Volodymyr Kaplunenko
"Nanomaterials and Nanotechnologies Ltd",
Ukraine
e-mail: kaplunenkov@gmail.com

Mykola Kosinov
"Nanomaterials and Nanotechnologies Ltd",
Ukraine
e-mail: nkosinov@ukr.net

LAWS OF HOMOGENEOUS CATALYSIS

Abstract. *The proton donor-acceptor mechanism of catalysis makes it possible to study catalytic reactions at the elementary particle level, which made it possible to obtain the laws of homogeneous catalysis. The main characteristics of homogeneous catalysis were obtained from the rate law. The main parameter in the formulas of the laws of homogeneous catalysis is the total electric charge obtained by the reactants. It was shown that in such dissimilar and different types of catalysis as homogeneous and field catalysis, the same mechanism of catalysis is realized. This mechanism is based on the transfer of electric charges to the reagents by means of protons and electrons. The proton and electron donor-acceptor mechanisms are charge-symmetric mechanisms of catalysis. The difference between the proton donor-acceptor mechanism of homogeneous catalysis and the electronic donor-acceptor mechanism of field catalysis lies in the electrical charge carriers. The donor-acceptor mechanism claims to be a universal mechanism of catalysis. The key factor leading to a decrease in the activation energy of a chemical reaction is the change in the charge state of the reactants. A generalized law is obtained, from which the laws of homogeneous and field catalysis follow as particular results.*

Keywords: laws of homogeneous catalysis, donor-acceptor mechanism of catalysis, protons in catalysis, electric charge of the proton, oxidation states of reactants, reactivity of substances, Faraday's constant.

1. Introduction

The history of catalysis shows that many fundamental discoveries in catalysis were made either accidentally or empirically by trying out a large number of substances. Despite the discovery of many types of catalysis, their laws remain undiscovered. The characteristics of the catalysts that set the values of the characteristics of catalysis have not been identified. This situation has given rise to the statement repeatedly encountered in the literature that "*catalysis is more of an art than a science*" [1 - 6]. The preparation of catalysts is also considered an art, and a successful catalyst is created by trial and error rather than by scientific analysis of the processes [4 - 6]. In this regard, the scientific substantiation of catalysis is an urgent task.

The aim of this article is to study homogeneous catalysis and to identify the characteristics of the catalyst that set the values of the characteristics of catalysis, as well as to obtain the laws of homogeneous catalysis. To solve these problems, the donor-acceptor mechanism of catalysis, which allows us to study catalytic reactions at the level of elementary particle interaction, is taken as the basis. Earlier adoption of this mechanism made it possible to obtain the laws of heterogeneous and field catalysis [7 - 10].

2. Studies of catalysis at the level of elementary particle interaction

In [7 - 10], the electronic donor-acceptor mechanism of catalysis was investigated instead of the generally accepted mechanism of the formation of intermediate compounds in catalysis. As a result, the laws of heterogeneous and field catalysis were obtained. This mechanism of catalysis with some modification can also be applied to homogeneous catalysis. In homogeneous catalysis, the proton donor-acceptor mechanism is implemented instead of the electronic mechanism. The proton and electronic donor-acceptor mechanisms of catalysis are very similar charge-symmetric mechanisms. These mechanisms of catalysis are implemented at the level of interaction of elementary particles. The difference between the proton donor-acceptor mechanism of homogeneous catalysis and the electronic donor-acceptor mechanism of field and heterogeneous catalysis consists only in the carriers of the electrical charge. Electrical charges in homogeneous catalysis are carried by protons. Electric charges in field and heterogeneous catalysis are carried by electrons [8, 10].

The model of donor-acceptor interaction between catalyst and reagents was first proposed by Thomas Martin Lowry in 1925-1928. He formulated the idea of a proton-donor-acceptor mechanism as applied to homogeneous catalytic processes. According to Lowry, homogeneous catalysis is caused by the alternating interaction of a reactant molecule with a catalyst. The mechanism consists in the addition by the reactant of a proton obtained from the donor catalyst and the subsequent transfer of protons to the catalyst-acceptor [12, 13].

The proton and the electron have much in common. Proton and electron are fundamental elementary particles. They have the same elementary electric charge (e = 1.602,176,634 × 10-19 C), which is a fundamental physical constant. In homogeneous catalysis, as in heterogeneous and field catalysis, the fundamental law of nature, the law of conservation of electric charge, is fulfilled.

The proton mechanism differs from the electron mechanism in that protons carry an electric charge of the opposite sign. Therefore, the electronic and proton-donor-acceptor mechanisms can be viewed as charge-conjugate or charge-symmetric mechanisms of catalysis. The donor function in the electronic donor-acceptor mechanism corresponds to the acceptor function in the proton donor-acceptor mechanism. The acceptor function in the electronic donor-acceptor mechanism corresponds to the donor function in the proton donor-acceptor mechanism.

The transfer of electrical charges by protons in homogeneous catalysis leads to a change in the oxidation state of the reactants [7 - 10]. This causes ionization of the reactants and, as a consequence, increases the reactivity of the substances, which leads to the acceleration of the chemical reaction [11].

The reaction of the type A+B = AB in the presence of a homogeneous Cat catalyst can be represented by the scheme shown in Fig. 1.

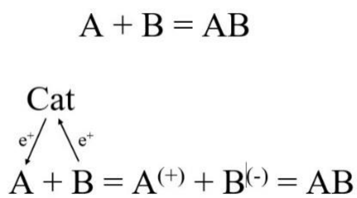

Fig. 1. *A, B - reagents. AB - reaction product.* $A^{(+)}$, $B^{(-)}$ *- ionized reagents. Cat - homogeneous catalyst.* e^{+} *- proton charge.*

The electric charge of the proton and the number of protons involved in the reaction are the main quantitative characteristics of the catalyst in homogeneous catalysis. These characteristics are the parameters in the formulas of the laws of homogeneous catalysis.

3. Laws of homogeneous catalysis

The following are the laws of homogeneous catalysis derived from the proton donor-acceptor mechanism of catalysis. A distinguishing quantitative feature of a homogeneous catalyst from a heterogeneous one is that the change in the oxidation degree of the catalyst does not exceed a value equal to one. This is due to the single proton charge. This feature is reflected in the formulas for the laws of homogeneous catalysis.

The 1st law of homogeneous catalysis (the law of the rate of homogeneous catalysis) has the form:

$$v_{Ho} = \frac{e \bullet n_a}{F \bullet (\tau_D + \tau_A) \bullet m_1 \bullet |z_1 - z_2|}; \qquad (1)$$

where: v_{Ho} *- catalytic reaction rate (mol/s); number of active centers of the catalyst; F - Faraday constant; e - electric charge of the proton;* τ_D *- time of the donor half-cycle of catalysis;* τ_A *- time of the acceptor half-cycle of catalysis;* z_1 *− degree of oxidation of the reactant in the initial product;* z_2 −

degree of oxidation of the reactant in the final product; m_1 - number of reactant atoms in the final product molecule.

The ratio for calculating the catalytic reaction yield (n_{Ho}) directly follows from the 1st law of catalysis:

$$n_{Ho} = \frac{e \bullet n_a \bullet t}{F \bullet (\tau_D + \tau_A) \bullet m_1 \bullet |z_1 - z_2|}; \qquad (2)$$

where: t - is the time of catalytic reaction.

The 2nd law of homogeneous catalysis has the form:

$$RON \bullet n_a = n_{Ho} \bullet m_1 \bullet |z_1 - z_2| \bullet N_A = n_{Ho} \bullet m_2 \bullet |q_1 - q_2| \bullet N_A; \qquad (3)$$

where: m_1 - is the number of atoms of the first reactant in the final product molecule; m_2 - is the number of atoms of the second reactant in the final product molecule; z_1 - is the oxidation degree of the first reactant in the initial substance; z_2 - is the oxidation degree of the first reactant in the final product; q_1 - is the oxidation degree of the second reactant in the initial substance; q_2 - is the oxidation degree of the second reactant in the final product; N_A is Avogadro number.

The second law of homogeneous catalysis is a direct consequence of one of the basic laws of nature - the law of conservation of electric charge.

The 3rd law of homogeneous catalysis (catalytic balance equation) has the form:

$$N_{kat}^{red} + N_{kat}^{ox} = m_1 \bullet N_1^{red} + m_2 \bullet N_2^{ox}; \qquad (4)$$

where: N_{kat}^{red} - number of elementary acts of reduction of the catalyst; N_{kat}^{ox} - number of elementary acts of oxidation of the catalyst; N_1^{red} - number of elementary acts of reduction of the first reactant; N_2^{ox} - number of elementary acts of oxidation of the second reactant; m_1 - number of atoms of the first reactant in the final product molecule; m_2 - number of atoms of the second reactant in the final product molecule.

Equations (1) and (2) can be represented as:

$$v_{Ho} = \frac{e \bullet n_a \bullet TOF}{F}; \qquad (5)$$

$$n_{Ho} = \frac{e \bullet n_a \bullet TON}{F}; \qquad (6)$$

The ratio for TOF and TON is as follows:

$$TOF = \frac{1}{(\tau_D + \tau_A) \bullet m_1 \bullet |z_1 - z_2|}; \qquad (7)$$

$$TON = \frac{t}{(\tau_D + \tau_A) \bullet m_1 \bullet |z_1 - z_2|}; \qquad (8)$$

Using the ROF and RON catalyst characteristics proposed in [7, 8], formulas (5) and (6) can be represented as:

$$v_{Ho} = \frac{e \bullet n_a \bullet ROF}{F \bullet \sigma}; \qquad (9)$$

$$n_{Ho} = \frac{e \bullet n_a \bullet RON}{F \bullet \sigma}; \qquad (10)$$

ROF and RON values can be calculated in two ways [7, 8]:

$$ROF = \frac{n_{Ho} \bullet N_A \bullet \sigma}{t \bullet n_a} = \frac{1}{\tau_D + \tau_A}; \qquad (11)$$

$$RON = \frac{n \bullet N_A \bullet \sigma}{n_a} = \frac{t}{\tau_D + \tau_A}; \qquad (12)$$

where: - Horiuchi stoichiometric number [14].

The laws of homogeneous catalysis are given in Table 1.

Table1.

Name	**The formula for the law of catalysis**	**Note**
1st law of homogeneous catalysis	$v_{Ho} = \frac{e \bullet n_a}{F \bullet (\tau_D + \tau_A) \bullet m_1 \bullet \lvert z_1 - z_2 \rvert}$	The rate law of homogeneous catalysis
2nd law of homogeneous catalysis	$RON \bullet n_a = n_{Ho} \bullet m_1 \bullet \lvert z_1 - z_2 \rvert \bullet N_A = n_{Ho} \bullet m_2 \bullet \lvert q_1 - q_2 \rvert \bullet N_A$	Law of Conservation in Homogeneous Catalysis
The 3rd law of homogeneous catalysis	$N_{kat}^{red} + N_{kat}^{ox} = m_1 \bullet N_1^{red} + m_2 \bullet N_2^{ox};$	Catalytic balance equation
Homogeneous catalysis	*Homogeneous catalysis is the initiation or acceleration of a chemical reaction in the presence of a catalyst, which is in the same phase as the reactants and products, by reducing the activation energy of the reaction by changing the oxidation state of the reactants during proton donor-acceptor interaction of the catalyst with the reactants.*	

The main characteristics of homogeneous catalysis and formulas for their calculation are given in Table 2.

Table 2.

Designation and name	Formulas	Note
n_{Ho} amount of product	$n_{Ho} = \frac{e \bullet n_a \bullet t}{F \bullet (\tau_D + \tau_A) \bullet m_1 \bullet \lvert z_1 - z_2 \rvert}$	
n_{Ho} amount of product	$n_{Ho} = \frac{e \bullet n_a \bullet TON}{F}$	
The rate of homogeneous catalysis	$v_{Ho} = \frac{e \bullet n_a \bullet TOF}{F}$	
TOF turnover frequency	$TOF = \frac{n_{Ho} \bullet N_A}{t \bullet n_a}$; $TOF = \frac{1}{(\tau_D + \tau_A) \bullet m_1 \bullet \lvert z_1 - z_2 \rvert}$	
TON turnover number	$TON = \frac{n_{Ho} \bullet N_A}{n_a}$; $TON = \frac{t}{(\tau_D + \tau_A) \bullet m_1 \bullet \lvert z_1 - z_2 \rvert}$	
ROF redox frequency	$ROF = \frac{n_{Ho} \bullet N_A \bullet \sigma}{t \bullet n_a} = \frac{1}{\tau_D + \tau_A}$	
RON redox number	$RON = \frac{n \bullet N_A \bullet \sigma}{n_a} = \frac{t}{\tau_D + \tau_A}$	

4. Homogeneous and field catalysis have common features

It is of interest to study such dissimilar and different types of catalysis as homogeneous and field catalysis in order to identify their common features. The main distinguishing feature between homogeneous and field catalysis is the catalyst used. These types of catalysis use catalysts that are radically different in nature. In homogeneous catalysis, the catalyst is a substance. Field catalysis uses a non-material catalyst, the field [9]. Despite their radical differences, these types of catalysis also have some features in common.

It follows from the law of field catalysis that in field catalysis the electronic donor-acceptor mechanism of catalysis is implemented. In field catalysis, the catalyst field acts as a generator of electrons when interacting with the reagents. The field external action on the reactants changes the oxidation state of the reactants. Field catalysts participate in the chemical reaction by field energy, which leads to the generation of electrons in the substance of the reactants. In this case, the role of an electron donor is performed by one of the reactants, and the role of an electron acceptor is performed by the other reactant. This makes it possible to reduce the activation energy of the reaction by changing the oxidation state of the reactants. The catalyst (field) itself remains unchanged and participates in the reaction only with its energy.

In homogeneous catalysis, the proton donor-acceptor mechanism of catalysis is realized. It follows from the laws of homogeneous catalysis (1) - (6) that the catalyst substance in homogeneous catalysis acts as a proton donor and acceptor when interacting with reagents. The protons of the catalyst transfer an electric charge to the reactants, which leads to a change in the oxidation state of the reactants and makes it possible to reduce the activation energy of the reaction. At the same time, the catalyst itself remains unchanged and participates in the reaction only by charge transfer via protons.

As we can see, in such different types of catalysis as homogeneous and field catalysis, a single donor-acceptor mechanism of catalysis is realized. The only slight difference is that in homogeneous catalysis, the carriers of electric charges to the reactants are protons, while in field catalysis they are electrons. At the same time, both protons and electrons carry equal electric charges. The common feature between homogeneous and field catalysis is the type of interaction between the catalyst and the reactants. This interaction is realized at the fundamental level, the level of elementary particles [9].

5. Generalized law of homogeneous and field catalysis

The consequence of the homogeneous mechanism of homogeneous and field catalysis is that the laws of these two types of catalysis also have common features. These common features can be seen in the above formula (2) for calculating the yield of the reaction of homogeneous catalysis and the formula for calculating the yield of the reaction of field catalysis:

$$n_{Fcat} = \frac{t \bullet e \bullet f_e \bullet E_{cat}}{F \bullet m_1 \bullet |z_1 - z_2| \bullet E_i}; \qquad (13)$$

where: n_{Fcat} is the reaction yield of field catalysis (mol); fe - is the frequency of field influence on the reactants; E_{cat} - is the field energy spent on catalysis during one cycle of catalysis (J); Ei - is the ionization energy of the reactants (J); F - is the Faraday constant; e - is the electric charge of the electron; z_1 – degree of oxidation of the reactant in the initial product; z_2 – degree of oxidation of the reactant in the final product; m_1 - is the number of reactant atoms in the final product molecule; t - is the reaction time.

In formula (2) of homogeneous catalysis, we see a dimensionless combination of quantities that includes the time of catalysis *t*, time of donor half-cycle of catalysis τ_D, and time of acceptor half-cycle of catalysis τ_A , as well as the number of n_a active centers of the catalyst:

$$N_{e,p} = N_p = \frac{n_a \bullet t}{(\tau_D + \tau_A)} \qquad (14)$$

This combination of values (14) is nothing but the total number of protons *Np* involved in homogeneous catalysis.

In formula (13) to calculate the yield of the reaction of field catalysis, we also see a dimensionless combination of quantities that includes the time of field catalysis, the frequency of field influence on the reagents f_e, the field energy charged to catalysis *Ecat*, and the ionization energy of the reagents *Ei*:

$$N_{e,p} = N_e = \frac{f_e \bullet E_{cat} \bullet t}{E_i} \qquad (15)$$

This combination of values (15) is nothing but the total number of electrons *Ne* obtained by the reactants during field catalysis. The number of electrons *Ne* and the number of protons *Np* are the same signs of *Ne,p* in the formulas of the laws of homogeneous and field catalysis. Equal amounts of electrons and protons carry equal amounts of electric charges.

Thus, for homogeneous and field catalysis, we can propose a single generalized formula for the law of catalysis speed v_{cat} and a single generalized formula for calculating the reaction yield of catalysis n_{cat},. The formulas include either the number of electrons *Ne* or the number of protons *Np* as a parameter. In formulae (14) and (15) this parameter is designated as *Ne,p*. As a result, we obtain such generalized formulas for the law of speed and yield of the catalysis reaction:

$$v_{Ho,F_{cat}} = \frac{e \bullet N_{e,p}}{F \bullet t \bullet m_1 \bullet |z_1 - z_2|} \qquad (16),$$

$$n_{Ho,F_{cat}} = \frac{e \bullet N_{e,p}}{F \bullet m_1 \bullet |z_1 - z_2|} \qquad (17)$$

The generalized formulas can be represented using the Avogadro N_A constant, taking into account the known relation of the fundamental physical constants $F = eN_A$.

$$v_{Ho,F_{cat}} = \frac{N_{e,p}}{N_A \bullet t \bullet m_1 \bullet |z_1 - z_2|} \qquad (18)$$

$$n_{Ho,F_{cat}} = \frac{N_{e,p}}{N_A \bullet m_1 \bullet |z_1 - z_2|} \qquad (19)$$

Formulas (16) and (17) at the value of *Ne,p* determined by formula (14) turn into relations (1) and (2), which are true for homogeneous catalysis.

$$v_{Ho} = \frac{e \bullet N_{e,p}}{F \bullet t \bullet m_1 \bullet |z_1 - z_2|} = \frac{e \bullet n_a}{F \bullet (\tau_D + \tau_A) \bullet m_1 \bullet |z_1 - z_2|}; \qquad (20)$$

$$n_{Ho} = \frac{e \bullet N_{e,p}}{F \bullet m_1 \bullet |z_1 - z_2|} = \frac{e \bullet n_a \bullet t}{F \bullet (\tau_D + \tau_A) \bullet m_1 \bullet |z_1 - z_2|} \qquad (21)$$

Formulas (16) and (17), with the value of *Ne,p* determined by formula (15), turn into relations that are true for field catalysis [11].

$$v_{Fcat} = \frac{e \bullet N_{e,p}}{F \bullet t \bullet m_1 \bullet |z_1 - z_2|} = \frac{e \bullet f_e \bullet E_{cat}}{F \bullet m_1 \bullet |z_1 - z_2| \bullet E_i}; \qquad \mathbf{(22)}$$

$$n_{Fcat} = \frac{e \bullet N_{e,p}}{F \bullet m_1 \bullet |z_1 - z_2|} = \frac{e \bullet f_e \bullet E_{cat} \bullet t}{F \bullet m_1 \bullet |z_1 - z_2| \bullet E_i}; \qquad (23)$$

The generalized formulas (16) and (17) include two fundamental physical constants: the elementary charge (*e = 1.602 176 634 × 10-19 C*) and the Faraday constant (*F = 96 485.332 12 . . . C* mol^{-1}) [16].

At the physical level, the main function of catalysts in homogeneous and field catalysis is the generation of elementary particles - protons or electrons [7 - 10] and changing the oxidation state of reactants with their participation. This mechanism, which is based on the transfer of electric charges to and from the reactants, is the key mechanism in catalysis. Due to this mechanism, the activation energy of the chemical reaction is reduced due to the electrical polarization of the reactants [11]. The table of oxidation degrees of chemical elements becomes a guiding document and a useful aid in catalysis [16].

As we can see, such dissimilar and different types of catalysis, as homogeneous and field catalysis, are subject to a common law. This gives one confidence that similar generalizations are possible for other types of catalysis.

6. Conclusion

Protons are the real participants in homogeneous catalysis. The electric charge of the proton is the active factor in homogeneous catalysis. Protons implement the mechanism to accelerate chemical reactions based on the donor-acceptor interaction of the catalyst and reagents. In the proton-donor-acceptor mechanism, the homogeneous catalyst acts as a proton donor and acceptor.

The adoption of the proton donor-acceptor mechanism of catalysis allowed the laws of homogeneous catalysis and the most important characteristics of homogeneous catalysis to be obtained. The main parameter in the formulas of the laws of homogeneous catalysis is the total electric charge obtained by the reactants.

Adopting a proton donor-acceptor mechanism for homogeneous catalysis allowed us to identify the characteristics of the catalyst and reagents that set the values of the characteristics of homogeneous catalysis. This allowed the dependence of the catalytic reaction rate to be described using a single equation.

The main function of a homogeneous catalyst is to transfer electric charges to the reactants via elementary particles - protons and change the oxidation state of the reactants with their participation. The change in the charge state of the reactants during the implementation of the proton donor-acceptor mechanism of homogeneous catalysis is the key factor that leads to a decrease in the activation energy of the chemical reaction.

7. Conclusions

1. The laws of homogeneous catalysis and relations for calculating the main characteristics of homogeneous catalysis have been obtained. This was made possible by the study of a deeper level of interaction in catalysis [9]. Instead of the interaction on the molecular and atomic level (the mechanism of intermediate compounds), the donor-acceptor mechanism which realizes the interaction on the elementary particle level has been investigated.

2. The most important characteristic of a homogeneous catalyst is the number of protons, through which the catalyst transfers electric charges to the reactants. The number of protons and the proton charge set the value of the characteristics of the catalysis.

3. The most important characteristics of the reactants are their oxidation states. The oxidation states of the reactants and the number of protons, together set the values of the characteristics of homogeneous catalysis. They are the parameters in the formulas of the laws of catalysis.

4. The change in the oxidation states of the reactants in homogeneous catalysis is the main result of the proton donor-acceptor mechanism of catalysis. It increases the reactivity of the reactants and leads to a decrease in the activation energy of the chemical reaction.

5. Such dissimilar and different types of catalysis as homogeneous and field catalysis have common features. They are united by the type of interaction between the catalyst and the reactants, which is realized at the level of interaction of elementary particles.

6. It has been shown that in homogeneous catalysis and field catalysis, a unified donor-acceptor mechanism of catalysis is implemented, the only difference being that in homogeneous catalysis the carriers of electric charges are protons, and in field catalysis, they are electrons.

7. The generalized law from which, as particular results, the laws of homogeneous and field catalysis follow.

8. The boundaries of applicability of the donor-acceptor mechanism of catalysis go far beyond homogeneous and field catalysis. The donor-acceptor mechanism claims to be a universal mechanism of catalysis. It makes it possible to consider catalytic processes not at the atomic and molecular level, but at the level of interaction of elementary particles.

9. The laws of catalysis show that a change in the oxidation state of substances under the action of catalysts leads to a change in their reactivity. The reactivity of the reactants is the greater the difference in their oxidation states.

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

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