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DECOMPOSITION OF HYDROGEN PEROXIDE - KINETICS AND REVIEW OF CHOSEN CATALYSTS

Abstract

Hydrogen peroxide is a chemical used in oxidation reactions, treatment of various inorganic and organic pollutants, bleaching processes in pulp, paper and textile industries and for various disinfection applications. It is a monopropellant, which, when purified, is self-decomposing at high temperatures or when a catalyst is present. Decomposing to yield only oxygen and water (disproportionation), hydrogen peroxide is one of the cleanest, most versatile chemicals available. The catalytic decomposition of hydrogen peroxide allows the use of various catalysts that will increase the rate of decomposition. Comparison and description of the most commonly used catalysts were presented in this review.

Key words

decomposition, hydrogen peroxide, catalysis, catalysts, silver catalyst, photocatalysis

Introduction

Hydrogen peroxide is a commonly used chemical compound with the formula H_2O_2 . In pure liquid form, it has a distinctive pale blue colour [1]. However, in a diluted solution, at room temperature, it appears as colourless mixture with a slightly sharp odour [2]. It doesn't ionise fully in water when it is dissolved. Solution of hydrogen peroxide is a weak acid, on the market it is usually offered in concentrations of 30, 50, and 70 percent by weight. Viscosity of H_2O_2 is slightly higher than water. Gaseous hydrogen peroxide is naturally present in the air in small amounts, which are created when ultraviolet rays strike oxygen in the presence of moisture [3]. Vapour of hydrogen peroxide is irritating to the respiratory tract and, what is more, it is corrosive to the skin and the eyes. Diluted solutions of H_2O_2 can be safely decomposed in the presence of proper enzymes known as catalase peroxidases, which are possessed by all aerobes.

Compounds with an oxygen–oxygen single bond are categorised as peroxides. Hydrogen peroxide represents the simplest type of this group of chemicals. It is characterized by its tendency to decompose, caused by the nature of its unstable peroxide bond. It must be stored with a stabilizer, such as acetanilide or similar organic materials. The solution is not flammable, regardless of its concentration. However, hydrogen peroxide is a strong oxidizing agent, due to its chemical structure and unpaired electrons [4].

Hydrogen peroxide can be used as a chemical intermediate, in the bleaching and deodorizing processes in pulp and the paper industry or in the textile industry [5]. H_2O_2 is also used in water treatment operations (substitute for chlorine in water and sewage treatment), exhaust air treatment, in production of foam rubber, organic chemicals (manufacture of glycerol) and in refining and cleaning metals. Concentrated hydrogen peroxide, or "high-test peroxide," has been also used to propel rockets [6]. In macroscale, it is known as a propellant of submarines and satellites. On the other hand, it is utilized for the propulsion of catalytic nanomotors in microscale [7]. In the food industry, H_2O_2 is applied in bleaching, oxidizing and neutralizing (in wine distillation) processes. It helps to control viscosity of starch and cellulose derivatives. In the form of carbamide peroxide it is commonly used for tooth whitening. At low concentrations (3-9%), hydrogen peroxide is found in many households as a hair and clothing bleach and for medical (anti-infective) applications. High reactivity gives H_2O_2 the ability to damage cellular macromolecules, including lipids, proteins and nucleic acids. Decomposing to only oxygen and water, hydrogen peroxide is said to be one of the most versatile and cleanest chemicals available. Hydrogen peroxide was discovered in 1818 by Louis Jacques Thenard. He reacted barium peroxide with nitric acid and then with hydrochloric acid. He managed to obtain pure hydrogen peroxide, which was called "oxygenated water". The addition of different substances, including blood, caused the new compound to decompose with the release of oxygen. Thenard's process was used until the middle of the 20th century.

Nowadays, production of hydrogen peroxide is mostly conducted according to the anthraquinone-auto oxidation process, where H_2O_2 is produced from hydrogen and atmospheric oxygen. The post-reaction mixture is then purified and concentrated. After appropriate stabilization, it is distributed as an aqueous solution. Different methods of derivation are electrolytic processes (aqueous sulphuric acid or acidic ammonium bisulfate is converted electrolytic ally) and auto oxidation of isopropyl alcohol [8].

Decomposition of hydrogen peroxide

Hydrogen peroxide is a very unique substance due to its molecular structure. It consists atoms of oxygen in oxidation state of -1 unlike many substances, where oxygen occurs in oxidation state of 0 or -2. This means that this substance can be used as both an oxidizing and a reducing agent, depending of pH of its solution. Due to those properties, particles of hydrogen peroxide can decompose via reaction of disproportionation as shown on Pic.1 [9].

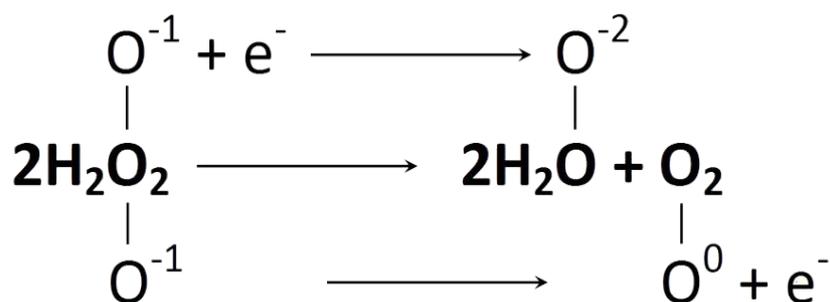


Fig. 1. Reaction of disproportionation of hydrogen peroxide

Source: Author's

Hydrogen peroxide is a quite stable substance in low and moderate pH. Although hydroxyl anions cannot trigger the reaction on their own [10], an increase of alkalinity in the environment of reaction up to a certain value may result in a rising rate of reaction. Unfortunately, the influence of pH on the rate of reaction is highly dependent on the purity of the solution. For example, in a low purity solution, pH may not affect it at all [11]. However, H_2O_2 easily decomposes when it is exposed to impurities or catalysts like metallic surfaces or yeast. Other factors that affect the rate of reaction are temperature, pressure, concentration of solution, type, activity and area of active catalytic surface of the catalyst, exposure to direct sunlight and presence of inhibitors [10].

Reaction of decomposition of hydrogen peroxide is very slow in moderate temperatures without the presence of a catalyst [11]. It can be sped up by increasing the temperature, which triggers the reaction of thermal decomposition. This reaction can be carried out in a liquid or vapour phase. Usually the rate of reaction is increased by adding catalysts to the reactor. Almost all types of catalyst can be used with this reaction: heterogeneous (e.g. silver, gold, iron), homogeneous (iodide or iron ions) and enzymes (catalase).

This reaction is exothermic. It produces large amount of heat ($\Delta H = -2884.5 \text{ kJ/kg H}_2\text{O}_2$ for pure compound), which further increases rate of reaction and makes reaction self-sustaining after phase of catalytic initiation [12]. The mechanism of reaction is dependent on the type of catalyst used. For example, for transition metal complexes two general types of mechanism have been postulated: radical mechanism proposed by Haber and Weiss and peroxide complex mechanism proposed by Kremer and Stein [13]. Some reaction mechanisms still need more research. For example, the exact mechanism of reaction catalysed by MnO_2 is still unknown. Thermal decomposition is a very complex reaction, one of the proposed models is based on 27 subordinate reactions [9]. Often the final mechanism of reaction is a combination of various other mechanisms, usually thermal decomposition in addition to the reaction with catalysts and impurities.

In many industries decomposition of hydrogen peroxide is used e.g. to produce sodium perborate and sodium percarbonate (bleaching agents in solid and liquid detergents). In textile industry hydrogen peroxide bleach and deodorize textiles, wood pulp, hair, fur, etc. The other applications of H_2O_2 are as source of organic and inorganic peroxides, rocket fuel, plasticizers, neutralizing agent in wine distillation, bleaching and oxidizing agent in foods, substitute for chlorine in water and sewage treatment, viscosity control for starch and cellulose derivatives [1]. Hydrogen peroxide is also present in pulp and paper industry, manufacture of glycerol, foam rubber, refining and

cleaning metals [2]. Because of antichlor, dyeing, electroplating, antiseptic epoxidation, hydroxylation, oxidation and reduction properties it is wide use in many industrial processes.

Silver catalyst

Silver is one of most effective heterogeneous catalysts for hydrogen peroxide decomposition. Due to the high influence of the catalytic active area on the rate of reaction usually only significantly modified geometric forms of silver are being used in this reaction. Generally, silver is used in metallic form or as an alloy. It is not used in ionic form, as it is prone to precipitation. The most popular forms used in industry or in laboratories are mesh and powder. Use of powder and nanoparticles is attractive due to their high surface area, but it may result in the necessity of using some additional process of separation of catalyst and solution. In other cases, a post reaction mixture may be contaminated [14]. Therefore, usually silver is used in the form of mesh or other geometric forms, which allows the forming of a bed. To restrain the cost of catalyst, sometimes silver is used in the form of a thin layer coating, instead of using pure silver. It does not affect rate of reaction, because reaction takes place only within the interphase layer.

Despite the fact that new catalysts for the reaction are developed every year, metallic catalyst based on silver are still very attractive. The main advantages of using silver are its high decomposition efficiency, compactness, uncomplicated process of fabrication, and multiple available form of catalyst [15]. Silver catalysts also have some disadvantages, like temperature limitations (melting temperature of silver – 961.8°C) and a complicated process of activation of catalyst. Another drawback of silver is catalyst loss of mass after several cycles of reaction. Mass loss is proportional to the squared area of catalyst [16]. This process is intensive for new and regenerated metal at the start of a process of decomposition. After some time, the concentration of ions of silver in the solution stabilizes due to the saturation of solution. These ions are inhibitors to silver degeneration. The most important disadvantage of silver is its vulnerability to surface poisoning. To counter that, the catalyst must be periodically regenerated, for example with nitric acid. Regeneration has one additional effect for the catalyst – it develops a surface of metal and increases the amount of active centres. It is crucial for new silver, where surface is undeveloped and smooth, which results in limited decomposition efficiency [17]. Silver is also quite expensive when compared to other catalysts, such as those based on iron.

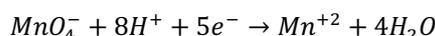
During the reaction of hydrogen peroxide decomposition catalysed by silver, four different reaction zones can be noticed, each dominant for a range of bulk solution temperatures. In low temperature zones, the rate of reactions is chemically controlled by properties of the catalyst. In nuclear boiling zone, due to the contact between catalysts and boiling liquid temperature of catalyst does not exceed the boiling point of the solution. The next zone is the film boiling zone, where the vapour phase is formed. This results in significant limitation of area of contact between reactant and catalysts and a noticeable decrease of the rate of reaction. The last zone is called the high temperature reaction zone, where the rate of reaction of decomposition is more dependent on the reaction of homogeneous thermal decomposition rather than heterogeneous catalysed reaction [18]. Due to the heterogeneous nature of the process of decomposition of H_2O_2 catalysed by silver exact mechanism of this reaction is still unknown, probably it is based on a radical mechanism.

Manganese compounds

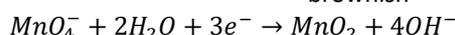
Potassium permanganate ($KMnO_4$) in standard conditions is a solid with density 2.7 g/cm³. It is not hygroscopic, in contrast to sodium permanganate. It has good water solubility, in 20°C it is 6,4g in 100cm³ of water. The colour of its solution is strictly dependent on its concentration; it varies from bright pink in diluted solutions to dark purple in saturated solutions. Solid $KMnO_4$ thermally decomposes in 240°C, as shown on following equation:



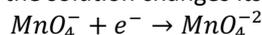
This substance has strong oxidizing properties, but mechanism of this oxidation reaction is strongly dependent on pH of the environment of the reaction. In acid solutions $KMnO_4$ is reduced to Mn^{+2} , the solution changes colour to pale pink:



In a weak alkaline or inert solution permanganate ion is reduced to manganese oxide, which appears in the form of a brownish sediment:



However, in a strong alkaline environment, the solution changes its colour to green:



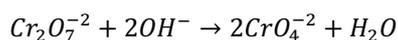
Potassium permanganate is being used in titration analysis - manganometry. It also has a strong bactericidal and fungicidal properties, diluted solution is used in oral rinsing.

The reaction of catalytic decomposition of hydrogen peroxide catalysed by potassium permanganate can be divided into three stages: a fast initial phase, an induction period and an autocatalytic step [25]. This reaction is autocatalytic, because product of reaction Mn^{2+} creates unstable complex with MnO_4^{-1} . Products of decomposition of this complex have better catalytic properties than the complex itself. Some studies are focused on obtaining porous cement slurry with the use of reaction decomposition of H_2O_2 with potassium based catalysts. Currently, aluminium powder is being used in this process, but the rate of reaction of producing gases is hard to control [22].

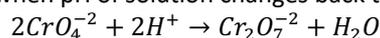
Manganese oxide (MnO_2) is widely used in heterogeneous reaction of decomposition of hydrogen peroxide thanks to its excellent catalytic efficiency in this reaction. It is usually used in the form of a powder thanks to its very high contact surface [21]. One of disadvantages of this catalyst is a necessity of immobilizing a manganese oxidize powder on solid base. The kinetics of catalytic decomposition hydrogen peroxide can be represented by pseudo first-order rate model [24]. Si-Hyun Do and the others [24] suggested a 15-tier mechanism of this reaction.

Potassium dichromate ($K_2Cr_2O_7$)

Potassium dichromate is an orange crystalline ionic solid. In water solutions, it is durable only in an acid environment, in alkali solutions its colour is changing from orange to yellow as a result of redox reaction of dichromate ion:



However, reverse reaction can occur when pH of solution changes back to acid:



Potassium dichromate is used as an oxidizing agent in various reactions in chemical industry and laboratory preparation, such as dye production, electrolysis, pyrotechnics, glassware, glues, dyeing, photography, lithography and the ceramic industry. The solution of potassium dichromate in sulfuric acid is being used to clean laboratory glass, soiled organic compounds and other compounds susceptible to oxidation. Nowadays its use is very limited, due to problematic disposal of wastes containing chromium compounds. Potassium dichromate, as other Cr (VI) compounds, is toxic, mutagenic and carcinogenic. It is also extremely dangerous to the environment.

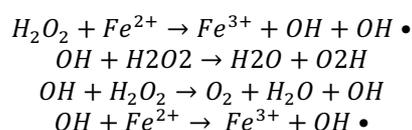
The mechanism of reaction of decomposition of hydrogen peroxide catalysed by potassium dichromate is complicated and a lot of research has been carried out over the last 100 years [1,2]. During those studies various Cr(VI)-peroxo complexes were observed at different pH and concentrations of solution, for example:

- at pH=4 - $Cr^{VI}(O)_5(H_2O)$ or $Cr^{VI}(O)(O_2)_2(OH_2)$. Their solution is called blue peroxochromic acid,
- at pH between 4 and 7 -violet deprotonated from of "peroxochromic acid" $[Cr^{VI}(O)(O_2)(OH)]^-$
- at least four Cr(V)-peroxocomplexes appearing at different pH and concentrations[19].

Potassium dichromate as a catalyst studied reaction has some advantages: it is easy to implement and the reaction rate can be controlled by H_2O_2 solution dilution [23].

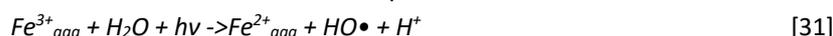
Iron oxide

Another known reaction based on the decomposition of hydrogen peroxide is Fenton's reaction, where a hydroxyl radical is created by iron ions. This reaction was suggested by Haber and Weiss in 1932, however their investigation was not focused on oxidation. Thanks to Fenton's theories about oxidation this process is now named after him [29]. One of the forms of catalyst used in that type of reaction is goethite, a pure crystalline iron oxide in aqueous medium- α -FeOOH. Many key reactions of this process are based on the transition aqueous hydrogen peroxide to the anion form OH_2^\bullet .



Profound research on that mineral was carried by Shu-Sung Lin and Mirat de Gurol[26]. It is worth mentioning that it is the most abundant crystalline iron oxide mineral in nature. However, this material is not the only one

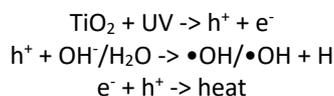
used as the source of iron oxide. Equally popular are hematite ($\alpha\text{-Fe}_2\text{O}_3$) and ferrihydrite ($(\text{Fe}^{3+})_2\text{O}_3 \cdot 0.5\text{H}_2\text{O}$) [28]. The most efficient of these three minerals is ferrihydrite thanks to its large inner surface that is associated with an increased rate of diffusion process. It is also worth mentioning that the iron oxide in the form of aqueous goethite is suitable for catalysing hydrogen peroxide decomposition even in highly concentrated solutions (about 1-10 mM). Compounds containing ferric ions, for example ferric salts, are also regarded as good catalysts, due to their high selectivity. Nevertheless, they can cause many problems since reactions of decomposition of catalyst and hydrogen peroxide are highly exoenergetic [27]. Despite this fact, iron oxides are commonly used in our daily life, in metallurgy, as a colorants or thermite component. Thanks to their super magnetic properties and the ability to overcome the blood-brain barrier they can also be used in medicines, as a contrast in MRI (Magnetic Resonance Imaging) [30]. Some scientists tried to overcome some of weaknesses of this reaction. About 20 years ago (Huston & Pignatello, 1999; Ruppert et al., 1993) it was found that combination of Fenton reaction and UV light radiation (nowadays it is called photo-Fenton process) has strongly accelerated the rate of degradation of a variety of pollutants, even if it is compared to the regular Fenton reaction. This is mainly due to the presence of photochemical reduction of Fe^{3+} back to Fe^{2+} . This reaction can be present as:



This reaction shows some strong advantages in comparison with regular Fenton reaction. First of all, the main weakness of classical Fenton reaction - self-blocking through the increasing amount of sludge is not present in photo-Fenton reaction. Secondly, chemical consumption is significantly lower, what results in lower cost of the process. To add more, due to negligible formation of sludge, there is no need to remove it, what reduces operation costs even more. [32]

Photocatalytic decomposition on TiO Surface

The next type of reaction of decomposition of hydrogen peroxide is a reaction involving use of UV light and TiO_2 surface [33] that has some photo catalytic properties as in the case of iron compounds in the photo-Fenton reaction. In this type of reaction when TiO_2 is irradiated light which has photon energy equal to or higher than its band-gap, an electron (e^-) can be excited from the valence band to the conduction band and leave a hole (h^+) in the valence band:



The presence of hydrogen peroxide in this reaction is essential, because as an electron acceptor it is responsible for the growth of photocatalytic efficiency and mineralization of losses of energy during UV radiation. Controlling the amount of H_2O_2 during this process is crucial. Although this process requires a large amount of hydrogen peroxide, too large of a dose may cause the accumulation of electrons on the hydrogen peroxide radicals and competitive adsorption of hydrogen peroxide instead of its decomposition [35]. In this type of process the amount of titanium dioxide is equally important because it can be used to control the rate of decomposition. Due to its vulnerability to fragmentation to the size of nanoparticles it has excellent photocatalytic properties, especially one of its catalytic form named anataz. However, that properties are highly dependent on the temperature and the specific surface area. Furthermore, TiO_2 may also be used in rutile form, which has very good mechanical and catalytic properties thanks to high chemical resistance and high refractive index.

Photocatalytic reactions on TiO_2 surface in presence of hydrogen peroxide are useful in the oxidised degradation or transformation of a wide range of pollutants for the treatment of drinking water, groundwater, wastewater and contaminated soils. It is also useful to get rid of hazardous compounds present in dyes [34]. However, most applications of titanium dioxide are still carried out at a laboratory rather than at industrial scales.

Hydrogen peroxide in living cells

Hydrogen peroxide naturally occurs in the human body as one of the by-products of biochemical metabolism of many different cells. For example, hydrogen peroxide is directly produced by some oxidise enzymes like glycolateoxidise or monoamine oxidise. Hydrogen peroxide is also created by the peroxisomal pathway for L-oxidation of fatty acids [36].

What is more, some popular beverages, including instant coffee, black tea and green tea, can contain hydrogen peroxide at low concentrations. Consumption of these beverages may conduct to diffusion of hydrogen peroxide

into the cells of the oral cavity and upper part of the gastrointestinal tract [36]. Another source of hydrogen peroxide in the gastrointestinal tract are oral bacteria which produce this substance [36].

Hydrogen peroxide could also be found in exhaled air of living organisms. It is uncertain whether the source of it is oral bacteria, phagocytes or other lung cells. People with lung diseases or cigarette smokers exhale more hydrogen peroxide than healthy ones[36].

Considering all the above it is not surprising that hydrogen peroxide is also present in the blood. But concentration of hydrogen peroxide in human blood has not been determined with satisfactory accuracy yet. There are some conventional methods of hydrogen peroxide detection but the absolute values remain uncertain [37]. The literature is full of data about the hydrogen peroxide level in blood but credibility of it is questionable due to the great variability of results [37].

To sum up, it seems that most human cells are exposed to hydrogen peroxide. H_2O_2 can have significant impact on aging processes. During decomposition of hydrogen peroxide with, for example, copper (I) and iron (II) ions as a catalyst, hydroxyl radical (OH^\bullet) is formed [38]. Free radicals are molecules, atoms or ions which have single, unpaired electron at the outer orbits. High activity individuals have this chemical and oxidize each compound with which they have contact in order to join or donate electrons. The objects of the attacks of free radicals in the human body are mainly compounds having double bonds in the molecules like proteins, DNA, polysaccharides, lipids (like cholesterol in the blood) or unsaturated fatty acids which are part of cell membranes [36].

Accumulation of hydrogen peroxide may have very bad influence on cells and can even kill them, so it has to be decomposed. The natural process of hydrogen peroxide decomposition is very slow. Therefore, in living organisms there has to be an acceleration mechanism for this reaction. It is widely known that reactions in cells are mostly accelerated by enzymes. In case of hydrogen peroxide decomposition, it occurs in the same way. One of such substance is an enzyme called Catalase, which lowers the energy of activation needed for decomposition [39].

Conclusion

Hydrogen peroxide is one of the cleanest, most versatile chemicals available. Due to its beneficial properties, it is used in a broad variety of application areas e.g.: in aseptic packaging as sterilizing agent, in cosmetics and medicine as an antimicrobial agent, in chemical synthesis as a powerful and environmentally benign oxidizing agent, in pulp, paper and textile industry as a versatile bleaching agent and also in transportation high concentrated, high purity hydrogen peroxide is used as a propellant. Hydrogen peroxide is the object of diverse studies due to its useful properties. Although there are many varied applications of this substance, its usage remains limited. The main reason for this is high production costs. Moreover, in lower concentration it is chemically unstable and requires addition of stabilizers, which change the kinetics of reaction of its decomposition. A separate direction of research concerning hydrogen peroxide is its presence in the human body, which can have a significant influence on biochemical reactions that occur in our cells. Therefore, this simple substance is more important than it seems and further studies concerning its production, usage and appropriate concentration measurement can make a breakthrough in many areas of our lives.

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