

An unprecedented DDQ–nickel(II)Salen complex interaction and X-ray crystal structure of nickel(II)Salen.DDH co-crystal



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ABSTRACT

The charge transfer interaction between DDQ (2,3-dichloro-5,6-dicyano-1,4-benzoquinone) and [Ni(Salen)] (Salen = bis(salicylaldehyde)ethylenediamine) complex was studied in the solution and solid state. The yellow color of [Ni(Salen)] dissolved in dichloromethane or chloroform was changed to green in the presence of the DDQ. The green precipitates of [Ni(Salen)]⁺DDQ⁻ charge transfer complex were collected via adding the *n*-hexane to the solution. Crystallization of charge transfer solid from dichloromethane/chloroform resulted in [Ni(Salen)] and DDH (DDH = 2,3-dichloro-5,6-dicyano-1,4-dihydroxybenzene), in which DDH was obtained from the catalytic reduction of DDQ via the oxidation of water. The structure of the new solid was determined by the X-ray crystallography. In this solid, DDH was combined with [Ni(Salen)] to create [Ni(Salen)].DDH co-crystals, in which there were the DDH molecules, were bonded to each other as a chain and bonded to the phenolic group of Salen Schiff base by the hydrogen bonding.

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1. Introduction

A large number of Schiff base ligands and their metal complexes have been investigated because of their interesting properties, such as catalytic activity for the preparative oxygenation of phenols [1], indols [2] and amines [3]. Beside these applications, some metal Schiff bases have been applied as ligands toward the tin compounds in the organic solvents [4–7].

The charge transfer interactions of DDQ and metal organic compounds have been investigated extensively. These interactions undergo complete electron transfer to form ML⁺DDQ⁻ salt through the outer sphere charge transfer of ML.DDQ complexes [8–13]. The DDQ is also applied for the catalytic dehydrogenation of coordinated Schiff bases [14].

Alternative energy sources are needed to be combined with energy storage. One of the best and clean energy sources is hydrogen, which can be prepared by the electrolysis of water. This method is cost consuming method and also needs a safe solid carrier for the storage of the H₂ molecules. In this direction, the DDQ is proposed as a hydrogen storage place in an electrocatalytic process [15].

Beside the ionic bonding, the hydrogen bonding interaction is used as the noncovalent interaction by crystal engineers. The

hydrogen bonding provides a directional bond, leading to designer solids [16].

The aims of the present work was studying DDQ and [Ni(Salen)] complex to investigate the charge transfer interaction between the complex as the donor and the DDQ as the acceptor. The DDQ and [Ni(Salen)] interaction result in forming the [Ni(Salen)]⁺DDQ⁻ salt. During the crystallization, [Ni(Salen)]⁺ absorbs H₂O to form the solvated cationic complex. The Ni(III) species gains the electron and DDQ⁻ abstracts the H⁺ from the coordinated water to form [Ni(Salen)] and DDH (Fig. 1). The strong hydrogen bonding interactions between [Ni(Salen)] and DDH lead to [Ni(Salen)].DDH co-crystal.

2. Experimental

2.1. Chemicals and apparatus

All chemicals and solvents used for synthesis were commercially available reagents. Infrared spectra were recorded as KBr discs on an FT-IR JASCO-680 spectrophotometer in the range of 4000–400 cm⁻¹. The elemental analysis was determined on a CHN-O-Heraeus elemental analyzer. UV–Vis spectra were recorded on a JASCO V-570 spectrophotometer in the range of 190–900 nm. The H NMR spectra were recorded by a Bruker Avance DPX 250 MHz spectrometer. X-ray diffraction studies were carried out by Bruker smart Apex II-2009 CCD.

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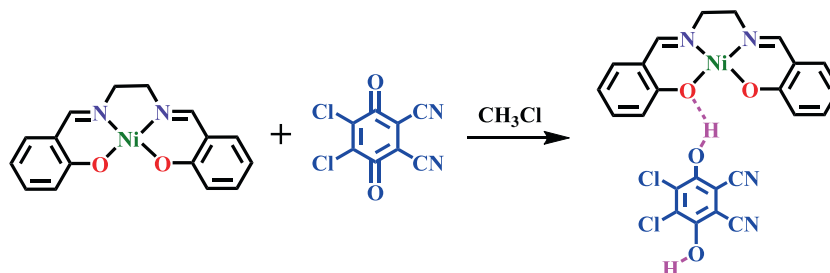


Fig. 1. The structure of [Ni(Salen)], DDQ and [Ni(Salen)].DDH complexes.

2.2. Synthesis of Schiff base and nickel complex

The Schiff base ligand, H₂Salen, was prepared according to the literature [17] by condensation between 1,2-ethylenediamine and 2-hydroxybenzaldehyde (2:1 mole ratio) in methanol and recrystallized by dichloromethane/methanol mixed solvent through the partial evaporation of dichloromethane. The [Ni(Salen)] complex was prepared according to a method described previously [18].

The general procedure for the synthesis of the [Ni(Salen)].DDQ adduct is as follows:

A dichloromethane solution containing an appropriate amount of DDQ (0.227 g, 1.0 mmol) was added to a solution (20 mL) of [Ni(Salen)] (0.325 g, 1.0 mmol) in dichloromethane. The color of solution was changed to green immediately. *n*-Hexane was added to the solution dropwise until the green precipitates were formed. The precipitates were filtered and washed with *n*-hexane/dichloromethane. Suitable crystals were formed by the recrystallization of precipitates in dichloromethane/chloroform. The needle crystals were formed during a week.

[Ni(Salen)].DDQ, 0.2CH₂Cl₂, Yield (80%). Anal. Calc. for C_{24.2}H_{14.4}Cl_{2.4}N₄O₄Ni: C, 51.08; H, 2.55; N, 9.85. Found: C, 51.07; H, 2.61; N, 9.98%. ¹H NMR (CDCl₃, δH) ([Ni(Salen)].DDH): 3.45 (s, 4H, CH₂CH₂), 5.06 (s, 2H, phenolic OH), 6.53–6.84 (t, 2H, aromatic protons), 6.89–6.92 (d, 2H, aromatic protons), 7.01–7.06 (d, 2H, aromatic protons), 7.11–7.26 (t, 2H, aromatic protons), and 7.53 (s, 1H, HC=N).

2.3. Crystal structure determination and refinement of [Ni(Salen)].DDH

The yellow plate crystal of [Ni(Salen)].DDH complex with a dimension of 0.21 × 0.13 × 0.09 mm was chosen and mounted on a glass fiber and used for data collection. Cell constants and an orientation matrix for data collection were obtained by least-squares refinement of diffraction data from 6879 unique reflections. Data were collected at a temperature of 100 K to a maximum 2θ value of 60.2° in a series of ω scans at 1° oscillations and integrated using the Stoe X-AREA software package. The numerical absorption coefficient, μ, for Mo Kα radiation was 0.710 mm⁻¹.

A numerical absorption correction was applied using X-RED and X-SHAPE [19] softwares. The data were corrected for Lorentz and Polarization effects. The structures were solved by direct methods [20] and subsequent difference Fourier maps and then refined on F² by a full-matrix least-squares procedure using anisotropic displacement parameters [21]. All hydrogen atoms were located in a difference Fourier map and then refined isotropically. Atomic factors were from International tables for X-ray crystallography. All refinements were performed using the X-STEP32 crystallographic software package [22]. A summary of the crystal data, experimental details and refinement results can be seen in Table 1 for [Ni(Salen)].DDH compound. A region of disordered electron

Table 1

Crystallographic data and structure refinements data for [Ni(Salen)].DDH.

Formula	C ₂₄ H ₁₆ Cl ₂ N ₄ NiO ₄
Formula weight	554.02
Temperature (K)	100(2)
Wavelength λ (Å)	0.71073
Crystal system	triclinic
Space group	P1
Crystal size (mm)	0.21 × 0.13 × 0.09
a (Å)	8.5663 (7)
b (Å)	11.1559 (9)
c (Å)	13.9130 (11)
α (°)	111.965 (1)
β (°)	103.449 (2)
γ (°)	92.788 (2)
V	1185.63 (17)
D _{calc} (g cm ⁻³)	1.552
Z	2
θ ranges for data collection	1.64–30.1
F(000)	564
Absorption coefficient	1.083
Index ranges	−12 ≤ h ≤ 12, −15 ≤ k ≤ 15, −19 ≤ l ≤ 19
Data collected	24736
Unique data (R _{int})	6879, (0.037)
Parameters/restraints	325/0
Final R ₁ , wR ₂ ^a (Observed data)	0.0421, 0.1220
Final R ₁ , wR ₂ ^a (All data)	0.0591, 0.1345
Goodness of fit (GOF) on F ² (S)	1.035
Largest difference peak and hole (e Å ⁻³)	1.77–0.35

^a R₁ = Σ||F_o| − |F_c||/Σ|F_o|, wR₂ = [Σ(w(F_o² − F_c²)²)/Σw(F_o²)]^{1/2}.

density, most probably, the disordered dichloromethane solvent molecule, was treated with the SQUEEZE routine in PLATON [23].

3. Results and discussion

3.1. IR characteristics

The IR spectra of [Ni(Salen)], DDQ, [Ni(Salen)].DDQ and [Ni(Salen)].DDH compounds studied in this work have been listed in Table 2.

The strong band of IR at about 1616 cm⁻¹ was assigned to C=N band characteristic of [Ni(Salen)] complex [24]. The ring skeletal vibrations (C=C) of Schiff base coordinated to the nickel ion were seen at 1451 and 1535 cm⁻¹. Important DDQ stretching frequencies containing nitrile (N≡C) and carbonyl (C=O) groups appeared in 2233 and 1674 cm⁻¹, respectively. Carbonyl stretching frequency of [Ni(Salen)].DDQ appeared at about 1675 cm⁻¹ while the nitrile vibrational frequency was seen at 2230 cm⁻¹ as a weak peak. In the crystalline product, [Ni(Salen)].DDH, the vibrational frequency of the carbonyl group disappeared completely and the nitrile stretching frequency was seen at 2230 cm⁻¹ as a medium peak.

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