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## Biological Significance of Keto Esters

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### Annotation:

Homogeneous asymmetric catalytic hydrogenation of prochiral carbonyl compounds is of great interest, since the resulting optically active secondary alcohols are valuable chiral blocks for fine organic synthesis. The substrates well studied in this reaction include  $\alpha$ - and  $\beta$ -ketoesters and their functional derivatives. Being less reactive,  $\gamma$ -ketoesters as prochiral substrates have not been studied practically.

**Keywords:** asymmetric hydrogenation, binuclear complexes, catalysis

In this regard, an important aspect is the search for effective catalytic systems for the reaction of asymmetric hydrogenation of  $\gamma$ -ketoesters based on the study of the regularities and mechanism of the reaction by kinetic and spectral methods. In addition, there is no general method in the literature for the synthesis of  $\gamma$ -alkyl- and  $\gamma$ -aryl-substituted lactones in enantiomerically pure form, which can be obtained in one stage by catalytic hydrogenation of  $\gamma$ -ketoesters. The possibility of recycling expensive chiral catalysts is also an important problem that has not yet been solved to date. This approach allows, on the one hand, reducing the cost of the process of obtaining  $\gamma$ -lactones and, on the other hand, meets the principles of "green" chemistry.

For the first time, a systematic study of homogeneous asymmetric hydrogenation of  $\gamma$ -ketoesters using ruthenium complexes with chiral bisphosphine ligands as catalysts was carried out. Highly efficient Ru(II)- and Ru(III)-containing catalytic systems for homogeneous asymmetric hydrogenation of esters of  $\gamma$ -ketocarboxylic acids with various substituents at the  $\gamma$ -carbonyl carbon atom have been created. It has been established that mineral acid additives lead to a sharp increase in the rate of asymmetric catalytic hydrogenation of  $\gamma$ -ketoesters with high enantiomeric purity of the resulting  $\gamma$ -lactones. The kinetics and mechanism of asymmetric catalytic hydrogenation of  $\gamma$ -ketoesters are investigated using isotopic and spectral methods; it is assumed that the catalytically active intermediates are binuclear Ru(II) hydride complexes formed as a result of H<sub>2</sub> heterolysis. A universal preparative method has been developed for obtaining enantiomerically pure  $\gamma$ -alkyl- and  $\gamma$ -aryl-substituted lactones based on the reaction of asymmetric hydrogenation of  $\gamma$ -ketoesters in the presence of catalytic systems [RuCl(BINAP)] - HCl and RuCl<sub>2</sub>(BINAP)-HCl. Using the example of asymmetric hydrogenation of methylvalerate, a reaction with repeated recycling of a metal-complex catalyst deposited on a solid organic polyelectrolyte was carried out for the first time.

A number of reviews have been devoted to the asymmetric catalytic hydrogenation of prochiral substrates, including ketoesters. In contrast to asymmetric hydrogenation using heterogeneous catalysts, when the catalysis is carried out surface complexes of metals such as nickel or platinum, the achievements of recent years in the field of homogeneous asymmetric hydrogenation are associated with the use of chiral complexes of ruthenium and rhodium. It was for outstanding achievements in this field that in 2001 W. Knowles and R. Noyori were awarded the Nobel Prize in Chemistry. As for the homogeneous asymmetric hydrogenation of ketoesters with catalysts based on other metals, such information is practically absent in the literature. In this review, we have summarized the literature data related to the reactions of homogeneous asymmetric hydrogenation of  $\alpha$ -, ( $\beta$ - and  $\gamma$ -ketoesters occurring with high enantioselectivity (at least 80% of it). Its lower values are given only for comparison. The review considers the

This conference will be organized in the USA on 10 the of October and the final proceeding will be provided on the 24th of October as a whole.

formation of effective metal-complex catalysts, the influence of the nature of chiral and achiral ligands, as well as reaction conditions on the conversion of initial ketoesters and enantioselectivity of asymmetric hydrogenation.

Highly efficient Ru(II)- and Ni(W)-containing catalytic systems for homogeneous asymmetric hydrogenation of esters of  $\gamma$ -keto-carboxylic acids with various substituents at the  $\gamma$ -carbonyl carbon atom have been created. Ru(COD)(MA) systems showed the highest activity and enantioselectivity in situ<sup>2</sup> - BINAP - HC1, [RUC12(DMOD)]<sup>2</sup> - BINAP - HC1 and RuCl<sub>3</sub>-BINAP-HC1, the latter system being used for the first time in asymmetric catalytic hydrogenation.

The kinetics of the reaction of asymmetric hydrogenation of methylevalinate - the simplest  $\alpha$ -ketoester - with hydrogen and deuterium in the presence of a catalytic Ru(COD)(MA) system has been studied<sup>2</sup> - BINAP - HC1. It is found that the kinetic order for H<sub>2</sub> and the catalyst is the first, and for the substrate and HC1 it changes from the first to zero with an increase in their initial concentrations. Kinetic isotopic effect is absent or insignificant.

Based on kinetic data and spectral study of metal-containing intermediates, a mechanism of catalytic hydrogenation of  $\alpha$ -ketoesters is proposed, including reversible stages of coordination of H<sub>2</sub> molecules by binuclear complexes of Ru(II), heterolysis of coordinated H<sub>2</sub> with the formation of active hydride intermediates and an irreversible stage of hydride transfer to the acid-activated keto group of the initial substrate.

A universal preparative method for obtaining enantiomerically pure  $\gamma$ -alkyl- and  $\gamma$ -aryl-substituted lactones based on an asymmetric catalytic hydrogenation reaction in the presence of Ru(COD)(MA) systems has been developed<sup>2</sup> - BINAP - HC1 hRUC13-BINAP-HC1.

Solid organic polyelectrolyte – poly (diallyldimethylammonium) was used for the first time in homogeneous metal-complex catalysis in order to recycle the catalyst and its repeated usehexafluorophosphate. It is shown that the activity of the catalyst does not decrease during three cycles, while the enantio selectivity remains high (98-99%) during at least five cycles.

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