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Yang group develops new mechanism for metal carbene radical cross coupling

In an effort to open the door to new and useful products, chemistry researchers are on the continual lookout for processes that unlock important molecules and the bonds that can put them together. Such is the case for UC Santa Barbara chemistry professor Yang Yang, who builds his research around discovering novel biocatalytic methods, processes that facilitate chemical reactions with biocatalysts from evolved natural proteins.

Published in the journal Nature Catalysis, Yang's research group and collaborators at the University of Pittsburgh and Florida State University have developed a new type of reaction, one that employs a mechanism that was until now unknown to both native enzymatic and synthetic chemistry.

"The goal was a new class of metal-carbene chemistry for carbon-carbon bond formation," Yang said. "This project is exciting because the field of transitional metal carbene chemistry has been developing for several decades, and yet we are able to come up with an entirely new mechanism for metal carbene-radical cross-coupling that is potentially general and useful."

Indeed, according to Yang, transition metal carbene chemistry — chemistry that involves the element carbon and the metals that exist in the middle "block" of the

periodic table — has a long history.

“It has many important applications, and mechanistically, it’s important because transition metal carbenes are very reactive intermediates,” he said. “Intermediates” are highly reactive molecular species that are produced during the process of a reaction for the purpose of enabling the next step in the reaction. Though fleeting in their existence, the reaction pivots around their presence.

Key to this reaction, according to the researchers, is the directed evolution of the metalloprotein catalyst, which is an engineered enzyme that has within its structure metal ions, in this case iron. It produces the iron carbene intermediate and the environment with which to control the highly reactive iron radical intermediate generated by the photocatalytic cycle and complete the “proton transfer” step that is fundamental to many organic chemistry reactions.

“If we didn’t work with this particular metalloprotein, this chemistry would essentially remain unknown,” Yang said.

This research, specifically the integration of photochemistry and metalloenzyme catalysis in a cooperative process, pushes the boundaries of transition metal carbene chemistry, according to Yang. This method for carbon-carbon bond forming reactions allows researchers to create molecules that have multiple stereogenic centers, which are fundamental to the structure and function of the often complex three-dimensional molecules found in products such as pharmaceuticals and agrochemicals.

“We think we have developed a fairly general type of carbon-carbon bond-forming reaction, so we’re generalizing this method to make a number of useful compounds.”

Research in this paper was also conducted by Huanan Wang (lead author), Chongtao Li and Xiao-Wang Chen in the Yang Lab at UCSB; Professor Peng Liu and Binh Khanh Mai at the University of Pittsburgh; and Rachel Weiss and Professor Bryan Kudisch at Florida State University.

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