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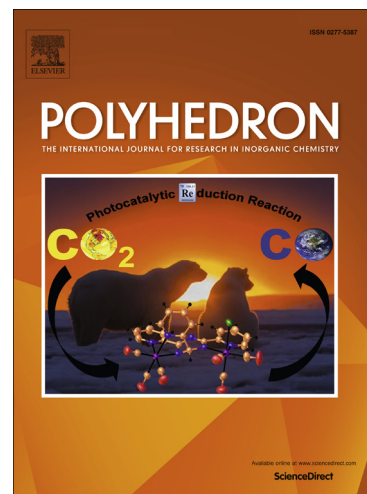
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Homoleptic, di- and trivalent transition metal complexes with monoanionic *N,N,O*-heteroscorpionate ligands: Potential redox mediators for dye-sensitized solar cells?

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Abstract

The reaction of bis(3,5-dimethylpyrazol-1-yl)acetic acid (Hbdmpza) with metal(II) acetates $M(\text{OAc})_2$ ($M = \text{Mn}, \text{Co}$ and Fe) yields a series of homoleptic first row transition metal complexes: $[\text{Mn}(\text{bdmpza})_2]$ (**1**), $[\text{Co}(\text{bdmpza})_2]$ (**2**) and $[\text{Fe}(\text{bdmpza})_2]$ (**3**). Subsequent oxidation results in the formation of the trivalent complexes $[\text{Mn}(\text{bdmpza})_2]\text{BF}_4$ (**4**), $[\text{Co}(\text{bdmpza})_2]\text{BF}_4$ (**5**) and $[\text{Fe}(\text{bdmpza})_2]\text{BF}_4$ (**6**). An unidentate binding of the carboxylate donor and thus κ^3 coordination of the ligand was determined by IR spectroscopy and single-crystal X-ray diffraction experiments, which also confirm the formation of the homoleptic complexes and the preservation of the geometry during oxidation. Furthermore, the magnetic measurements exhibit that all complexes are in high-spin state with exception of the low-spin complex $[\text{Co}(\text{bdmpza})_2]\text{BF}_4$ (**5**). The high-spin complex $[\text{Mn}(\text{bdmpza})_2]\text{BF}_4$ (**4**) displays a Jahn-Teller distortion. In addition, the divalent and trivalent complexes show promising spectroscopic and electrochemical properties regarding their suitability as potential redox mediators for dye-sensitized solar cells (DSSCs).

Keywords. Heteroscorpionate ligand, Bis(pyrazol-1-yl)acetic acid, Transition metal complexes, Redox mediator, DSSC

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