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Oxidant or Catalyst for Oxidation? A Study of How Structure and Disorder Changes Selectivity for Direct vs. Catalytic Oxidation Mediated by Manganese (III,IV) Oxides

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Structural type and disorder have become important questions in catalyst design with the most active catalysts often noted to be “disordered” or “amorphous”. To quantify the effects of disorder and structural type systematically, a test set of manganese (III,IV) oxides was developed and their reactivity as oxidants and catalysts tested against three substrates; methylene blue, hydrogen peroxide and water. We find that disorder destabilises the materials thermodynamically making them stronger chemical oxidants but not necessarily better catalysts. For the disproportionation of H2O2 and the oxidative decomposition of methylene blue- MnOx mediated direct oxidation competes with catalytically-mediated oxidation, making the most disordered materials the worst catalysts. Whereas, for water oxidation the most disordered materials and the strongest chemical oxidants are also the best catalysts. Even though the manganese (III,IV) oxide materials were able to oxidize both methylene blue and peroxides directly, the same materials were able to act as catalysts for the oxidation of methylene blue in the presence of peroxides. This implies that effects of electron transfer timescales are important and strongly affected by structural type and disorder. This is discussed in the context of catalyst design.

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