Biochemical Production Using DNA Catalysts

Tim Coogan (University of Massachusetts), Eric Hall (Hoover High School), Wenya Lu & Dr. Keith Woo (Iowa State University)

Introduction

Traditionally, chemicals used in industry have been produced using petroleum-based starting materials. The techniques used in doing this have long been established and the processes are generally very efficient. Unfortunately, we have a finite supply of petroleum which to manufacture all of the chemicals industry requires. For this reason, research has begun which may lead to the discovery of a biorenewable source (such as cellulose and related materials) that could be used to create the most commonly used industrial chemicals.

Additionally, research has also started into the process of transforming these common biorenewable starting materials into chemicals of industrial importance. The overarching goal of this research is to discover a biocatalyst which will convert glucose into an intermediates chemical, and couple it with a chemical catalyst which will convert the intermediate into an industrially-significant compound. (CBIRC, 2008)

The focus of our summer research corresponds to the second portion of the CBIRC research goal. Our goal is to create and optimize a DNA-based chemical catalyst which can be used to facilitate chemical transformations. (Woo, 2008)

Background & Methods

The model reaction used was the Mesozioki-Heck reaction, as its mechanism is well-known and understood. (Figure 2) This reaction begins with an unsaturated halide and an alkene and uses a palladium catalyst to produce a substituted alkene. Our experiment called for the addition of a short (40 bp) single-stranded DNA oligo to the alkyl halide reactant and a biotin marker to be added to the alkene (the biotin allows for monitoring of product formation). Previous work in the Woo lab has shown that this reaction system works well when an additional Pd catalyst is present, however substantially less activity is seen when the DNA-bound alkyl halides are combined with the alkene without the Pd catalyst. In addition, the DNA base sequence is thought to have a significant effect on the efficiency of the reaction.

Our work in this summer focused on optimizing the selection process used for determining which DNA sequences most effectively catalyze the Mesozioki-Heck reaction. This process (referred to as SELEX (Figure 1) - Systematic Evolution of Ligands by Exponential Enrichment) has narrowed the DNA pool from 10^14 sequences down to only fourteen. When the reaction substrates are bound to one of these fourteen sequences a substantial increase in catalytic activity has been seen. Specifically, we tested the fourteen unique DNA sequences in a Mesozioki-Heck reaction using TC-1-1 and TC-1-4 as the substrates. (Figure 3) We did this in an attempt to confirm the success of the model reaction while having the DNA bound to our reaction of interest. As a comparison, we also ran the reaction using the same substrates, but without the DNA bound to them. Both were done with a palladium acetate catalyst present.

Literature Cited


Acknowledgements

I would like to thank Wenya Lu and Dr. Keith Woo for allowing me to learn so much from them this summer. I also appreciate the expertise of my lab co-worker, Tim Coogan. As always, I appreciate the support of Dr. Ashah Lechem-Ackerman. Thanks also to Craig Walker for his help in preparation for our workshop experiences. Lastly, thanks to CBIRC, the Iowa Energy Center, and the National Science Foundation (Grant No. EE0-0813570) for funding my research experience.