

The aim of this project is to thoroughly investigate the decomposition path of ruthenium catalysts in olefin metathesis. We plan to explore the possible reaction pathways of commonly used and novel metathesis catalysts using computational methods.

The olefin metathesis is one of the most useful tools in modern organic synthesis. In this process, to precisely create new C = C double bonds, the catalysts based on transition metals are needed. The most popular group of these complexes are based on ruthenium, and are called Grubbs and Hoveyda-Grubbs catalysts. However, despite the great utility of this reaction and the stability of the catalysts, it has been found that undesirable reactions occur during this process, such as catalyst degradation. In spite of the fact that the decomposition of ruthenium catalysts has been repeatedly discussed in the literature, there are no clearly defined mechanisms of its course. Due to the high popularity of metathesis reactions and wide availability of the catalysts, need for solving problems related to the side reactions of metathesis appears to be evident. Scientists have noted before, that except expected reaction products, the undesired products of metathesis and olefin isomerization products are also formed. In addition, the turnover number of the catalysts used in the reaction decreases, which is probably related to degradation of catalysts during metathesis.

The presented project will be implemented using computational techniques - modern density functional theory (DFT) methods. We will build new complexes based on the crystal structures of known ruthenium catalysts. Next, we will calculate the optimal geometries and electronic properties of the created ruthenium complexes as well as the Gibbs free energy of individual reaction stages.

Understanding the mechanisms of decomposition will allow us to explain the occurring side reactions, as well as to uncover the weak points of currently used catalysts. The results obtained in this research will help fully assess the possible degradation pathways of ruthenium catalysts and develop an overall methodology that will be used in the future to design new catalysts. Moreover, the acquired knowledge will allow design of new, more efficient catalysts, which will be more resistant to degradation reactions.