Crystalline Peroxosolvates

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Peroxosolvates - adducts of hydrogen peroxide and molecules or salts formed by hydrogen bonding.

hydrogen peroxide | two-component crystals | periodic DFT computations | hydrogen bond | hydrogen bond enthalpy and energy | peroxosolvate | mixed pharmaceutical forms | perhydrate | adduct

1. Introduction

Crystalline peroxosolvates, adducts of hydrogen peroxide, were first introduced by Tanatar who synthesized sodium percarbonate $Na_2CO_3·1.5H_2O_2$ [1] and urea perhydrate (percarbamide) $CH_4N_2O·H_2O_2$ [2]. These compounds are the two most widely used solid peroxocompounds with annual production in the millions of tons[3]. Sustainable, nontoxic, and minimal hazard processing trends combine to intensify the use of hydrogen peroxide in diverse fields and the same trends are responsible for the perpetually growing use of peroxosolvates[3][4][5]. Peroxosolvates are now used for bleaching, disinfection, and oxidation; as chemical reagents in household commodities, cosmetics, pharmaceuticals, and washing powders; in industrial environmental processes such as remediation, bioremediation, and oxygen production; and as explosive ingredients and reagents for chemical synthesis [3][4][5][6]. In general, hydrogen peroxide release from peroxosolvates tends to lower the pH, whereas hydroperoxo- and peroxo-complexes tend to increase the pH [6][7][8][9][10]. Thus, peroxosolvates are considered safer and more economic as aqueous hydrogen peroxide decomposes at high pH.

Cambridge Structural Database (CSD)[11] and the Inorganic Crystal Structure Database (ICSD)[12] contain information on 134 peroxosolvates. This is several orders of magnitude less than the number of crystalline hydrates that exist in these databases[11][13]. A significant number of peroxosolvates (44 adducts) were synthesized and structurally characterized at the Kurnakov Institute of General and Inorganic Chemistry of Russian Academy of Sciences (IGIS RAS).

2. Chemical Composition of Crystalline Peroxosolvates

All 134 crystalline peroxosolvates known to date can be divided into three main groups depending on the chemical nature of the coformer (Figure 1):

1) salts of inorganic and carboxylic acids,

2) amino acids, peptides and related zwitterions, and

3) molecular compounds with a lone electron pair on nitrogen and/or oxygen atoms.

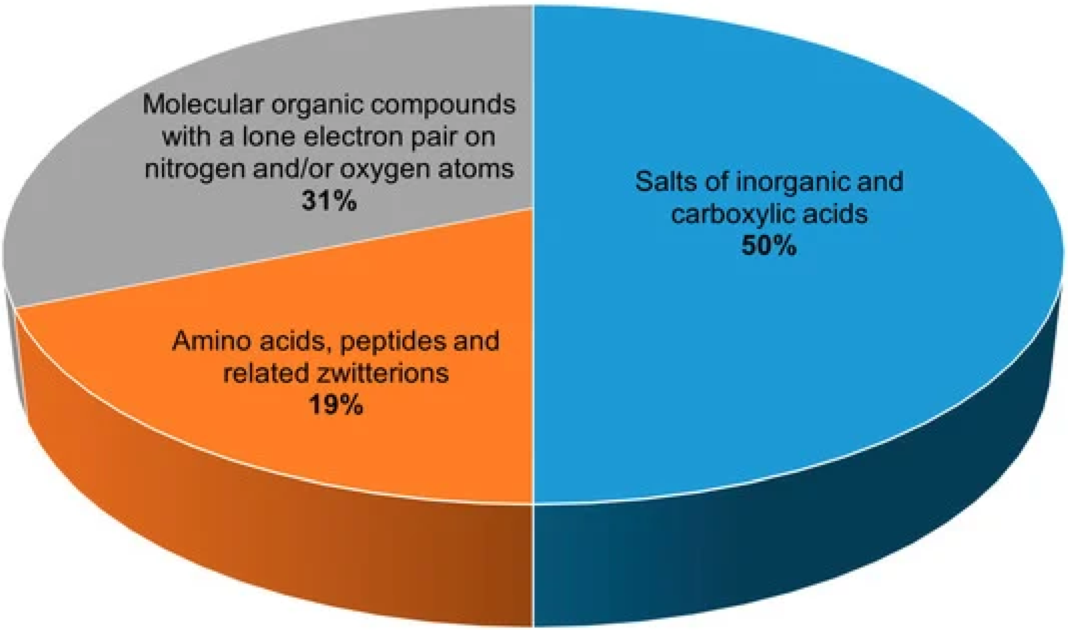

Figure 1. Distribution of peroxosolvates by the chemical nature of the coformer.

Intermolecular H-bonds play a structure-directing role in the considered crystals.

The largest number of structurally characterized peroxosolvates, 67 compounds, are adducts of hydrogen peroxide and salts of inorganic and carboxylic acids with various cations. Among the salts of carboxylic acids that form stable peroxosolvates [14][15][16][17][18], oxalates can be distinguished in the composition of six compounds. The salts of inorganic acids that form adducts with hydrogen peroxide are very diverse. In addition to fluorides [19][20][21], chlorides, and bromides[22][23], a number of peroxosolvates of alkali metal and ammonium carbonates are known[24][25][26]. The latter include the commercially demanded sodium peroxocarbonate synthesized by Tanatar[1]. This class of compounds should include peroxosolvates of complex anions, which can formally be attributed to the salts of the corresponding complex acids, for example, peroxovanadates[27][28][29][30][31][32][33][34], peroxoniobates[35][36][37], peroxotantalates[38], uranyl peroxo complexes[39], peroxotellurates[40], and platinum complexes[41][42][43]. Peroxosolvates of metal peroxides[44][45][46][47] can formally belong to the specified class of peroxosolvates of salts of inorganic acids if hydrogen peroxide is considered as a diacid.

The next group in terms of the number of compounds (42 compounds) are peroxosolvates formed by molecular organic compounds with a lone electron pair(s) on the nitrogen and/or oxygen atom(s). The main representatives of this group of crystalline hydrogen peroxide adducts are organophosphorus compounds containing the P=O functional group [48][49][50][51][52][53][54][55] and nitrogen-containing heterocyclic compounds[56][57][58][59][60][61][62][63][64][65], in particular N-oxides[66][67][68][69][70][71][72][73], obtained as a result of the oxidation reaction of the corresponding compounds with hydrogen peroxide. Urea peroxosolvate[74] is used as a solid source of hydrogen peroxide, and, along with 1,4-diazabicyclo[2.2.2]octane (DABCO) peroxosolvate[75], is used in organic syntheses to obtain anhydrous hydrogen peroxide solutions.

Separately, it is worth highlighting the third group, which includes amino acids, peptides, and related zwitterions (25 peroxosolvates). Peroxosolvates of a number of proteionogenic l-amino acids (serine, threonine, leucine, isoleucine, tyrosine, glycine, and phenylalanine)[76][77] and non-proteinogenic amino acids (gamma-aminobutyric acid, beta-alanine, and sarcosine) were obtained and structurally characterized at the Kurnakov Institute of General and Inorganic Chemistry RAS[77][78]. The class of zwitterions related to amino acids that form peroxosolvates includes pyridine carboxylic acids: nicotinic, isonicotinic, and picolinic acids[79], as well as 2-aminonicotinic acid[66]. Peroxosolvates of cyclic dipeptides—diglycine, disarcosine, and dialanine—are an example of the nonoxidative interaction of concentrated hydrogen peroxide and a peptide fragment[80].

The CSD analysis revealed the necessary properties of co-former peroxosolvates, which are promising compounds for the synthesis of new crystalline peroxosolvates[56]:

1) Hydrogen peroxide should not participate in redox reactions with coformers.

2) They must be sufficiently soluble in protic solvents to carry out the crystallization process.

3) Coformers should have the ability to form H-bonds, primarily as proton acceptors.

4) Compounds with pronounced acidic properties do not form peroxosolvates [14], since in such compounds the proton-acceptor groups are protonated.

5) Coformers should exhibit amphoteric or basic properties. Strong bases deprotonate hydrogen peroxide and form peroxide or hydroperoxide as ionic or complex moieties (ZnO_2 [81], $NH_4^+OOH^-$ [82], or $[Sn(OOH)_6]^{2-}$ [7]).

A significant part of the 134 peroxosolvates available in the structural databases, namely 40 structures, contain incomplete or erroneous data. Some crystal structures contain unlocalized hydrogen atoms and errors in the O-O bond lengths and H-O-O-H and O-O-H angles in the hydrogen peroxide molecule. This is due to the fact that during the preparation of these compounds, H_2O_2 was used as an oxidizing agent or ligand to obtain the corresponding peroxo complexes; therefore, the mass content of hydrogen peroxide in the obtained crystals is low. The chemical composition of 94 crystal structures of peroxosolvates with objectively localized protons and free of structural errors is presented in Supplementary Materials Table S1. These 94 crystal structures are the subject of the review, as they allow one to analyze the topology of H_2O_2 hydrogen-bonded networks.

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