Title

Air-Stable Cobalt(III) and Chromium(III) Complexes as Single-Component Catalysts for the Activation of Carbon Dioxide and Epoxides

Abstract

Cobalt(III) and chromium(III) salophen chloride complexes were synthesized and tested for the cycloaddition of carbon dioxide (CO2) with epoxides to obtain cyclic carbonates. The cat1, cat2, cat4, and cat5 complexes presented high catalytic activity without cocatalysts and are solvent-free at 100 °C, 8 bar, and 9 h. At these conditions, the terminal epoxides (1a-1k) were successfully converted into the corresponding cyclic carbonates with a maximum conversion of ~99%. Moreover, cat5 was highlighted due to its capability of opening internal epoxides such as limonene oxide (1l) with a 36% conversion to limonene carbonate (2l), and from cyclohexene oxide (1m), cyclic trans-cyclohexene carbonate (2m) and poly(cyclohexene carbonate) were obtained with 15% and 85% selectivity, respectively. A study of the coupling reaction mechanism was proposed with the aid of electrospray ionization mass spectrometry (ESI-MS) analysis, confirming the single-component behavior of the complexes through their ionization due to epoxide coordination. In addition, crystallographic analysis of cat1 single crystals grown in a saturated solution of pyridine helped to demonstrate that the substitution of chloride ion by pyridine ligands to form an octahedral coordination occurs (Py-cat1), supporting the proposed mechanism. Also, a recyclability study was performed for cat5, and a total turnover number of 952 was obtained with only minor losses in catalytic activity after five cycles.

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Index Keywords

Carbonates; Carbonation; Catalyst activity; Chlorine compounds; Chromium compounds; Cobalt compounds; Coordination reactions; Electrodeposition; Electrospray ionization; Mass spectrometry; Monoterpenes; Olefins; Pyridine; Single crystals; Synthesis (chemical); acetaminosalol; carbon dioxide; carbonic acid; carbonic acid derivative; chloride; chloride ion; chromium; cobalt; cyclohexane oxide; epoxide; limonene; pyridine; solvent; Air stable; Chloride complexes; Chromium III; Co catalysts; Cyclic carbonates; Cycloadditions; Cyclohexenes; Salophen; Single-component catalysts; Synthesised; article; catalysis; catalyst; controlled study; cycloaddition; Diels Alder reaction; electrospray mass spectrometry; ionization; reaction analysis; turnover number; Carbon dioxide

Chemicals/CAS

acetaminosalol, 118-57-0; carbon dioxide, 124-38-9, 58561-67-4; carbonic acid, 3812-32-6, 463-79-6; chloride, 16887-00-6; chromium, 16065-83-1, 7440-47-3, 14092-98-9; cobalt, 7440-48-4; cyclohexane oxide, 286-20-4; limonene, 138-86-3, 5989-27-5; pyridine, 110-86-1

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