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Free Radical Mechanism for Metal Ion Catalysed Autoxidation of Sulphur Dioxide in Atmosphere Aqueous Medium

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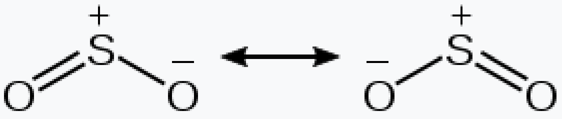
ABSTRACT: A free-radical reaction is any chemical reaction involving free radicals. This reaction type is abundant in organic reactions. Two pioneering studies into free radical reactions have been the discovery of the triphenylmethyl radical by Moses Gomberg (1900) and the lead-mirror experiment[1] described by Friedrich Paneth in 1927. In this last experiment tetramethyllead is decomposed at elevated temperatures to methyl radicals and elemental lead in a quartz tube. The gaseous methyl radicals are moved to another part of the chamber in a carrier gas where they react with lead in a mirror film which slowly disappears. Sulfur dioxide (IUPAC-recommended spelling) or sulphur dioxide (traditional Commonwealth English) is the chemical compound with the formula SO_2. It is a toxic gas responsible for the odor of burnt matches. It is released naturally by volcanic activity and is produced as a by-product of copper extraction and the burning of sulfur-bearing fossil fuels. In atmosphere, There, it reacts with water to form clouds of sulfuric acid, and is a key component of the planet's global atmospheric sulfur cycle and contributes to global warming.

KEYWORDS: free radical, metal ion, autoxidation, sulphur dioxide, aqueous medium, atmosphere, methyl radicals

I.INTRODUCTION

SO_2 is a bent molecule with C_{2v} symmetry point group. A valence bond theory approach considering just s and p orbitals would describe the bonding in terms of resonance between two resonance structures.[9]



Two resonance structures of sulfur dioxide

The sulfur–oxygen bond has a bond order of 1.5. There is support for this simple approach that does not invoke d orbital participation.[9] In terms of electron-counting formalism, the sulfur atom has an oxidation state of +4 and a formal charge of +1.[1-8]

Sulfur dioxide is found on Earth and exists in very small concentrations in the atmosphere at about 15 ppb.[10]

On other planets, sulfur dioxide can be found in various concentrations, the most significant being the atmosphere of Venus, where it is the third-most abundant atmospheric gas at 150 ppm. There, it reacts with water to form clouds of sulfuric acid, and is a key component of the planet's global atmospheric sulfur cycle and contributes to global warming.[11] It has been implicated as a key agent in the warming of early Mars, with estimates of concentrations in the lower atmosphere as high as 100 ppm,[12] though it only exists in trace amounts. On both Venus and Mars, as on Earth, its primary source is thought to be volcanic. The atmosphere of Io, a natural satellite of Jupiter, is 90% sulfur dioxide[13] and trace amounts are thought to also exist in the atmosphere of Jupiter. The James Webb Space Telescope has observed the presence of sulfur dioxide on the exoplanet WASP-39b, where it is formed through photochemistry in the planet's atmosphere.[14]

As an ice, it is thought to exist in abundance on the Galilean moons—as subliming ice or frost on the trailing hemisphere of Io,[15] and in the crust and mantle of Europa, Ganymede, and Callisto, possibly also in liquid form and readily reacting with water.[16]

Sulfur dioxide is primarily produced for sulfuric acid manufacture (see contact process). In the United States in 1979, 23.6 million metric tons (26 million U.S. short tons) of sulfur dioxide were used in this way, compared with 150,000 metric tons (165,347 U.S. short tons) used for other purposes. Most sulfur dioxide is produced by the combustion of elemental sulfur. Some sulfur dioxide is also produced by roasting pyrite and other sulfide ores in air.[17] Sulfur dioxide is the product of the burning of sulfur or of burning materials that contain sulfur:[15]

$^1\!/_8\ S_8 + O_2 \rightarrow SO_2, \Delta H = -297$ kJ/mol

To aid combustion, liquified sulfur (140–150 °C, 284-302 °F) is sprayed through an atomizing nozzle to generate fine drops of sulfur with a large surface area. The reaction is exothermic, and the combustion produces temperatures of 1000–1600 °C (1832–2912 °F). The significant amount of heat produced is recovered by steam generation that can subsequently be converted to electricity.[17]

The combustion of hydrogen sulfide and organosulfur compounds proceeds similarly. For example:

$H_2S + {}^3\!/_2\ O_2 \rightarrow SO_2 + H_2O$

The roasting of sulfide ores such as pyrite, sphalerite, and cinnabar (mercury sulfide) also releases SO_2:[18]

$2\ FeS_2 + {}^{11}\!/_2\ O_2 \rightarrow Fe_2O_3 + 4\ SO_2$
$ZnS + {}^3\!/_2\ O_2 \rightarrow ZnO + SO_2$
$HgS + O_2 \rightarrow Hg + SO_2$
$2\ FeS + {}^7\!/_2\ O_2 \rightarrow Fe_2O_3 + 2\ SO_2$

A combination of these reactions is responsible for the largest source of sulfur dioxide, volcanic eruptions. These events can release millions of tons of SO_2.

Sulfur dioxide can also be a byproduct in the manufacture of calcium silicate cement; $CaSO_4$ is heated with coke and sand in this process:

$2\ CaSO_4 + 2\ SiO_2 + C \rightarrow 2\ CaSiO_3 + 2\ SO_2 + CO_2$

Until the 1970s, commercial quantities of sulfuric acid and cement were produced by this process in Whitehaven, England. Upon being mixed with shale or marl, and roasted, the sulfate liberated sulfur dioxide gas, used in sulfuric acid production, the reaction also produced calcium silicate, a precursor in cement production.[19]

On a laboratory scale, the action of hot concentrated sulfuric acid on copper turnings produces sulfur dioxide.

$Cu + 2\ H_2SO_4 \rightarrow CuSO_4 + SO_2 + 2\ H_2O$

Tin also reacts with concentrated sulfuric acid but it produces tin(II) sulfate which can later be pyrolyzed at 360°C into tin dioxide and dry sulfur dioxide.

$Sn + H_2SO_4 \rightarrow SnSO_4 + H_2$
$SnSO_4 \rightarrow SnO_2 + SO_2$

II.DISCUSSION

In chemistry, a radical, also known as a free radical, is an atom, molecule, or ion that has at least one unpaired valence electron.[1][2] With some exceptions, these unpaired electrons make radicals highly chemically reactive. Many radicals spontaneously dimerize. Most organic radicals have short lifetimes.[23]

A notable example of a radical is the hydroxyl radical (HO·), a molecule that has one unpaired electron on the oxygen atom. Two other examples are triplet oxygen and triplet carbene (:CH_2) which have two unpaired electrons.

Radicals may be generated in a number of ways, but typical methods involve redox reactions, Ionizing radiation, heat, electrical discharges, and electrolysis are known to produce radicals. Radicals are intermediates in many chemical reactions, more so than is apparent from the balanced equations.

Radicals are important in combustion, atmospheric chemistry, polymerization, plasma chemistry, biochemistry, and many other chemical processes. A majority of natural products are generated by radical-generating enzymes. In living organisms, the radicals superoxide and nitric oxide and their reaction products regulate many processes, such as control of vascular tone and thus blood pressure. They also play a key role in the intermediary metabolism of various biological compounds. Such radicals can even be messengers in a process dubbed redox signaling. A radical may be trapped within a solvent cage or be otherwise bound.[25]

Autoxidation (sometimes auto-oxidation) refers to oxidations brought about by reactions with oxygen at normal temperatures, without the intervention of flame or electric spark.[1] The term is usually used to describe the gradual degradation of organic compounds in air at ambient temperatures. Many common phenomena can be attributed to autoxidation, such as food going rancid,[2] the 'drying' of varnishes and paints, and the perishing of rubber.[3] It is also an important concept in both industrial chemistry and biology.[4] Autoxidation is therefore a fairly broad term and can encompass examples of photooxygenation and catalytic oxidation.28

The common mechanism is a free radical chain reaction, where the addition of oxygen gives rise to hydroperoxides and their associated peroxy radicals (ROO•).[5] Typically, an induction period is seen at the start where there is little activity; this is followed by a gradually accelerating take-up of oxygen, giving an autocatalytic reaction which can only be kept in check by the use of antioxidants. Unsaturated compounds are the most strongly effected but many organic materials will oxidise in this way given time.

Although autoxidation is usually undesirable it has been exploited in chemical synthesis. In these cases the term 'autoxidation' is often used more broadly to include spontaneous reactions with oxygen at elevated temperatures, such as in the Cumene process.35

III.RESULTS

The free radical chain reaction is sometimes referred to as the Bolland-Gee mechanism[6][7] or the basic autoxidation scheme (BAS)[8] and was originally based on the oxidation of rubbers,[9] but remains generally accurate for many materials. It can be divided into three stages: initiation, propagation, and termination.[10] The initiation step is often ill-defined and many agents have been proposed as radical initiators.[11] The autoxidation of unsaturated compounds may be initiated by reactions with singlet oxygen[12] or environmental pollutants such as ozone and NO_2.[13] Saturated polymers, such as polyolefins would be expected to resist autoxidation, however in practise they contain hydroperoxides formed by thermal oxidation during their high temperature moulding and casting, which can act as initiators.[14][15] In biological systems reactive oxygen species are important. For industrial reactions a radical initiator, such as benzoyl peroxide, will be intentionally added.33

All of these processes lead to the generation of carbon centred radicals on the polymer chain (R•), typically by abstraction of H from labile C-H bonds. Once the carbon-centred radical has formed, it reacts rapidly with O_2 to give a peroxy radical (ROO•). This in turn abstracts an H atom from a weak C-H bond give a hydroperoxide (ROOH) and a fresh carbon-centred radical. The hydroperoxides can then undergo a number of possible homolytic reactions to generate more radicals,[8] giving an accelerating reaction. As the concentration of radicals increases chain termination reactions become more important,40 these reduce the number of radicals by radical disproportionation or combination, leading to a sigmoid reaction plot. In general, chemical reactions occur faster in the presence of a catalyst because the catalyst provides an alternative reaction mechanism (reaction pathway) having a lower activation energy than the non-catalyzed mechanism. In catalyzed mechanisms, the catalyst usually reacts to form an intermediate, which then regenerates the original catalyst in the process.[8][9][10][11]

As a simple example occurring in the gas phase, the reaction $2\ SO_2 + O_2 \rightarrow 2\ SO_3$ can be catalyzed by adding nitric oxide. The reaction occurs in two steps:

$2\ NO + O_2 \rightarrow 2\ NO_2$ (rate-determining)
$NO_2 + SO_2 \rightarrow NO + SO_3$ (fast)

The NO catalyst is regenerated. The overall rate is the rate of the slow step[11]

$v = 2k_1[NO]^2[O_2]$.

An example of heterogeneous catalysis is the reaction of oxygen and hydrogen on the surface of titanium dioxide (TiO_2, or titania) to produce water. Scanning tunneling microscopy showed that the molecules undergo adsorption and dissociation.56 The dissociated, surface-bound O and H atoms diffuse together. The intermediate reaction states are: HO_2, H_2O_2, then H_3O_2 and the reaction product (water molecule dimers), after which the water molecule desorbs from the catalyst surface.[12][13]

IV.CONCLUSIONS

An added substance that lowers the rate is called a reaction inhibitor if reversible and catalyst poisons if irreversible.[1] Promoters are substances that increase the catalytic activity, even though they are not catalysts by themselves.[43]

Inhibitors are sometimes referred to as "negative catalysts" since they decrease the reaction rate.[44] However the term inhibitor is preferred since they do not work by introducing a reaction path with higher activation energy; this would not lower the rate since the reaction would continue to occur by the non-catalyzed path. Instead, they act either by deactivating catalysts or by removing reaction intermediates such as free radicals.[44][8] In heterogeneous catalysis, coking inhibits the catalyst, which becomes covered by polymeric side products.[57]

The inhibitor may modify selectivity in addition to rate. For instance, in the hydrogenation of alkynes to alkenes, a palladium (Pd) catalyst partly "poisoned" with lead(II) acetate ($Pb(CH_3CO_2)_2$) can be used.[45] Without the deactivation of the catalyst, the alkene produced would be further hydrogenated to alkane.[46][47]

The inhibitor can produce this effect by, e.g., selectively poisoning only certain types of active sites. Another mechanism is the modification of surface geometry. For instance, in hydrogenation operations, large planes of metal surface function as sites [58] of hydrogenolysis catalysis while sites catalyzing hydrogenation of unsaturates are smaller. Thus, a poison that covers the surface randomly will tend to lower the number of uncontaminated large planes but leave proportionally smaller sites free, thus changing the hydrogenation vs. hydrogenolysis selectivity. Many other mechanisms are also possible.

Promoters can cover up the surface to prevent the production of a mat of coke, or even actively remove such material (e.g., rhenium on platinum in platforming). They can aid the dispersion of the catalytic material or bind to reagents.[59]

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