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CATALYSIS: A FUNDAMENTAL PHENOMENON AT THE INTERFACE BETWEEN SCIENCES AND DISCIPLINES

Abstract. *The article presents a study of previously obtained laws of catalysis for their fundamental and interdisciplinary nature. It follows from the laws of catalysis that catalysis belongs to the class of chemical and physical phenomena. The laws of catalysis and the formulas for calculating TOF and TON are combinations of quantities of chemical and physical nature. The main mystery of catalysis for many years has been the undisclosed role of oxidation states in the mechanism of catalysis. The field of application of oxidation states in catalysis has expanded. The oxidation states have been used for a new purpose as parameters in the laws and formulas of catalysis. It has been shown that in catalysis it is necessary to consider not only the oxidation states of the catalyst, but also the oxidation states of the reactants. The concept of oxidation states as quantitative values proved to be the main missing link that made it possible to obtain the laws of catalysis. The key role of the oxidation states of the catalyst and reagents in the donor-acceptor mechanism of catalysis has been demonstrated. The list of oxidation states of chemical elements known in chemistry can be applied as a tool for the selection of catalysts. The role and place of electric charges in the mechanism of catalysis and in the laws of catalysis have been shown. A new field of application of Faraday's constant in chemistry is outlined. In addition to its well-known use in the law of electrolysis and in the Nernst and Goldman equations, the Faraday constant has marked itself in catalysis and is included as an interaction constant in the laws of catalysis. The signs of the fundamentality of catalysis are given and the place of the laws of catalysis in the family of fundamental physical laws is shown. The laws of catalysis complement the family of fundamental laws of Nature.*

Key words: *heterogeneous catalysis, homogeneous catalysis, laws of catalysis, universal law of catalysis, field-induced catalysis, catalysts, donor-acceptor mechanism of catalysis, electric charge, oxidation states in catalysis, Faraday's constant in catalysis.*

1. Introduction

The history of catalysis goes back more than 200 years. It began long before the generalization of Berzelius, J. J. [1] with the articles of Kirchhoff, S. K., Thenard, L., Davy, H., Döbereiner, J.W., Faraday, M. [2 - 6]. Currently, the study of catalysis has become a global field of activity, in which

many thousands of researchers around the world are involved. Despite the long history of catalysis, the scientific aspects of this phenomenon are underdeveloped [7 - 17]. In 2 centuries of research on catalytic phenomena, the general mechanism inherent in different types of catalysis has not been revealed. A general theory of catalysis has not been created and the laws of catalysis have not been discovered [7, 8, 9]. This holds back the development of industrial catalysis. There is a paradoxical situation in catalysis: a widespread and long-known phenomenon does not have a strict scientific substantiation. The reason for this unusual situation in modern science should be sought in the undisclosed nature of catalysis and the striking variety of catalytic processes [10, 17].

The history of catalysis shows that many fundamental discoveries in catalysis were made either accidentally or empirically by enumerating a large number of substances in extensive and expensive experiments. No quantitative theory of catalysis has been created that can predict the catalytic properties of substances and calculate the rates of catalytic reactions [8 - 10]. Although the number of different catalysts is in the thousands, the general mechanism of their catalytic action remains undisclosed. Successful "*recipe*" of catalysts have been created by trial and error, rather than by scientific analysis of the processes involved [8, 9].

Such a situation does not add faith to the scientificity of catalysis. In the specialized literature on catalysis, one can encounter statements that "*catalysis is more of an art than a science*" [8 - 12] or "*the creation of a general theory of heterogeneous catalysis is hardly possible*" [13]. Such a pessimistic viewpoint on the scientific status of catalysis is not conducive to the development of the science of catalysis.

The wide spread of catalysis in nature (biochemical reactions, crystallogenes of diamonds in nature, the phenomenon of photosynthesis, and nitrogen fixation) points to the objective nature of catalytic processes and to the fact that catalysis is subject to objective natural laws. The science of catalysis has yet to discover these laws. The discovery of objective laws of catalysis will give impetus to the development of industrial catalysis. With the laws of catalysis in place, the catalytic phenomenon can become a scientific field in its own right. In this case, the intuitive choice of industrial catalysts will be replaced by a scientific choice.

The huge amount of experimental material accumulated during the long history of catalysis and the extraordinary variety of catalytic phenomena need to be generalized and systematized. There is an urgent need to identify common features in a large family of different types of catalysis. There is a need for the disclosure of general principles of the functioning of catalytic systems. There is a need to discover the natural laws of catalysis and to apply them in industrial catalysis.

These problems are not tasks in one action. For example, to discover the laws of catalysis, a chain of interdependent problems must be solved. The laws of catalysis are mathematical models of processes that are implemented in the mechanism of catalysis. This means that success in discovering the laws of catalysis depends to a large extent on discovering the actual mechanism of catalysis. In turn, the correct choice of the mechanism of catalysis depends to a large extent on the correct definition of the nature of catalysis, i.e., it is necessary to answer the question: *to what science should catalysis be referred?* The correct definition of the nature of catalysis is the first and main link in the strategy of searching for the laws of catalysis.

2. Catalysis - a chemical-physical phenomenon at the interface between sciences and disciplines

In no science have so many failed attempts been made to develop a scientific theory to explain just one widespread phenomenon as has occurred in catalysis. Examples of failed theoretical concepts of catalysis include the Berzelius' theory, the Mendeleev's theory, the adsorption theory of catalysis by Faraday and Schwab's, the Sabatier's theory, Taylor's theory of active centers, Balandin's multiplet theory, Kobozev's theory of active ensembles, Dowden and Volkenstein's electronic theory of catalysis, Semenov and Voevodsky's chain theory of catalysis, Roginsky's theory, Langmuir's catalysis theory, etc.

One of the reasons that catalysis has no general scientific theory and the laws of catalysis have not been discovered is that catalysis has been tried to be studied within the framework of one science, either chemistry or physics. This is evidenced by the listed theories of catalysis, which can be divided into two groups - physical theories and chemical theories. The physical theories of catalysis include the theories of catalysis by Faraday, Schwab, Dowden, Wolkenstein, and Langmuir. They relied on the physical nature of catalysis. Other theories relied on the chemical nature of catalysis.

Neither chemical nor physical theories led to a general theory of catalysis. Neither chemical nor physical theories led to the discovery of a unified mechanism for catalytic processes. None of the known theories led to the discovery of the laws of catalysis. Physicists paid more attention to physical processes in catalysis and underestimated the role and place of chemical processes. Chemists paid more attention to chemical processes in catalysis, and underestimated the role and place of physical processes. The modest results of only physical and only chemical theories of catalysis can be explained by the fact that catalysis is a complex interdisciplinary phenomenon and its nature remains undiscovered. The description of catalysis is beyond the scope of a single science [18 - 21]. For this reason, its study within the framework of physics or chemistry alone could not lead to the discovery of the laws of catalysis. Catalysis needs a generalization of known theoretical concepts and their integration into an interdisciplinary quantitative theory of catalysis.

3. The interdisciplinary nature of the laws of catalysis.

The laws of catalysis were obtained in [22 - 32]. Let us examine the laws of catalysis for their fundamental and interdisciplinary nature.

The rate laws of heterogeneous, homogeneous, field, and combined catalysis have the form:

$$v_{He} = \frac{e \bullet n_a \bullet |k_1 - k_2|}{F \bullet (\tau_D + \tau_A) \bullet m_1 \bullet |z_1 - z_2|} \quad (1)$$

$$v_{Ho} = \frac{e \bullet n_a}{F \bullet (\tau_D + \tau_A) \bullet m_1 \bullet |z_1 - z_2|} \quad (2)$$

$$v_{Fcat} = \frac{e \bullet f_e \bullet E_{cat}}{F \bullet m_1 \bullet |z_1 - z_2| \bullet E_i} \quad (3)$$

$$v_{Comb} = \frac{e \bullet n_a \bullet (|k_1 - k_2| + \dots + |r_1 - r_2|)}{F \bullet (\tau_D + \tau_A) \bullet m_1 \bullet |z_1 - z_2|} \quad (4)$$

where: v_{He} - reaction rate of heterogeneous catalysis (mol/s); v_{Ho} - reaction rate of homogeneous catalysis (mol/s); v_{Fcat} - reaction rate of field catalysis (mol/s); v_{Comb} - reaction rate of

combined catalysis (mol/s); n_a - number of catalyst active centers; F - Faraday constant; e - electric charge of electron; τ_D - time of donor half-cycle of catalysis; τ_A - time of acceptor half-cycle of catalysis; f_e - frequency of field influence on reagents; E_{cat} - field energy spent on catalysis during one cycle of catalysis (J); E_i - ionization energy of reactants (J); k_1 - initial oxidation state of catalyst; k_2 - final oxidation state of catalys; r_1 - initial oxidation state of catalyst; r_2 - final oxidation state of the catalyst; z_1 - oxidation state of the reactant in the initial product; z_2 - oxidation state of the reactant in the final product; m_1 - number of reactant atoms in the molecule of the final product; t - catalytic reaction time.

By generalizing these laws, a universal law of the catalytic rate was obtained for different types of catalysis [32]. The universal law of catalysis has the form:

$$v_{cat} = \frac{Q_{\Sigma}}{F \bullet t \bullet m_1 \bullet |z_1 - z_2|} \quad (5)$$

where: Q_{Σ} - is the value of the total electric charge transferred to the reactants during the catalytic reaction.

From the universal law of catalysis (5), the laws of heterogeneous, homogeneous, combined, and field catalysis follow as private results (Fig. 1.). The equations of the laws of catalysis rate and the values of chemical and physical nature included in them show that these are chemical-physical laws. They are represented by a combination of chemical and physical quantities (Fig. 1).

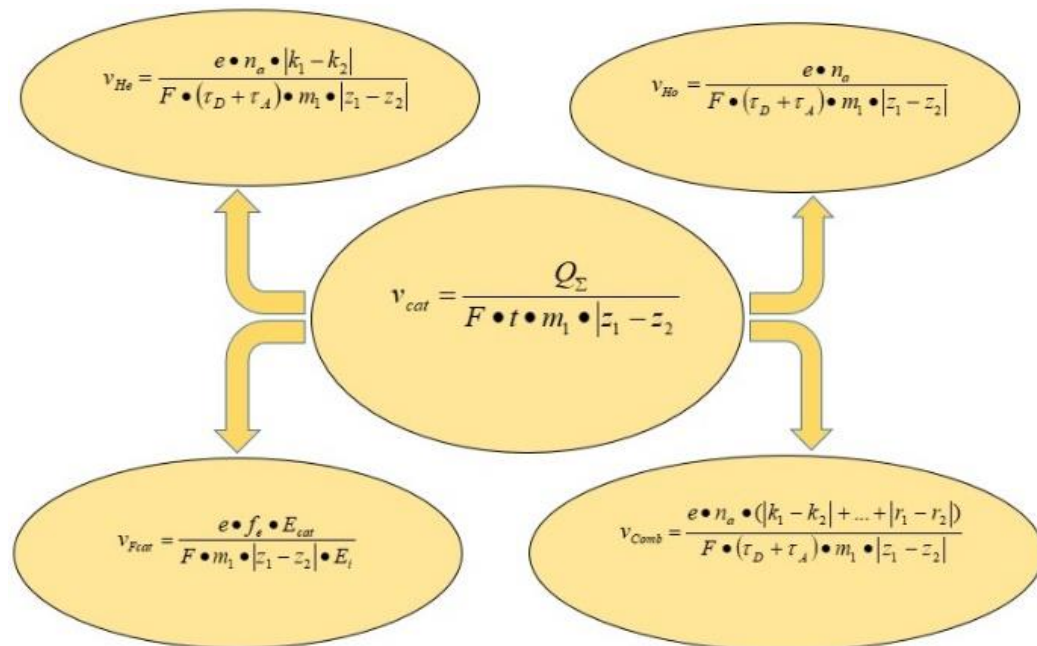


Fig. 1. The laws of catalysis rate are chemical-physical laws.

The laws of catalysis are quantitative mathematical models of chemical-physical processes that are realized in a catalytic reaction. Ratios for the reaction yield of heterogeneous, homogeneous, field, and combined catalysis are derived from the catalysis rate laws [27 - 32]:

$$n_{He} = \frac{e \bullet n_a \bullet t \bullet |k_1 - k_2|}{F \bullet (\tau_D + \tau_A) \bullet m_1 \bullet |z_1 - z_2|} \quad (6)$$

$$n_{Ho} = \frac{e \bullet n_a \bullet t}{F \bullet (\tau_D + \tau_A) \bullet m_1 \bullet |z_1 - z_2|} \quad (7)$$

$$n_{Fcat} = \frac{t \bullet e \bullet f_e \bullet E_{cat}}{F \bullet m_1 \bullet |z_1 - z_2| \bullet E_i} \quad (8)$$

$$n_{Comb} = \frac{e \bullet n_a \bullet (|k_1 - k_2| + \dots + |r_1 - r_2|) \bullet t}{F \bullet (\tau_D + \tau_A) \bullet m_1 \bullet |z_1 - z_2|} \quad (9)$$

From the universal law of the rate of catalysis, a generalized relationship for the yield of the catalytic reaction is derived:

$$n_{cat} = \frac{Q_{\Sigma}}{F \bullet m_1 \bullet |z_1 - z_2|} \quad (10)$$

From the generalized law (10), as private results, the reaction yields of heterogeneous, homogeneous, combined, and field catalysis follow (Fig. 2). It was shown in [32] that Faraday's law of electrolysis directly follows from the generalized law of catalysis (10) as a private result, in addition to the laws of catalysis. This may indicate that electrolysis belongs to the class of catalytic processes.

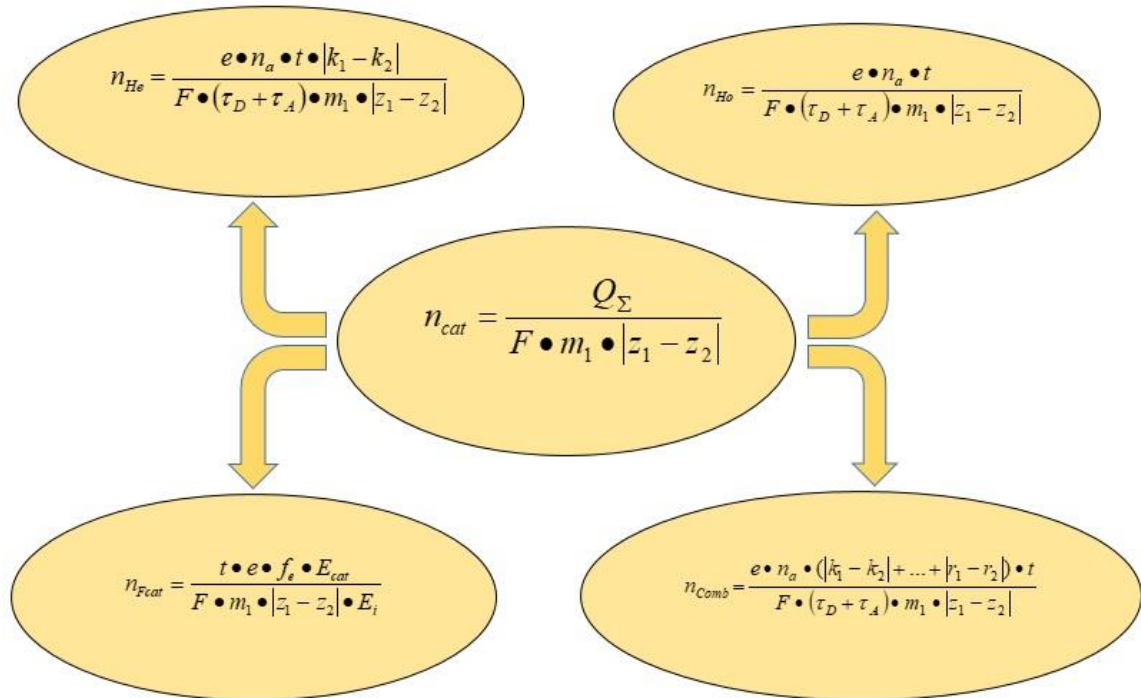


Fig. 2. Chemical-physical formulas of the catalysis reaction yield.

The catalysis reaction yield equations (6) - (10) and the values included in them show that they are chemical-physical laws. They, as well as the laws of the rate of catalysis (1) - (5), are represented by a combination of chemical and physical quantities.

4. The chemical-physical formulas of TOF and TON

From the laws of catalysis rate in [22 - 32], the ratios for calculating the TOF and TON were derived. The formulas for calculating the TOF in heterogeneous, homogeneous, field, and combined catalysis are as follows:

$$TOF_{He} = \frac{|k_1 - k_2|}{(\tau_D + \tau_A) \bullet m_1 \bullet |z_1 - z_2|} \quad (11)$$

$$TOF_{Ho} = \frac{1}{(\tau_D + \tau_A) \bullet m_1 \bullet |z_1 - z_2|} \quad (12)$$

$$TOF_{Fcat} = \frac{f_e \bullet E_{cat}}{m_1 \bullet |z_1 - z_2| \bullet E_i} \quad (13)$$

$$TOF_{Comb} = \frac{|k_1 - k_2| + \dots + |r_1 + r_2|}{(\tau_D + \tau_A) \bullet m_1 \bullet |z_1 - z_2|} \quad (14)$$

From the universal law of the rate of catalysis (5), a generalized formula for calculating the TOF is obtained, which has the form:

$$TOF_{cat} = \frac{Q_\Sigma}{e \bullet n_a \bullet t \bullet m_1 \bullet |z_1 - z_2|} \quad (15)$$

From the generalized formula for TOF (15), as private results, follow formulas for calculating the TOF of heterogeneous, homogeneous, combined, and field catalysis (Fig. 3.).

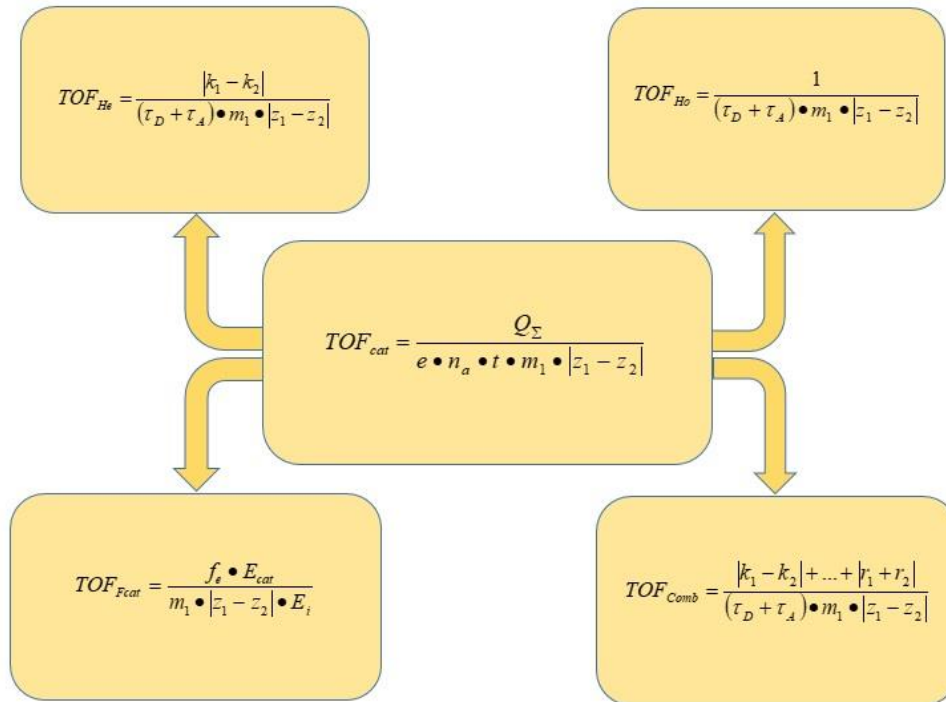


Fig. 3. Chemical-physical formulas for calculating the TOF .

The formulas for calculating the TOF are a combination of chemical and physical quantities.

The formulas for calculating TON in heterogeneous, homogeneous, field, and combined catalysis are as follows:

$$TON_{He} = \frac{|k_1 - k_2| \cdot t}{(\tau_D + \tau_A) \cdot m_1 \cdot |z_1 - z_2|} \quad (16)$$

$$TON_{Ho} = \frac{t}{(\tau_D + \tau_A) \cdot m_1 \cdot |z_1 - z_2|} \quad (17)$$

$$TON_{Fcat} = \frac{t \cdot f_e \cdot E_{cat}}{m_1 \cdot |z_1 - z_2| \cdot E_i} \quad (18)$$

$$TON_{Comb} = \frac{(|k_1 - k_2| + \dots + |r_1 + r_2|) \cdot t}{(\tau_D + \tau_A) \cdot m_1 \cdot |z_1 - z_2|} \quad (19)$$

The generalized formula for calculating the TON is as follows:

$$TON_{cat} = \frac{Q_\Sigma}{e \cdot n_a \cdot m_1 \cdot |z_1 - z_2|} \quad (20)$$

From the generalized formula TON (20), as private results, follow formulas for calculating the TON of heterogeneous, homogeneous, combined and field catalysis (Fig. 4.).

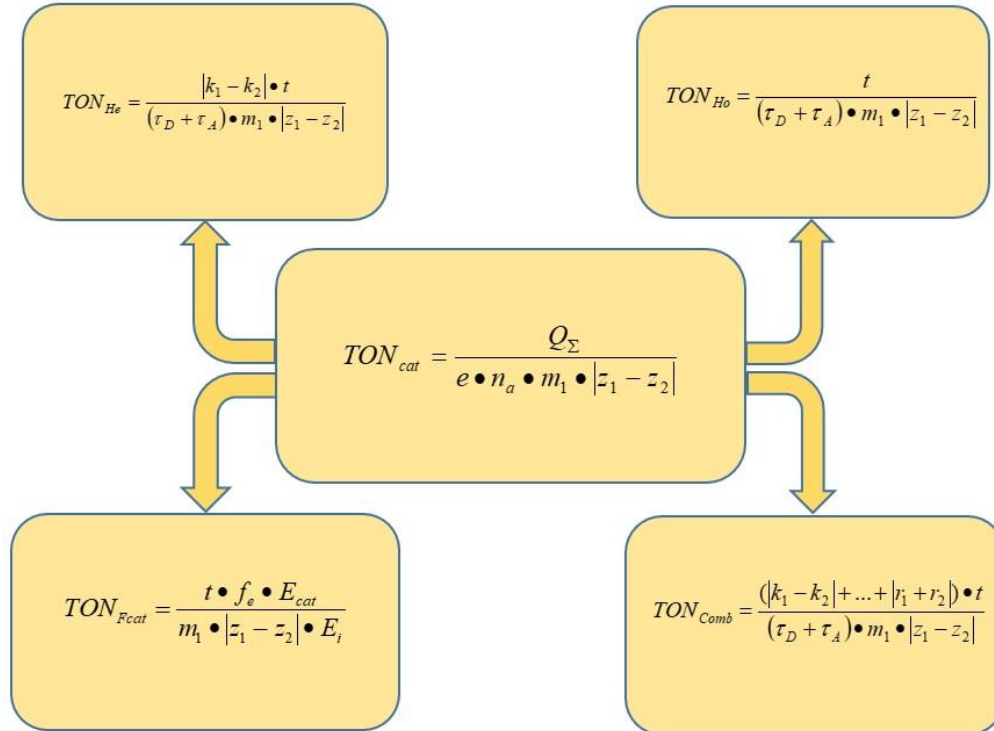


Fig. 4. Chemical-physical formulas for calculating the TON .

All equations for calculating the TON are represented by a combination of chemical and physical quantities.

5. Peculiarities of mathematical formulas of the laws of catalysis

To clarify the nature of catalysis, it is necessary to determine which science it should be referred to. The formulas of the laws of catalysis provide a clue. The mathematical representation of the laws of catalysis demonstrates the presence of quantities of chemical and physical nature in them. Among the physical magnitudes in the laws of catalysis are the fundamental physical constants - Faraday's constant F and the electron charge e . The universal law of catalysis (5) and generalized formulas of catalysis (15) and (20) include the electric charge [32]. These are electrical quantities. Electrical quantities are representatives of the physical component of the complex mechanism of catalysis. On the one hand, this may indicate the electrical nature of catalysis. On the other hand, the presence of chemical quantities in the laws and formulas of catalysis is evidence of the chemical nature of catalysis. The simultaneous presence of chemical and physical quantities is evidence of the interdisciplinary nature of catalysis. The presence of chemical and physical quantities side by side in the laws of catalysis is a consequence of the fact that the general mechanism of catalysis is a sequence of physical and chemical stages. Each stage (chemical and physical) contributes to the overall mechanism of catalysis. Certain quantities of chemical or physical nature are associated with each stage, which are the parameters in the laws of catalysis. The laws of catalysis are mathematical models of the mechanism of catalysis. Consider the relationship between chemical and physical quantities in the laws of catalysis. This will allow us to understand the nature of catalysis.

6. Chemical and physical quantities in the laws of catalysis

Fig. 5 shows the chemical and physical quantities included in the universal law of catalysis rate.

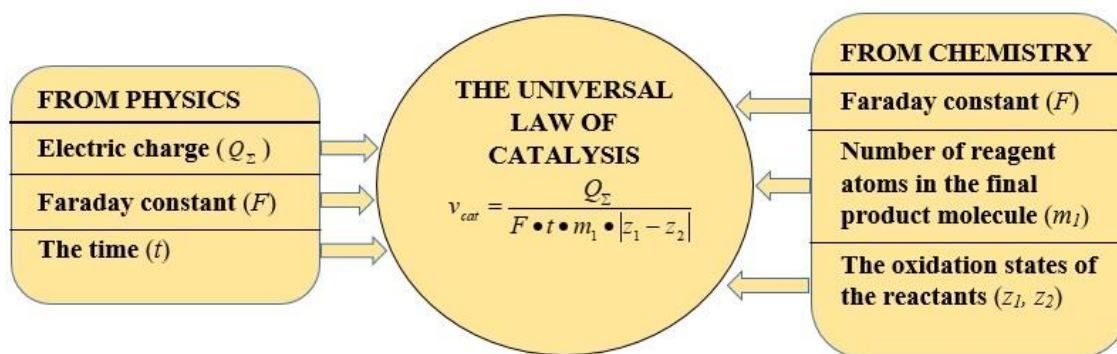


Fig. 5. Chemical and physical quantities in the universal law of catalysis rate.

Fig. 6 shows the chemical and physical quantities included in the law of speed of heterogeneous catalysis.

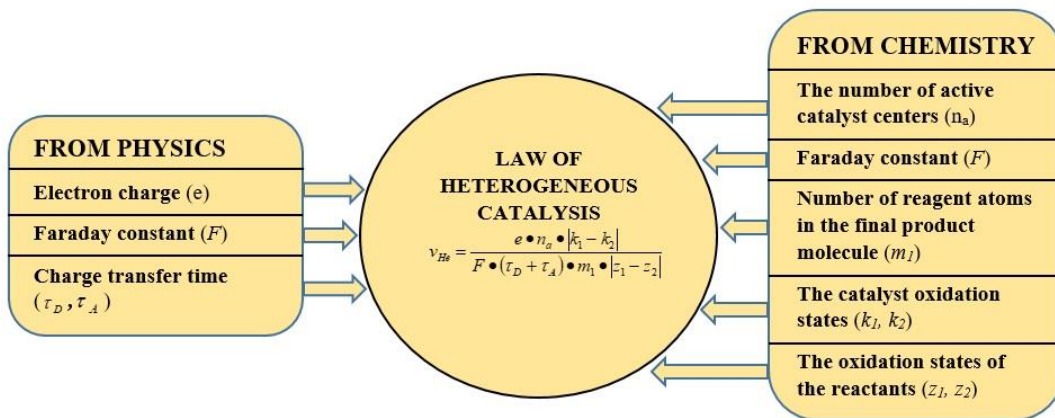


Fig. 6. Chemical and physical quantities in the rate law of heterogeneous catalysis

FIG. 7 shows the chemical and physical quantities included in the rate law of homogeneous catalysis.

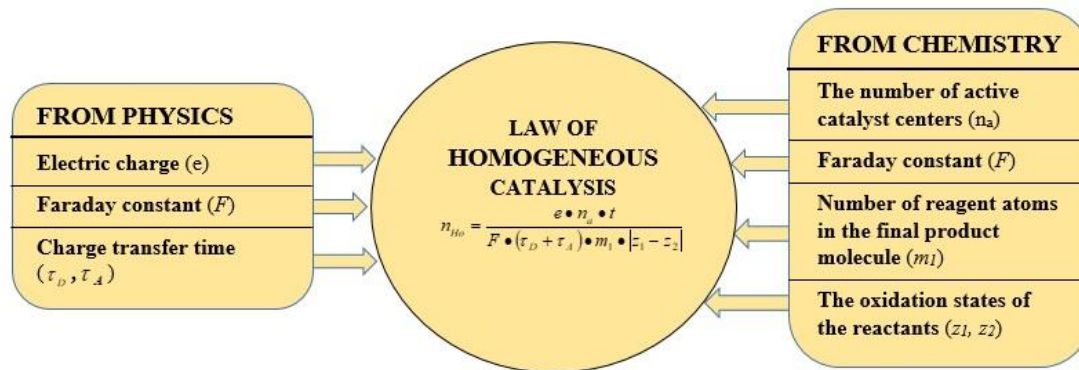


FIG. 7. Chemical and physical quantities in the homogeneous catalysis rate law.

FIG. 8 shows the chemical and physical quantities included in the field catalysis productivity law.

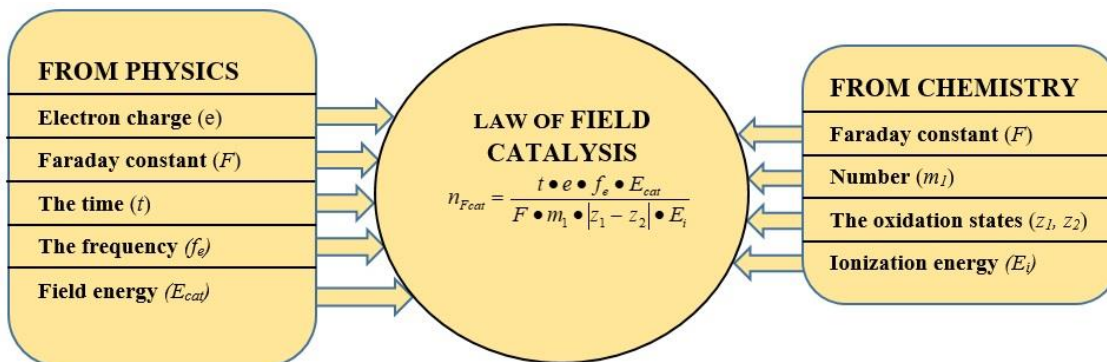


FIG. 8. Chemical and physical quantities in the field catalysis productivity law

From Fig. 5 to Fig. 8 we can see that chemical quantities predominate in the laws of catalysis. Despite the fact that in field catalysis the catalyst is not represented by a chemical substance, but by a physical field [27], the share of chemical quantities even in this law of catalysis is significant (Fig. 8).

7. Chemical and physical quantities in TOF and TON

Let's consider in what ratio there are chemical and physical quantities in the TOF formulas. Fig. 9 shows chemical and physical quantities included in the generalized TOF formula.

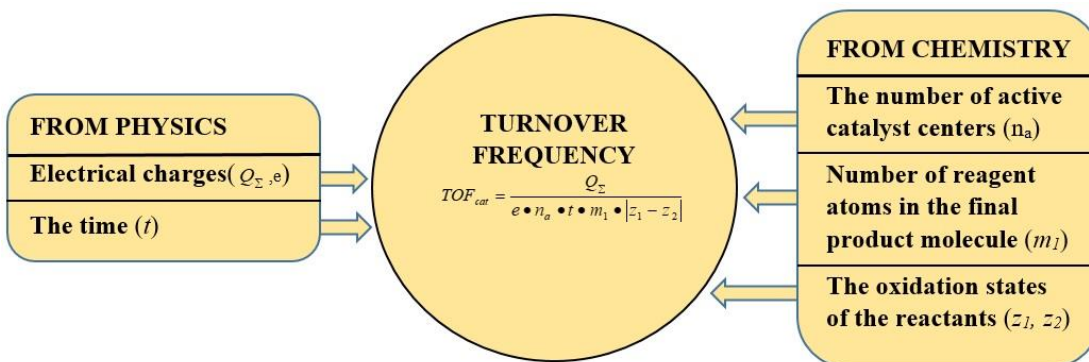


FIG. 9. Chemical and physical quantities in the generalized TOF formulae

FIG. 10 shows the chemical and physical quantities included in the TOF formula of heterogeneous catalysis.

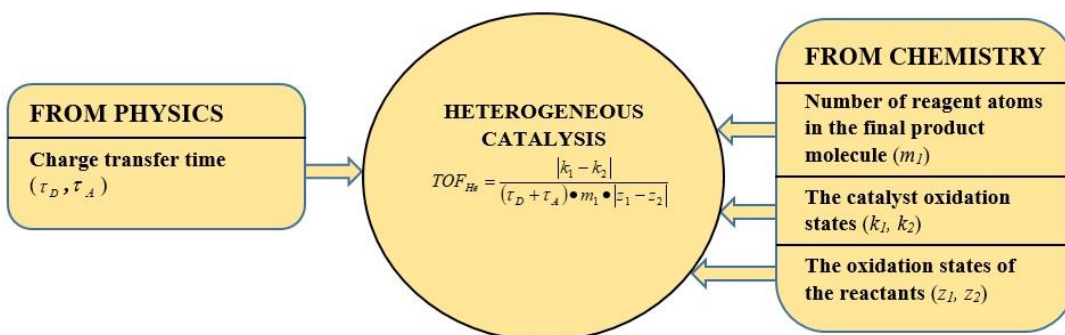


FIG. 10. Chemical and physical quantities in the TOF formula of heterogeneous catalysis

FIG. 11 shows the chemical and physical quantities included in the TOF formula of homogeneous catalysis.

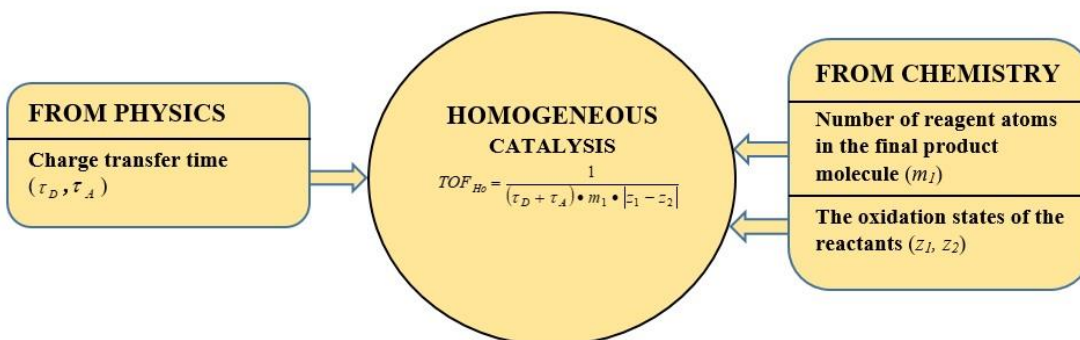


FIG. 11. Chemical and physical quantities in the TOF homogeneous catalysis formula

FIG. 12 shows the chemical and physical quantities included in the TOF field catalysis formula.

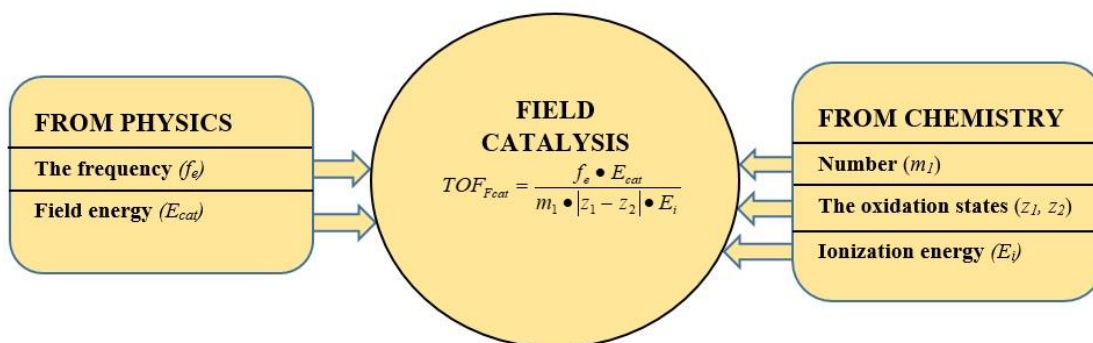


FIG. 12. Chemical and physical quantities in the TOF field catalysis formula

FIG. 13 shows the chemical and physical quantities included in the TOF formula of combined catalysis.

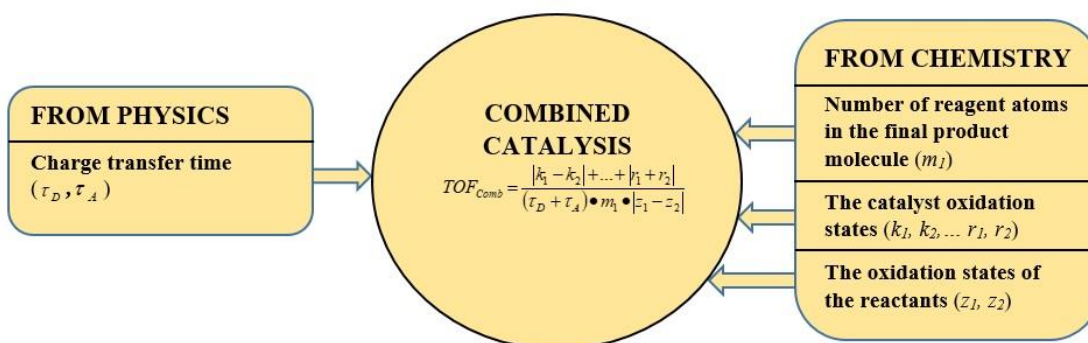


FIG. 13. Chemical and physical quantities in the TOF formula of combined catalysis

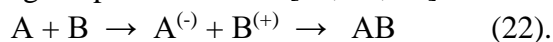
From Fig. 9 to Fig. 13, we can see that the TOF formulas are represented by a combination of chemical and physical quantities with a predominance of chemical quantities. Similarly, chemical and physical quantities are represented in the TON formulas (16) - (20).

8. Chemical and physical processes in the mechanism of catalysis

To determine the nature of catalysis, it is necessary to find out the distribution of chemical and physical processes in the mechanism of catalysis. A chemical reaction that takes place without a catalyst can be represented by the following scheme:



The same reaction in the presence of a catalyst is accelerated due to donor-acceptor interaction of the catalyst with reagents [27, 28, 29, 30, 31, 32]. The catalytic reaction can be represented by the following simplified scheme [27, 31, 32]:



Reagents A and B have a change in the oxidation state during donor-acceptor interaction with the catalyst (Fig. 14). Since electrical charges are released in the donor-acceptor interaction, there must be particles that accept charges. This follows directly from the law of conservation of electric charge. The charge-accepting particles are reagents A and B. Ionized reagents $A^{(-)}$ and $B^{(+)}$ easily enter

into chemical reactions. The reaction between ions $A^{(-)}$ and $B^{(+)}$ requires much less activation energy and is faster.

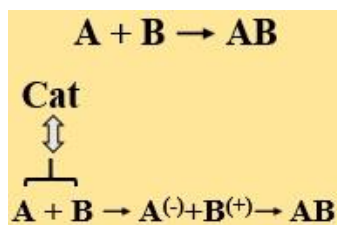


Fig. 14: Schematic of the donor-acceptor mechanism of catalysis. A, B - reagents. AB - reaction product. $A^{(-)}$, $B^{(+)}$ - ionized reagents. Cat - the catalyst.

The mechanism of catalysis is a multistage process. It is a sequence of processes of chemical and physical nature. In all kinds of catalysis (Fig. 15) the single donor-acceptor mechanism of catalysis is realized [22-32]. The process of catalysis is represented by both physical and chemical stages. All types of catalysis are characterized by the same type of catalysis mechanism.

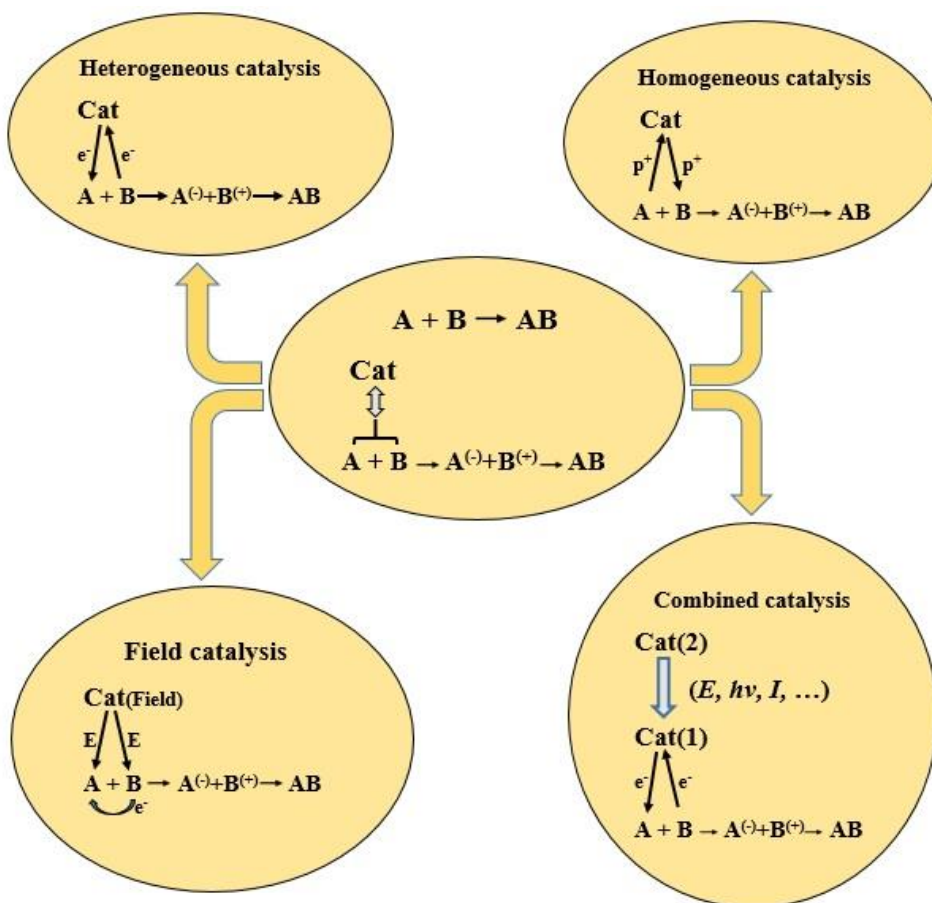


Fig. 15. Schematic representation of the main physical and chemical stages of the donor-acceptor mechanism of catalysis. A, B - reagents; AB - reaction product; $A^{(-)}$, $B^{(+)}$ - ionized reagents; Cat - catalyst; e - electrons; p - protons; E - electric field; $h\nu$ - photons; I - electric current.

In heterogeneous catalysis, in the donor-acceptor interaction of the catalyst with the reactants, electrical charges are transferred from the catalyst to the reactants by electrons. This effect of the

catalyst on the reactants leads to a change in the oxidation state of the reactants. The intermediate states of the reactants and the catalyst in a catalytic reaction are their states in a changed oxidation state. In homogeneous catalysis, electrical charges are transferred from the catalyst to the reactants by protons. This effect of the catalyst leads to a change in the oxidation state of the reactants. In field catalysis, the ionization of reactants occurs under the influence of an electric field. The electric field acts as a non-material catalyst [27]. This field effect leads to a change in the oxidation state of the reactants. In combined catalysis, the change in the oxidation state of the reactants occurs from the combined action of two or more catalysts.

The example of heterogeneous catalysis (Fig. 16) shows schematically the main physical and chemical stages of the catalysis process.

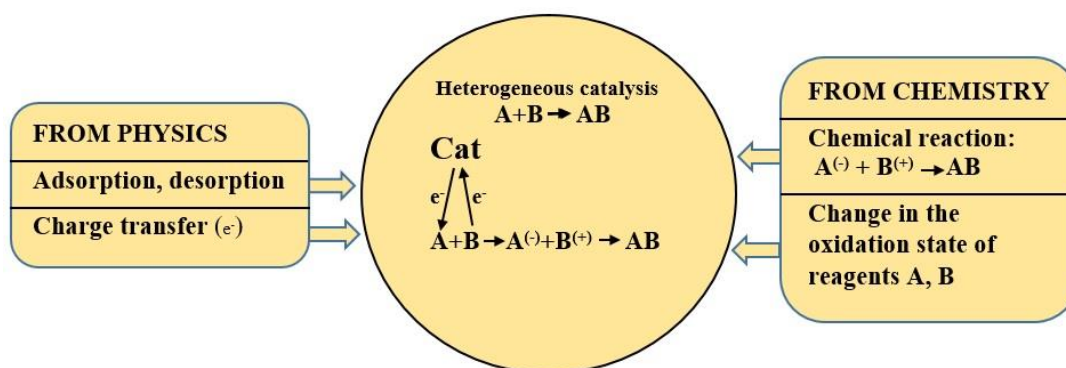


Fig. 16. Physical and chemical steps in the mechanism of heterogeneous catalysis
The catalyst in heterogeneous catalysis acts as an intermediary in electron transfer.

The example of field catalysis (Fig. 17) shows schematically the main physical and chemical stages of the catalysis process.

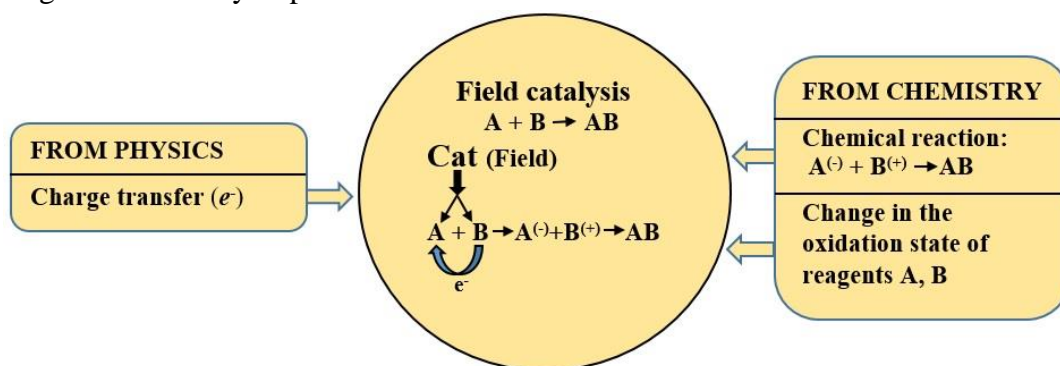


Fig. 17. Physical and chemical steps in the field catalysis mechanism

The catalyst (field) in field catalysis creates conditions for direct electron transfer between the reactants.

In the formulas of the laws of catalysis (1) to (10) and in the formulas of TOF and TON (11) to (20), the proportion of chemical quantities prevails over physical ones. The mechanism of catalysis is also a sequence of chemical and physical stages. The large proportion of chemical quantities in the laws of catalysis and in the characteristics of catalysis allows us to apply terminology with an emphasis on chemistry and to use the wording "chemical-physical" instead of "physical-chemical".

9. Electric charge is an active factor in catalysis and a parameter in the laws of catalysis

Both the laws of catalysis and the donor-acceptor mechanism of catalysis vividly represent the electric component. In the laws of catalysis, these are the fundamental physical constants - the electron charge e and Faraday's constant F . In the mechanism of catalysis, this is the transfer of electric charges to the reagents. This process is accompanied by a change in the oxidation state of the catalyst and reactants. In the universal law of catalysis (5) and in the generalized formulas of TOF (15) and TON (20) it is the total electric charge Q_{Σ} . Such a wide representation of electric quantities in the laws and formulas of catalysis makes us consider the role and place of electric charges and electric fields in catalysis in more detail.

The total electric charge Q_{Σ} transmitted (received) by the reactants during a reaction is included as a parameter in the universal law of catalysis (5) and in the formulas (15), (20) of TOF and TON. - is the amount of electricity that is transferred to the reactants during the catalytic reaction. The total electric charge Q_{Σ} is a dynamic characteristic of the catalytic process. For example, for heterogeneous catalysis, the value of the total electric charge Q_{Σ} is directly related to the total number of electrons transferred from the catalyst to the reactant during all donor half-cycles of catalysis [27, 32].

Formulas for calculating the total charge Q_{Σ} in heterogeneous, homogeneous, field, and combined catalysis are presented below (Fig. 18):

$$Q_{\Sigma} = Q_{He} = \frac{e \cdot n_a \cdot |k_1 - k_2| \cdot t}{(\tau_D + \tau_A)} \quad (23)$$

$$Q_{\Sigma} = Q_{Ho} = \frac{e \cdot n_a \cdot t}{(\tau_D + \tau_A)} \quad (24)$$

$$Q_{\Sigma} = Q_{Fcat} = \frac{e \cdot f_e \cdot E_{cat} \cdot t}{E_i} \quad (25)$$

$$Q_{\Sigma} = \frac{e \cdot n_a \cdot (|k_1 - k_2| + \dots + |r_1 - r_2|) \cdot t}{(\tau_D + \tau_A)} \quad (26)$$

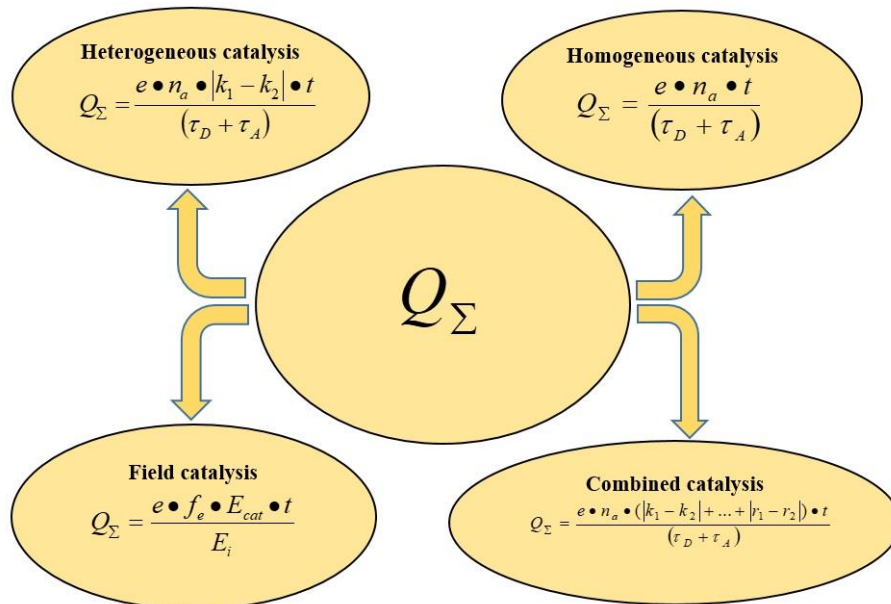


Fig. 18. Formulas for calculating the total charge Q_{Σ} in different types of catalysis.

Substituting appropriate values into the universal law of catalysis (5) formula (5) turns into the laws of heterogeneous (1), homogeneous (2), field (3), and combined catalysis (4) (Fig. 19). Note that it was shown in [32] that the law of Faraday's electrolysis directly follows from the formula of the universal law of catalysis (5) in addition to the laws of catalysis.

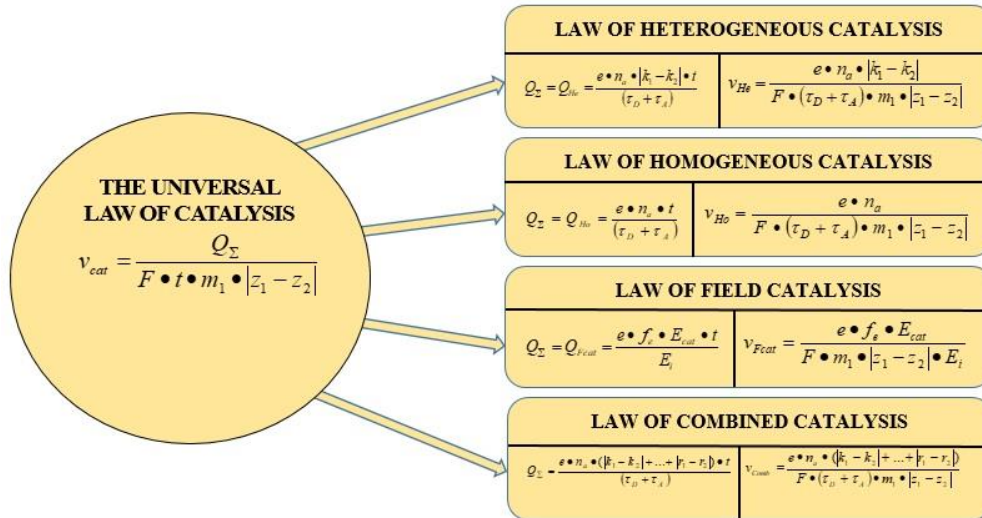


Fig. 19. Reduction of the universal law of catalysis to the partial laws of catalysis at the corresponding values of Q_{Σ} .

Substituting the corresponding values Q_{Σ} into the generalized TOF formula (15), formula (15) is transformed into TOF of heterogeneous (11), homogeneous (22), field (33), and combined catalysis (14) (Fig. 20).

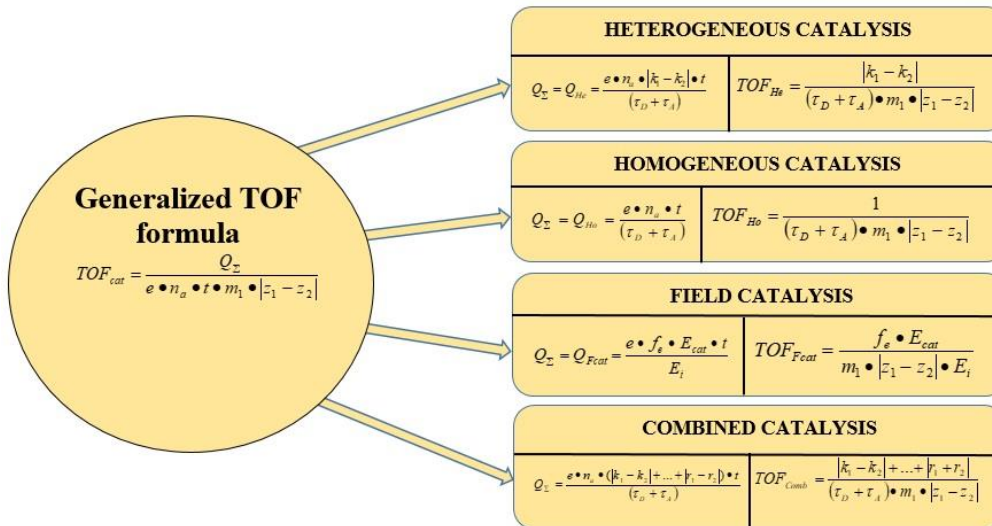


Fig. 20: Reduction of the generalized TOF formula to private tof at the corresponding values of Q_{Σ} .

Fig. 21 shows a summary table of the laws of catalysis.

Name	Formula	Note
Universal law of the rate of catalysis	$v_{cat} = \frac{Q_{\Sigma}}{F \cdot t \cdot m_1 \cdot z_1 - z_2 }$	Q_{Σ} - total electric charge
Law of the rate of heterogeneous catalysis	$v_{ho} = \frac{e \cdot n_a \cdot k_1 - k_2 }{F \cdot (\tau_D + \tau_A) \cdot m_1 \cdot z_1 - z_2 }$	$Q_{\Sigma} = \frac{e \cdot n_a \cdot k_1 - k_2 \cdot t}{(\tau_D + \tau_A)}$
Law of the rate of homogeneous catalysis	$v_{ho} = \frac{e \cdot n_a}{F \cdot (\tau_D + \tau_A) \cdot m_1 \cdot z_1 - z_2 }$	$Q_{\Sigma} = \frac{e \cdot n_a \cdot t}{(\tau_D + \tau_A)}$
Field catalysis rate law	$v_{fc} = \frac{e \cdot f_e \cdot E_{cat}}{F \cdot m_1 \cdot z_1 - z_2 \cdot E_i}$	$Q_{\Sigma} = \frac{e \cdot f_e \cdot E_{cat} \cdot t}{E_i}$
Law of the rate of combined catalysis	$v_{comb} = \frac{e \cdot n_a \cdot (k_1 - k_2 + \dots + r_1 - r_2)}{F \cdot (\tau_D + \tau_A) \cdot m_1 \cdot z_1 - z_2 }$	$Q_{\Sigma} = \frac{e \cdot n_a \cdot (k_1 - k_2 + \dots + r_1 - r_2) \cdot t}{(\tau_D + \tau_A)}$
The yield of the catalytic reaction	$n_{cat} = \frac{Q_{\Sigma}}{F \cdot m_1 \cdot z_1 - z_2 }$	Q_{Σ} - total electric charge
Catalytic reaction yield of heterogeneous catalysis	$n_{ho} = \frac{e \cdot n_a \cdot t \cdot k_1 - k_2 }{F \cdot (\tau_D + \tau_A) \cdot m_1 \cdot z_1 - z_2 }$	$Q_{\Sigma} = \frac{e \cdot n_a \cdot k_1 - k_2 \cdot t}{(\tau_D + \tau_A)}$
Catalytic reaction yield of homogeneous catalysis	$n_{ho} = \frac{e \cdot n_a \cdot t}{F \cdot (\tau_D + \tau_A) \cdot m_1 \cdot z_1 - z_2 }$	$Q_{\Sigma} = \frac{e \cdot n_a \cdot t}{(\tau_D + \tau_A)}$
Catalytic reaction yield of field catalysis	$n_{fc} = \frac{t \cdot e \cdot f_e \cdot E_{cat}}{F \cdot m_1 \cdot z_1 - z_2 \cdot E_i}$	$Q_{\Sigma} = \frac{e \cdot f_e \cdot E_{cat} \cdot t}{E_i}$
Catalytic reaction yield of combined catalysis	$n_{comb} = \frac{e \cdot n_a \cdot (k_1 - k_2 + \dots + r_1 - r_2) \cdot t}{F \cdot (\tau_D + \tau_A) \cdot m_1 \cdot z_1 - z_2 }$	$Q_{\Sigma} = \frac{e \cdot n_a \cdot (k_1 - k_2 + \dots + r_1 - r_2) \cdot t}{(\tau_D + \tau_A)}$

Fig. 21. Summary table of catalysis laws

Fig. 22 shows a summary table of the TOF and TON formulas

Name	Formula	Note
Generalized formula for TOF	$TOF_{cat} = \frac{Q_{\Sigma}}{e \cdot n_a \cdot t \cdot m_1 \cdot z_1 - z_2 }$	Q_{Σ} - total electric charge
TOF formula for heterogeneous catalysis	$TOF_{Ho} = \frac{Q_{\Sigma}}{e \cdot n_a \cdot t \cdot m_1 \cdot z_1 - z_2 } = \frac{ k_1 - k_2 }{(\tau_D + \tau_A) \cdot m_1 \cdot z_1 - z_2 }$	$Q_{\Sigma} = Q_{Ho} = \frac{e \cdot n_a \cdot k_1 - k_2 \cdot t}{(\tau_D + \tau_A)}$
TOF formula for homogeneous catalysis	$TOF_{Ho} = \frac{Q_{\Sigma}}{e \cdot n_a \cdot t \cdot m_1 \cdot z_1 - z_2 } = \frac{1}{(\tau_D + \tau_A) \cdot m_1 \cdot z_1 - z_2 }$	$Q_{\Sigma} = Q_{Ho} = \frac{e \cdot n_a \cdot t}{(\tau_D + \tau_A)}$
TOF formula for field catalysis	$TOF_{Fcat} = \frac{Q_{\Sigma}}{e \cdot n_a \cdot t \cdot m_1 \cdot z_1 - z_2 } = \frac{f_e \cdot E_{cat}}{m_1 \cdot z_1 - z_2 \cdot E_i}$	$Q_{\Sigma} = Q_{Fcat} = \frac{e \cdot f_e \cdot E_{cat} \cdot t}{E_i}$
TOF formula for combined catalysis	$TOF_{Comb} = \frac{Q_{\Sigma}}{e \cdot n_a \cdot t \cdot m_1 \cdot z_1 - z_2 } = \frac{ k_1 - k_2 + \dots + r_1 + r_2 }{(\tau_D + \tau_A) \cdot m_1 \cdot z_1 - z_2 }$	$Q_{\Sigma Comb} = \frac{e \cdot n_a \cdot (k_1 - k_2 + \dots + r_1 + r_2) \cdot t}{(\tau_D + \tau_A)}$
Generalized formula for TON	$TON_{cat} = \frac{Q_{\Sigma}}{e \cdot n_a \cdot m_1 \cdot z_1 - z_2 }$	Q_{Σ} - total electric charge
TON formula for heterogeneous catalysis	$TON_{Ho} = \frac{ k_1 - k_2 \cdot t}{(\tau_D + \tau_A) \cdot m_1 \cdot z_1 - z_2 }$	$Q_{\Sigma} = \frac{e \cdot n_a \cdot k_1 - k_2 \cdot t}{(\tau_D + \tau_A)}$
TON formula for homogeneous catalysis	$TON_{Ho} = \frac{t}{(\tau_D + \tau_A) \cdot m_1 \cdot z_1 - z_2 }$	$Q_{\Sigma} = \frac{e \cdot n_a \cdot t}{(\tau_D + \tau_A)}$
TON formula for field catalysis	$TON_{Fcat} = \frac{t \cdot f_e \cdot E_{cat}}{m_1 \cdot z_1 - z_2 \cdot E_i}$	$Q_{\Sigma} = \frac{e \cdot f_e \cdot E_{cat} \cdot t}{E_i}$
TON formula for combined catalysis	$TON_{Comb} = \frac{(k_1 - k_2 + \dots + r_1 + r_2) \cdot t}{(\tau_D + \tau_A) \cdot m_1 \cdot z_1 - z_2 }$	$Q_{\Sigma} = \frac{e \cdot n_a \cdot (k_1 - k_2 + \dots + r_1 + r_2) \cdot t}{(\tau_D + \tau_A)}$

Fig. 22. Summary table of TOF and TON formulas

As we can see, electric charges are very widely represented in the mechanism of catalysis, in the laws of catalysis, and in the formulas of catalysis. The total electric charge obtained (given up) by a reactant during a catalytic reaction is a parameter that sets quantitative characteristics of catalysis. Let's take a closer look at the influence of electric charges and electric fields in catalysis.

10. Catalysis induced by an electric field

The electrical nature of catalysis and the electrical mechanism of catalysis was first discussed by Berzelius immediately after the discovery of catalysis. In his concept of catalysis based on the catalytic force, he noted that "the catalytic force must consist in some influence on the polarity of the atoms which it increases, decreases or changes so that in fact it is based on the excitation of electrical relations. These electrical relationships have so far eluded our research" [33, 34]. About the catalytic force, "which affects the polarity of atoms... based on the excitation of electrical relations," Berzelius spoke long before the discovery of the electron [29]. The lack of a material carrier of electricity in the science of the time did not allow Berzelius to create a quantitative theory of catalysis. Ostwald [35], Pisarzhevsky [36], Roginsky [37] and Volkenshtein [38] further developed the idea of the electrical mechanism of catalysis.

L. W. Piszczek put forward the hypothesis of direct transitions of electrons between reactants and the hypothesis of the determining role of electrons in catalytic reactions. Like Berzelius, he believed that catalysis is realized under the action of electric forces. L. V. Piszczek believed that in a catalytic reaction, the atoms of reactants are ionized. The resulting ions of one reactant meet the opposite ions of the second reactant and enter into a chemical reaction with them. The reaction between ions requires much less activation energy than between molecules and proceeds much faster [39].

Despite the great efforts of scientists to study the electrical mechanism of catalysis, all attempts to create a physical (electrical, electronic) theory of catalysis have been unsuccessful [17, 38]. This problem could not be solved within the framework of physics alone.

In recent years, the number of works investigating electric mechanisms of catalysis and electric fields in catalysis has increased dramatically [40-85]. It has been discovered that the electric field plays a key role in various types of catalysis [81, 82, 83]. Studies have appeared in which electric fields are considered to be catalysts of chemical reactions. They concluded that "*electric fields open up a new way to catalyze chemical reactions.*" [45 - 48]. Some articles refer to the electric field as a class of reagents and call it a "*smart reagent*" [67 - 85].

A new trend is forming in catalysis, which is based on the use of electric fields instead of material catalysts, and a new type of catalysis has appeared in chemistry: *electric field catalysis* [27, 31, 32, 55, 57, 78, 79]. The immaterial field catalyst has many advantages [49]. Conventional material catalysts are effective only for specific chemical reactions. Field induced catalysis has broader capabilities. Studies have shown that field-induced catalysis can accelerate a wide class of reactions. Moreover, in catalysis controlled by an external electric field, the reaction rate and selectivity of chemical reactions can be adjusted at will [42, 49, 50, 78, 80].

As we can see, material catalysts are not the only and obligatory attribute of catalysis. An immaterial substance, the electric field, can also act as a catalyst. In [27], the electric field was referred not to reagents, but to the class of catalysts, and the laws of field catalysis were obtained. The class of real catalysts has been extended to include field catalysts [27]. The assignment of fields to catalysts forces a radical revision of the modern concept of catalysis and the search for new explanations of the mechanism of catalytic reactions. The generally accepted mechanism of catalysis based on the formation of intermediate compounds of the catalyst with reagents is not suitable for field catalysis.

11. The influence of an external electric field in catalysis

It is well known that the electric field interacts only with electrically charged particles. The scheme of donor-acceptor mechanism of catalysis (Fig. 15) shows that such objects are electrons, protons, and ionized reagents. In Fig. 23, an example of a heterogeneous catalytic reaction, we can see objects that are influenced by the electric field. These objects are electrons and ionized reactants.

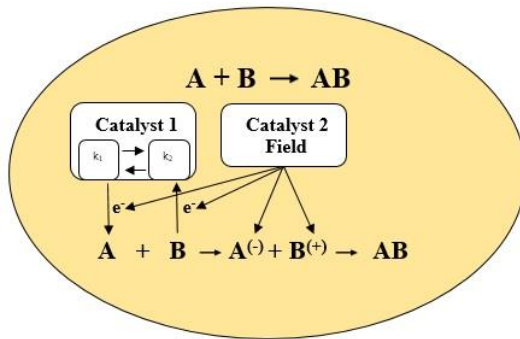


Fig. 23. Mechanism of electric field influence on heterogeneous catalytic reaction. A, B - reagents; AB - reaction product; k_1 - initial oxidation state of the catalyst; k_2 - final oxidation state of the catalyst; $A^{(-)}$ - reagent A in the changed oxidation state; $B^{(+)}$ - reagent B in the changed oxidation state; e^- - electrons.

12. Role of spontaneous electric fields in catalysis

In addition to the direct action on the reactants and on the catalyst, the influence of internal electric fields on the catalytic activity and selectivity has been revealed [43, 52]. The acceleration of chemical reactions is realized not only due to an external field, but also due to spontaneous electric fields, which manifest themselves in all types of catalysis [52]. Spontaneous electric fields have received little attention in catalysis, and their influence has been underestimated. It turned out that their influence on the catalysis rate cannot be neglected [52, 75]. Since spontaneous electric fields are present in all types of catalysis, [52] concluded that *electrostatic phenomenology can be inherent in all types of catalysis*. It is noted in [84] that any heterogeneous process should be considered as having parallels with electrochemical processes.

13. Chemical and physical quantities in total charge formulas .

Consider how quantities of chemical and physical nature are represented in the total electric charge Q_Σ formulas . Fig. 24 shows the chemical and physical quantities included in the formula for the total electric charge Q_Σ in heterogeneous catalysis.

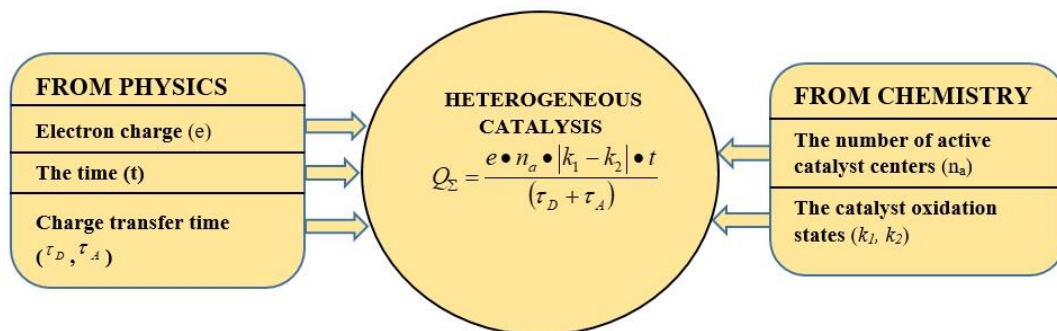


FIG. 24. Chemical and physical quantities in the formula for the total electric charge Q_Σ in heterogeneous catalysis.

FIG. 25 shows the chemical and physical quantities included in the formula of the total electric charge Q_Σ in the homogeneous catalysis.

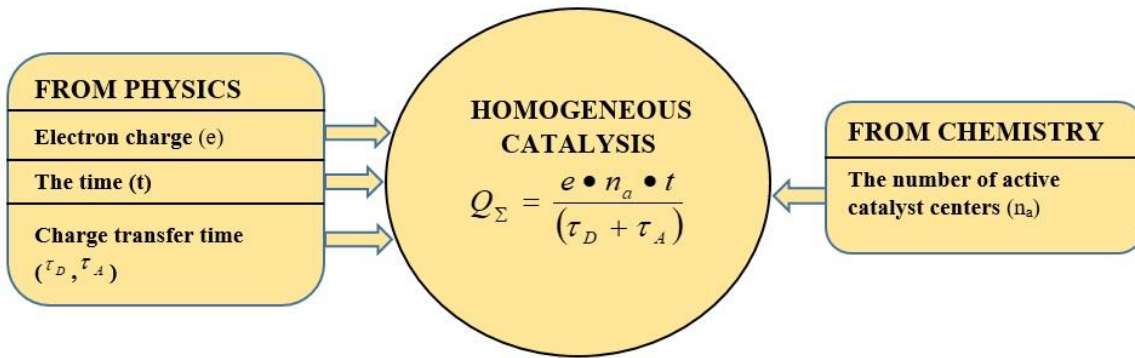


FIG. 25. Chemical and physical quantities in the formula for total electric charge Q_{Σ} in homogeneous catalysis.

FIG. 26 shows the chemical and physical quantities included in the formula for the total electric charge Q_{Σ} in field catalysis.

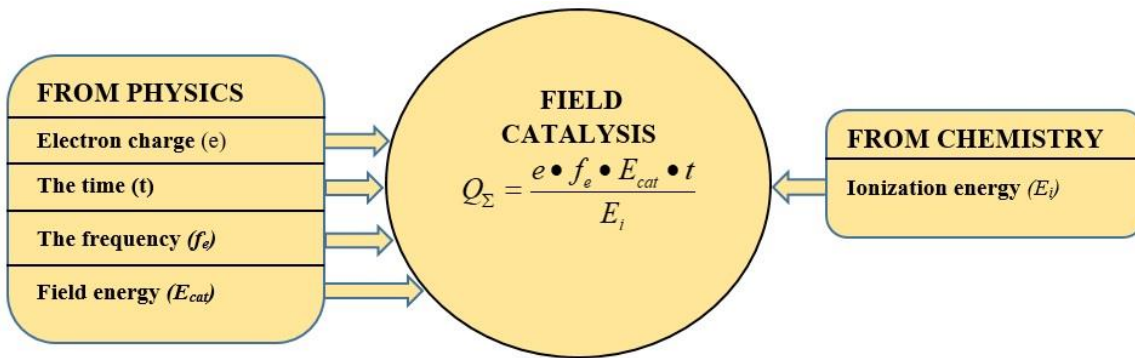


FIG. 26. Chemical and physical quantities in the formula for total electric charge Q_{Σ} in field catalysis.

FIG. 27 shows the chemical and physical quantities included in the formula for the total electric charge Q_{Σ} in the combined catalysis.

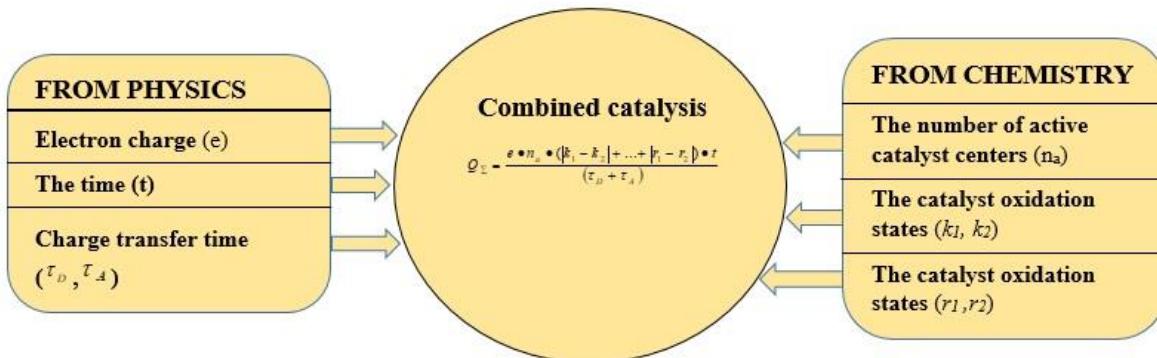


FIG. 27. Chemical and physical quantities in the formula for total electric charge Q_{Σ} in combined catalysis.

The value of the total electric charge Q_{Σ} depends on quantities of chemical and physical nature (Fig. 24 to Fig. 27), The formulas for calculating the charge Q_{Σ} for all types of catalysis are combinations of chemical and physical quantities.

14. The electrical or chemical nature of catalysis?

As we can see, electrical phenomena and electrical quantities are very convincingly represented in catalysis. Confirmation of the essential role of electrical phenomena in catalysis is:

- The large number of examples of reactions in which a physical (electric) field acts either as a catalyst or activates a catalytic reaction.
- high sensitivity of catalytic reactions to electric fields, both external and internal.
- the presence of the total electric charge as a parameter in the laws and formulas of catalysis.
- The presence of fundamental physical constants in the laws and formulas of catalysis: the electron charge and Faraday's constant.

One may get the wrong impression that catalysis has an electric nature and belongs to the class of physical phenomena. The history of catalysis has shown that this is not the case. The history of catalysis has shown that all attempts to create a physical (electric, electronic) theory of catalysis were unsuccessful [17, 38]. The physical component is only one side of catalysis as an interdisciplinary phenomenon. The chemical component of the phenomenon of catalysis is also presented no less convincingly. In the laws of catalysis and in the catalysis formulas (1) - (20), the share of chemical quantities prevails over physical ones. Even the value of the total electric charge Q_{Σ} depends on quantities of chemical nature. Among the quantities of chemical nature, all of the catalysis laws and formulas (1) through (20) include oxidation states.

15. The main secret of catalysis

In the laws and formulas of catalysis, a special role belongs to the oxidation states. Moreover, the oxidation states are included in the laws and formulas of catalysis (1) - (20) as quantitative values. This circumstance requires an explanation. In chemistry, the oxidation states are considered to be quantities that have no physical meaning. This circumstance has been a great obstacle to obtaining the laws of catalysis and clarifying the nature of catalysis. In the long history of catalysis, oxidation states have not been used in catalysis as quantitative quantities. The laws and formulas of catalysis (1) to (20) show that without taking into account the oxidation states of the catalyst and the reactants, it is impossible to obtain quantitative characteristics of catalysis. The concept of oxidation states as quantitative quantities proved to be fruitful and made it possible to obtain a new TOF formula [28], and then the laws of catalysis [29 - 32]. The oxidation states turned out to be that missing key link in catalysis, the use of which allowed the laws of catalysis to be obtained. Here we come to the main mystery of the catalytic phenomenon. This mystery consists in the special role of oxidation states in catalysis.

In [22 - 32], the oxidation states are used in catalysis for a new purpose. In catalysis, the oxidation states act not as auxiliary, but as the main quantitative quantities. In the mechanism of catalysis, the oxidation state of the reactants and the oxidation state of the catalyst are equally important. The unraveling of the main secret of catalysis required an extended interpretation of the concept of the oxidation state in chemistry. In order for catalysis to become a scientific field in its own right, the following must be accepted:

1. The concept of the oxidation state is fundamental to the science of catalysis.
2. The universal donor-acceptor mechanism of catalytic reactions is based on the oxidation state concept.

3. The oxidation states of a catalyst are its most important chemical characteristics, so they can be used as quantitative quantities.

4. The oxidation states of reactants are the most important chemical characteristics of reactants, so they can be used as quantitative quantities.

An important feature of the laws and formulas of catalysis (1) - (20) is that they include not only the oxidation states of the catalyst, but also the oxidation states of the reactants as quantitative values. The oxidation states of the reactants are just as important in catalysis as the oxidation states of the catalyst. This is the first time we have encountered a situation in which the oxidation states of the reactants act as parameters in the laws and formulas of catalysis. While the oxidation states of the catalyst have long attracted the attention of catalysis researchers, there have been no in-depth studies of the influence of the oxidation states of reactants in catalysis. More precisely, we were unable to find such studies in the entire history of catalysis. For some reason, the oxidation state of reactants did not receive the attention of scientists. They were not given the attention they deserved, although their influence in a catalytic reaction is just as significant as that of the oxidation state of the catalyst.

The oxidation states of the catalyst and reagents turned out to be those missing links, thanks to which opportunities for solving many problems of catalysis are opened. They are the missing links not only for solving problems of catalysis. The oxidation states are the missing link for another problem in chemistry: the problem of the reactivity of substances. The problems of catalysis are part of this larger problem of chemistry. As N. N. Semenov, the author of the chain theory of catalysis, noted, *the question of the theory of catalysis lies in the context of the more general problem of the reactivity of substances* [17]. The peculiarity of catalytic reactions is the change in the reactivity of reagents under the influence of catalysts. The concept of the oxidation state is not only the key to the main secret of catalysis, it is also the key to the main secret of the reactivity of chemical substances.

16. The concept of oxidation states in chemistry and in catalysis

From the laws and formulas of catalysis (1) to (20), it follows that the main contribution to the mechanism of catalysis is made by the chemical component, in which the oxidation states of the catalyst and reagents play a determining role [27, 28, 29, 30, 31, 32]. The oxidation states of the catalyst and reactants also play a determining role in the catalysis mechanism.

Despite the fact that the oxidation state of substances is widely used in chemistry, it is considered to be an auxiliary conventional value that has no physical meaning [86-89]. There has long been much debate among scientists about the meaning of the oxidation state in chemistry [88, 90 - 93]. The oxidation state is used in chemistry for such purposes [91 - 93]:

1. As a descriptor.
2. As a parameter in chemical nomenclature.
3. As a variable for tabulation or plotting.
4. As a value related to d-electron configurations of transition-metal (TM) ions in compounds.
5. As a formalized basis for balancing redox equations.

There is no consensus that the oxidation state should be considered a conventional value that has no physical meaning. Moreover, some authors point out that the oxidation state is universal and fundamental. [94, 95, 96]. There is a well-known opinion of Linus Pauling, who said, *"If scientific progress continues, the next generation may have a theory of valence that is sufficiently precise and*

powerful to permit chemistry to be classed along with physics as an exact science" [94,95]. In [94, 96] it is explicitly stated that the concept of oxidation state is fundamental to the science of catalysis. This shift in emphasis on the fundamental status of the oxidation state in chemistry offers great opportunities for future catalysis research. Even more opportunities in catalysis research are offered by the management of the oxidation state of the catalytic active centers [96, 98]. The concept of oxidation state is most actively explored in catalysis using transition metals, which can easily reach different oxidation states [99, 100].

For example, the Fischer-Tropsch process cyclically changes the oxidation state of nickel. Under the influence of hydrogen, nickel lowers its oxidation state from +2 to 0. The resulting nickel interacts with carbon monoxide to form carbide, where the nickel is again oxidized to state +2. The oxidation reaction of hydrocarbons on vanadium V₂O₅ catalyst involves electron transfer between the reacting molecules and the catalyst in the catalysis mechanism. In this case, the oxidation state of vanadium is changed [101, 102]. Changes in the oxidation states of the active centers of the catalyst occur due to the transfer of electrons between the catalyst and the reactants [103].

Interest in the oxidation state in catalysis arose quite a long time ago [104 - 107]. Even in early studies of the effect of the oxidation state on chemical reactions, it was concluded that the activity and selectivity of the catalytic process depend on the oxidation state of the catalyst [106 - 107].

Recently, the number of papers investigating the influence of the oxidation state of a catalyst on the efficiency of catalysis has increased dramatically [108 - 112].

The emphasis in these works is on the fact that oxidation states can help predict the reactivity of substances and their chemical activity. In [96] it is noted that "*valence chemistry is of fundamental importance in catalysis science,*" and "*management of the oxidation state provides the key to future catalysis research*".

Despite the large number of examples of catalytic reactions in which the oxidation state plays a key role, no in-depth studies of the role and place of the oxidation state in catalysis have been conducted. The role of oxidation states in catalysis has been underestimated. To this day, the oxidation state remains in chemistry "*an auxiliary conventional value that has no physical meaning.*" [86 - 89].

In studies of the effect of the oxidation state on the efficiency of catalysis, the main focus has been on the oxidation state of the catalyst [94 - 112]. The influence of the oxidation state of the reactants fell out of favor. It was shown in [22-32] that the oxidation states of the reactants should be considered in catalysis on a par with the oxidation states of the catalyst. It was only possible to obtain laws and formulae for catalysis by taking into account the oxidation states of both the catalyst and the reactants. The concept of the oxidation state of the catalyst and reactants was so fruitful that it made it possible to obtain new formulas for calculating the TOF [28] and the laws of catalysis [22-32].

In the mechanism of catalytic reactions, different types of catalysis have revealed such common features [22 - 32]:

- a) In catalytic reactions there is a cyclic change in the oxidation state of the catalyst;
- b) in catalytic reactions there is a change in the oxidation state of the reactants;
- c) catalytic processes occur with the participation of electrically charged particles;
- d) the mechanism of catalysis involves the transfer of electric charges;

The concept of the oxidation state is "*involved*" in these general features. In order to realize such functions, catalysts must have a special property - they must easily change the oxidation state within a wide range.

17. The role of the oxidation state in the catalysis mechanism

In [22 - 32], the concept of the oxidation state in catalysis was expanded. The oxidation states have been applied in a new way. The oxidation states of the catalyst and reagents in catalysis act not as auxiliary, but as basic quantitative quantities. The concept of the oxidation state plays a key role in the donor-acceptor mechanism of catalysis. The oxidation states are such important characteristics of the catalyst substance and reactants that they are decisive in the catalysis mechanism [22 - 32].

Figure 28 shows changes in the oxidation state of the catalyst and reagents A and B in the example of heterogeneous catalysis. In the reaction there is a cyclic change in the oxidation state of the active centers of the catalyst from state k_1 to state k_2 and back. There is a unidirectional change in the oxidation state of reagents A and B. Reagent A goes to the oxidation state $A^{(-)}$. Reagent B goes to the oxidation state $B^{(+)}$. The change in the oxidation state of the catalyst and reactants is due to electron transfer in the donor-acceptor mechanism of heterogeneous catalysis.

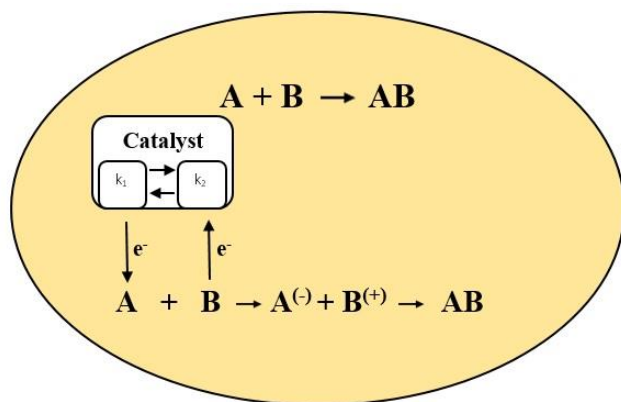


Fig. 28: Change in the oxidation state of the catalyst and reactants in the donor-acceptor mechanism of heterogeneous catalysis. A, B - reactants; AB - reaction product; k_1 - initial oxidation state of the catalyst; k_2 - final oxidation state of the catalyst; $A^{(-)}$ - reagent A in the changed oxidation state; $B^{(+)}$ - reagent B in the changed oxidation state; e^- - electrons.

The intermediate states of reagents A and B are their states in an altered oxidation state $A^{(-)}$ and $B^{(+)}$. The intermediate state of the catalyst is the oxidation state of the active centers in the modified oxidation state k_2 . The oxidation state of the active centers of the catalyst changes cyclically between the oxidation states k_1 and k_2 .

18. Oxidation states in the laws of catalysis

The oxidation states of the catalyst and reactants proved to be the main secret in the mechanism of catalysis, the use of which allowed the laws of catalysis [22 - 32]. The oxidation states of the catalyst and reagents set the values of the catalysis characteristics [27, 28, 29, 30, 31, 32] and act as parameters in the laws of catalysis and in the TOF and TON formulas [28, 29]. Fig. 29 conventionally

shows the extension of the fields of application of oxidation states beyond their known application. Instead of auxiliary conditional quantities that have no physical meaning, the oxidation states in catalysis act as quantitative quantities.

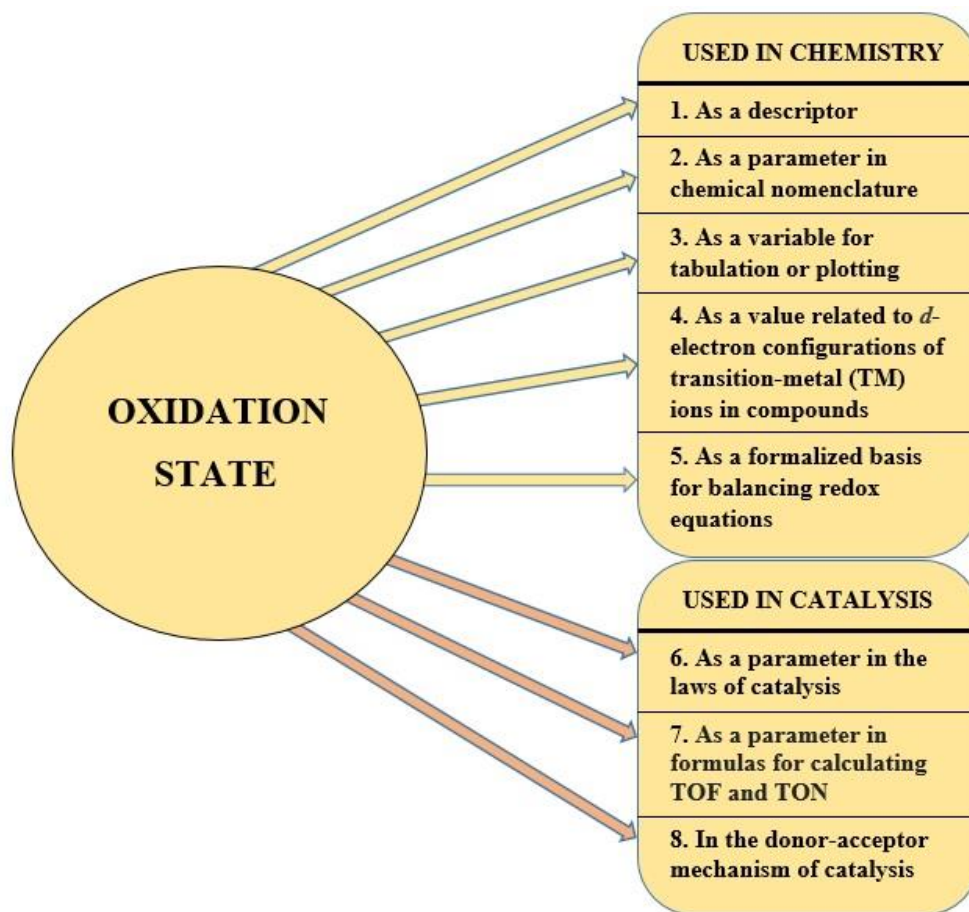


Fig. 29: Extension of the application of oxidation states to the field of catalysis.

Despite the great interest of scientists in the concept of oxidation states in catalysis [94 - 112], oxidation states have not been used as quantitative quantities in catalysis. At the same time, their role as quantitative quantities in catalysis is evident in the mechanism of catalysis itself. A change in the oxidation state of reactants is directly related to the donor (acceptor) function of the interaction of the catalyst with the reactants. The change in the oxidation state of the catalyst and reagent sets the value of the electric charge transferred (received) in the donor (acceptor) half-cycle of catalysis [27 - 32]. The presence of oxidation states in the laws and formulas of catalysis (1) - (20) takes on a physical meaning. For example, for heterogeneous catalysis, the amount of change in the oxidation state is directly related to the number of electrons transferred (received) by the active center in the donor (acceptor) halfcycle of catalysis.

In our opinion, such concepts as the oxidation state of the catalyst, the oxidation state of the reactants, the change in the oxidation state, and the range of change in the oxidation state should become central concepts of catalysis. Oxidation states have a direct relationship to the mechanism of

catalysis. The donor-acceptor mechanism of catalysis is based on the transfer of electrical charges and the change in the oxidation state of the catalyst and reactants. The concept of the oxidation state is the key to understanding the nature of catalysis and the key to unlocking the laws of catalysis that scientists have been searching for years.

19. List of oxidation states of chemical elements - a tool for selecting catalysts

The known list of oxidation states of chemical elements in chemistry becomes a basic aid and tool in the selection of catalysts. The oxidation states of the elements are important characteristics of chemical elements. They are summarized in a table [113]. The list of oxidation states provides important information for catalysis about the most stable oxidation states. It shows the ranges of changes in the oxidation states of the elements and the distribution of the spectrum of oxidation states within the range. The list of oxidation states of chemical elements is useful for describing, understanding and predicting chemical reactivity.

20. Faraday's constant in the laws of catalysis

It is known that the Faraday constant is included in the formula of Faraday's law of electrolysis, in the Nernst equation and in the Goldman equation. This constant is a physicochemical constant. It is a linking constant between physical and chemical quantities - the electron charge and Avogadro's number. Faraday's constant is the electric charge of one mole of electrons. Its value is 96485.3321233100184 C/mol.

It was shown in [27-32] that the presence of Faraday's constant in the laws and formulas of chemistry is not limited to Faraday's law of electrolysis, the Nernst equation and the Goldman equation. It is included in the equations of the laws of catalysis (1) - (4) and in the universal law of catalysis (5). Fig. 30 conventionally shows the extension of the application of Faraday's constant beyond the known laws and equations of chemistry and its extension to the field of catalysis.

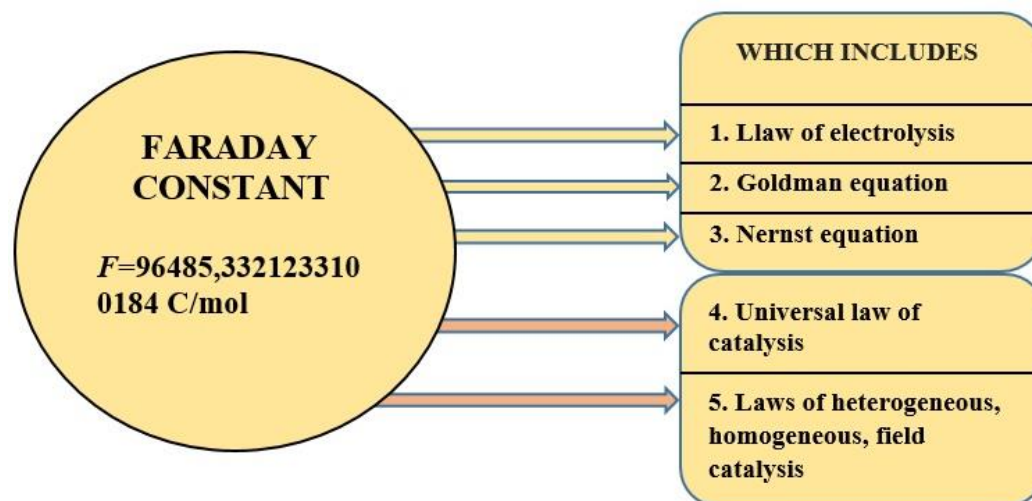


Fig. 30. Extension of the application of Faraday's constant to the field of catalysis.

The fact that Faraday's physical-chemical constant is included in the laws of catalysis testifies in favor of the interdisciplinary nature of catalysis. The fundamental status of Faraday's constant points to the fundamental status of catalysis as a chemical-physical phenomenon.

21. Signs of the fundamentality of catalysis

Catalysis is increasingly being categorized as a class of fundamental phenomena [29, 32, 114 - 117]. As evidence of the fundamental status of catalysis, the following signs can be cited:

1. In catalytic reactions, fundamental interactions at the level of elementary particles are realized.

2. The equations of the laws of catalysis include the fundamental constants: Faraday's constant, the electron charge, Avogadro's number.

3. All types of catalysis are subject to a universal law, from which the laws of heterogeneous, homogeneous, combined, and field catalysis follow as particular results.

4. In all types of catalysis, a single donor-acceptor mechanism of catalysis is realized. The donor-acceptor interaction is a particular case of the fundamental interaction of electric charges. The constant of donor-acceptor interaction in catalysis is the Faraday constant.

5. In catalytic reactions, the basic law of nature, the law of conservation of electric charge, is fulfilled.

Fig. 31 shows signs that indicate the fundamental status of catalysis.

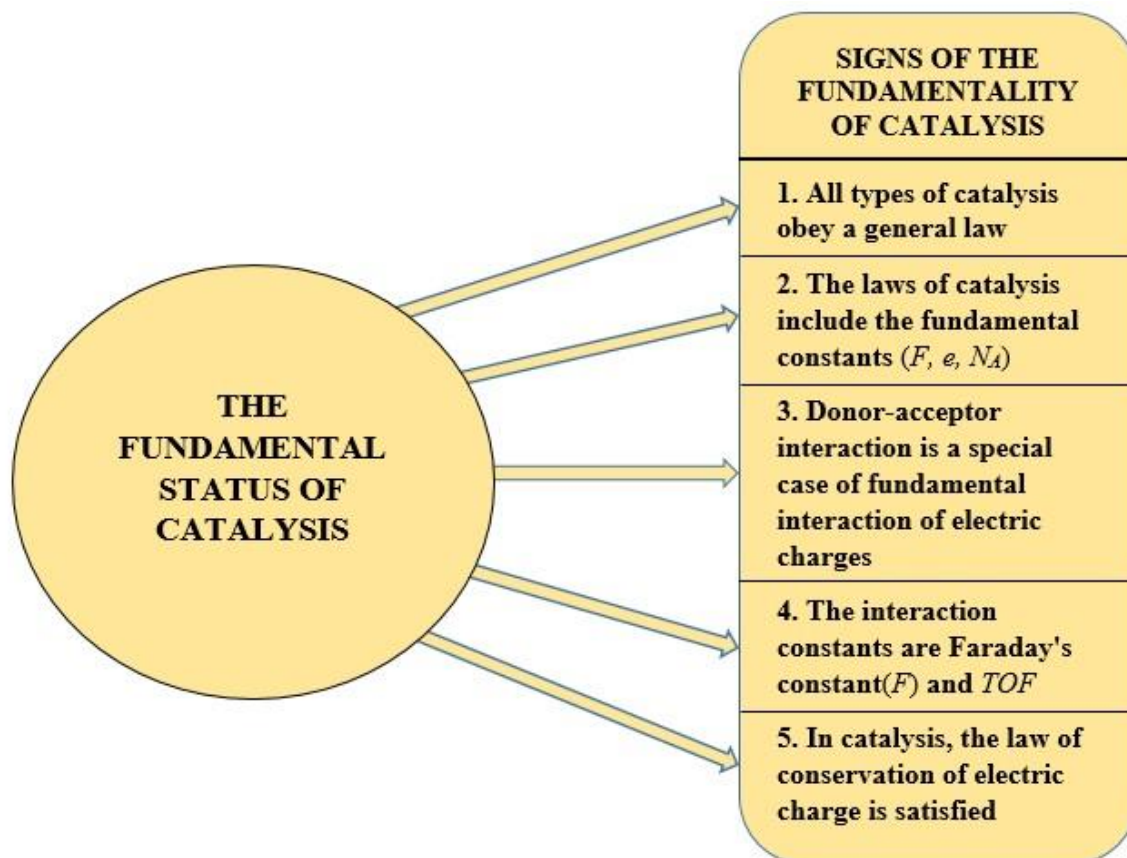


Fig. 31. Signs of the fundamentality of catalysis

22. The laws of catalysis complement the family of fundamental laws of Nature.

As shown above, catalysis has signs of a fundamental chemical and physical phenomenon. In this connection, it is of interest to determine the place of catalysis in the family of fundamental laws of Nature.

The laws of catalysis have much in common with the fundamental physical laws. Catalysis, like fundamental physical phenomena, is characterized by a certain type of interaction. In catalysis, the donor-acceptor interaction of the catalyst and the reactants is realized. Like the fundamental physical laws, the laws of catalysis have an interaction constant. The interaction constant in the laws of catalysis is the Faraday constant. Fundamental physical laws are quantitative mathematical models of fundamental physical processes. The laws of catalysis are also quantitative mathematical models of catalytic processes. The difference between the laws of catalysis and the fundamental physical laws is their interdisciplinary nature. For this reason, the laws of catalysis are represented by a combination of chemical and physical quantities. Physical laws are represented by a combination of physical quantities. As far as we know, the laws of catalysis are the first representatives of interdisciplinary laws represented by quantitative mathematical models.

In Fig. 32 there is a comparative table, which shows that the universal law of catalysis and the fundamental laws of Nature have many common features. Newton's law of gravity, Coulomb's law, Faraday's law of electrolysis and Ohm's law are taken for comparison. The universal law of catalysis complements the family of fundamental physical laws.

	Law of gravity	Coulomb's law	Faraday Laws of electrolysis	Ohm's Law	The universal law of catalysis
Type of interaction	Gravitational	Electro-magnetic	Electro-magnetic	Electro-magnetic	Donor-acceptor
Characteristic	Force (F)	Force (F)	Performance (n_F)	Current (I)	Catalysis reaction speed (v)
Acting factor	Gravitational charge	Eelectrical charge	Electric current (I)	Electric current (I)	Eelectrical charge (Q_z)
Law Formula	$F = G \frac{m_1 \cdot m_2}{r^2}$	$F = k \frac{q_1 \cdot q_2}{r^2}$	$n_F = \frac{I \cdot t}{F \cdot z_1}$	$I = \frac{U}{R}$	$v_{cat} = \frac{Q_z}{F \cdot t \cdot m_1 \cdot z_1 - z_2 }$ $v_{cat} = \frac{e \cdot n_a \cdot TOF_{cat}}{F}$
Interaction constant	G	k	F (Faraday constant)	R (Resistance constant)	1) TOF - donor-acceptor interaction constant 2) F - Faraday constant
Interaction constant formula		$k = \frac{1}{4\pi\epsilon_0}$	$F = e \cdot N_A$	$R = \frac{\rho \cdot l}{S}$	1) $TOF_{cat} = \frac{Q_z}{e \cdot n_a \cdot t \cdot m_1 \cdot z_1 - z_2 }$ 2) $F = e \cdot N_A$
The value of the interaction constant	From formula: $G = \frac{F \cdot r^2}{m_1 \cdot m_2}$	From formulas: $k = \frac{F \cdot r^2}{q_1 \cdot q_2}$ $k = \frac{1}{4\pi\epsilon_0}$	From formulas: $F = \frac{I \cdot t}{n_F \cdot z_1}$ $F = e \cdot N_A$	From formulas: $R = \frac{U}{I}$ $R = \frac{\rho \cdot l}{S}$	From formulas: 1) $TOF_{cat} = \frac{v_{cat} \cdot N_A}{n_a}$ $TOF_{cat} = \frac{Q_z}{e \cdot n_a \cdot t \cdot m_1 \cdot z_1 - z_2 }$ 2) $F = e \cdot N_A$
Theory	Gravitation theory	Electro magnetic theory	Electrolysis theory	Electric circuit theory	Donor-acceptor theory of catalysis

Fig. 32. The universal law of catalysis has common features with the fundamental physical laws.

In Fig. 33 there is a comparative table, which shows that private laws of catalysis have common features sfundamental physical laws.

	Law of heterogeneous catalysis	Law of homogeneous catalysis	Law of field catalysis	Law of combined catalysis
Type of interaction	Donor-acceptor	Donor-acceptor	Donor-acceptor	Donor-acceptor
Characteristic	Catalysis reaction speed (v)	Catalysis reaction speed (v)	Catalysis reaction speed (v)	Catalysis reaction speed (v)
Acting factor	Eelectrical charge $Q_z = \frac{e \cdot n_a \cdot \bar{k}_1 - \bar{k}_2 \cdot t}{(\tau_D + \tau_A)}$	Eelectrical charge $Q_z = \frac{e \cdot n_a \cdot t}{(\tau_D + \tau_A)}$	Eelectrical charge $Q_z = \frac{e \cdot f_e \cdot E_{cat} \cdot t}{E_i}$	Eelectrical charge $Q_z = \frac{e \cdot n_a \cdot (\bar{k}_1 - \bar{k}_2 + \dots + r_1 - r_2) \cdot t}{(\tau_D + \tau_A)}$
Law Formula	$v_{Hc} = \frac{e \cdot n_a \cdot \bar{k}_1 - \bar{k}_2 }{F \cdot (\tau_D + \tau_A) \cdot m_1 \cdot z_1 - z_2 }$	$v_{Ho} = \frac{e \cdot n_a}{F \cdot (\tau_D + \tau_A) \cdot m_1 \cdot z_1 - z_2 }$	$v_{Fcat} = \frac{e \cdot f_e \cdot E_{cat}}{F \cdot m_1 \cdot z_1 - z_2 \cdot E_i}$	$v_{Comb} = \frac{e \cdot n_a \cdot (\bar{k}_1 - \bar{k}_2 + \dots + r_1 - r_2)}{F \cdot (\tau_D + \tau_A) \cdot m_1 \cdot z_1 - z_2 }$
Interaction constants	1) TOF 2) F - Faraday constant	1) TOF 2) F - Faraday constant	1) TOF 2) F - Faraday constant	1) TOF 2) F - Faraday constant
Interaction constants formulas	1) $TOF_{cat} = \frac{v_{cat} \cdot N_A}{n_a}$ $TOF_{Ho} = \frac{ \bar{k}_1 - \bar{k}_2 }{(\tau_D + \tau_A) \cdot m_1 \cdot z_1 - z_2 }$ 2) $F = e \cdot N_A$	1) $TOF_{cat} = \frac{v_{cat} \cdot N_A}{n_a}$ $TOF_{Ho} = \frac{1}{(\tau_D + \tau_A) \cdot m_1 \cdot z_1 - z_2 }$ 2) $F = e \cdot N_A$	1) $TOF_{cat} = \frac{v_{cat} \cdot N_A}{n_a}$ $TOF_{Fcat} = \frac{f_e \cdot E_{cat}}{m_1 \cdot z_1 - z_2 \cdot E_i}$ 2) $F = e \cdot N_A$	1) $TOF_{cat} = \frac{v_{cat} \cdot N_A}{n_a}$ $TOF_{Comb} = \frac{ \bar{k}_1 - \bar{k}_2 + \dots + r_1 - r_2 }{(\tau_D + \tau_A) \cdot m_1 \cdot z_1 - z_2 }$ 2) $F = e \cdot N_A$
Theory	Donor-acceptor theory of catalysis	Donor-acceptor theory of catalysis	Donor-acceptor theory of catalysis	Donor-acceptor theory of catalysis

Fig. 33. The laws of catalysis and the fundamental physical laws have common features.

The laws of catalysis are represented by quantitative mathematical models. This is similar to the fundamental physical laws, which are also represented by quantitative mathematical models. The laws of catalysis combine chemical and physical quantities within a single formula, whereas the formulae of the fundamental physical laws consist only of physical quantities. This is their difference. The difference stems from the interdisciplinary status of the laws of catalysis. Figure 34 conventionally shows the extension of the family of fundamental laws of Nature and their addition to the laws of catalysis.

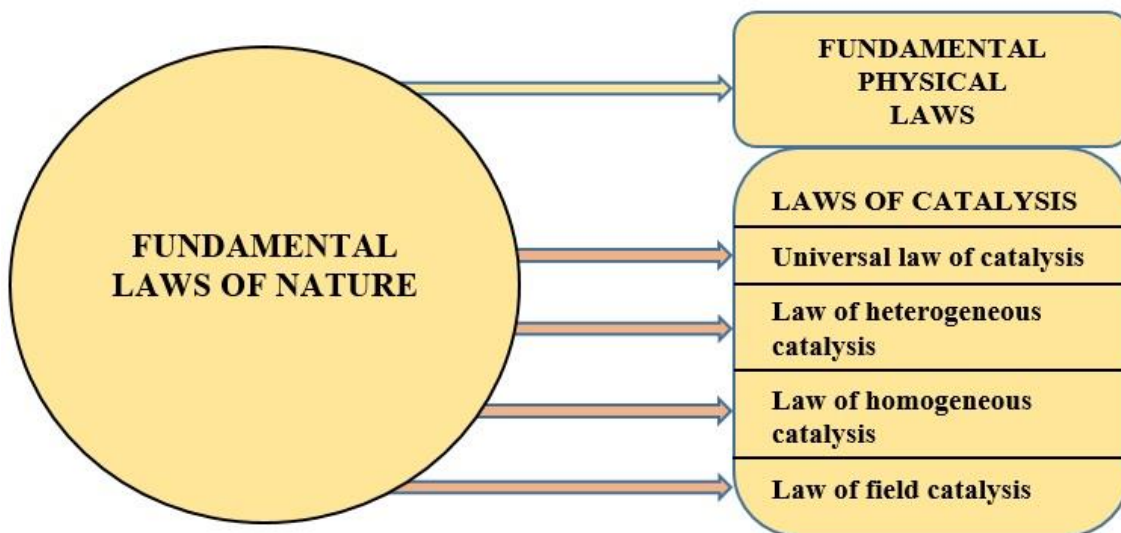


Fig. 34. The laws of catalysis complement the family of fundamental laws of Nature.

23. Conclusion

Catalysis research has now become a global activity, involving many thousands of researchers around the world [118, 119]. Catalysis is a fundamental phenomenon and has an interdisciplinary character. It occupies a place at the junction of two sciences: chemistry and physics. The laws of catalysis are a confirmation of the interdisciplinary character of catalysis and its fundamental status. The interdisciplinary nature of catalysis is indicated by the fact that the laws of catalysis are represented by a combination of chemical and physical quantities. In the laws of catalysis, the fundamental constants (Faraday's constant, electron charge) and the oxidation states of the catalyst and reagents occupy a special place. The well-known use of Faraday's fundamental constant in chemistry has been extended to the field of catalysis.

The main secret of catalysis, which remained undisclosed throughout its history, turned out to be the special role of the oxidation states of the catalyst and reactants in the mechanism of catalysis. The oxidation states were the main missing link in the mechanism of catalysis, the use of which made it possible to obtain the laws of catalysis. We applied the oxidation states to a new purpose as parameters in the laws of catalysis. In catalysis, the oxidation states act not as auxiliary, but as the main quantitative values. It turned out that in catalysis, it is equally necessary to take into account not only the oxidation states of the catalyst, but also the oxidation states of the reactants.

The laws of catalysis complement the family of fundamental laws of Nature. With the advent of the laws of catalysis, the catalytic phenomenon is capable of becoming a scientific field in its own right, "*not just an interdisciplinary mixture of chemistry and physics.*" [7].

24. Conclusions

1. For many years, the main secret of catalysis was the undisclosed role of the oxidation rates of the catalyst and reagents in the mechanism of catalysis. The oxidation states are used for a new purpose and appear in the laws and formulas of catalysis as quantitative values and characteristics of the catalyst and reagents.

2. Catalysis belongs to the class of fundamental phenomena and is interdisciplinary in nature.

3. The laws of catalysis and formulas for calculating TPO and TON are combinations of quantities of chemical and physical nature.

4. The universal law of the rate of catalysis is represented by a combination of quantities of chemical and physical nature:

$$v_{cat} = \frac{Q_{\Sigma}}{F \cdot t \cdot m_1 \cdot |z_1 - z_2|}$$

Fig. 35. Universal law of the rate of catalysis.

5. From the universal law of the rate of catalysis as special cases follow the rate laws of heterogeneous, homogeneous and field catalysis:

$$v_{He} = \frac{e \cdot n_a \cdot |k_1 - k_2|}{F \cdot (\tau_D + \tau_A) \cdot m_1 \cdot |z_1 - z_2|}$$

$$v_{Ho} = \frac{e \cdot n_a}{F \cdot (\tau_D + \tau_A) \cdot m_1 \cdot |z_1 - z_2|}$$

$$v_{Fcat} = \frac{e \cdot f_e \cdot E_{cat}}{F \cdot m_1 \cdot |z_1 - z_2| \cdot E_i}$$

Fig. 36. Rate laws of heterogeneous, homogeneous and field catalysis.

6. A generalized formula for the yield of a catalytic reaction is represented by a combination of quantities of chemical and physical nature:

$$n_{cat} = \frac{Q_{\Sigma}}{F \cdot m_1 \cdot |z_1 - z_2|}$$

FIG. 37: Generalized formula for the yield of a catalytic reaction.

7. From the generalized catalytic reaction yield formula, as special cases, follow the reaction yield formulas of heterogeneous, homogeneous, field and combined catalysis:

$$n_{He} = \frac{e \cdot n_a \cdot t \cdot |k_1 - k_2|}{F \cdot (\tau_D + \tau_A) \cdot m_1 \cdot |z_1 - z_2|}$$

$$n_{Ho} = \frac{e \cdot n_a \cdot t}{F \cdot (\tau_D + \tau_A) \cdot m_1 \cdot |z_1 - z_2|}$$

$$n_{Fcat} = \frac{t \cdot e \cdot f_e \cdot E_{cat}}{F \cdot m_1 \cdot |z_1 - z_2| \cdot E_i}$$

$$n_{Comb} = \frac{e \cdot n_a \cdot (|k_1 - k_2| + \dots + |r_1 - r_2|) \cdot t}{F \cdot (\tau_D + \tau_A) \cdot m_1 \cdot |z_1 - z_2|}$$

Fig. 38. Reaction yields of heterogeneous, homogeneous, field and combined catalysis.

8. The generalized formula for calculating the TOF is represented by a combination of quantities of chemical and physical nature:

$$TOF_{cat} = \frac{Q_{\Sigma}}{e \cdot n_a \cdot t \cdot m_1 \cdot |z_1 - z_2|}$$

Fig. 39. Generalized formula for calculating the TOF.

9. From the generalized formula for the calculation of TOF, as special cases, follow formulas for the calculation of TOF in heterogeneous, homogeneous and field catalysis:

$$TOF_{He} = \frac{|k_1 - k_2|}{(\tau_D + \tau_A) \cdot m_1 \cdot |z_1 - z_2|}$$

$$TOF_{Ho} = \frac{1}{(\tau_D + \tau_A) \cdot m_1 \cdot |z_1 - z_2|}$$

$$TOF_{Fcat} = \frac{f_e \cdot E_{cat}}{m_1 \cdot |z_1 - z_2| \cdot E_i}$$

Fig. 40. Formulas for calculating TOF in heterogeneous, homogeneous, and field catalysis.

10. The parameter in the generalized laws of catalysis is the total electric charge transferred to the reactants during the catalytic reaction. The formulas for calculating the total electric charge in heterogeneous, homogeneous, field, and combined catalysis are as follows:

$$Q_{He} = \frac{e \cdot n_a \cdot |k_1 - k_2| \cdot t}{(\tau_D + \tau_A)}$$

$$Q_{Ho} = \frac{e \cdot n_a \cdot t}{(\tau_D + \tau_A)}$$

$$Q_{Fcat} = \frac{e \cdot f_e \cdot E_{cat} \cdot t}{E_i}$$

$$Q_{Comb} = \frac{e \cdot n_a \cdot (|k_1 - k_2| + \dots + |r_1 - r_2|) \cdot t}{(\tau_D + \tau_A)}$$

Fig. 41. Formulas for calculating the electric charge carried on the reactants in heterogeneous, homogeneous, field and combined catalysis.

11. a chemical reaction that proceeds without a catalyst according to the scheme: $A+B \rightarrow AB$, in the presence of a catalyst will proceed according to the scheme: $A+B \rightarrow A(-) + B(+) \rightarrow AB$, (Fig. 42).

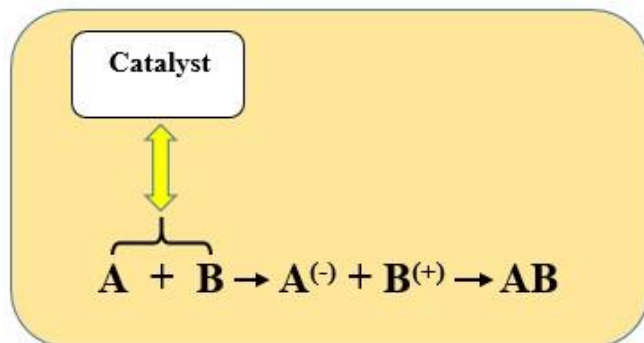


Fig. 42. Scheme of the chemical reaction in the presence of the catalyst.

12. The table of oxidation states of chemical elements becomes a guiding document and a useful aid in catalysis.

13. The donor-acceptor mechanism is the universal mechanism of catalysis. This mechanism is implemented in all types of catalysis.

14. The intermediate states of reactants and catalyst in a catalytic reaction are their states in an altered oxidation state.

15. The known use of Faraday's constant in chemistry is extended to the field of catalysis. In addition to its known use in the law of electrolysis and in the Nernst and Goldmann equations, the Faraday constant is included as an interaction constant in the laws of catalysis.

16. The laws of catalysis complement the family of fundamental laws of Nature.

17. With the emergence of the laws of catalysis, the catalytic phenomenon is able to become an independent scientific field.

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“Interdisciplinary knowledge of solid-state physics, chemistry, biology, and medicine, coupled with frontier engineering are needed to discover new catalysis science.”

“Междисциплинарные знания в области физики твердого тела, химии, биологии и медицины, в сочетании с передовой инженерией необходимы для открытия новой науки о катализе”

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Electrochemical Promotion of Catalysis: From Discovery to Fundamentals to Applications

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Электрохимическое продвижение катализа (ЕРОС) представляет собой электрохимически индуцированный каталитический эффект, который соответствует *in situ* обратимой модификации каталитического поведения катализатора металла или оксида металла - электродов, нанесенных на твердые электролиты или смешанные ионно-электронные проводники (МИЕС) при поляризации интерфейса электрод/электролит или МИЕС. Эта работа освещает ключевые ориентиры в ЕРОС, начиная с момента его открытия. В-первых, краткая история экспериментальной работы, которая привела к довольно неожиданному открытию ЕРОС при использовании в качестве катализаторов металлических пленочных электродов, нанесенных на $ZrO_2(Y_2O_3)$, а O^{2-} дирижер, представлен.