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Recent developments in non-enzymatic catalytic oxidative kinetic resolution of secondary alcohols

Hélène Pellissier

Aix Marseille Univ, CNRS, Centrale Marseille, iSm2, Marseille, France

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ABSTRACT

The goal of this review is to collect the recent developments in non-enzymatic catalytic oxidative kinetic resolutions of secondary alcohols reported since the beginning of 2011. It is divided into four sections, dealing successively with manganese-catalysed oxidative kinetic resolutions of secondary alcohols, palladium-catalysed oxidative kinetic resolutions of secondary alcohols, oxidative kinetic resolutions of secondary alcohols catalysed by other metals and organocatalysed oxidative kinetic resolutions of secondary alcohols.

Contents

1. Introduction

Among a wide number of methodologies for preparing chiral compounds,[1] the kinetic resolution still constitutes the most efficient and practical approach especially in industry.[2] Along with widely employed kinetic resolutions based on the use of enzymes, transition metal-mediated and more recently developed organocatalysed kinetic resolutions have gained popularity over the last few decades due to the progress made in the discovery of novel chiral catalysts. So far, many procedures based on catalytic non-enzymatic kinetic resolution have been developed, providing high enantioselectivities and conversions. The first one was reported by Fajans and Bredig in 1908, dealing with the decarboxylation of camphor-3-carboxylic acid performed with chiral alkaloids.[3] Then in 1981, a milestone occurred when Sharpless reported the powerful kinetic resolution of allylic alcohols performed with $Ti(OiPr)_4$/tartrate ligand/*tert*-butylhydroperoxide as reagent,[4] which has been widely developed ever since.[5] Indeed, many reactions based on a kinetic resolution strategy have been achieved

E-mail address: h.pellissier@univ-amu.fr.

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Abbreviations

Ar	aryl
Bn	benzyl
Boc	*tert*-butoxycarbonyl
Bz	benzoyl
Cbz	benzyloxycarbonyl
cod	cyclooctadiene
Cy	cyclohexyl
ee	enantiomeric excess
MS	molecular sieves
Naph	naphthyl
nbd	norbornadiene
NBS	*N*-bromosuccinimide
NEDA	1,2-di(1-naphthyl)-1,2-ethanediamine
Pent	pentyl
PIFA	phenyl iodonium bis(trifluoroacetate)
r.t.	room temperature
s	selectivity factor
salen	*N,N′*-ethylenebis(salicylideneiminato)
TBHP	*tert*-butyl hydroperoxide
TBS	*tert*-butyldimethylsilyl
TCCA	trichloroisocyanuric acid
Tf	trifluoromethanesulfonyl
Tol	tolyl

with high efficiency,[6] such as the oxidative kinetic resolution of alcohols,[6,7] allowing a practical approach to these chiral pivotal intermediates in asymmetric synthesis, pharmaceutical, agrochemical and fine chemical industries.[8] This review collects the recent developments in non-enzymatic catalytic oxidative kinetic resolution of secondary alcohols reported since the beginning of 2011 since, to my knowledge, this field was previously reviewed in 2011.[6a] It must be noted that the special area of non-enzymatic and not especially oxidative catalytic kinetic resolution of diols was the object of a digest paper reported by Zheng in 2018 including only two references ≥2011,[6d] and that of asymmetric salen manganese-catalysed oxidative kinetic resolutions of alcohols was reviewed in 2017 by Ahmad et al. containing only eight references ≥2011 among which seven ≤2014.[9] Moreover in 2016, Petersen reported a review on general chiral Brønsted acid-catalysed kinetic resolutions but it included no example of oxidative kinetic resolution of secondary alcohols.[10] The present review includes oxidative kinetic resolutions of secondary alcohols catalysed by all kinds of chiral catalysts, spanning from chiral complexes derived from all types of metals to chiral organocatalysts. It is divided into four sections according to the nature of the catalysts used to promote these reactions. Indeed, the four sections deal successively with manganese-catalysed reactions, palladium-catalysed reactions, reactions catalysed by other metals and organocatalysed reactions. It must be noted that special kinetic resolutions, such as parallel kinetic resolutions,[11] stoichiometric kinetic resolutions,[12] and dynamic kinetic resolutions[13] are not included in this review.

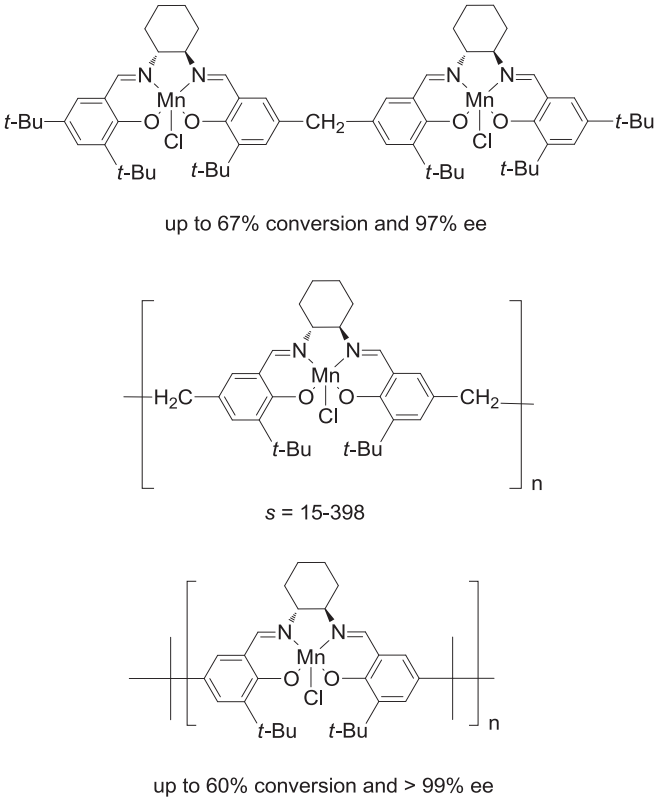
2. Manganese-catalysed oxidative kinetic resolutions of secondary alcohols

2.1. Using PhI(OAc)$_2$ as oxidant

Many chiral salen manganese complexes have been successfully applied in the presence of PhI(OAc)$_2$ as oxidant to promote oxidative kinetic resolutions of secondary alcohols,[9] since the first efficient report by Sun et al., in 2003, allowing enantioselectivities of up to 88% ee to be achieved by using water as solvent.[14] Ever since, a number of different types of chiral salen manganese complexes have been successfully investigated by several groups, spanning from monomeric complexes to dimeric,[15] macrocyclic, polymeric,[16] and supported ones.[17] Several of these catalysts are depicted in Scheme 1.

In 2013, Abdi et al. reported the synthesis of novel macrocyclic chiral salen manganese complexes to be evaluated in oxidative kinetic resolutions of racemic secondary alcohols having different steric environment.[18] The process was performed at 15 °C with PhI(OAc)$_2$ as oxidant and KBr as additive in a mixture of dichloromethane and water as biphasic solvent. It was catalysed by optimal macrocyclic complex **1** at 2 mol% of catalyst loading, which provided variable enantioselectivities (7–99% ee) combined with generally good conversions (53–64%), as shown in Scheme 2. Notably, the best enantioselectivities (93–99% ee) were achieved in the oxidative kinetic resolution of aliphatic alcohols while secondary benzylic alcohols provided lower enantioselectivities (7–84% ee). It must be noted that catalyst **1** was easily recovered through simple precipitation from hexane and could be recycled up to seven times without loss in both enantioselectivity and activity.

Earlier in 2011, Yin and Tan synthesised novel ionic liquid-bridged chiral dimeric salen manganese complexes to be investigated in comparable reactions.[19] Indeed, *N,N*-dialkylimidazolium ionic liquids were used to bridge chiral dimeric salen



Scheme 1. Recent examples of dimeric and polymeric salen complexes employed in oxidative kinetic resolutions of secondary alcohols by using PhI(OAc)$_2$ as oxidant.