm^3 K mol^{-1}$ (9.95 μ_B) for antiferromagnetically coupled Mn_3Cr_2 ($S_T = 9/2$). The abrupt increase in $\chi_M T$ around 70 K suggests the onset of three-dimensional magnetic ordering. The decrease of $\chi_M T$ below 50 K results from a saturation of the χ_M value.

Low-field magnetization measurements for the dimetallic compound confirm the long-range magnetic ordering below 69 K to produce a ferrimagnet (Figure 4). The field dependence of magnetization M shows a sharp increase with applied field and rapid saturation. The saturation magnetization $M_S = 8.7\ \mu_B$ is close to the expected value of 9 μ_B, adding support to the assumption that the adjacent Mn^{II} and Cr^{III} centers are antiferromagnetically coupled (Figure 5). The magnetic hysteresis loop at 2 K (Figure 5, insert) is typical of soft magnets showing a remnant magnetization of $1.01 \times 10^4\ cm^3 G mol^{-1}$ and a weak coercive field $H_c = 28$ G.

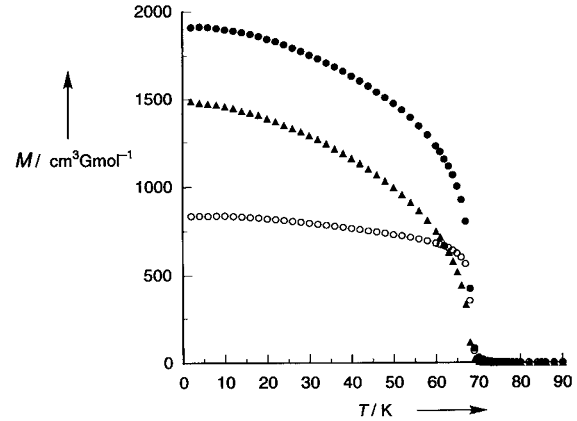

Figure 4. Plots of magnetization M versus T at 5 G: field-cooled magnetization (FCM; ●), remnant magnetization (RM; ▲), zero-field magnetization (ZFCM; ○).

It should be noted that $[Mn(en)]_3[Cr(CN)_6]_2 \cdot 4H_2O$ is similar to one type of Prussian-Blue family, $A^{II}_3[B^{III}(CN)_6]_2 \cdot xH_2O$.[6, 14] We presume that this type of Prussian-Blue analogue has a similar 3D network structure with an $A(H_2O)_2^{2+}$ site in place of the $[Mn(en)]^{2+}$ site in the present compound.

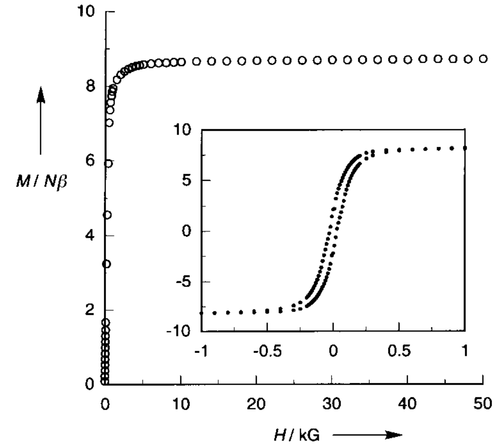

Figure 5. Field dependence of magnetization M ($N\beta$) at 2 K. Insert: hysteresis loop ($M/N\beta$ versus H) at 2 K.

In conclusion, $[Mn(en)]_3[Cr(CN)_6]_2 \cdot 4H_2O$ is a 3D ferrimagnet showing magnetic ordering below 69 K. To the best of our knowledge, the magnetic phase-transition temperature is the highest among the structurally characterized molecular-based magnets.

Experimental Section

To a solution of $MnCl_2 \cdot 4H_2O$ (0.6 mmol) and en (0.6 mmol) in water (15 mL) was added an aqueous solution (10 mL) of $K_3[Cr(CN)_6]$ (0.4 mmol) under argon at room temperature, and the resulting yellow mixture was allowed to stand to give light green prismatic crystals. They were collected by suction filtration and washed with water. All the operations for the synthesis were carried out in the dark to avoid the decomposition of $K_3[Cr(CN)_6]$. Yield 53 mg, 32 %. Elemental analysis (%): calcd for $C_{18}H_{32}N_{18}Cr_2Mn_3O_4$: C 25.94, H 3.87, N 30.25, Cr 12.48, Mn 19.78; found: C 25.92, H 3.74, N 30.20, Cr 12.19, Mn 19.49; IR: $\tilde{\nu} = 2152\ cm^{-1}$ (C≡N).

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Keywords: chromium · magnetic properties · manganese · network structures

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[12] Crystal data for $[Mn(en)]_3[Cr(CN)_6]_2 \cdot 4H_2O$ at 298 K: light green prisms, $C_{18}H_{32}N_{18}Cr_2Mn_3O_4$, $M_r = 833.37$, crystal size $0.2 \times 0.2 \times 0.2$ mm, monoclinic, space group $C2/c$ (No. 15), $Z = 4$, $a = 24.505(7)$, $b = 11.323(4)$, $c = 14.810(4)$ Å, $\beta = 120.69(2)^\circ$, $V = 3533(1)$ Å^3, $\rho_{calcd} = 1.566$ g cm^{-3}, $\mu(Mo_{K\alpha}) = 16.90$ cm^{-1}. Data collection: Rigaku AFC-5S diffractometer with graphite-monochromated $Mo_{K\alpha}$ radiation ($\lambda = 0.71069$ Å). The data were collected using the ω-2θ scan technique to a maximum 2θ value of 55.0° at a scan speed of 8.0° min^{-1} (in omega). The cell parameters were determined by 25 reflections in the 2θ range of $29.07^\circ \leq 2\theta \leq 29.94^\circ$. Over the course of the data collection, the standard intensities decreased by 5.5%. Intensity data were corrected for Lorentz polarization, which effected the Ψ scan absorption. The structure was solved by direct methods and expanded using Fourier techniques. The non-hydrogen atoms were refined anisotropically. Hydrogen atoms were included but not refined. The refinement converges with $R = \Sigma||F_o| - |F_c||/\Sigma|F_o| = 0.033$ and $R_w = \{\Sigma w(|F_o| - |F_c|)^2/\Sigma w|F_o|^2\}^{1/2} = 0.042$ ($w = 1/\sigma^2(F_o)$) for 3032 reflections ($I > 3.00\sigma(I)$). Further details on the crystal structure investigation may be obtained from the Fachinformationszentrum Karlsruhe, D-76344 Eggenstein-Leopoldshafen, Germany (fax: (+49) 7247-808-666; e-mail: crysdata@fiz-karlsruhe.de), on quoting the depository number CSD-410501.

[13] The magnetic measurements of the sample were carried out between 2 and 300 K using a Quantum Design MPMS-XL5R SQUID magnetometer. The data was corrected for the contribution of the sample holder and diamagnetism estimated from Pascal's constants.

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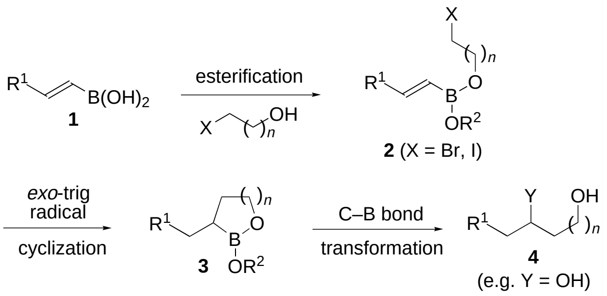
The First Boron-Tethered Radical Cyclizations and Intramolecular Homolytic Substitutions at Boron**

Robert A. Batey* and David V. Smil

Silicon-tethered radical cyclizations, first reported by Nishiyama et al.[1] and Stork et al.,[2] are a useful strategy for the construction of C–C bonds. Numerous syntheses have incorporated these processes, usually for hydroxymethylations of allylic alcohols,[1–3] and these reactions account for approximately half of the publications in the field of silicon-tethered chemistry. The success of temporary silicon connections in radical chemistry, together with our ongoing interest in developing new reactions of organoboron compounds,[4] led us to consider employing boron in an analogous tethering role. Narasaka et al. used phenylboronic acid to tether dienes and dienophiles together through O-B-O linkages in Diels–Alder reactions,[5] but there are no examples of this strategy in free-radical chemistry. Carboni et al. demonstrated both intermolecular free-radical additions to alkenylboranes and radical cyclizations with alkenylboranes.[6] We now report the first boron-tethered radical cyclizations for the synthesis of 1,3-, 1,4-, and 1,5-diols. The general strategy employs the covalent C-B-O linkage of boronic esters as a tether (Scheme 1).[7] We envisaged that the ease of synthesis of the precursor boronic acids **1** and esters **2** and the synthetic versatility of the C–B bond[8] in the cyclic products **3** would significantly expand the scope of tethered-radical processes.



Scheme 1. General strategy for boron-tethered radical cyclizations of alkenylboronic esters **2**.

The requisite (*E*)-alkenylboronic acids **1** were obtained in good yields by hydroboration of the corresponding alkynes with the dimethyl sulfide complex of dibromoborane[9] (R^1 = alkyl) or catecholborane[10] (R^1 = aryl). Treatment of **1** with 2-bromoethanol, 2-iodoethanol, or 3-bromopropanol in THF in the presence of 4 Å molecular sieves at room temperature over 24 h readily afforded the (*E*)-alkenylboronic esters **2**. The precursors **2** were then subjected to free-radical conditions by using Corey's catalytic tributylstannane method.[11] Heating **2a–f** (substrates with *n*-alkyl or aryl substituents at the β-alkenyl position) to 55 °C in THF for 48 h, in the presence of the radical initiator dimethyl-2,2′-azobisisobutyrate (DAB),[12] gave the boracycles **3**, which were not isolated, but immediately oxidized with trimethylamine *N*-oxide (TMANO)[13] to the 1,3- or 1,4-diols **4a–f** (Table 1), that is, the products of 5- or 6-*exo*-trig radical cyclization, respectively. No products resulting from 6-*endo*-trig or 7-*endo*-trig cyclization were isolated or observed in the crude reaction mixture, and this indicates an *exo*:*endo* selectivity of greater than 95:5.[14] Initial attempts to perform these cyclizations in refluxing THF gave lower yields (10–40% below those with optimized conditions) due to competing direct reduction of the C–Br bond of **2**. Ionic reduction of **2** by the sodium cyanoborohydride co-reductant in the absence of radical initiator was demonstrated to be increasingly competitive

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