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Coordination chemistry of manganese and iron with N,O-donor ligands: oxidation catalysis and magnetochemistry of clusters

Godbole, M.D.

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STELLINGEN

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Coordination Chemistry of Manganese and Iron with N,O-Donor Ligands: Oxidation Catalysis and Magnetochemistry of Clusters

1. The proposal by Berkessel *et al.* that “Mn TMTACN/Ascorbic acid system is a remarkably efficient catalysts for epoxidation” is not correct, because the studies reported in the paper are for the “MnTMTACN/Sodium ascorbate” system and not for the Mn TMTACN/Ascorbic acid system. The difference between catalytic activities imparted by a carboxylic acid and a carboxylate salt as an additive could be completely opposite.

Berkessel, A., Sklorz, C. A.; Tet. Lett. (1999) 40, 7965-7968.

This thesis.

2. In publications concerning catalytic asymmetric epoxidation reactions many important experimental data are often missing, which is particularly annoying when the product epoxides are not commercially available.

Martinez, A., Hemmert, C., Meunier, B.; J. Catalysis (2005) 234, 250-255

Palucki, M., McCormick G. J., Jacobsen E. N.; Tet. Letters (1995) 36, 5457-5460

3. The reports by Gilbert versus de Boer *et al.* are contrasting in the sense that both use the same catalytic system, Mn-TMTACN, however, Gilbert *et al.* use basic pH conditions for good epoxidation activity and de Boer *et al.* report the high epoxidation activity when using strongly acidic additives.

Gilbert, B. C., Smith J. R. L., Payeras A. M. I., Oakes J., Prats R. P. I.; J. Mol. Cat. A: Chemical (2004) 219, 265–272

de Boer, J. W.; Brinksma, J.; Browne, W. R.; Meetsma, A.; Alsters, P. L.; Hage, R.; Feringa, B. L.; J. Am. Chem. Soc. (2005) 127, 7990-7991.

4. The conclusion reported by Pietikainen: “Basicity is a sufficient property for catalytic epoxidation activity by manganese complexes” is correct and it not only applies to bases such as carboxylate or carbonate salts, but also to substituted imidazoles.

Pietikainen, P., Tetrahedron (1998), 54, 4319-4326.

This thesis.

5. The structural investigation of high-valent manganese-salen complexes using Raman, XANES, and EXAFS spectroscopy in combination with UV-VIS and IR methods by Feth et al. provides a clear evidence of species that are “NOT” involved in the catalytic epoxidation mechanism.
Feth, M. P., Bolm, C., Hildebrand, J. P., Kohler, M., Beckmann, O., Bauer, M., Ramamonjisoa, R., Bertagnolli, H., Chem. J. (2003), 9, 1348-1359
6. Very small changes in the ligand environment such as the stereochemistry of the ligands around the metal center may cause a considerable difference in the effective catalytic epoxidation activity by metal complexes.
This thesis.
7. ESI-MS analyses in combination with deuterated solvents is a very useful tool for determination of solution structures of metal complexes.
This thesis.
8. The awe-inspiring beauty and the very frustrating fact of the Mn,Fe/acetate clusters is that their “self-assembly” cannot be controlled.
This thesis.
9. An Indian in a Dutch country, at first ‘likes’ the Dutch weather being cold, cloudy and wet; then tries to get used to it in time; and after a while starts complaining about the Dutch weather exactly like the native people around.
10. Four years of a PhD study in the Netherlands teaches not only useful skills in chemistry, but in addition may deliver survival skills like painting the house, building furniture and to raise the curtains.
11. Living in a new country, with new people and in a new culture, broadens the mind, increases tolerance to experiences of life and creates bonds between people that used to be thousands of miles apart.
12. An easy recipe for developing an RSI syndrome is the following: “try to make ‘good’ pictures of ‘high’ quality from the obtained X-ray crystal structures”. The chances of getting the syndrome are directly proportional to the number of crystal structures obtained.

Meenal D. Godbole
Leiden, 12th January 2006