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THE MECHANISM OF ULTRA-WEAK LUMINESCENCE IN BIOLOGICAL SYSTEMS

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The weak luminescence which accompanies biological processes has now for a long time attracted the attention of investigators. In the twenties and thirties of this century A.G. Gurvich (1934) used a biological detector to discover the weak ultraviolet radiation of living cells and tissues which, as the author's data showed, exerted a stimulating effect on the process of cell division and which he called mitogenetic radiation. Isolated attempts were later undertaken to study the luminescence of biological objects by means of modified Geiger counters sensitive to ultraviolet radiation (Rodionov, Frank, 1936; Audubert, 1938; Becher, 1957).

Research on the processes which go on with the participation of the excited states of biologically important compounds (photosynthetic pigments, proteins, and nucleic acids) gave us the impetus to study the ultra-weak luminescence occurring in biological systems as the result of photochemical reactions, as well as with dark processes.

From the very beginning of the investigation the main difficulty was the lack of suitable methods for recording luminescence. Therefore the first stage of our work was to create the proper measuring equipment. Since the sensitivity of a photomultiplier is principally limited by the dark current level the most radical method to raise sensitivity seemed to us to be to cool the photomultiplier.

A photomultiplier cooled with liquid nitrogen was employed by Arnold and Strehler (Strehler, 1954) to record the weak luminescence

of photosynthesizing organisms after they had first been illuminated. The equipment utilized by these authors has unfortunately not been described in sufficient detail (Strehler, 1951).

In 1959 we developed a sensitive device for measuring weak luminescence. It consisted of a photomultiplier cooled with liquid nitrogen and an impulse counter. This apparatus helped detect the luminescence of bean roots, dry proteins irradiated with ultraviolet, and other biological objects. In all cases studied the luminescence lay in the visible region of the spectrum. The different emissions in the biological systems were probably of differing characters and were distinguished by extremely low strength; hence we called them "ultra-weak luminescence of biological systems" (Vladimirov, Litvin, 1959).

The present paper gives the most general system of the mechanism of the processes responsible for the ultra-weak luminescence of biological systems. The basis of the concepts which are developed is the set of experimental findings which indicate the existence of a profound similarity between the mechanism of the ultra-weak luminescence in biological objects and the mechanism of luminescence which accompanies inverse photochemical reactions.

WEAK LUMINESCENCE IN INVERSE PHOTOCHEMICAL PROCESSES (PHOTOCHEMOLUMINESCENCE)

1. Chemoluminescence in Photochemical Transformations of Chlorophyll

In the course of investigating the mechanism of the primary processes of photosynthesis we resorted to studying luminescence during photochemical reactions of chlorophyll in model systems, i.e., in the reaction of reversible photoreduction of chlorophyll (Krasnovskiy's reaction) and in the chlorophyll photooxidation reaction. In both cases we detected ultra-weak luminescence which occurred during processes that were reversible with respect to the direct photochemical reaction (Litvin, Vladimirov, et al., 1960). The figure diagrams the processes underlying this phenomenon. The absorption of a light quantum by a chlorophyll molecule X, i.e., the transition $X \longrightarrow X^*$, is accompanied by transfer of the molecule into the triplet system X^V and formation of a reduced intermediate form with the nature of an ion-radical $^{\cdot}X^-$. The reduction was accomplished by transfer of an electron from a donor molecule AH. The donor may be ascorbic acid, phenylhydrazine, a solvent, etc. A proton is then added and a stable photochemical product -- a "secondary" photoreduced form XH -- is created.

Luminescence occurs during the inverse dark reactions during regeneration of the original chlorophyll molecule from the photoreduced form. For it to be generated, however, it is essential that oxygen

reacting with the primary photoreduced form be present ($^{\cdot}X^{-} + O_2 \longrightarrow X +$ $+ h\nu$, the transition $^{\cdot}X^{-} \longrightarrow X^{\nu}$). The spontaneous inverse reaction proceeding in the absence of oxygen and the reaction of the photoreduced form with the pigment apparently is not accompanied by perceptible luminescence (the transition $^{\cdot}X^{-} \longrightarrow X$). The inference that it is precisely the primary photoreduced form (chlorophyll ion-radical) which is responsible for the luminescence is based on the fact that luminescence rises drastically in the presence of ammonia, which, as has been previously demonstrated, causes a rapid transformation of the secondary photoreduced form XH into the ion-radical $^{\cdot}X^{-}$ (Krasnovskiy, Brin, 1953).

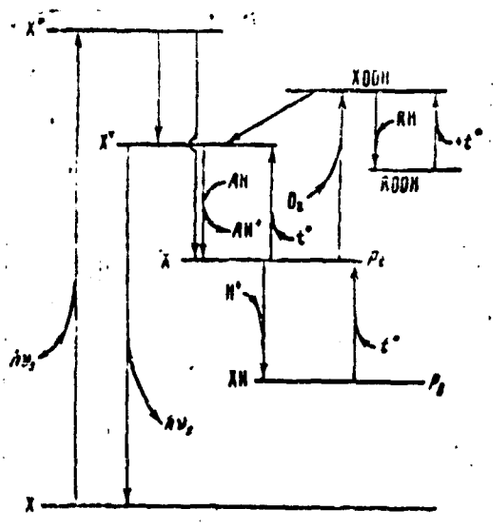
Kinetic studies indicated that the luminescence decays in step with regeneration of chlorophyll from the primary photoreduced form (Litvin, Vladimirov, et al., 1960).

The underlying reaction in the processes of chlorophyll chemoluminescence during chlorophyll photoreduction is therefore that of oxidation of the active ion-radical of the pigment of the primary photoreduced form by the oxygen of the air.

In this respect the process of chemoluminescence in inverse oxidation of the photoproduct of chlorophyll resemble the other chemoluminescence reactions; the participation of oxygen almost always proves to be necessary for de-excitation of a quantum. The question remains open whether the role of oxygen is limited to its furnishing energy to the system or whether this role is that its unexcited molecule is in the triplet state so that the reaction product immediately is found in the triplet (excited) state; then the subsequent transition of the molecule into the basic state will be accompanied by de-excitation of the quantum (White, 1961).

In the case of chemoluminescence with oxidation of the chlorphyll ion-radical by atmospheric oxygen the measurements of the spectral composition of the emission show that the act of luminescing is associated with the radiative transition in the chlorophyll molecule (spectral region of 680-730 millimicrons), although on the basis of existing data it is difficult to form an opinion of what the exact excited level is (triplet or singlet) from which the light quantum is emitted.

We also detected afterglow in the case of photooxidation of chlorophyll in a pyridine solution. The intermediate product responsible for the luminescence in this case is probably a compound like a peroxide forming during the photochemical reaction with participation of the chlorophyll (Tumerman et al., 1962). It is a familiar fact that in the reaction of peroxide with chlorophyll there arises a luminescence whose intensity may be very high, e.g., in the reaction of chlorophyll with benzoyl peroxide. The process of sensitized storage of peroxide



Molecular Mechanism of Ultra-Weak Luminescence of Biological Systems

X, X*, X^{V} -- Molecules in basic, excited singlet, and triplet states; $^{\cdot}X^{-}$ -- ion-radical (e.g., primary photoreduced form of chlorophyll); AH -- molecule of reducing agent (e.g., ascorbic acid); XH -- stable photoreduced product (e.g., secondary "red" form of reduced chlorophyll); XOOH, ROOH -- peroxides (e.g., of chlorophyll and lipids); P_t -- radical responsible for protein thermoluminescence; P_b -- more stable (nonradical?) product responsible for "fading" of protein luminescence; $h\nu$ -- quantum of visible or ultraviolet radiation; O_2 -- molecular oxygen; t° -- heating. Luminescence in inverse photochemical reactions is caused by accumulation of $^{\cdot}X^{-}$ by the direct photochemical process $X \longrightarrow X^{*} \longrightarrow {}^{\cdot}X^{-}$. Luminescence in "dark" biochemical reactions ("biochemoluminescence") probably involves accumulation of $^{\cdot}X^{-}$ by processes of cell metabolism

in the photooxidation of chlorophyll possibly involves oxidation of ion-radical photoreduced form; the electron donor in this case may be a solvent (transitions $^{\cdot}X^{-} \longrightarrow [XOOH] \longrightarrow X^{V} \longrightarrow X + h\nu$ and $XOOH \rightleftarrows ROOH$ in the figure).

Proceeding from the results of investigations made of chlorophyll solutions we endeavored to clarify the nature of weak luminescence occurring when dry chloroplast films are illuminated (Litvin, Vladimiro, et al., 1960). It was established that there would be no afterglow and "thermoluminescence" in the films if the specimens were illuminated in an oxygen-free medium or at a low temperature (-55°C). These findings indicate that chloroplast afterglow and thermoluminescence are likewise caused by the chemoluminescence of chlorophyll photoproducts, not by the purely physical process of storing energy in semiconducting electron traps, as proposed by Arnold et al. (Arnold, Sherwood, 1957).

The chloroplast thermoluminescence effect may be explained by formation of a peroxide product during illumination. This product is relatively stable at room temperature and subsequently decomposes on heating (see Tumerman et al., 1962 and the transitions $ROOH^{t} \longrightarrow XOOH \longrightarrow X^{V} \longrightarrow \longrightarrow X + h\nu$ in the figure).

The protracted afterglow of living photosynthetic organisms (photosynthetic luminescence) detected by Strehler and Arnold (Strehler, Arnold, 1951) may also be regarded as the chemoluminescence of chlorophyll photoproducts in inverse photosynthesis reactions. The mechanism of these processes in vivo may be understood on the basis of the above-analyzed system.

2. Luminescence of Aromatic Acids and Proteins

Ultraviolet irradiation of frozen aromatic amino-acid solutions (at -196°C) causes processes like the above-described in these systems (Roshchupkin, Vladimirov, 1964). The action of light causes pile-up of two photochemical reaction products corresponding to the products P_t and P_b in the figure. Heating of the previously ultraviolet-irradiated specimen to 80-83°K leads to disappearance of product P_t and de-excitation of a quantum from triplet level X^V -- "low-temperature thermoluminescence" (transition $P_t \longrightarrow X^V$ in the figure). The same transition may be effected by replacing heat treatment of the specimen with additional illumination in visible light ("light-induced afterglow"). In more prolonged illumination of the specimen in ultraviolet light the light-resistant photostable product P_b is accumulated which is transformed into the original amino-acid molecule only at higher temperatures (150-200°K). This process is accompanied by an increase in the specimen's photoluminescent intensity (transitions $P_b \longrightarrow X$ in the figure). This last transition ($P_b \longrightarrow X$) is not attended by luminescence -- which at first glance may seem very strange, for if the transition $P_b \longrightarrow P_t$ takes place, then it would seem that this should be followed by the transition $P_t \longrightarrow X^V$ accompanied by the de-excitation of a phosphorescence quantum. The explanation of the lack of luminescence is most probably that ordinary phosphorescence is extinguished. This is sharply marked at a temperature of 150-200°K, for the transition $X^V \longrightarrow X$ becomes non-emittive. It seems to us that the significance of this fact goes beyond the limits of the given specific case. It is possible that the reason for the chemoluminescence yields in most reactions at room temperature is not in the formation of a too small number of molecules in the excited state, but in the low quantum yield of the phosphorescence. The mechanism of the temperature quenching of chemoluminescence is therefore similar to temperature quenching of phosphorescence. From this, of course, it does not yet follow that at ordinary temperatures chemoluminescence must have a negative temperature coefficient since when the temperature is raised there is at the same time a sharp increase in the rate of the reaction responsible for formation of the excited molecules.

The main difference between the luminescence mechanisms of chlorophyll and amino acids in inverse photochemical reactions is apparently that the products of photochemical reaction in amino acids and proteins preserve a substantially larger part of the energy of the absorbed

quantum. The level of the energy corresponding to photoproduct P_t is in this case only slightly lower than the excited level of the original molecule. A slight heating is sufficient to cause the transition $P_t \longrightarrow$ $\longrightarrow X^V$ which is accompanied by de-excitation of the quantum. At room temperature there is no accumulation of product P_t at all. The energy level of photoproduct $^{\cdot}X^-$ is for chlorophyll apparently situated substantially beneath the level of excited molecule X^V. The product $^{\cdot}X^-$ is stable even at room temperature; and the transition $^{\cdot}X^- \longrightarrow X^V$, which is accompanied by luminescence, requires additional energy, which is furnished by oxidation of the ion-radical by atmospheric oxygen. The possibility is not excluded that oxygen may also play a certain role in the case of protein thermoluminescence.

In conclusion it is to be noted that recombination of the photoproducts may be rendered more difficult not merely as the result of a temperature drop, but also because of steric difficulties occurring in the solid phase (without these difficulties the afterglow could not be registered). It is probable that a similar phenomenon may explain the afterglow in dry proteins which we discovered at room temperature (Vladimirov, Litvin, 1959) and which was later investigated by S.V. Konev and M.A. Katibnikov (1961, 1962). The great duration and low intensity of the afterglow in photosynthetic organisms may also be caused by the spatial distribution of the photoreaction products in the chloroplast structures. This mechanism limiting the velocity of inverse recombination processes may be very important in raising the efficiency of photosynthesis.

ULTRA-WEAK LUMINESCENCE IN BIOLOGICAL SYSTEMS IN DARK PROCESSES (BIOLUMINESCENCE)

By means of devices for measuring ultra-weak luminescence it has by now been possible to discover and investigate the luminescence of bean roots (Veselovskiy et al., 1963; Vladimirov, Litvin, 1959; Gasanov et al., 1963), of rat liver homogenates and pulp (Vladimirov et al., 1962; Vladimirov, L'vova, 1964; Polivoda, Sakamova, 1962; Tarusov et al., 1961, 1961a), and that accompanying such enzymatic reactions as decomposition of hydrogen peroxide by catalase and peroxidase in isolated enzymatic systems or in homogenates (Vladimirov et al., 1962).

The mechanism of the processes leading to quantum de-excitation obviously differs in different cases and is still rather unstudied at present. The little which is known of the nature of these processes, however, makes it possible to assume that luminescence in dark processes is of common nature with the luminescence occurring during inverse photochemical reactions, for it likewise involves the oxidation of greatly reduced products and probably the subsequent decomposition of peroxide. This assumption may be illustrated by the example of luminescence developing in rat liver homogenates and pulp, where the process has been

studied in greatest detail (Vladimirov, L'vova, 1964).

In the given case luminescence grew in time and passed through four successive phases. The characteristics of each of the phases with an indication of the process determining the course of the process in each of its stages are given in the table.

The most important thing is that the energy storage preceding quantum de-excitation result from oxidation of substance X of unknown nature by molecular oxygen and that the energy accompanying de-excitation be liberated during decomposition of the oxidized product XO_2. In this respect the luminescence mechanism resembles luminescence in the transitions $^{*}X^{-} \longrightarrow XOOH \longrightarrow X^{V} \longrightarrow X + h\nu$ in the case of photochemoluminescence (see figure).

We unfortunately know nothing about the nature of substance X which is responsible for luminescence. It is probable that it is a strong reducing agent which, however, under ordinary physiological conditions reacts rather slowly with oxygen. The low velocity of this process is apparently explained by two reasons: the high energy barrier of the reaction and by the structural obstacles (spatial distribution of metabolites). This explanation may be corroborated by the following facts: (1) luminescence is not evolved until the cell structures are broken down, i.e., there is a "latent period" in luminescence development (see table); (2) under conditions of cell structure stabilization (in 0.25 M of sucrose + Versene [a chelating agent]) the development of luminescence is inhibited; (3) the oxidation reaction of substance X has an unusually high activation energy -- 57 kcal/mole (Vladimirov, L'vova, 1964).

It is very probable that the existence of such energy and space barriers in the living cell is not a random phenomenon, but a specifically developed mechanism preventing the squandering of valuable chemical energy to be found in the labile intermediate products.

Among the boundless number of cell-metabolism intermediate products two compounds seem to us to be the most probable participants in the processes which are accompanied by ultra-weak luminescence -- nicotinamide-adenine-dinucleotide (NAD) and flavin nucleotides. Derivatives of nicotinamide and flavins possess great luminescence in the 400-550 millimicron region, i.e., in the region characteristic of ultra-weak luminescence. The oxidizing-reducing reactions of these compounds pass through a stage of ion-radicals (semiquinones). Flavins may participate in chemoluminescence processes (Strehler, Shoup, 1953) and they enter into the composition of systems responsible for bioluminescence (Strehler, 1954). The luminescence which we discovered in a suspension of yeast after previous illumination (photochemoluminescence) is also possibly associated with flavins (Vladimirov et al., 1962). This luminescence

Main Features of Development of Ultra-Weak Luminescence in Rat Liver Homogenates and Pulp

Phase of Luminescence Development	Features in Course of the Process	Nature of Process Responsible for Given Phase
Latent period (phase of delayed development of luminescence)	May occur in absence of O_2. Suppressed in presence of 0.25 M saccharose + 10^{-3} M of Versene	Elution of substances from tissue atructures
Phase of growing luminescence	Seen only in presence of oxygen. Suppressed in presence of KCN and ascorbic acid. Increases drastically on heating; activation energy E_A = 57 kcal/mole	Accumulation of oxidized product $X + O_2 \longrightarrow XO_2$
Phase of luminescence decay	Greatly activated by KCN. Suppressed by methylmercaptan and ascorbic acid	Decomposition of oxidized product and de-excitation of quantum $XO_2+Y \longrightarrow Z+h\nu$
Residual luminescence	Slightly activated on heating. Observed only in presence of oxygen	Slow oxidation of unknown substances present in excess, and subsequent reactions

is closely associated with the vital processes of the cells and disappears when the cells are inactivated by heating. The spectral region of the exciting light which induces chemoluminescence and the region of luminescence coincide respectively with the region of absorption and of fluorescence in flavins (Vladimirov et al., 1962).

The assumption that the very same substances (flavin and possibly NAD) may be regarded as the main participants also in the luminescence developing in darkness and in photochemoluminescence is very plausible if the data on electron paramagnetic resonance derived by Commoner and Lippincott (1958) are taken into consideration. According to the research by these authors the formation of free flavin radicals (semiquinones) occurring in the course of dark reactions in the electron transport system is significantly intensified when yeast suspensions are illuminated.

It may be assumed that ion-radicals of substances which are participants in the cell system of electron transport (e.g., reduced flavin enzyme) during the interaction with oxygen form energy-rich compounds

like peroxides which decompose and de-excite a quantum of chemoluminescence. Gibson and Hastings (1962) used chemoluminescence in the presence of luminol to study the formation of peroxides in autooxidation of flavin enzymes.

From the viewpoint of the final result -- the affect of illumination -- it makes no difference whether the formation of ion-radicals capable of autooxidation results from the illumination or during the dark reactions.

LUMINESCENCE IN ENZYMATIC PROCESSES

The autooxidation of powerfully reduced compounds in a biological system leads not only to useless waste of valuable chemical energy, but may cause direct harm to the organism in addition because the peroxide compounds formed in this process are generally poisonous. Perhaps this is precisely the reason why such enzymes as catalase and peroxidase, which specialize in the decomposition of peroxides, exist. We have demonstrated that in the reaction of hydrogen peroxide with catalase, peroxidase, tissue homogenates, and blood luminescence occurs (Vladimirov et al., 1962). It is possible that spontaneous luminescence of liver homogenates likewise does not take place without the participation of these enzymes in the concluding stage of the process, decomposition of the peroxide compounds which accumulated as the result of autooxidation reactions.

It is therefore essential to note that spontaneous decomposition of hydrogen peroxide or its decomposition under the effect of an agent like $KMnO_4$ did not result in the appearance of such strong luminescence as did enzymatic decay (Vladimirov et al., 1962). Probably the path taken by peroxide decomposition is not without significance for the appearance of luminescence. It may be thought that luminescence here, too, occurs during the reaction of intermediate free radicals with atmospheric oxygen which, as is known, is needed for luminescence in the reaction of hydrogen peroxide decomposition by iron-containing compounds in the presence of luminol (White, 1961).

It seems to us that the luminescence which occurs in biological systems under the effect of ionizing radiation and the luminescence in abiological systems have many features in common with the above-considered processes (see reports by B.N. Tarusov, A.I. Zhuravlev, R.F. Vasil'yev, and so on).

THE BIOLOGICAL SIGNIFICANCE OF ULTRA-WEAK LUMINESCENCE

A distinction must be made between two aspects of the meaning of research on ultra-weak luminescence. It may be assumed that ultra-weak luminescence has a definite and probably very important physiological significance in the totality of the vital processes. It is not excluded

that this effect is one of the manifestations of an as yet unknown and fundamentally mechanism of cell process regulation using radiation as a means of communication within the limits of one or several cells (Gurvich, 1934), but it seems more probable to us that radiation by itself is ordinarily useless to an organism and indicates only the inability of a cell completely to keep stored energy from being spent unproductively (under normal conditions the degree of this "leakage" is obviously very insignificant in the total energy balance of the cell).

On the other hand the existence of luminescence indicates that an excited molecule has been formed as the result of a chemical reaction. The perturbation of the electron orbit, hidden in the usual chemical process, acquires the visible form of the jump of the electron between adjacent energy levels. The de-excited quantum in one way or another carries with itself certain information about the energy processes which are unfolding in the electron shells of the participants in the biochemical process. Let this information at first be negligible, as negligible and the strength of the luminescence itself -- the investigators who have devoted themselves to the thankless task of studying ultra-weak luminescence in biological systems may console themselves with the hope that the first ray of light is beginning to penetrate an entirely new field -- the field of study of "dark" biological processes on the submolecular level in the concepts of electron orbits and jumps. It is hard to say what place measurements of ultra-weak luminescence will occupy in research in the future, but it seems unquestionable to us that they are preparing the soil for more fundamental study.

CONCLUSIONS

The results which have been amassed by now in study of the ultra-weak luminescence of biological systems indicates that the molecular mechanism of this luminescence is of a common nature with the mechanism of luminescence in inverse photochemical reactions. Fundamental to biochemoluminescence is the reaction of oxidation of greatly reduced product (probably an ion-radical) with the subsequent decomposition of peroxides:

$$\text{Reduced substance (ion-radical)} + O_2 \longrightarrow \text{peroxide} \xrightarrow{+\text{ reducing agent}} \longrightarrow \text{oxidized product in triplet state} \longrightarrow \text{oxidized product} + h\nu.$$

Storage of peroxides in the living organism is impeded by energy and space barriers, but when these compounds are nevertheless formed by "short-circuiting" the chain of oxidizing reactions they are enzymatically decomposed with concomitant luminescence. Study of the mechanism of ultra-weak luminescence in biological systems is one of the few

approaches to the new field of submolecular biology, where biological processes are regarded from the viewpoint of events occurring in the electron shells of molecules.

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