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Perspective on Cryochemistry

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Perspective

It is counterintuitive that chemical reactions can be accelerated by freezing, but this amazing phenomenon was discovered as early as the 1960s. In frozen systems, the increase in reaction rate is caused by various mechanisms and the freeze concentration effect is the main reason for the observed acceleration. Some accelerated reactions have great application value in the chemistry synthesis and environmental fields; at the same time, certain reactions accelerated at low temperature during the storage of food, medicine, and biological products should cause concern. The study of reactions accelerated by freezing will overturn common sense and provide a new strategy for researchers in the chemistry field. In this review, we mainly introduce various mechanisms for accelerating reactions induced by freezing and summarize a variety of accelerated cryochemical reactions and their applications.

Early attention to frozen systems was mainly based on the practical problem of food storage in refrigerators. In recent years, scientists have paid more attention to cryosphere atmospheric chemistry and atmospheric-related freezing reactions on polar regions and utilizing special low-temperature environments to develop more ways to deal with pollutants. At the same time, with the development of biopharmaceuticals and reagents, a series of studies have been carried out on their stable and continuous storage as frozen systems. However, in the field of chemistry, low temperatures are often not the preferred experimental conditions. In a frozen state, the rate of chemical reactions is decreased or even inhibited. The phenomenon that the reaction rate increases when the temperature decreases is counterintuitive, but some reactions in solution can be accelerated by freezing. This phenomenon has been described since the 1960s.

Although the mechanism of reaction acceleration in a frozen system is not fully comprehended, the freeze concentration effect is considered the main factor in many cases. Freeze-concentrated solution is seen as a reaction field. The freeze concentration effect is the result of the conversion of water into relatively high purity ice crystals during freezing. Given ice is intolerant to impurities, and foreign atoms cannot enter ice lattice, all non-aqueous components escape to the freeze-concentrated solution and are concentrated in a reduced liquid phase. Crystallized solvents form in non-aqueous media in a frozen state. This leaves liquid pockets of highly concentrated solutes where reactions occur at a faster rate. Freeze concentration can produce a supersaturated solution in which the reactants can even be concentrated in 0.1% of the original liquid volume. The acceleration caused by the concentration effect is greater than the inhibition caused by freezing, thus the final manifestation is that the chemical reaction is accelerated by freezing.

Since the 1960s, it has been known that freezing can accelerate certain chemical reactions. For example, the reaction rate of ethylene chlorohydrin and sodium hydroxide in was increased by 1000-fold in a frozen solution. This phenomenon could be explained by the freeze concentration effect: at 5°C, 0.001 M initial reactant concentration, 99.9% of the frozen solution was solid, and the reaction would proceed at 1000 times the initial rate in the remaining 0.1% of liquid. Besides, the study on the stability of penicillin amide bonds has found that β-lactam bonds were unusually cleaved in frozen systems. Penicillin solutions containing imidazole lost antimicrobial activity when stored at −18°C but not at 38°C. The imidazole-catalyzed cleavage of the β-lactam bonds of penicillin only occurs in frozen systems at temperatures between −5°C and −30°C, not in super cooled systems. The concentration effect and the high proton mobility in ice promote the general base catalysis of β-lactam cleavage and rapid proton transfer in nucleophilic reactions. The structures of ice-like water containing "cavities" provide a rapid proton transport mechanism. During this period, the discovery of many accelerated reactions in frozen systems aroused the interest of researchers. The traditional concept that reactions were inhibited at low temperature might thus be subverted and freezing might be considered as a new option for some reactions that were difficult to carry out at room or high temperature.

The accelerated cryoreactions can be classified into two categories: one is to break the traditional concept that chemical reactions can be hindered at low temperature. On the contrary, a known reaction is accelerated apparently as a result of freezing. Usually, this type of reaction is explained by the freeze concentration effect. However, this kind of abnormally accelerated reaction should be taken seriously in the preservation of food, medicine and biological products. The other is new reactions that cannot proceed at room/ high temperature, but can occur unexpectedly upon lowering of temperature. Freezing is a necessary condition for this kind of reaction to occur. This type of reaction is usually seen in peptide synthesis.

In conclusion, studying chemical reactions accelerated by freezing will overturn common sense, and provide novel knowledge for researchers in the chemistry field. This article mainly introduces the mechanism of accelerated cryoreactions and some accelerated cryoreaction examples to provide new exploration conditions for the reactions that are difficult to carry out at room or high temperature.

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