Synthesis, Crystal Structures, and Density Functional Theory Studies of Two Salt Cocrystals Containing Meldrum’s Acid Group

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ABSTRACT: Two salt cocrystals, C_{11}H_{14}N_4O_8 (DDD) and C_{17}H_{26}N_2O_8 (MDD), were synthesized and their structures were determined by single-crystal X-ray diffraction. DDD is made up of one (C_{11}H_{14}O_8)^{−} anion, one (C_{6}H_{10}N_2)^{+} cation, and one S,6-dimethyl-1H-benzo[d]imidazole molecule. MDD consists of one (C_{17}H_{26}O_8)^{+} cation and one (C_{13}H_{13}O_8)^{−} anion. DDD and MDD belong to the monoclinic, P21/c space group and triclinic, P-1 space group, respectively. A 1D-chained structure of DDD was constituted by N···N and N···O hydrogen bonds. However, a 1D-chained structure of MDD was bridged by N···H···O hydrogen bonds. Their density functional theory-optimized geometric structures with a B3LYP/6-311G(d,p) basis set fit well with those of crystallographic studies. By calculating their thermodynamic properties, the correlation equations of C_{E}^{	ext{exp}} S^\circ m, H^\circ m, and temperature T were obtained. By comparing the experimental electronic spectra with the calculated electronic spectra, it is found that the PBE-PBE/6-311G(d,p) method can simulate the UV–Vis spectra of DDD and MDD. In addition, the fluorescence spectra in the EtOH solution analysis show that the yellowish-green emission occurs at 570 nm (λ_{ex} = 310 nm) for DDD and the purplish-blue emission occurs at 454 nm (λ_{ex} = 316 nm) for MDD.

1. INTRODUCTION

As an important kind of N-heterocycle, imidazole and its derivatives have been widely applied in various fields including medicinal chemistry,8 bio-organic chemistry,9 fuel cells and solar cells,10 mild steel corrosion,11 electrocatalysts,12 agriculture,13 industry,14 organic light-emitting diodes (OLEDs),15,16 colorimetric and fluorometric chemosensors,17 and luminescent materials.18 As another kind of N-heterocycle, benzimidazole and its derivatives have also received significant attention owing to their potential applications in fluorescent sensors,13,14 fluorescent probes,15 chemodosimeters,16 OLEDs,17 and phosphorescent organic light-emitting diodes (PhOLEDs).18 Recently, Meldrum’s acid plays an increasingly important role in the synthesis of pharmacologically active compounds such as dual inhibitors of AChE and BChE in the treatment of Alzheimer’s disease,19 antibacterial agents,20 anticancer agents,21 and antioxidant agents.22 In particular, C-5-substituted derivatives of Meldrum’s acid have attracted considerable interest owing to their structure and unique properties. Different synthetic routes toward the C(5)-position of Meldrum’s acid have been reported.23–26 Based on the above facts, a series of C-5-substituted derivatives of Meldrum’s acid were prepared by our group during the past 10 years.27–30 However, to the best of our knowledge, most of the reported preparation methods are step by step, and each intermediate needs to be isolated and purified. In this work, two salt cocrystals containing Meldrum’s acid group are available via a simple one-pot eco-friendly method. Furthermore, less theoretical calculations or vibrational analyses have been carried out on Meldrum’s derivatives coupled with a benzo[d]imidazole or imidazole nucleus. As a part of ongoing research, two new compounds (Chart 1), S,6-dimethyl-1H-benzo[d]imidazol-3-ium-5-((2,2-dimethyl-4,6-dioxo-1,3-dioxan-5-ylidene)methyl)-2,2-dimethyl-4,6-dioxo-1,3-dioxan-5-ide,5,6-dimethyl-1H-benzo[d]imidazole (DDD) and 2-methyl-1H-imidazol-3-ium-5-((2,2-dimethyl-4,6-dioxo-1,3-dioxanylidenemethyl)-2,2-dimethyl-4,6-dioxo-1,3-dioxan-5-ide (MDD), were obtained by reacting Meldrum’s acid with 5,6-dimethyl-1H-benzo[d]imidazole and 2-methyl-1H-imidazole in the mixture of trimethoxymethane and ethanol. Their crystal structures, vibrational frequencies, thermodynamic properties, and electronic spectra along with density functional theory (DFT) or time-dependent DFT (TD-DFT) calculational results are also reported.

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2. EXPERIMENTAL AND THEORETICAL METHODS

2.1. Physical Methods. IR data were obtained on a Nicolet 5700 FT-IR Spectrometer. The UV–Vis absorption and fluorescence spectra were collected on a TU-1901 spectrometer and an RF-5301PC fluorospectrophotometer, respectively. The C, H, and N atoms of the two compounds were recorded on an Elementar Vario EL III elemental instrument. The NMR (¹H and ¹³C) spectra in CDCl₃ were obtained on a Bruker AVANCE III HD instrument (400 MHz).

2.2. Preparation of Two Cocrystals. The synthetic method of the two compounds was the same as that in our earlier report. Briefly, a mixture of Meldrum’s acid (0.72 g, 5 mmol) and trimethoxymethane (0.635 g, 6 mmol) was dissolved in EtOH (20 mL) and refluxed at 70 °C for 2−2.5 h. Then, 2-methyl-1H-imidazole (0.41 g, 5 mmol) was added to the reaction solution and the mixture continued refluxing for another 5 h. The solution was cooled, and the product was filtered, washed, dried, and recrystallized at room temperature in EtOH to collect red block-shaped crystals of MDD. Yield, 48.5%. Found: C, 53.62%; H, 5.38%; N, 7.41%. Calc. for C₁₇H₂₀N₂O₈: C, 53.68%; H, 5.30%; N, 7.37%. m.p.: 172.6−173.0 °C. FT-IR(KBr) cm⁻¹: 1718, 1678 (C=O), 1456 (C−N), 1275, 1190 (C−O).

¹H NMR (400 MHz, CDCl₃, δ ppm): 7.52 (s, 1H), 7.00 (s, 1H), 5.45 (s, 1H), 3.53 (s, 3H), 2.73 (s, 1H), 2.18 (s, 1H), 1.76 (s, 6H), 1.25 (s, 6H). ¹³C NMR (125 MHz, CDCl₃, δ ppm): 150, 118, 101, 94, 26, 19.

The preparation method of DDD was the same as that of MDD except that 5,6-dimethyl-1H-benzo[d]imidazole (0.73 g, 5 mmol) replaced 2-methyl-1H-imidazole (0.41 g, 5 mmol) to get a red powder (DDD). Yield, 28.5%. m.p.: 173.8−174.5 °C. Found: C, 63.58%; H, 5.85%; N, 9.52%. Calc. for C₃₁H₃₄N₄O₈: C, 63.04%; H, 5.80%; N, 9.49%. FT-IR(KBr) cm⁻¹: 1698, 1633 (C==O), 1449 (C−N), 1272, 1197 (C−O).

2.3. X-ray Diffraction Analysis of DDD and MDD. The X-ray data of DDD and MDD were collected on a Spider Rapid IP (Rigaku, Japan) detector. The molecular structures of the two compounds were ascertained by SHELXL-2015 and SHELXT-2015. The H atoms of the two compounds were placed in calculated positions and refined using the riding coordinates with C−H distances of 0.93−0.97 Å and N−H distances of 0.86 Å. \( U_{iso}(H) = 1.2U_{eq}(C) \) for aromatic/amide H atoms, and \( U_{iso}(H) = 1.5U_{eq}(C) \) for methyl H atoms.

2.4. Computational Methods. DFT calculations of DDD and MDD were performed with the Gaussian 09 package using a hybrid functional, namely, B3LYP or PBEPBE at the basis set 6-311G(d,p). TD-DFT calculations were used to predict the electronic spectra of the two compounds. The calculated vibrational frequencies and electronic spectra were obtained at the B3LYP/6-311G(d,p) level and the PBEPBE/6-311G(d,p) level.

3. RESULTS AND DISCUSSION

3.1. Crystal Structures of the Two Salt Cocrystals. The key parameters of the two salts are summarized in Table 1. Their molecular structures with hydrogen atoms are shown in Figure 1.

As shown in Figure 1, the molecular structure of DDD includes one \( \text{C}_{13}\text{H}_{13}\text{O}_{8}^- \) anion, one \( \text{C}_{6}\text{H}_{13}\text{N}_{2}^+ \) cation, and one 5,6-dimethyl-1H-benzo[d]imidazole molecule. However, MDD consists of one \( \text{C}_{13}\text{H}_{13}\text{O}_{8}^- \) and one \( \text{C}_{6}\text{H}_{13}\text{N}_{2}^+ \). The
The central C(7) atom is bridged by two Meldrum’s acid moieties, which forms the \((\text{C}_5\text{H}_7\text{O}_3)^-\) anion in the two compounds.

As shown in Table 2, bond lengths C7-C8 (1.384 (3) Å) and C7-C3 (1.374 (3) Å) (DDD) and C6-C7 (1.380 (2) Å) and C7-C8 (1.398 (2) Å) (MDD) are both larger than the C=C double bond and shorter than the C-C single bond, which form a conjugated system. Their bond angles (C3-C7=C8 (128.51 (19)°) (DDD) and C6-C7=C8 (130.91 (13)°) (MDD)) also both resemble that of our earlier report (131.19 (10)°). The two 1,3-dioxane rings of the \((\text{C}_5\text{H}_7\text{O}_3)^-\) anion in the two compounds are both in a distorted envelope conformation, with their puckering parameters as follows: for ring 1 (O1, O2, C1-C4) in

### Table 1. Crystal Structure Details for DDD and MDD

|       | DDD                  | MDD                  |
|-------|----------------------|----------------------|
| formula | \(\text{C}_5\text{H}_7\text{N}_2\text{O}_4\) | \(\text{C}_6\text{H}_8\text{N}_2\text{O}_4\) |
| CCDC   | 1817941              | 2014484              |
| color/shape | red/block           | red/block           |
| \(M_t\) | 380.35               | 380.35               |
| crystal system, space group | monoclinic, \(P2_1/c\) | triclinic, \(P\) |
| \(a\), \(b\), \(c\) (Å) | 13.2783 (6), 16.0197 (6), 14.3536 (6) | 9.936 (2), 10.556 (2), 10.880 (2) |
| \(\alpha\), \(\beta\), \(\gamma\) (°) | 90, 103.2430 (10), 90 | 113.33 (3), 100.27 (3), 11.39 (3) |
| crystal size (mm) | 0.1 x 0.8 x 0.04 | 0.25 x 0.18 x 0.10 |
| wavelength (Å) | 0.71073              | 0.71073              |
| \(\theta\) ranges (°) | 3.06-27.484        | 3.476-27.47         |
| V (\(Å^3\)) | 3135.5 (2)           | 893.0 (3)           |
| \(Z\) | 4                    | 2                    |
| \(F(000)\) | 1248                | 400                  |
| \(D_0\) (g·cm\(^{-3}\)) | 1.251              | 1.415                |
| \(-h\), \(h-l\), \(k-l\), \(l\) | -17, 17/+-20, 21/-18, 18, 13 | -12, 12/-13, 13/-14, 13, 13 |
| total, unique, and \([I > 2\sigma(I)]\) reflections | 30,494, 7196, 2955 | 8284, 3960, 3295 |
| no. of reflections, restraints, parameters | 7196, 0, 396 | 3960, 0, 244 |
| \(R(int)\) | 0.0462              | 0.0393              |
| \(R_c\), \(wR_c\), \(S\) | 0.0541, 0.1311, 0.890 | 0.0539, 0.1383, 1.108 |
| \((\Delta\rho)_{	ext{min}}\) (\((\Delta\rho)_{	ext{max}}\) (\(e/Å^3\)) | 0.290, -0.194 | 0.386, -0.356 |

### Table 2. Main Bond Lengths (Å) and Bond Angles (°) by X-ray and DFT Calculations for DDD and MDD

|       | DDD                      | MDD                      |
|-------|--------------------------|--------------------------|
| bond  | exp. calc. | bond  | exp. calc. |
| \(C7-C8\) | 1.384 (3) 1.405          | \(C7-C8\) | 1.398 (2) 1.399 |
| \(C3-C7\) | 1.374 (3) 1.385          | \(C6-C7\) | 1.380 (2) 1.383 |
| \(O5-C9\) | 1.214 (3) 1.204          | \(O7-C4\) | 1.2096 (19) 1.202 |
| \(O6-C11\) | 1.222 (3) 1.237          | \(O8-C5\) | 1.2211 (17) 1.223 |
| \(O8-C11\) | 1.350 (3) 1.367          | \(O6-C5\) | 1.3559 (19) 1.361 |
| \(O8-C13\) | 1.425 (4) 1.446          | \(O6-C3\) | 1.4450 (17) 1.448 |
| \(O7-C9\) | 1.367 (3) 1.391          | \(O5-C4\) | 1.3634 (2) 1.384 |
| \(O7-C10\) | 1.432 (3) 1.418          | \(O5-C3\) | 1.433 (2) 1.422 |
| \(N(1)-C(14)\) | 1.339 (3) 1.324 | \(N1-C14\) | 1.374 (3) 1.381 |
| \(N(1)-C(15)\) | 1.377 (3) 1.389 | \(N1-C16\) | 1.330 (2) 1.332 |

### Table 3. Intra- and Intermolecular Interactions and π⋯π Stacking Interactions of DDD and MDD

| D–H⋯A | symmetry | D–A (Å) | \(\angle\)D–H–A (°) |
|-------|----------|---------|-------------------|
| **DDD** |          |         |                   |
| N1–H1⋯O2 | 1 – x, y, z | 1 – z | 2.723 (5) 167 |
| N2–H2⋯O7 | 1 – x, y, z | 2 – x, y, 1/2 | 2.953 (6) 115 |
| N2–H2⋯O8 | 2 – x, y, 1/2 | 1 – z | 2.833 (6) 157 |
| C3g–C3g | x, y, z | 2.467 (9) |
| **MDD** |          |         |                   |
| N4–H4⋯O4 | 1 – x, y, 1/2, 1/2 | 1 – z | 2.754 (2) 157.6 |
| N1–H1⋯O6 | intra | 2.743 (3) 171.3 |
| N3–H3⋯N2 | intra | 2.685 (2) 174.2 |

*3C ring denotes ring N1, N2, and C14–C16.*

Figure 1. ORTEP drawings of DDD and MDD with 30% probability thermal ellipsoids.
(3) Å, respectively. However, the \((\text{C}_9\text{H}_{10}\text{N}_2)^+\) cation and \(\text{C}_9\text{H}_{11}\text{N}_2\) molecule are linked by N–H···N intramolecular interactions, and the distances of N3···N2 are 2.685 (2) Å. The bond angle of N3–H3···N2 is 174.2°. The 1D-chained structure of DDD was connected by N–H···O intermolecular interactions (Figure 2). The 1D chain further constitutes the 3D-net structure (Figure 3).

**3.2. Optimization of Molecular Geometry for MDD and DDD.** The optimized geometric structures of the two compounds are carried out using DFT at the B3LYP/6-311G(d,p) level, and the values are also listed in Table 2.

As shown in Table 2, most of the predicted bond lengths and angles are slightly larger than the ones measured in the experiments. It is likely because the experimental parameters of the two compounds are described in the solid state; however, the predicted values are obtained in the gas phase. Comparing the predicted values with the experimental ones, it can be found that the maximum differences in bond lengths and bond angles are 0.025 Å and 3.60° (DDD) and 0.011 Å and 1.97° (MDD), respectively, indicating that the predicted results are satisfactory and the B3LYP/6-311G(d,p) level is suitable to simulate the two crystal structures.

**3.3. Thermodynamic Properties.** Three main thermodynamic properties (capacity \((C^0_{p,m})\), entropy \((S^0_{m})\), and enthalpy \((H^0_{m})\)) of MDD and DDD are listed in Table 4. As shown in Table 4, all the values of thermodynamic parameters increase with the temperature rising from 100.0 to 1000.0 K, which is mainly due to the enhancement of the two molecular vibrations when the temperature rises.

The correlation equations of \(C^0_{p,m} S^0_{m} H^0_{m}\) and temperature \(T\) are as follows:

\[
\begin{align*}
C^0_{p,m} = 44.478 + 1.459T - 6.163 \times 10^{-4} T^2, \\
S^0_{m} = 25135 \\
H^0_{m} = 4.78 \times 10^{22} \times 10^{-4} T^2
\end{align*}
\]

The correlation equations are obtained using the least squares method. The values of the correlation coefficient are given in Table 4.
3.4. Electronic Analysis. The experimentally obtained absorption spectra of the two compounds in EtOH and the calculated spectra in the gas state using the TD-DFT method at the PBE0-PBE/6-311G(d,p) level are shown in Figure 4.

The two compounds both present two absorption bands at 245 and 379 nm (DDD) and at 241 and 377 nm (MDD): the first absorption band is owing to the $\pi \rightarrow \pi^*$ transition of the $(C_9H_{11}N_2)^+$ cation or $(C_4H_7N_2)^+$ cation and the second band is due to the $n \rightarrow \pi^*$ transition of the $(C_{13}H_{13}O_8)^-$ anion. The results resemble those of the literature reported (243 and 384). The calculated spectra of the two compounds were also found to exhibit two bands at 268 and 352 nm (DDD) and at 265 and 342 nm (MDD), which illustrates that the PBE0-PBE/6-311G(d,p) method can simulate the experimental electronic spectra. The charge densities of four frontier molecular orbitals of the two compounds are listed in Figure 5. The HOMO-1 and HOMO electrons are mostly localized on both the 1,3-dioxane ring and C6-C7=C8 bonds of the $(C_{13}H_{13}O_8)^-$ anion in the two compounds; however, the LUMO and LUMO+1 electrons are localized on the
Two new salt cocrystals, C$_7$H$_{14}$N$_4$O$_4$ (DDD) and C$_{17}$H$_{36}$N$_2$O$_4$ (MDD), were synthesized and characterized by elemental analysis, infrared, UV–Vis, and NMR ($^1$H and $^13$C) spectroscopy and single-crystal X-ray diffraction. Their crystal structures show that DDD belongs to the monoclinic, $P2_1/c$ space group, and MDD belongs to the triclinic, P-1 space group. In DDD, the (C$_{13}$H$_{12}$O$_4$)$^-$ anion, (C$_{6}$H$_{15}$N$_2$)$^{+}$ cation, and C$_6$H$_{10}$N$_2$ molecule were linked by N–H···O and N–H···N molecular interactions. However, in MDD, the (C$_{13}$H$_{12}$O$_4$)$^-$ anion and (C$_{6}$H$_{14}$N$_2$)$^{+}$ cation were bridged by N–H···O intermolecular interactions. The optimized geometric structure method at the PBEPBE/6-311G(d,p) level is suitable to simulate the molecular structures of DDD and MDD. The calculated results of the two compounds with the TD-DFT method at the PBEPBE/6-311G(d,p) level are in accordance with experimental values.

### ASSOCIATED CONTENT

Accession Codes
CCDC 1817941 (DDD) and 2014484 (MDD) contain the supplementary crystallographic data for this paper. These data can be obtained free of charge at www.ccdc.cam.ac.uk/conts/retrieving.html [or from the Cambridge Crystallographic Data Centre (CCDC), 12 Union Road, Cambridge CB2 1EZ, UK; fax: +44(0)1222-336033; email: deposit@ccdc.cam.ac.uk].

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**Author Contributions**
W.Z. designed the experiment, synthesized the two compounds, and wrote the draft. X.W. provided the funds. Y.Z. calculated the vibration spectra and electronic spectra. All authors have read and agreed to the published version of the manuscript.

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