

Supporting Information

The preparation of the N3 dye followed the general procedure described in *JACS* **1993**, *115*, 6382. A typical procedure is given below.

cis-Dichloro-*N,N*-bis(2,2'-bipyridyl-4,4'-dicarboxylicacid)ruthenium(II)dihydrate, $\text{RuL}_2\text{Cl}_2 \cdot 2\text{H}_2\text{O}$ (where L = 2,2'-bipyridyl-4,4'-dicarboxylicacid) was synthesized by mixing 1.544 g $\text{RuCl}_3 \cdot 3\text{H}_2\text{O}$ (5.9 mmol), 1.84 g 2,2'-bipyridyl-4,4'-dicarboxylic acid (11.8 mmol), and 1.66 g LiCl (0.40 mmol) in 25 ml DMF (reagent grade). The solution was refluxed with stirring for 8 h. The solution was cooled to room temperature, combined with 50 ml acetone, and cooled at 0 °C overnight. The mixture was then filtered and the solid material was dissolved in 50 mL CH_2Cl_2 . The solution was extracted three times with 50 ml H_2O , dried over MgSO_4 , filtered, and rotary evaporated to dryness.

N3, *cis*-Di(thiocyanato)-*N,N*-bis(2,2'-bipyridyl-4,4'-dicarboxylicacid)ruthenium(II) dihydrate, $[\text{RuL}_2(\text{NCS})_2] \cdot 2\text{H}_2\text{O}$ was synthesized by dissolving 200 mg of $\text{RuL}_2(\text{Cl})_2 \cdot 2\text{H}_2\text{O}$ (0.28 mmol) prepared as described above in 30 ml of DMF in the dark. Approximately 20 ml of 0.1 M aqueous NaOH was added followed by the addition of 49.9 mg NaSCN (0.616 mmol) dissolved in 1 ml of H_2O . The reaction mixture was heated to reflux under argon for 6 hours with stirring. Next, the reaction mixture was allowed to cool, and the solvent was removed. The material was dissolved in a minimal amount of H_2O and filtered using a glass crucible. The pH of the filtrate was lowered to 2.0 using HCl and the solution was cooled in the refrigerator overnight. Finally, the solid was filtered and rinsed sequentially with H_2O , acetone:ether solution (1:10), and anhydrous ether, and the N3 product was dried under vacuum. Spectroscopic characterization was satisfactory.