

The dataset contains data for this study that were measured at the University of Leeds and the University of Rennes:

X-ray Crystallographic data:

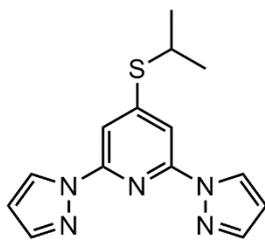
- Structures of **1[BF₄]₂·MeNO₂** (phase 1) at thirteen temperatures between 120-230 K (CCDC 1976560-1976565, 1976567-1976572 and 1976574).
- Structure of **1[BF₄]₂·MeNO₂** (phase 2) at 40 K (CCDC 1976613), and unit cell parameters showing LIESST relaxation of the phase 3 irradiated crystal.
- Structures of **1[BF₄]₂·yMe₂CO** at twelve temperatures between 110-220 K (CCDC 1976575-1976582, 1976586-1976589).
- Structures of **1[BF₄]₂·yMe₂CO** at 20 K, before and after photoexcitation (CCDC 1976566 and 1976573).
- Structures of **1[BF₄]₂·MeCN** at thirteen temperatures between 120-230 K, measured in both cooling and warming temperature ramps (CCDC 1976590-1976612 and 1976614-1976615).
- Structures of **1[BF₄]₂·H₂O** at eleven temperatures between 150-260 K, (CCDC 1976616-1976626).
- Structures of **1[ClO₄]₂·MeNO₂** at 10 K, before and after photoexcitation (CCDC 1976559 and 1976585).
- Structures of **1[ClO₄]₂·H₂O** at 10 K, before and after photoexcitation (CCDC 1976583 and 1976584).

Hirshfeld surface calculations (*CrystExplorer* output files) for the full asymmetric unit and the individual cation, anion and solvent residues in:

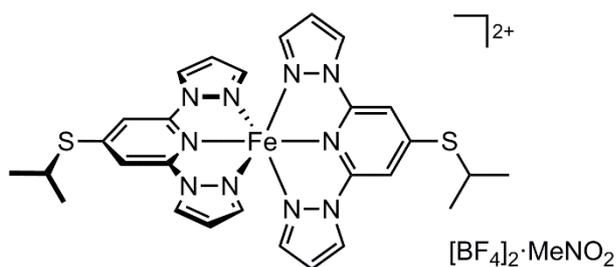
- High-spin **1[BF₄]₂·MeCN** at 15 K.
- High-spin and low-spin **1[BF₄]₂·MeCN** at 85 K.
- High-spin and low-spin **1[BF₄]₂·MeCN** at 165 K.
- High-spin and low-spin **1[BF₄]₂·H₂O** at 20 K.
- High-spin and low-spin **1[BF₄]₂·H₂O** at 220 and 200 K, respectively.
- High-spin and mixed spin **1[ClO₄]₂·MeNO₂** at 10 K.
- High-spin and low-spin **1[ClO₄]₂·H₂O** at 10 K.

Computational data in the study belong to co-author Sergi Vela at the EPFL, Lausanne.

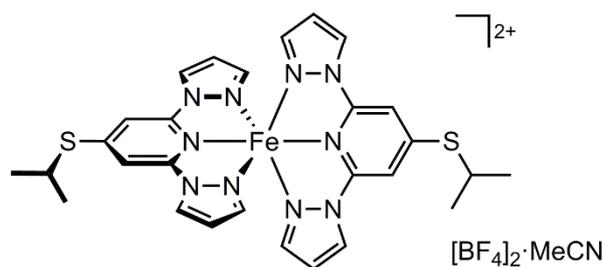
Compounds referred to in this dataset



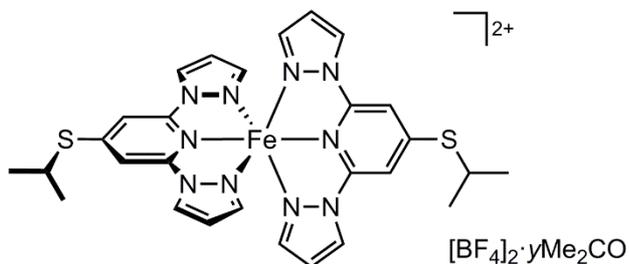
L
4-(Isopropylsulfanyl)-2,6-di(pyrazol-1-yl)pyridine
C₁₄H₁₅N₅S



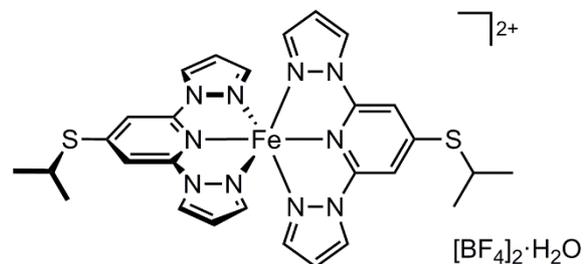
1[BF₄]₂·MeNO₂
Bis[4-(isopropylsulfanyl)-2,6-di(pyrazol-1-yl)pyridine]-
iron(II) ditetrafluoroborate nitromethane solvate
C₂₉H₃₃B₂F₈FeN₁₁O₂S₂



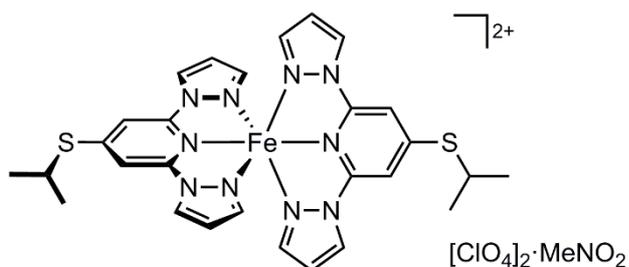
1[BF₄]₂·MeCN
Bis[4-(isopropylsulfanyl)-2,6-di(pyrazol-1-yl)pyridine]-
iron(II) ditetrafluoroborate acetonitrile solvate
C₃₀H₃₃B₂F₈FeN₁₁S₂



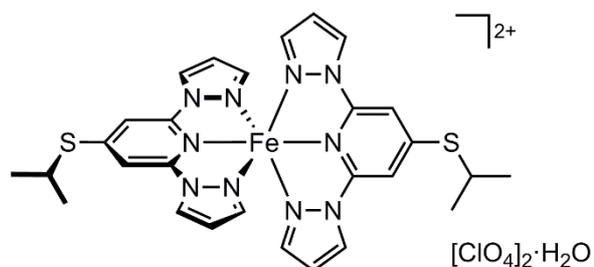
1[BF₄]₂·yMe₂CO
Bis[4-(isopropylsulfanyl)-2,6-di(pyrazol-1-yl)pyridine]-
iron(II) ditetrafluoroborate acetone (1/0.75) solvate
C_{30.25}H_{34.50}B₂F₈FeN₁₀O_{0.75}S₂



1[BF₄]₂·H₂O
Bis[4-(isopropylsulfanyl)-2,6-di(pyrazol-1-yl)pyridine]-
iron(II) ditetrafluoroborate hydrate
C₂₈H₃₂B₂F₈FeN₁₀OS₂

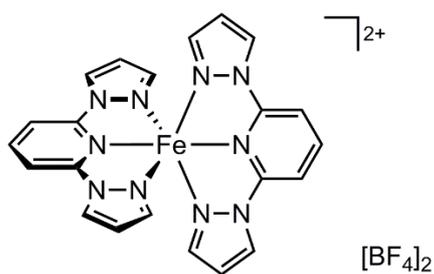


1[ClO₄]₂·MeNO₂
Bis[4-(isopropylsulfanyl)-2,6-di(pyrazol-1-yl)pyridine]-
iron(II) diperchlorate nitromethane solvate
C₂₉H₃₃Cl₂FeN₁₁O₁₀S₂

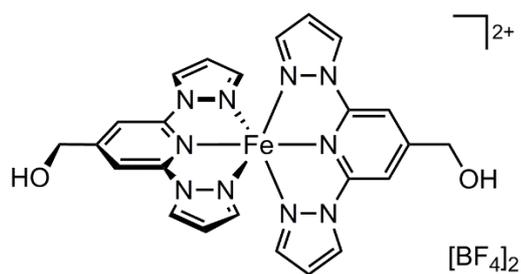


1[ClO₄]₂·H₂O
Bis[4-(isopropylsulfanyl)-2,6-di(pyrazol-1-yl)pyridine]-
iron(II) diperchlorate hydrate
C₂₈H₃₂Cl₂FeN₁₀O₉S₂

Compounds referred to in this dataset (continued)



Bis[2,6-di(pyrazol-1-yl)pyridine]iron(II)
ditetrafluoroborate
 $C_{22}H_{18}B_2F_8FeN_{10}$



Bis[4-(hydroxymethyl)-2,6-di(pyrazol-1-yl)pyridine]-
iron(II) ditetrafluoroborate
 $C_{24}H_{22}B_2F_8FeN_{10}O_2$