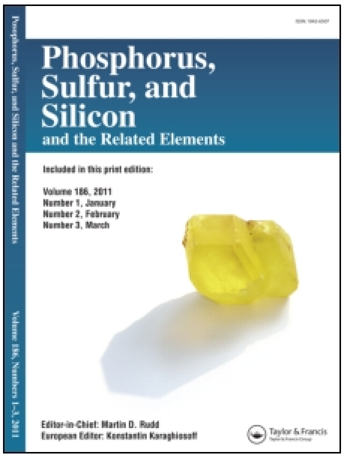

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Supramolecular Catalysis of Phosphoryl Transfer

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Supramolecular Catalysis of Phosphoryl Transfer

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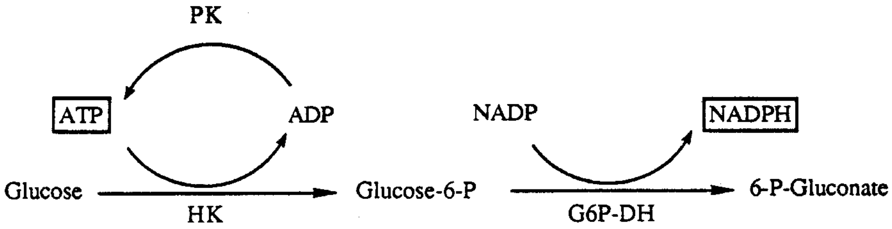
Reactivity and catalysis represent major features of the functional properties of supramolecular systems. Molecular receptors bearing appropriate functional groups may bind selectively to a substrate, react with it, and release the products. Supramolecular reactivity and catalysis thus involve two main steps: recognition of the substrate followed by transformation of the bound species into products. The design of efficient and selective molecular catalysts may give mechanistic insight into the elementary steps of catalysis, provide new types of chemical reagents, and produce models of reactions effected by enzymes that reveal factors contributing to enzymatic catalysis.

Bond cleavage reactions have been extensively studied in this respect. A further step lies in the design of systems capable of inducing bond formation, which would thus effect synthetic reactions as compared to degradative ones. In order to realize "bond-making" rather than "bond-breaking" processes, the presence of several binding and reactive groups is essential. Such is the case for coreceptor molecules in which subunits may cooperate for binding and transformation of the substrates; as a consequence, they should be able to perform co-catalysis by bringing together substrate(s) and cofactor(s) and mediating reactions between them within the supramolecular complex.

These two opposite aspects of chemical reactivity and catalysis have been realized in the supramolecular catalysis of phosphoryl transfer by macrocyclic polyamines: – bond cleavage in ATP hydrolysis; – bond formation in pyrophosphate synthesis, ATP synthesis and substrate phosphorylation. These protoenzymatic processes are respectively of proto-ATPase and proto-kinase nature.

Finally, the coupling of an "artificial enzyme" to a natural enzymatic system has been realized. ATP generated in situ from ADP by the macrocyclic protokinase is used up in a sequence of two enzymatic reactions to produce NADPH along the process:



PK: Macrocyclic protokinase
HK: Hexokinase
G6P-DH: Glucose-6-phosphate dehydrogenase.