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Enzymes are biological catalysts which greatly accelerate the rates of chemical reactions, oftentimes by many orders of magnitude over the uncatalyzed reaction. The remarkable catalytic rate enhancement afforded by enzymes derives ultimately from the structure and chemical properties of the enzyme active sites, which allow enzymes to selectively bind to their substrates and to stabilize high energy chemical species and unstable intermediates along the reaction coordinate. To enhance their catalytic ability, many of the chemical functionality and reactivity that are not accessible by the twenty canonical amino acids and, hence, coenzymes serve to greatly enhance the diversity of chemical reactions that can be mediated by enzymes. The work described in this dissertation focuses on mechanistic studies of two enzymes that use common coenzymes in unusual ways. In the first section of this work, studies will focus on the type II isopentenyl diphosphate:dimethylallyl diphosphate isomerase (IDI-2), an essential enzyme in isoprenoid biosynthesis. The IDI-2 catalyzed reaction involves no net redox change, raising questions as to the role of the flavin in the chemical mechanism. The chemical mechanism of IDI-2 will be interrogated with a combination of spectroscopic studies and biochemical techniques. Our studies suggest that the flavin coenzyme of IDI-2 may employ a novel mode of flavin-dependent catalysis involving acid/base chemistry. In the second section of this dissertation, attention will be directed to ACCD, an enzyme that plays a role in regulating the production of the potent plant hormone, ethylene. ACCD is a pyridoxal-5P-phosphate (PLP)-dependent enzyme that catalyzes a C-C bond cleavage event that is unique among the catalytic cycles of PLP-dependent enzymes. Altogether, our mechanistic studies of IDI-2 and ACCD help to illustrate the catalytic diversity of common coenzymes, and demonstrate that some enzymes have evolved to exploit readily available coenzymes for atypical reactions.

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