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Articles

A Convenient Nitroxyl Radical Catalyst for the Selective Oxidation of Primary and Secondary Alcohols to Aldehydes and Ketones by O_2 and H_2O_2 under Mild Conditions

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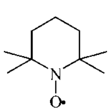
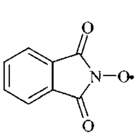
Abstract:

A new macrocyclic tetrafunctional nitroxyl radical, I, developed by Ciba Specialty Chemicals, is a particularly effective catalyst in combination with Mn(II) and Co(II) or Cu(II) nitrates for the selective oxidation of primary and secondary alcohols to the corresponding aldehydes and ketones by air or O_2 under mild conditions (ambient temperature and pressure) or H_2O_2. A distinctive feature of I is the possibility of easy recovery and recycles, due to its low solubility, particularly as ammonium salt, in most organic solvents, which makes it especially useful for practical applications.

Introduction

The oxidation of primary and secondary alcohols to the corresponding aldehydes and ketones is a basic transformation of great interest for general synthetic purposes and for practical applications, and a large variety of oxidising reagents has been utilised successfully. Environmentally reliable catalytic systems in combination with cheap oxidants, such as O_2 or H_2O_2, are desirable for practical purposes. Organic persistent nitroxyl radicals such as TEMPO (2,2,6,6-tetramethylpiperidin-1-oxyl)[1] or nonpersistent radicals such as PINO (phthalimido-*N*-oxyl, generated in situ from *N*-hydroxyphthalimide)[2] have been shown to be particularly effective catalysts for the selective aerobic oxidation of primary and secondary alcohols to the corresponding aldehydes and ketones.


TEMPO


PINO

The use of persistent nitroxyl radicals, such as TEMPO, for the oxidation of alcohols has been largely explored by a variety of procedures.[3] A convenient method, involving a two-phase system (CH_2Cl_2/H_2O), sodium hypochlorite as oxidant and bromide ion as cocatalyst, has been developed by Montanari and co-workers;[4] a modified "Montanari process" has been recently utilised for the production of fine chemicals.[5] Several metal salt complexes were also utilised in combination with TEMPO for the aerobic oxidation of alcohols: cupric salts were effective with benzylic alcohols, while they proved to be less effective with nonbenzylic alcohols;[6] $RuCl_2(PPh_3)_3$ is an efficient catalyst at 100 °C, and a more complex catalyst[7] involving $CuBr{\cdot}Me_2S$, perfluoroalkylated bipyridine, and perfluorooctane−chlorobenzene biphasic solvent system, was utilised at 90 °C.

Recently we have reported in a preliminary communication[9a] that a catalytic system, involving $Mn(NO_3)_2$ in combination with $Co(NO_3)_2$ or $Cu(NO_3)_2$, is particularly effective for the aerobic oxidation of carbonyl compounds under very mild conditions (cyclohexanone is oxidised to adipic acid by oxygen with high selectivity at ambient temperature and pressure), but it is completely inert towards alcohols; it becomes very effective for the aerobic oxidation of primary and secondary alcohols in combination with TEMPO, which however inhibits the further oxidation of aldehydes and ketones. This procedure appears particularly convenient for the synthesis of aldehydes and ketones from the corresponding alcohols, due to the simple and mild

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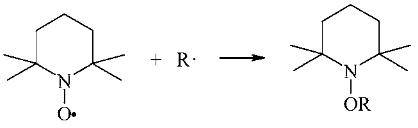
conditions (the oxidation takes place at room temperature and atmospheric pressure of air or O_2), the high selectivity, and the cheap metal salt catalyst.[1]

TEMPO has two important functions: it generates the oxoammonium salt, which is responsible for the alcohol oxidation and inhibits the further oxidation of aldehydes and ketones, which occurs by free-radical chain processes under the same conditions in the absence of TEMPO; this last, being persistent, quickly traps a variety of free radicals (eq 1).

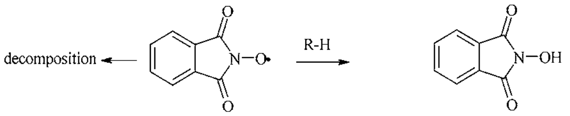


(1)

The advantage of using TEMPO, compared to the aerobic oxidation involving the PINO radical,[2] is related to the general character for the oxidation of benzylic and nonbenzylic primary alcohols to the corresponding aldehydes, while the use of *N*-hydroxyphthalimide, which generates in situ the PINO radical, is limited to benzylic alcohols (with nonbenzylic derivatives the oxidation of primary alcohols leads to carboxylic acids, even at low conversions).

This different behaviour of the two nitroxyl radicals, TEMPO and PINO, was explained[10] by the bond dissociation enthalpies of the O−H bond of the corresponding *N*-hydroxyderivatives (69.9 kcal/mol for the *N*-hydroxy-2,2,6,6-tetramethylpiperidine and 88.1 kcal/mol for the *N*-hydroxyphthalimide). Consequently PINO can induce free-radical chain processes, the selectivity of which is determined by polar, enthalpic, and captodative effects in the hydrogen abstraction step, while TEMPO inhibits such processes.

The advantage of using *N*-hydroxyphthalimide concerns its low solubility in several solvents, which facilitates the recovery and the recycle of the catalyst that are more difficult with TEMPO. PINO, however, is much less stable than TEMPO, and it can decompose in competition with the regeneration of *N*-hydroxyphthalimide, particularly with less reactive substrates (a value of 0.1 s^{-1} for the first-order self-decay of PINO has been determined[10] at 25 °C); a key step in the catalysis by N-hydroxyphthalimide is, in fact, the hydrogen abstraction by the PINO radical (eq 2).

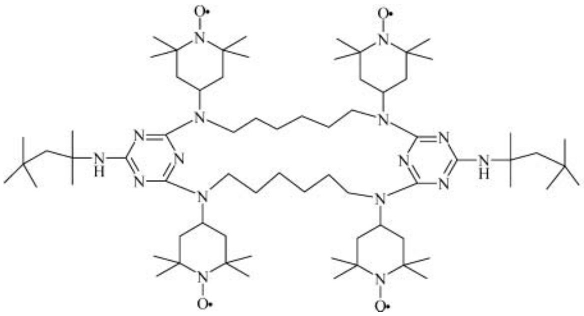


(2)

Recently a Ciba patent[11] has reported a stable and persistent polynitroxyl radical, characterised by low solubility in many organic solvents and therefore suitable to be easily recovered and recycled, making particularly convenient its utilisation as catalyst for the aerobic oxidations of practical interest. Moreover, the presence of amino groups makes the recovery of the catalyst as the ammonium salt still easier, considering that the catalysis is effective only in acidic medium, as it will be discussed later on.

Results and Discussion

The radical of structure **I**, as reported in the Ciba patent, has been investigated as catalyst for the aerobic oxidation of alcohols.

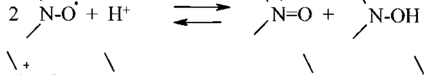


I

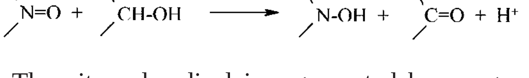
I has been prepared by oxidation of the amine precursor (N−H → N−O•), which is a commercial product, CHIMASSORB 966, sold by Ciba Specialty Chemicals. The oxidation of the amine precursor has been carried out according to the procedures described for the other tetramethylpiperidine derivatives, involving the use of hydrogen peroxides or peracetic acid as oxidants.

The tetranitroxyl radical **I** has a low solubility in most organic solvents, and its use as heterogeneous catalyst for the oxidation of alcohols is effective with sodium hypochlorite,[11] but it gave poor results in combination with Mn(II) and Co(II) or Cu(II) nitrates for the aerobic oxidation because of poor solubility. **I**, however, is soluble in acidic medium, due to the presence of amino groups and is particularly soluble in acetic acid, which is a suitable solvent according to the procedure for the aerobic oxidation catalysed by TEMPO and metal salts. Thus, the nitroxyl radical **I** in combination with Mn(II) and Co(II) or Cu(II) nitrates catalyses the aerobic oxidation of primary and secondary alcohols to the corresponding aldehydes and ketones in acetic acid solution with high selectivity under mild conditions (room temperature and atmospheric pressure of oxygen or air). The results with a variety of alcohols are reported in Table 1. Blank experiments have shown that in the absence of the radical **I** or the metal salts, no substantial oxidation occurs.

The reaction mechanism involves the disproportionation of the nitroxyl radical, catalysed by the acidic medium (eq 3), with the formation of the oxoammonium salt, which is known to be the actual oxidant of the alcohol (eq 4).



(3)



(4)

The nitroxyl radical is regenerated by oxygen and the metal salt catalytic system (eq 5).

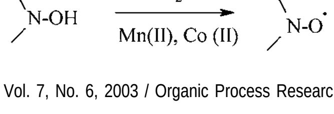


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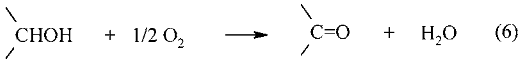
Table 1. **Aerobic oxidation of alcohols to aldehydes and ketones, catalysed by the nitroxyl radical I in combination with Mn(II) and Co(II) nitrates**[a]

alcohol	**I**/ mol %	procedure	reaction time/h	T/°C	yield/ %
$PhCH_2OH$	0.5	A	2	20	99
$PhCH_2OH$	1.2	A	1	20	97
$PhCH_2OH$	2.5	A	0.6	20	98
$PhCH_2OH$[b]	1.2	A	1	20	97
$PhCH_2OH$	2.5	B	2	20	96
$PhCH_2OH$[b]	2.5	B	2	20	93
$PhCH_2OH$	2.5	A	0.3	40	97
$PhCH_2OH$	1.2	A	0.5	40	95
$PhCH_2OH$	0.5	A	1	40	98
$PhCH_2OH$[b]	2.5	C	1	20	99
p-MeO-C_6H_4-CH_2OH	1.2	A	0.8	20	99
p-MeO-C_6H_4-CH_2OH[b]	1.2	A	1	20	94
p-MeO-C_6H_4-CH_2OH	1.2	B	4	20	97
p-MeO-C_6H_4-CH_2OH[b]	2.5	C	1	20	99
m-MeO-C_6H_4-CH_2OH	2.5	B	3	20	95
p-Cl-C_6H_4-CH_2OH	2.5	A	3	20	95
p-Cl-C_6H_4-CH_2OH	2.5	B	3	20	94
p-Cl-C_6H_4-CH_2OH[b]	2.5	C	3	20	94
o-NO_2-C_6H_4-CH_2OH	2.5	B	5	20	92
m-NO_2-C_6H_4-CH_2OH	2.5	B	5	20	94
p-NO_2-C_6H_4-CH_2OH	2.5	A	2	40	98
p-NO_2-C_6H_4-CH_2OH	2.5	B	5	20	97
p-NO_2-C_6H_4-CH_2OH[b]	2.5	C	5	20	96
m-C_6H_4-$(CH_2OH)_2$	2.5	B	4	20	90
Ph-CHOH-CH_3	1.2	A	1	25	96
Ph-CHOH-CH_3[b]	1.2	C	1	20	98
furfuryl alcohol	1.2	B	3	20	95
furfuryl alcohol[b]	1.2	C	1	20	97
2-(hydroxymethyl)pyridine	2.5	A	1.5	40	98
cinnamyl alcohol	1.2	A	2	40	96
2-methyl-pentanol	2.5	A	1	25	97
2-nonanol	2.5	A	1	25	93
cyclohexanol	2.5	A	1.5	40	99
cyclooctanol	2.5	A	2	40	92
cyclododecanol	2.5	A	2.5	40	94

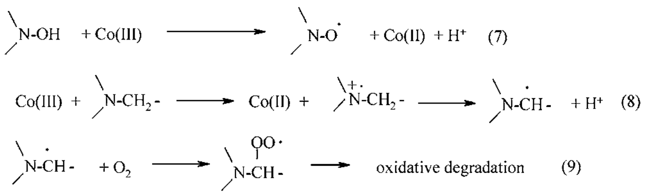
[a] Procedure A with oxygen, Procedure B with air. [b] Cu(II) nitrate instead of Co(II) nitrate.

The overall stoichiometry of the reaction is given by eq 6.



On the other hand, the further aerobic free-radical oxidation of aldehydes and ketones, which easily occurs under the same conditions in the absence of **I**, is completely inhibited by the presence of **I**, which quickly traps intermediate free radicals (eq 1). The oxidation rate is not only affected by the temperature but also by the concentration of the catalyst. This behaviour is useful because it allows regulating the oxidation rate by the concentration of the nitroxyl, which is more stable at lower reaction temperature. The amount of the catalyst is relatively unimportant, as concerns its cost contribution on the overall process, because it can be easily recovered and recycled.

Attempts to recover and recycle the catalyst from acetic acid solution gave only moderate results; the recovery of the catalyst is relatively simple by evaporating acetic acid and utilising a suitable solvent, such as methyl-*tert*-butyl ether or ethyl acetate, which dissolves the reaction product (aldehydes and ketones), while the insoluble catalyst is easily recovered. However, the recovered catalyst has partially lost the activity. Our hypothesis is that the catalyst undergoes a partial degradation during the aerobic oxidation; in particular, the methylene groups in the α-position to the nitrogen are very sensitive to oxidation. It is likely the Co(III) and the Mn(III) salts intermediates of the oxidation contribute to regenerate the nitroxyl radical from hydroxylamine derivative (eq 7), formed in eqs 3 and 4. Unfortunately, they may also oxidise the amino groups by an electron-transfer process (eqs 8 and 9), leading to the oxidative degradation of the catalyst.

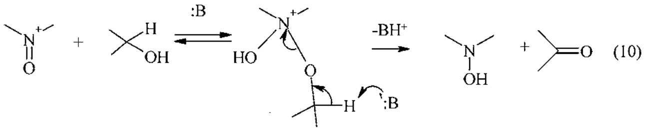


To overcome this possible oxidative degradation we thought it right to add a strong acid to the reaction medium to protonate more extensively the amino groups of the catalyst; the protonated amino groups are quite inert towards the electron-transfer oxidation (eq 8) and the subsequent oxidative degradation (eq 9).

Indeed, the overall processes was considerably improved by carrying out the oxidation in acetic acid in the presence of a suitable amount of p-toluenesulphonic acid:

(i)The higher acidity favours the disproportionation of the nitroxyl radical (eq 3), making the overall process faster. However, an excess of acid must be avoided to prevent the esterification of the alcohol substrate in esters of the acetic acid, the products which are inert toward the aerobic oxidation.

(ii) The protonation of the amino groups of the catalyst increases the electrophilic character of the oxoammonium salt and therefore increases the rate of the addition of the alcohol (eq 10), which is the rate-determining step of the oxidation.[12]



Recent results,[12] concerning the oxidation of substituted benzyl alcohols catalysed by nitroxyl radicals, show that electron-donating groups activate and electron-withdrawing groups deactivate the reaction in agreement with the formation of the complex between the oxoammonium salt and the alcohol as the rate-determining step.

(iii) The most relevant effects of increasing the acidity, however, are related to the fact that the oxidative degradation of the catalyst is practically inhibited and the recovery and recycle of the protonated catalyst with substantially unchanged activity is much easier.

Several groups have addressed the problem of the recycling nitroxyl radicals, which are rather expensive, by

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(12) Minisci, F.; Recupero, F.; Cecchetto, A.; Gambarotti, C.; Punta, C.; Faletti, R.; Paganelli, R.; Pedulli, G. F. *Eur. J. Org. Chem.* Submitted.

designing heterogeneous variants of TEMPO.[13−17] In all these cases, however, the catalysts are active by using NaOCl as oxidant, while they appear rather inactive for the aerobic oxidation. Thus, the nitroxyl radical **I** appears to be, as far as we know, the only catalyst suitable for the aerobic oxidation of alcohols under mild conditions and for an easy recycling.

In general, if the catalyst are soluble, it is not easy to recover them from the final reaction mixture. Otherwise, in the aerobic oxidation, if the catalyst is not in solution, the reaction does not proceed as well as in homogeneous conditions. The new nitroxyl radical **I** combines both the solubility in the reaction solvent (acetic acid) and the feasibility to recover the catalyst from the reaction mixture after removing the solvent.

The catalytic system can be utilised for the aerobic oxidation of cycloalkanes to dicarboxylic acids, which are compounds of particular industrial interest; in particular adipic, suberic and 1,12-dodecandioc acids (from cyclohexane, cyclooctane, and cyclododecane) are valuable starting materials for the synthesis of polyamides and other polymeric materials. A general industrial process for the production of dicarboxylic acids involves the autoxidation of cycloalkanes to mixtures of cycloalkanols and cycloalkanones at low conversion to have high selectivity, followed by the oxidation of those mixtures with nitric acid, which has, however, some drawbacks, resulting mainly from the corrosivity of nitric acid, and environmental problems, due to the formation of nitrogen oxides.

An alternative approach is provided by the aerobic oxidation of the mixtures of cycloalkanols and cycloalkanones catalysed by the nitroxyl radical developed by Ciba Specialty Chemicals (in combination with Mn(II) and Co(II), or Cu(II) nitrates) according to the procedure reported in this work, the separation of the nitroxyl catalyst **I** and the final aerobic oxidation of the pure cycloalkanones to dicarboxylic acids by the same metal salt catalytic system (Mn(II) and Co(II) or Cu(II) nitrates) under very mild conditions (room temperature and atmospheric pressure of oxygen), following the procedure recently reported by this research group.[9]

After oxygen, hydrogen peroxide is a particularly convenient oxidant of organic compounds for several reasons:

(i) high oxidation potential ($E^\circ = 1.77$V),

(ii) formation of water as reduction product, thus minimising the environmental drawbacks which are often involved with other oxidants, including a cheap oxidant such as sodium hypochlorite,

(iii) the low cost,

(iv) the low molecular mass.

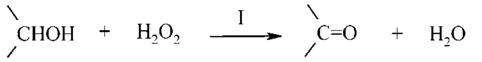
Table 2. **Oxidation of alcohols to aldehydes and ketones by H_2O_2, catalysed by the nitroxyl radical I in combination with Mn(II) and Co(II) nitrates**

alcohol	**I**/mol %	reaction time/h	*T*/°C	yield/%
$PhCH_2OH$	7.5	8	25	97
$PhCH_2OH$	5.0	12	25	98
$PhCH_2OH$	2.5	16	25	96
$PhCH_2OH$	1.2	2	50	93
$PhCH_2OH$[a]	2.5	8	25	-
$PhCH_2OH$[b]	2.5	8	25	-
p-MeO-C_6H_4-CH_2OH	1.2	1.5	50	96
p-Cl-C_6H_4-CH_2OH	1.2	2	50	93
p-Me-C_6H_4-CH_2OH	1.2	2	50	98
p-NO_2-C_6H_4-CH_2OH	2.5	2	50	94
2-nonanol	2.5	8	50	98
cyclohexanol	2.5	5	50	96
1-heptanol	2.5	6	50	92

[a] By using the couple Cu(II)/ Mn(II) nitrates. [b] By using the couple Cu(II)/ Co(II) nitrates.

Compared to oxygen, the use of aqueous hydrogen peroxide is especially beneficial, since its process requires relatively simple reactor design of the production plants, which can result in convenience for the production of moderate amounts of fine chemicals. However a significant drawback is represented by the high activation energy required for the oxidation of many organic compounds by hydrogen peroxide so that catalysis is often necessary for the reaction to occur.

We have investigated the possibility of catalysing the hydrogen peroxide oxidation of alcohols by **I** in combination with the same metal salts (Mn(II) and Co(II) or Cu(II) nitrates) utilised for the aerobic oxidation. The results (Table 2) show that the Mn(II)/Co(II) nitrates couple is an effective catalyst in combination with **I** for the oxidation of alcohols by hydrogen peroxide (eq 11), while the couples Mn(II)/ Cu(II) or Co(II)/Cu(II) give poor results. It appears that the copper salt catalyses the decomposition of H_2O_2 without involving the oxidation of the alcohol.



$$>CHOH + H_2O_2 \xrightarrow{I} >C{=}O + H_2O \quad (11)$$

In the absence of the nitroxyl radical **I** or of the Mn(II)/ Co(II) nitrates couple no substantial oxidation occurs, suggesting that also with hydrogen peroxide the actual oxidant of the alcohol is the oxoammonium salt (eq 4), which is continuously regenerated by the combination of hydrogen peroxide and the metal salt catalysis. Also in this case the oxidation rate is affected by the concentration of **I**. The nitroxyl catalyst **I** can be recovered and recycled from acetic acid, without loss of activity, as described for the aerobic oxidation processes.

Experimental Section

All the starting materials and catalysts were commercial products and used without further treatment. The nitroxyl radical **I** was prepared according to the Ciba patent.[11]

General Procedures for the Aerobic Oxidation of Alcohols. *Procedure A.* A solution of 6 mmol of the alcohol

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(13) Fey , T.; Fischer, H.; Bachmann, S.; Albert, K.; Bolm, C. *J. Org. Chem.* **2001**, *66*, 8154.

(14) Brunel, D.; Lentz, P.; Sutra, P.; Deroide, B.; Fajula, F.; Nagy, J. B. *Stud. Surf. Sci. Catal.* **1999**, *125*, 237.

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(16) Ciriminna, R.; Blum, J.; Avnir, D.; Pagliaro, M. *Chem. Commun.* **2000**, 1441.

(17) Dijksman, A.; Arends, I. W. C. E.; Sheldon, R. A. *Chem. Commun.* **2000**, 271−272; *Synlett* **2001**, 102−104.

in 35 mL of acetic acid, each specific amount of the nitroxyl catalyst **I** reported in Table 1, and 0.25 mol % of Mn(II) and Co(II) or Cu(II) nitrates, was stirred at the specified temperature under oxygen atmosphere at ambient pressure for the times reported in Table 1. The acetic acid was then evaporated at ca. 50 Torr, and the reaction products were extracted with methyl-*tert*-butyl ether (MTBE) or with ethyl acetate (the nitroxyl radical **I** is insoluble in these solvents and can be recovered and recycled). All the reaction products are known, and GC and GC−MS analyses were performed. The products were identified through a comparison of the analytical data with those of authentic samples and the yields were estimated by GC with the internal standard technique. In few cases the reaction products were isolated in experiments on larger scale (60 mmol of alcohol) simply by evaporating the solvent and obtaining a purity > 98%. The recovered catalyst **I** was recycled with partial loss of activity.

Procedure B. The experiment was carried out as for the procedure A, except that the stirred reaction solution was maintained under air at atmospheric pressure.

Procedure C. A solution of 92.6 mmol of the alcohol, 2.25 mmol of the nitroxyl radical catalyst **I**, 4.5 mmol of *p*-toluenesulphonic acid, 1.8 mmol of Mn(II) nitrate and 1.8 mmol of Co(II) nitrate, in 100 mL of acetic acid was stirred at room temperature for 3 h under air at atmospheric pressure. The analysis, the isolation of the reaction products, and the recovery of the catalyst were carried out according to procedure A. The nitroxyl catalyst has been recycled for four times without significant loss of activity, as *p*-toluenesulphonic acid salt. No additional *p*-toluenesulphonic acid was required for the recycles. After the catalyst **I** was recycled four times, the yield of benzaldehyde with respect to the benzyl alcohol was 95%.

General Procedure for the Oxidation of Alcohols by Hydrogen Peroxide. A solution of the alcohol (6 mmol), 30% aqueous H_2O_2 (15 mmol) in 35 mL of acetic acid, the amount of the nitroxyl catalyst **I** reported in Table 2 and 0.25 mol % of Mn(II) and Co(II) nitrates was stirred at the specified temperature for the time reported in Table 2. The workup, analysis, and recovery of the catalyst **I** were carried out according to procedure A. The catalyst **I** was recycled five times in the oxidation of benzyl alcohol without significant loss of activity: the oxidation yield of the benzyl alcohol with respect to benzaldehyde was 96%

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