



# Charge-transfer host system composed of 9,10-bis(3,5-dihydroxyphenyl)-anthracene and methylviologen

Yoshitane Imai<sup>a,\*</sup>, Takafumi Kinuta<sup>a</sup>, Kensaku Kamon<sup>a</sup>, Nobuo Tajima<sup>b</sup>, Tomohiro Sato<sup>c</sup>,  
Reiko Kuroda<sup>c,d</sup>, Yoshio Matsubara<sup>a,\*</sup>

<sup>a</sup> Department of Applied Chemistry, Faculty of Science and Engineering, Kinki University, 3-4-1 Kowakae, Higashi-Osaka, Osaka 577-8502, Japan

<sup>b</sup> First-Principles Simulation Group, Computational Materials Science Center, NIMS, Sengen, Tsukuba, Ibaraki 305-0047, Japan

<sup>c</sup> JST ERATO-SORST Kuroda Chiromorphology Team, 4-7-6 Komaba, Meguro-ku, Tokyo 153-0041, Japan

<sup>d</sup> Department of Life Sciences, Graduate School of Arts and Sciences, The University of Tokyo, 3-8-1 Komaba, Meguro-ku, Tokyo 153-8902, Japan

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## ABSTRACT

By using 9,10-bis(3,5-dihydroxyphenyl)anthracene as an electron donor and 1,1'-dimethyl-4,4'-bipyridinium dichloride as an electron acceptor, a spontaneously resolved charge-transfer (CT) complex is formed. This CT complex can include *n*-alkyl alcohol molecules as guests, and the DRS of this CT complex change with the type of component guest molecules.

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## 1. Introduction

The properties of organic compounds in the solid state are different from those in the solution state, because molecules are densely packed and are strongly influenced by the neighboring molecules in the solid state. Therefore, the use of many solid-state chiral supramolecular organic host compounds for chiral molecular recognition and enantioselective reactions has been reported.<sup>1</sup> There has been an increase in the demand for modulating the size and shape of the chiral cavity of such chiral host systems in order to incorporate various guest molecules. In order to meet these demands, supramolecular organic host systems composed of two or three types of organic molecules have been developed.<sup>2</sup> If a solid-state molecular recognition system is composed of two or more types of organic molecules, the property of molecular recognition may be easily controlled by changing the component molecules without additional synthesis steps. We have focused on employing donor–acceptor interactions as intermolecular forces for constructing and controlling the supramolecular host structure.<sup>3</sup>

Recently, we have developed strongly colored chiral charge-transfer (CT) host systems composed of chiral 1,1'-bi-2-naphthol as an electron donor and 1,1'-dibenzyl-4,4'-bipyridinium dichloride (BVCl<sub>2</sub>, benzylviologen) as an electron acceptor.<sup>4</sup> This CT complex can include alcohol molecules as guests. However, this chiral CT host system has been produced by using a chiral component molecule (chiral 1,1'-bi-2-naphthol). Chiral functional organic materials are typically prepared by using chiral molecules. However, in general, most chiral molecules are not easily available and are more expensive than achiral molecules. Therefore, chiral CT host systems prepared by a preferential crystallization method by using achiral component molecules can be used for the industrial production of chiral materials without using any chiral auxiliaries.

In this paper, we report the formation of a novel CT host system by using electron donor and acceptor molecules. As an electron donor, 9,10-bis(3,5-dihydroxyphenyl)anthracene (**1**), which is one of the most important component organic molecules used for preparing supramolecular complexes,<sup>5</sup> was used. As electron acceptors, three types of diquaternary salts of 4,4'-bipyridine (viologen), namely, 1,1'-diphenyl-4,4'-bipyridinium dichloride (PVCl<sub>2</sub>, phenylviologen), BVCl<sub>2</sub>, and 1,1'-dimethyl-4,4'-bipyridinium dichloride (MVCl<sub>2</sub>, methylviologen), were used. In general, it is not easy to predict the structure of supramolecular complexes prepared using new component molecules. Therefore, it is also confirmed

\* Corresponding authors. Tel.: +81 06 6730 5880x5241; fax: +81 06 6727 2024 (Y.I.).

E-mail addresses: [y-imai@apch.kindai.ac.jp](mailto:y-imai@apch.kindai.ac.jp) (Y. Imai), [y-matsu@apch.kindai.ac.jp](mailto:y-matsu@apch.kindai.ac.jp) (Y. Matsubara).

that this study is a significant advancement in the design of novel two-component supramolecular host systems (Chart 1).

## 2. Results and discussion

The inclusion of ethanol (EtOH), which is a simple *n*-alkyl alcohol, as guest molecules is first studied. The inclusion of EtOH is attempted by crystallization from EtOH solution (3–5 mL) containing **1** (10 mg, 0.025 mmol) and each of the three types of viologen molecules (0.025 mmol) separately. Each EtOH solution is left to stand at room temperature for 5–7 days. As a result, colored complexes—**I** (8 mg) from **1**/PVCl<sub>2</sub> system; **II** (11 mg), **1**/BVCl<sub>2</sub> system; and **III** (7 mg), **1**/MVCl<sub>2</sub> system—are obtained.<sup>6</sup> The colors of these CT complexes depend on the type of viologen molecule, that is, **I**, **II**, and **III** are brown, purple, and orange in color, respectively. The colors of these CT complexes are quite different from those of the component solids (**1**, BVCl<sub>2</sub>, and PVCl<sub>2</sub>: white solids; and MVCl<sub>2</sub>: light yellow solid).

In order to investigate the effect of inclusion of the guest molecules, <sup>1</sup>H NMR analysis is performed on the obtained crystals. It is found that **I** and **II** obtained from the **1**/PVCl<sub>2</sub> and **1**/BVCl<sub>2</sub> systems include no guest EtOH molecules. On the other hand, **III** obtained from the **1**/MVCl<sub>2</sub> system includes EtOH molecules as guests. Although good quality crystals of complexes **I** and **II** were not obtained, a crystal suitable for X-ray crystallographic analysis was obtained from **1**/MVCl<sub>2</sub> system. Then, to study the mechanism of the formation of this CT host system, the crystal structures of **III**, as shown in Figure 1, are investigated by X-ray crystallographic analysis.

Interestingly, although all the used component molecules are achiral, the space group of this crystal is *P*2<sub>1</sub>2<sub>1</sub>2. That is, this CT host system is a spontaneous resolution system. The stoichiometry of **III** is **1**/MVCl<sub>2</sub>/EtOH=1:1:1. Molecules of **1** (blue molecules in Fig. 1) are connected by chloride ions (green balls in Fig. 1) through hydrogen bonds, and two types of 1D-structure units are formed along the *c*-axis (Fig. 1a). One is composed of molecules of **1** wherein the torsion angles between benzene and anthracene rings are 87.9° and 107.5° (dotted red rectangle, Fig. 1b). The other is composed of molecules of **1** wherein the torsion angles between the benzene and anthracene rings are 92.6° and 106.4° (dotted blue rectangle, Fig. 1b). Moreover, a 2D-layered network structure along the *b*- and *c*-axes is formed by the self-assembly of these two types of 1D-structure units by anthracene–benzene edge-to-face interactions (red arrows in Fig. 1b, 2.95 Å).<sup>7</sup> Disordered guest EtOH molecules (red molecules in Fig. 1) are included in the cavity formed by the self-assembly of these 1D-structure units in the 2D-layered network structure. In this case, a hydrogen bond and CH–π interaction between EtOH and host component molecule are not observed.<sup>7</sup> Methylviologen ions (MV<sup>2+</sup>, green molecules in Fig. 1c and d) connect these 2D-layered network structures (dotted red rectangle, Fig. 1c and d) by CT interactions along the *a*-axis and form this crystal (Fig. 1c and d). The distances between the center of the six-membered ring of MV<sup>2+</sup> and the center of the nearest six-membered ring of **1** are 3.62 Å and 3.89 Å.

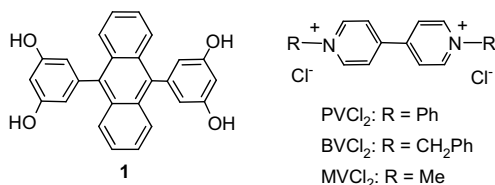
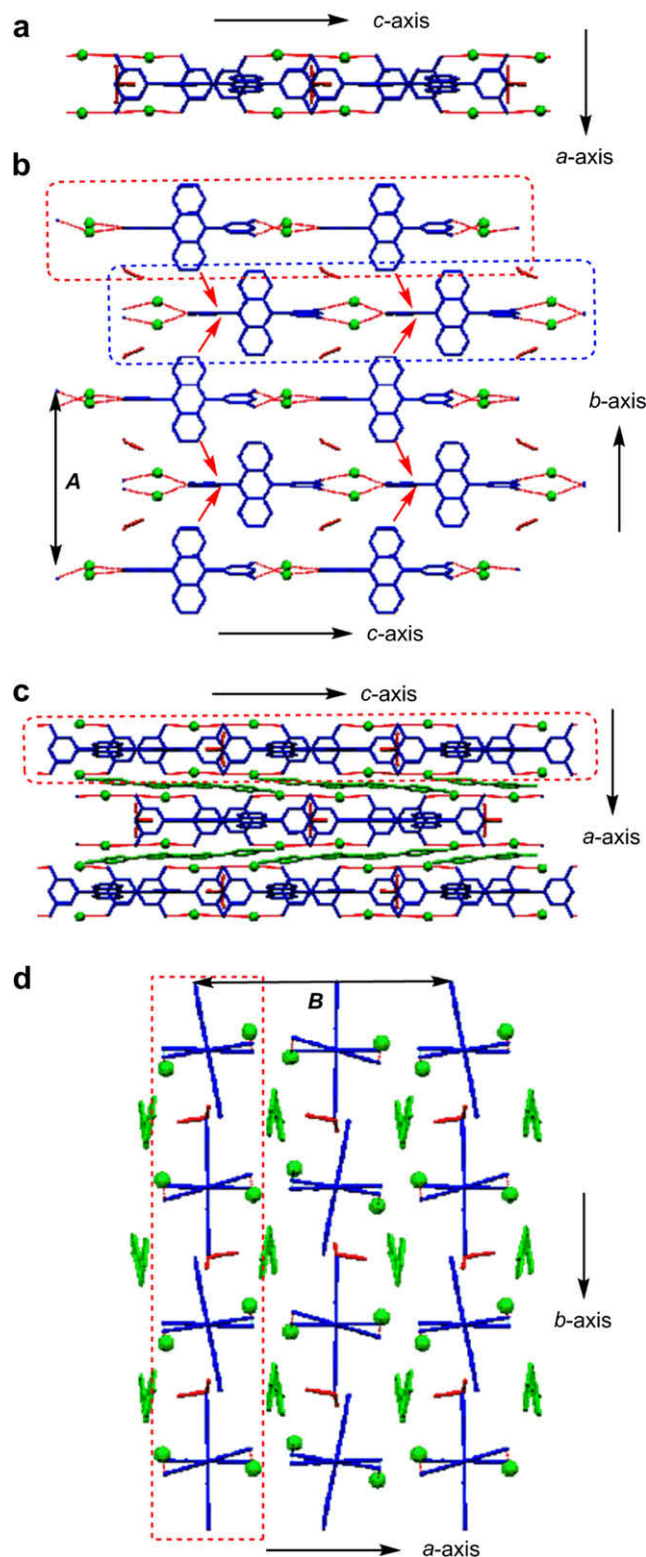
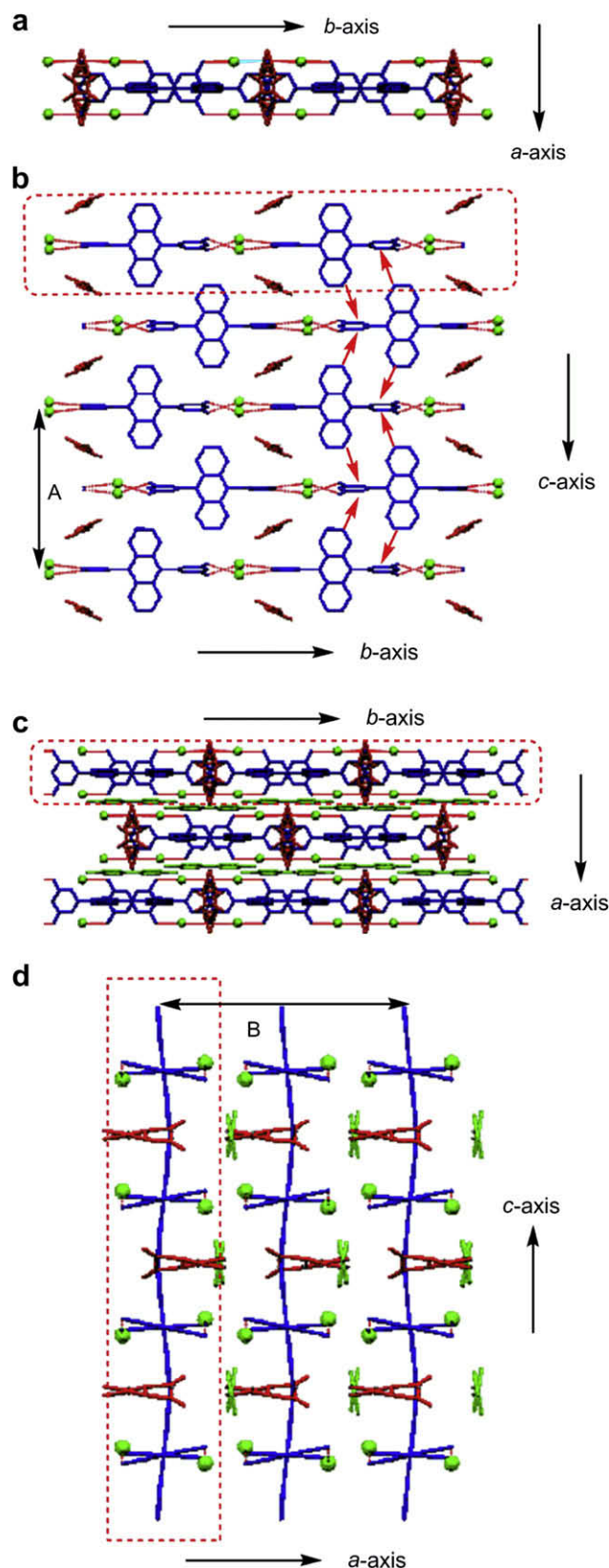


Chart 1.



**Figure 1.** Crystal structure of **III**. **1**, MV<sup>2+</sup>, and EtOH are represented as blue-, green-, and red-colored molecules, respectively. Chloride ions are represented as green balls. (a) Extracted 2D-layered structure unit observed along the *b*-axis. (b) Extracted 2D-layered structure unit observed along the *a*-axis. MV<sup>2+</sup> is squeezed. Dotted red and blue rectangles show a 1D-structure unit. Red arrows show anthracene–benzene edge-to-face interactions. (c) Packing diagram observed along the *b*-axis. Dotted red rectangle shows a 2D-layered structure unit. (d) Packing diagram observed along the *c*-axis. Dotted red rectangle shows a 2D-layered structure unit.



**Figure 2.** Crystal structure of **V**. **1**,  $MV^{2+}$ , and *n*-BuOH are represented as blue-, green-, and red-colored molecules, respectively. Chloride ions are represented as green balls. (a) Extracted 2D-layered structure unit observed along the *c*-axis.  $MV^{2+}$  is squeezed. Dotted red rectangle shows a 1D-structure unit. Red arrows show anthracene–benzene edge-to-face interactions. (c) Packing diagram observed along the *c*-axis. Dotted red rectangle shows a 2D-layered structure unit. (d) Packing diagram observed along the *b*-axis. Dotted red rectangle shows a 2D-layered structure unit.

Next, in order to study the guest inclusion ability of the **1**/ $MVCl_2$  system, two other *n*-alkyl alcohols—*n*-propanol (*n*-PrOH) and *n*-butanol (*n*-BuOH)—are used as guests. Using the same procedure, we attempt to include the guest molecules into the **1**/ $MVCl_2$ -CT host system by crystallization from the two guest solutions. A mixture of **1** and  $MVCl_2$  is dissolved in the two guest solutions and left to stand at room temperature. After 5–7 days, orange colored CT complexes **IV** obtained from the *n*-PrOH solution (5 mg) and **V** obtained from the *n*-BuOH solution (8 mg)<sup>6</sup> are obtained by crystallization from each guest solution. However, since **IV** obtained from the *n*-PrOH solution is not a good quality crystal, the X-ray analysis was unsatisfactory.

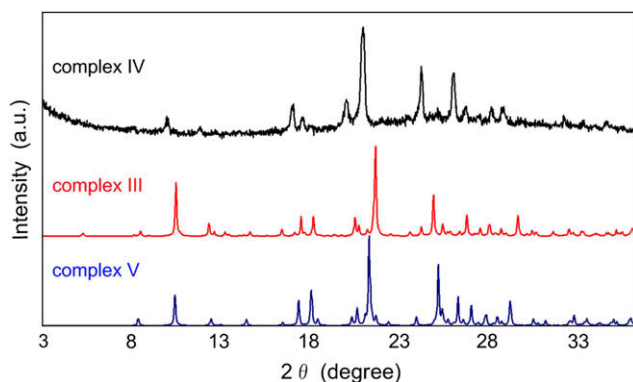
The crystal structures of **V** obtained from the *n*-BuOH solution are shown in Figure 2.

Interestingly, the space group of obtained crystal is  $C222_1$ . The stoichiometry of complex **V** is **1**/ $MVCl_2$ /*n*-BuOH=1:1:1. The crystal structure of **V** is similar to that of **IV**. In this complex too, a 1D-structure unit composed of molecules of **1** (blue molecules in Fig. 2) and chloride ions (green balls in Fig. 2) is formed along the *b*-axis (Fig. 2a). This 1D-structure unit is composed of one type of molecules of **1** wherein the torsion angles between the benzene and anthracene rings are  $93.1^\circ$  and  $109.3^\circ$ . The self-assembly of these 1D-structure units (dotted red rectangle, Fig. 2b) by anthracene–benzene edge-to-face interactions (red arrows in Fig. 2b, 2.98 Å) results in the formation of a 2D-layered network structure along the *b*- and *c*-axes (Fig. 2b). Although the guest *n*-BuOH molecules (red molecules, in Fig. 2) are disordered, they are included in the cavity formed by the self-assembly of these 1D-structure units in the 2D-layered network structure (dotted red rectangle, Fig. 2c and d) without a hydrogen bond and CH– $\pi$  interaction between *n*-BuOH and host component molecule.<sup>7</sup> Methylviologen ions ( $MV^{2+}$ , green molecules in Fig. 2c and d) connect these 2D-layered network structures by CT interactions along the *a*-axis and form this crystal (Fig. 2c and d). The distance between the center of the six-membered ring of  $MV^{2+}$  and the center of the nearest 6-membered ring of **1** is 3.72 Å.

Since the structural information of **IV** obtained from the *n*-PrOH solution could not be obtained, X-ray powder diffraction analysis of **IV** is carried out, and the results are compared with the X-ray powder diffraction analysis results of **III** and **V** (Fig. 3).

The X-ray powder diffraction pattern of **IV** is similar to that of **III** and **V**. Moreover,  $^1H$  NMR analysis reveals that the host–guest ratio of this complex is **1**/ $MVCl_2$ /*n*-PrOH=1:1:1. This stoichiometry is also the same as that of **III** and **V**. From these results, it is concluded that **IV** is structurally similar to **III** and **V**.

The sizes of the cavities of **III** and **V** are compared, and it is found that they depend on the type of included guest molecule; when the guest molecule is changed from EtOH to *n*-BuOH, the distance between the 1D-structure units in the 2D-layered network structure [A, Figs. 1b and 2b] reduces from 14.85 to 14.70 Å. On the other



**Figure 3.** X-ray powder diffraction patterns of **III**–**V**. Patterns of **III** and **V** are calculated using X-ray crystallographic analysis data.



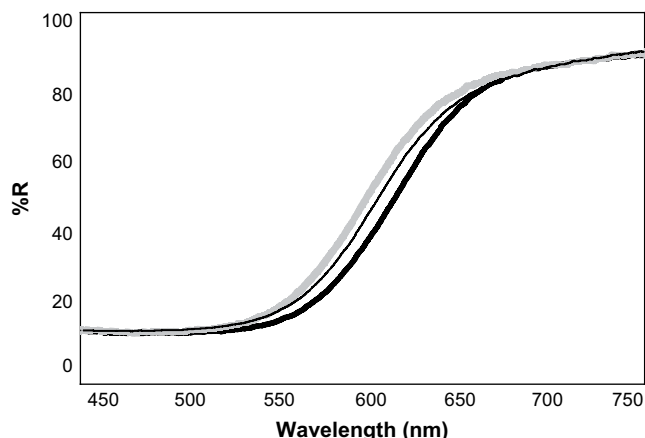


Figure 4. DRS of **III** (black line), **IV** (gray line), and **V** (narrow black line).

hand, the distance between the 2D-layered network structures [**B**, Figs. 1d and 2d] increases from 13.82 to 14.09 Å. This shows that this CT host system includes guest molecules by tuning the distance between the 1D-structure units and that between the 2D-layered network structures. Moreover, these results suggest that spontaneous resolution of **1**/MVCl<sub>2</sub>-CT host system is mainly caused not by intermolecular interactions between guest *n*-alkyl alcohol molecule and host component molecules but by steric and electric repulsions between **1** and MVCl<sub>2</sub>.

The diffuse reflectance spectra (DRS) of **III**–**V** are measured and shown in Figure 4.

As the colors of these crystals are similar, i.e., orange, as expected, the DRS of these CT complexes differ only slightly. Exactly stating, crystal **III** absorbs slightly longer wavelengths than **V**. It is well known that the color of a CT complex depends primarily on the intermolecular distance between the donor and acceptor molecules. The X-ray crystallographic analyses show that the distances between the donor and the acceptor molecules are not different markedly in the two crystals (3.63 Å in **III** and 3.71 Å in **V**), which is consistent with the similar colors of the two crystals.

### 3. Conclusion

Novel spontaneously resolved CT host system composed of **1** and MVCl<sub>2</sub> molecules is successfully created. This CT host crystal can include *n*-alkyl alcohols as guests by tuning the packing distance between the shared 1D-structure units and that between the 2D-layered network structures. Moreover, the DRS of this CT crystal change with the type of component guest molecules. This CT host system may be useful in designing novel host systems that can be used in asymmetric reactions and for molecular recognition.

## 4. Experimental

### 4.1. General methods

All reagents were used directly as obtained commercially. Component molecules **1** and viologen derivatives were purchased from Tokyo Kasei Kogyo Co. Guest solutions were purchased from Wako Pure Chemical Industry.

### 4.2. Formation of supramolecular CT host complex

Compound **1** (10 mg, 0.025 mmol) and each of the three types of viologen molecules (0.025 mmol) were dissolved in guest *n*-alkyl

alcohol (3–5 mL). After 5–7 days, colored complexes were deposited and collected. The total weight of all the crystals obtained in a batch is 7–11 mg.

### 4.3. X-ray crystallographic study of crystal

X-ray diffraction data for single crystals were collected using *BRUKER APEX*. The crystal structures were solved by the direct method<sup>8</sup> and refined by full-matrix least-squares using *SHELX97*.<sup>9</sup> The diagrams were prepared using *PLATON*.<sup>10</sup> Absorption corrections were performed using *SADABS*.<sup>11</sup> Non-hydrogen atoms were refined with anisotropic displacement parameters, and hydrogen atoms were included in the models in their calculated positions in the riding model approximation. *Crystallographic data for III*: C<sub>26</sub>H<sub>18</sub>O<sub>4</sub>·C<sub>12</sub>H<sub>14</sub>N<sub>2</sub>Cl<sub>2</sub>·C<sub>2</sub>H<sub>6</sub>O, *M*=697.62, orthorhombic, space group *P*2<sub>1</sub>2<sub>1</sub>2, *a*=13.8192(12), *b*=14.8487(13), *c*=17.0490(15) Å, *V*=3498.4(5) Å<sup>3</sup>, *Z*=4, *D*<sub>c</sub>=1.325 g cm<sup>−3</sup>, *μ*(Mo *Kα*)=0.233 mm<sup>−1</sup>, 21,881 reflections measured, 4558 unique, final *R*(*F*<sup>2</sup>)=0.0581 using 4348 reflections with *I*>2.0σ(*I*), *R*(all data)=0.0604, *T*=130(2) K. CCDC 714754. *Crystallographic data for V*: 0.5C<sub>26</sub>H<sub>18</sub>O<sub>4</sub>·0.5C<sub>12</sub>H<sub>14</sub>N<sub>2</sub>Cl<sub>2</sub>·0.5C<sub>4</sub>H<sub>10</sub>O, *M*=362.84, orthorhombic, space group *C*222<sub>1</sub>, *a*=14.0895(12), *b*=17.1652(14), *c*=14.6979(12) Å, *V*=3554.7(5) Å<sup>3</sup>, *Z*=8, *D*<sub>c</sub>=1.356 g cm<sup>−3</sup>, *μ*(Mo *Kα*)=0.233 mm<sup>−1</sup>, 15,435 reflections measured, 2337 unique, final *R*(*F*<sup>2</sup>)=0.0503 using 2109 reflections with *I*>2.0σ(*I*), *R*(all data)=0.0582, *T*=105(2) K. CCDC 714755. These data can be obtained free of charge via [www.ccdc.cam.ac.uk/conts/retrieving.html](http://www.ccdc.cam.ac.uk/conts/retrieving.html) (or from the Cambridge Crystallographic Data Centre, 12, Union Road, Cambridge CB21EZ, UK; fax: +44 1223 336 033; [deposit@ccdc.cam.ac.uk](mailto:deposit@ccdc.cam.ac.uk)).

### 4.4. Measurement of DRS of CT complexes

DRS of crystals were measured with a *HITACHI U-4000 Spectrometer*.

### 4.5. Measurement of XRD spectrum of CT complex

DRS of crystals were measured with a *HITACHI U-4000 Spectrometer*.

### 4.6. <sup>1</sup>H NMR analysis of CT complexes

<sup>1</sup>H NMR spectra were recorded with a Varian Mercury M300 spectrometer in dimethylsulfoxide-*d*<sub>6</sub> using tetramethylsilane as an internal standard (300 MHz).

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## Supplementary data

Supplementary data associated with this article can be found in the online version, at [doi:10.1016/j.tet.2009.02.044](https://doi.org/10.1016/j.tet.2009.02.044).

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