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Photocatalysis as a phenomenon: Aspects of terminology

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Abstract

The paper considers the terminology used in the application of phenomena generalized under the very wide term "photocatalysis". It is, however, necessary to distinguish more clearly the phenomena of different nature related to both photochemistry and catalysis. An approach to a more correct definition of terms in this area is given. © 1997 Elsevier Science B.V.

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This issue of *Catalysis Today* is devoted completely to photocatalysis and related phenomena, now well known and well recognized by the scientific community [1]. The biogenic photocatalytic phenomenon like natural photosynthesis of plants were known to people even in the prehistoric period (indeed, without realizing the intrinsic mechanisms of plant growth). Abiogenic photocatalytic phenomenon was recognized, possibly, since the beginning of studies on photochemical phenomena, the term "photocatalysis" being introduced into the glossary of science in the early 1930s.

Since then the term "photocatalysis" is used in scientific literature, and for years there was no need for it to be defined more accurately. Usually this term represented the division of chemistry studying catalytic reactions proceeding under the action of light. Therewith, the totality of phenomena connected at the same time with both photochemistry and catalysis was referred to as belonging to the sphere of photocatalysis. For a long period of time such phenomena seemed to be mainly "exotic", and interested only the narrow group of specialists. Nevertheless, over the last 15–20 years the interest towards the natural photosynthesis and chemical methods of solar energy transformation connected with photosynthesis greatly developed our knowledge in this direction and revealed a variety of phenomena related to both photochemistry and catalysis [1]. Therewith many of the phenomena listed appeared to differ qualitatively by their nature and, generally speaking, are to be placed in different divisions of chemistry. For example, it occurred possible to distinguish such phenomena as catalysis of photochemical reactions, photoactivation of catalysts, photochemical activation of catalytic processes, etc. Further suggested is an analysis of terminology in the area of phenomena associated with both photochemistry and catalysis [2,3]. This analysis does not claim to be exhaustive and certainly does not require to follow strictly the suggested terminology, but still it seems to be quite important for creation and unification of the language for

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communication of the representatives of various scientific schools consolidated by the term "photocatalysis".

First of all it should be noted that we really do not know any serious attempts to make a strict definition of the term "photocatalysis". In particular, the word "photocatalysis" is mentioned neither in the latest edition of the "Great Soviet Encyclopedia" (volume 27 appropriate to this word was issued in 1977), nor in the latest edition of the Russian "Encyclopedic Dictionary of Chemistry" (1983). As was noted above, this term is often used to denote either the division of chemistry studying the catalytic reactions proceeding under the action of light, or the overall totality of phenomena connected both with photochemical and catalytic processes. For example, one of the nomenclature commissions of the IUPAC (International Union of Pure and Applied Chemistry) acted in such a way defining the term "photocatalysis" is as follows: "Photocatalysis is catalytic reaction involving light absorption by a catalyst or a substrate" [4]. The same definition of the IUPAC has been given in the last issue of its glossary of terms used in photochemistry [5]. However, simultaneously, here a complementary definition of a photo-assisted catalysis has been given: "Catalytic reaction involving production of a catalyst by absorption of light".

In some cases the attempts were made to oppose the phenomena of "photocatalysis" and "photosynthesis" on the basis of thermodynamic analysis of chemical transformations occurring in the process discussed; thus, it was suggested to attribute to "photosynthesis" the processes where thermodynamic potential of the reaction products is higher than the potential of reagents, and to "photocatalysis" – the processes where potential of the reaction products is lower [6]. Attempts are known to introduce for a quantitative criterion of the phenomenon the "number of turns" of a photocatalyst in the course of the process. However, it is evident that the basic definitions of a physico-chemical phenomenon must not depend upon thermodynamic and other qualitative properties of the processes.

Apparently, the conventional simplified terminology will be still widely used in the scientific literature. Nevertheless, this terminology has some evident drawbacks. In particular, the usage of simplified terminology immediately gives rise to the question as to what is the "photocatalyst" in the process of photocatalysis.

In our opinion, a semantic analysis of the terms becomes much more fruitful and definitions – more unambiguous if one starts not from the notion "photocatalysis", but just from the notion "photocatalyst". Note that productivity of this approach has been demonstrated also when defining the notions "catalysis" and "catalyst". Classical definitions formulated by academician Boreskov [7], the incontestable Russian authority on the science of catalysis, are based just on the primarity of the definition of the notion "catalyst": "Catalysis is a change in the rate of chemical reactions in the presence of substances (catalysts) which come into intermediate chemical interactions with the reactants, but restore their chemical composition after each cycle of the intermediate interactions. Reactions with the participation of catalysts are called catalytic". A similar definition of the phenomenon of catalysis through the definition of the notion "catalyst" was also formulated by another eminent representative of the Russian science of catalysis Krylov [8].

Note that the given definition is universal in the sense that it allows gradually, as the mechanism of the particular catalytic process will be better understood, to refine step by step the content of the notion "catalyst" for this process. For example, separating in the substance "heterogeneous catalyst" a sequentially inactive carrier, an active component of the catalyst, an active center of the catalyst, etc., until it becomes possible to describe an elementary act of the catalytic reaction.

Following this approach and starting from the fact that classical and evident phenomenon relating to photocatalysis is the natural plant or bacterial photosynthesis, we put forward the following definition: photocatalyst is a substance that is able to produce, by absorption of light quanta, the chemical transformations of the reaction participants, repeatedly coming with them into the intermediate chemical interactions and regenerating their chemical composition after each cycle of such interactions [1,2,9].

Such a definition, along with the definition of the notion "catalyst" made by Boreskov, enables to refine the sense of this term as the object is better understood.

In particular, for the natural photosynthesis[2] it will gain the following hierarchy of the content of the term "photocatalyst":

Plant (photosynthesizing bacteria)
↓
Photosynthesizing cell
↓
Chloroplast
↓
Photosynthesizing reaction center of Photosystem I or II
↓
Chlorophyll molecule (dimer of molecules) performing the initial photoseparation of the charges

In our opinion, the above cited definition of the term "photocatalyst" is rather convenient also for the artificial photocatalytic systems, since in the construction of these systems the substance (and sometimes even a molecule) that is acting as a photocatalyst is usually apparent. Therewith, starting from the methodology of Boreskov [7] and Krylov [8], the following definition of the phenomenon of photocatalysis is given:

Photocatalysis is a change in the rate of chemical reactions or their generating under the action of light in the presence of the substances (photocatalysts) that absorb light quanta and are involved in the chemical transformations of the reaction participants, repeatedly coming with them into intermediate interactions and regenerating their chemical composition after each cycle of such interactions. (Similar definition of this term is also made in [13].)

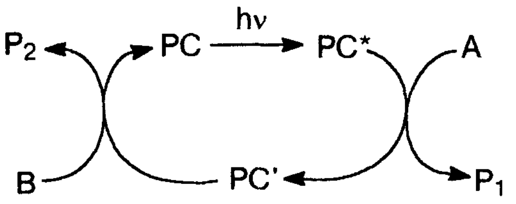
An appropriate scheme of the phenomenon of photocatalysis and of the photocatalyst (PC) action for the typical photocatalytic reaction

$$A + B \xrightarrow[PC]{h\nu} P_1 + P_2, \qquad (1)$$

in this case may be presented in the following steps:

$$\begin{aligned} &PC + h\nu \rightarrow PC^*, \\ &PC^* + A \rightarrow PC' + P_1, \qquad (2) \\ &PC' + B \rightarrow PC + P_2, \end{aligned}$$

or in the following generalized form with explicit display of the cyclic nature of the chemical interactions of a photocatalyst:



(3)

Here PC^* is the form of a photocatalyst induced by light quanta (usually electronically excited), PC' is the form of a photocatalyst after the chemical interaction of its excited form with the initial reagent A. Evidently, the light quantum serves here as a sort of reagent and thus the quantum yield of the products in a typical photocatalytic reaction cannot exceed a unit when taking into account the light absorbed by the photocatalyst. (Curiously, in [14] for denoting a photocatalyst it is suggested to use the terms "photoassister" or "pseudocatalyst", since the electron-induced form of PC can aid in carrying no more than one cycle of the chemical transformations.)

Note that in schemes (2) and (3) it is supposed that the simplest concrete case of the regeneration of a PC forms by the second initial reagent B, while in concept the other ways of regeneration of this form are possible as well.

Alongside with schemes (1) and (2) of the typical photocatalytic reactions, one could also suggest even more simple schemes for a primitive photocatalytic reaction like

$$A \xrightarrow[PC]{h\nu} P.$$

The corresponding mechanism of such photocatalytic reaction would be either

$$\begin{aligned} &PC + h\nu \rightarrow PC^*, \\ &A + PC^* \rightarrow P + PC, \qquad (4) \end{aligned}$$

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[2] It can be noted that the notion "photosynthesis" also has no commonly accepted definition. Most often this notion is treated as applied only to natural photosynthesis, for example: "Photosynthesis is the production by the superior plants, algae and photosynthesizing bacteria, of complex organic substances from the plain compounds (e.g., carbon dioxide and water) at the cost of light energy absorbed by chlorophyll and other photosynthesizing pigments" [10] (see also [11]). Interestingly, even in such classical monographs on the natural photosynthesis as [12], the direct definition of this notion was avoided.

or

$$A + PC \rightarrow [A \cdot PC],$$
$$[A \cdot PC] + h\nu \rightarrow [A \cdot PC]^*, \qquad (5)$$
$$[A \cdot PC]^* \rightarrow P + PC.$$

One should note that scheme (4) implies, in contrast to the below cited schemes (6)–(10) of photosensitization phenomena, a direct chemical transformation of substrate A during the interaction with the excited (thermodynamic nonequilibrium) form of PC rather than transfer of electronic excitation from PC^* onto A. A peculiarity of scheme (5) is absorption of light not by the PC itself but by its photosensitive complex with the reaction substrate. Schemes (4) and (5) may take place for some homogeneous photocatalytic reactions assisted by complexes of transition metals (see review [3]) as well as for some localized heterogeneous photocatalytic reactions of adsorbed reactants on isolator oxides, etc.

The above treatise of the phenomenon of photocatalysis allows to distinguish it from the conventional or "simple" photochemical processes where the chemical transformations are carried out after the electron excitation or photoionization as a result of absorption of light quanta immediately by the participants of the chemical transformations (by the substrate molecules) [15,16].

In this case, for example, of the variants of a simple photochemical (not photocatalytic) proceeding of the reaction, similar to (1),

$$A + B \xrightarrow{h\nu} P_1 + P_2, \qquad (6)$$

would be presented by the schemes

$$A \xrightarrow{h\nu} A^*,$$
$$A^* \rightarrow P_1 + C, \qquad (7)$$
$$C + B \rightarrow P_2$$

or

$$A \xrightarrow{h\nu} A^*,$$
$$A^* + B \rightarrow P_1 + P_2, \qquad (8)$$

where A^* is a form of the light absorbing substance A induced by the light quanta (electronically excited or photoionized); C is an intermediate product of the photochemical transformation of substance A. Note that the schemes of the "simple" photochemical reactions in the generalized form do not include the cyclic transformations of the excited substance of the type indicated in scheme (3).

Definition of the term "photocatalyst" given above allows to separate the phenomenon of photocatalysis also from the totality of the more universal photocatalysis phenomena of photosensitization of chemical reactions.[3] Actually, even though the definition given to the phenomenon of photosensitization is rather similar in a sense to the foregoing definition of photocatalysis (in [17], e.g., a rather proper definition is cited: "Photosensitized reactions are carried out under the action of light absorbed not by the reacting substances themselves, but by the molecules of another compound – sensitizer", see also [4]), it is generally implied that the effect of the photosensitizing agent resides in the fact that the molecules of sensitizer excited by light may initiate the reaction via *transfer of the energy of excitation* (emphasized by Parmon) to the molecule's reagents, with the result that the latter pass into excited state [17] (see also [4,16]). Therefore chemical interaction of the excited particles of a photosensitizer S with the reacting agent is considered merely as a nontypical case of a photosensitizer action. It should also be noted that when defining the term "photosensitizer" it is not nearly always emphasized that a photosensitizer is not consumed in the overall photosensitized process. So often we have the cases where the term "sensitizer" is taken to mean the agent initiating under the action of light the transformations in question, this agent being consumed therewith (see, e.g., [18]). Probably, here the term "photoinitiator" seems to be more appropriate [4].

Thus, when examining the photosensitized reaction

$$A + B \xrightarrow[S]{h\nu} P_1 + P_2, \qquad (9)$$

totally identical to reaction (1) in terms of the formal equation notation, in case of the typical mechanism of

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[3] In this paper the phenomenon of photosensitization is considered in a broad sense, but not as the more particular phenomenon of sensitization of a system to a definite part of the light spectrum, attainable in certain cases through the change in photophysical properties of some elements of the system.

its occurrence the following scheme is implied:

$$S \xrightarrow{h\nu} S^*,$$

$$S^* + A \rightarrow S + A^*, \qquad (10)$$

and further similar to schemes (7) and (8); the mechanism represented by schemes (2) and (3) is supposed to be nontypical. The most "classical" examples of the photosensitized reactions are the reactions initiated by mercury vapor excited by UV-light and by the triplet-excited molecules of the aromatic [2,14,16].

Sequence (10) enables to represent the transformations of the excited states S^* as a cyclic scheme similar to (3), having as its product an electronically excited state of the substrate molecule, but with substantial difference: regeneration of the initial state S is attained via a physical process – deactivation of the excited state. Interestingly, transfer of the electron excitation may occur in an "exciplex" – an intermediate complex of the electron-induced particle of photosensitizer with the molecule to which excitation is transferred, i.e., by way of chemical interaction of photosensitizer and substrate (see, e.g., [19]).

Therefore, the phenomenon of photocatalysis may be defined also as a special case of the phenomenon of photosensitization, when the excited particles of a photosensitizer do not ensure the transfer of excitation, but initiate the chemical transformations through the chemical interactions with the molecules of reagents [1–3].

The most typical version of the elementary chemical interaction of the excited particles of photosensitizer (photocatalyst) is the transfer of an electron from these particles to the reagent molecule (and inversely) and thus providing some redox reactions [1–3,9,14,16,17,20]. The appropriate scheme of the photosensitizer–photocatalyst operation may be presented in the following form:

$$PC \xrightarrow{h\nu} PC^*,$$

$$PC^* + A \rightarrow PC^{\pm} + A^{\mp}, \qquad (11)$$

$$PC^{\pm} + B \rightarrow PC + B^{\pm},$$

that is currently conventional also for the sequence of elementary reactions initiating the chain of secondary chemical transformations in the natural photosynthesis.

Obviously, the agent photocatalyst may include a photosensitizer (in a general sense of this term) as one of the components of a photocatalyst, if the notion "photocatalyst" applies to the integral, i.e., comprising several components, subunit of the system. For example, if in case of the natural photosynthesis the notion "photocatalyst" is applied to the photosynthesizing reaction center, then the molecules of the "antenna" chlorophyll belonging to this reaction center [12] are evidently the photosensitizers in respect to the molecules (dimers of molecules) of chlorophyll, performing the primary photoseparation of charges, i.e., to the molecules photocatalysts of more deep level in the hierarchy of notions.

The definitions of photocatalysis and photosensitized reactions cited above allow to formulate rather correct definitions also for some other phenomena associated concurrently with photochemistry and catalysis.

Among such phenomena worthy of mention are the following.

Photoinduced (photoinitiated) catalysis is the initiating of chemical transformations, through the formation in the system under the action of light quanta (photochemical processes) the substances, which are catalytically active even without the light quanta affecting them. (In [4,5] such reactions are suggested to be generalized by the term "photoassisted".)

When writing the process of photoinduced catalysis by the common equation

$$A + B \underset{K}{\overset{h\nu}{\rightarrow}} P_1 + P_2,$$

similar to Eqs. (1) and (9), the sequence of the reactions occurring therewith principally differs from schemes (2), (3), (10) and may, e.g., be the following:

$$K \xrightarrow{h\nu} K^*,$$

$$K^* \rightarrow K', \qquad (12)$$

$$A + B \underset{K'}{\rightarrow} P_1 + P_2,$$

where by symbol K′ is meant the "traditional" "dark" catalyst formed through a photochemical transformation of substance K (see also [1,14]). The qualitative criterion enabling to recognize easily the processes associated with the photoinduced catalysis, is the possibility of attaining here the quantum yields considerably exceeding a unit when taking into account the light absorbed by the substance K which is the precursor of the catalyst K′.

Reactions represented by scheme (12) are familiar. The most representative examples are the reactions of hydrogenation and isomerization of olefins catalyzed by the products of photochemical destruction of the transition metal carbonyl complexes (see, e.g., reviews [3,13,14,21]). Extremely numerous are the examples of photoproduction of colloids or photoprecipitation of the particles of catalytically active metals from their salts and complexes (see, e.g., a review [22]).

Very close to the phenomenon of photoinduced (photoinitiated) catalysis is the phenomenon of the catalyst photoactivation (photomodification) that may be defined as increasing (alteration) of the catalytic activity (selectivity) of the substance after it was affected by light quanta. Formally, a sequence of processes occurring therewith may be written by a scheme similar to (12). Typical examples of the catalysts photoactivation are the above mentioned photoprecipitation of noble metals over semiconductor suspensions through photoreduction of salts or complex compounds of metals in question (see, e.g., reviews [22,23]), as well as photogeneration on the surface of heterogeneous photocatalysts or oxide carriers of catalytically active centers containing, e.g., the ions of transition metals at the "unusual" oxidation degrees or the active structures (see, e.g., review [23]).

It is also known that the phenomenon of catalysis (promotion) of photochemical reactions may be defined as the raising (alteration) of the efficiency of the photochemical reactions via an intermediate interaction of photochemically active particles with certain compounds that play the role of "catalysts" (promoters) of the appropriate chemical transformations of the former [13,14]. An attempt to formulate the definition of a catalyst of photochemical reactions was made in [13]: "A catalyst of the photochemical reaction is a substance that manifests itself in expression for the quantum yield of reaction from a certain excited state at the degree exceeding its coefficient in the stoichiometric equation".

Processes of this type are also familiar. As particular examples one can mention an increase in ability to photochemical oxidation of copper(I) cations in aqueous solutions through formation of halide complexes of these cations [24,25] and at the same time alteration of chemical activity of some organic compounds through protonation (deprotonation) of their molecules, formation of Lewis complexes, etc. [13,26]. "Catalysis" of photochemical reactions is made possible also due to the effect on the secondary chemical processes that changes the resulting quantum yields of the ultimate products of chemical and photochemical transformations [13].

The phenomenon of redox photocatalysis is in close proximity to the phenomenon of photoelectrochemical transformations (photoelectrocatalysis) (see, e.g., [2,4,5,27,28]) that usually has the same physico-chemical nature. The role of the photocatalyst in the latter case is played by a photoelectrode, which is a semiconductor, as a rule. Evidently, as an essential distinction of these phenomena it may be considered only the fact that at photoelectrochemical (photoelectrocatalytic) transformations it is possible to distinguish the system "electrode–electrolyte" and a rather explicit electrical circuit (generally macroscopic) for the charge transfer; for photocatalysis it is generally unattainable. Conditionality of the distinctions in question becomes evident when considering the mechanism of the semiconductive photocatalysts action: in case of highly dispersed particles of such photocatalysts the phenomenon is closer to "photocatalysis", and for the larger ones it is closer to "photoelectrolysis" (see, e.g., [22]).

Since the term "photosynthesis" (see above) gained wide acceptance when applied not only to natural but also to artificial systems and objects, it would be proper to interpret this term generally as a synthesis of more complex compounds from the less complex ones under the action of the light quanta. In such a case, photosynthesis is not necessarily associated with the photocatalytic transformations, but have to include photochemical transformations as a possible element. Nevertheless, in practice the processes combined by the terms like "artificial photosynthesis" yet appear to be bound just with the photocatalytic processes in the exact meaning of this notion (see, e.g., [22,29]).

Naturally, the phenomena listed above do not exhaust all the variety of the processes concerning overlaps in the area of both photochemistry and catalysis. Undeniably, to the sphere of heterogeneous photocatalysis are attracted all the phenomena connected with the surface photochemistry (for instance, its photoreduction or photo-oxidation [30]), as well as the phenomena of photoabsorption and photodesorption (see, e.g., [31–33]). It is pertinent to note also that when using rigid quanta of light, beginning probably with the region of vacuum UV, the phenomenon of photocatalysis may be distinguished from the phenomenon of radiation catalysis only by convention. In the radiation catalysis the primary interaction of the irradiation with the catalyst is generally its ionization rather than electronic excitation.

In quantitative description of the photocatalytic reactions the widely used terms are "photocatalytic activity" of a system and "quantum yield of a photocatalytic process".

Quantum yield of a photocatalytic process may be defined as the quantum yield of a photochemical process, i.e., as the number of molecules of this process, products formed per one quantum of light with the given energy, that is absorbed by a photocatalyst (see also [34–36]).

Due to the complexity of photocatalytic systems and often impossibility or conditionality of separation in the system of subunits of the "real" photocatalyst and the portion of light absorbed by these subunits, the expediency of using the term "photocatalytic activity of a system" or "photocatalytic efficiency" become clear. This term may be defined as the number of molecules of the photocatalytic process, products formed per one quantum of light with the given energy, that are incident on the photocatalytic system. Obviously, as it is in "traditional" catalysis, the rise in photocatalytic activity of a system may be caused by an improvement of the system as a whole. More sophisticated approaches to quantitative characterization of the photocatalyst action may be found in [35,36].

When completing the analysis of terminology in the area of photocatalysis, note that in all the definitions listed above, the effect is emphasized not just on the light but on the quanta of light. The necessity for such specification is due to the fact that in the case of light with high density of flux it is possible that not only photochemical (quantum) processes would proceed but also the induced thermochemical ones as a result of the system heating through absorbing the energy of light. Often this occurs on exposure to the IR-light or pulse laser emission [32]. These conditions of the chemical systems functioning are wide spread also for the real catalytic converters of concentrated fluxes of solar energy, especially when catalysts are directly affected by the dense light fluxes (see, e.g., [37]). In this connection in certain cases it is evidently worthwhile to use the terms like "thermophotocatalysis" or "light initiated thermocatalytic phenomena" which emphasizes that the phenomenon of (photo)catalysis occurs in circumstances when the system is heated by light too.

This issue of *Catalysis Today* presents only a few examples of modern research in many mentioned directions of photocatalysis and light-driven related catalytic phenomena. However, one could expect that even these few examples can show how large the area of photocatalytic studies is now, spreading from solution of particular environmental problems to the global problems of the Earth's atmosphere as well as the problem of efficient transformation of solar light.

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