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ELECTRIC FIELD - INDUCED CATALYSIS. LAWS OF FIELD CATALYSIS

***Abstract.** The article explores a new type of catalysis - electric field catalysis. The laws of field catalysis are given. The characteristics of the electric field are determined, which set the values of the characteristics of the field catalysis. Field catalysis and field catalyst do not fit into the traditional definition of catalysis and catalyst, which may require a revision of the terminology of catalysis. The field is a more versatile catalyst compared to material catalysts, both in terms of its application to a wider range of chemical reactions, and in the ability to control the rate and selectivity. It is shown that a common donor-acceptor mechanism of catalysis is realized in heterogeneous and field catalysis. Generalized formulas are obtained, from which, as partial results, the laws of heterogeneous and field catalysis follow. New definitions of catalyst and field catalysis are given. The class of material catalysts has been expanded and supplemented with field catalysts.*

***Keywords:** electric field catalysis, laws of field catalysis, electric field-immaterial catalyst, donor-acceptor mechanism of catalysis, overcoming the Sabatier maximum, oxidation states of reagents, reactivity of substances, Faraday constant.*

1. Introduction

Despite the successes achieved in catalysis, the general mechanism of catalytic reactions still remains undiscovered and the laws of catalysis are not discovered. The terminology of catalysis remains incomplete and, in some cases, contradictory.

The definition of catalysis, which was given by G.K. Boreskov, is known: “*catalysis is the initiation or acceleration of chemical reactions in the presence of substances (catalysts) that repeatedly enter into intermediate chemical interaction with reagents and restore their chemical composition after each cycle of such interaction*” [1 , 2]. In this definition, the class of chemical reaction catalysts is limited to substance catalysts only,

which "enter into an intermediate chemical interaction with reagents and restore their chemical composition after each cycle of such interaction."

Known definition of catalyst given by Boudart "A catalyst is a substance that transforms reactants into products, through an uninterrupted and repeated cycle of elementary steps in which the catalyst is changed through a sequence of reactive intermediates, until the last step in the cycle regenerates the catalyst in its original form" [3 , 4]. In Boudart's definition, a catalyst is represented by a substance.

The IUPAC Golden Book defines a catalyst as follows: "A catalyst is a substance that increases the rate of a reaction without changing the overall standard change in the Gibbs energy of the reaction. A catalyst is both a reactant and a reaction product" [5]. And in this formulation, the catalyst is defined as a substance.

At the same time, catalysis never ceases to amaze with the variety of types of catalytic reactions. Over a long period of research on catalysis, a new type of physical influence on chemical reactions with the help of an external electric field has been revealed, which is proposed to be considered a physical catalyst or pseudocatalyst [6, 7]. The practice of catalysis has shown that the variety of catalysts is not limited to material catalysts. An immaterial object - a field - can also act as a catalyst. Since the field is neither a reactant, nor a reaction product, nor a substance [6], the traditional definition of a catalyst as a substance is not a universal definition. The field, as a catalyst or pseudo-catalyst, is beyond the scope of generally accepted definitions. Both the definition of catalysis given by Boreskov, and the definition of a catalyst given by M. Boudart and the IUPAC definition of a catalyst cannot be extended to field catalysis. As we can see, the practice of catalysis is far ahead of the well-known ideas about catalysis and catalysts.

2. Electron-induced catalysis

It was shown in [8, 9] that electrons are the main active factor in heterogeneous catalysis, and the catalyst-substance implements a donor-acceptor interaction with the reagents, in which the oxidation state of the reagents changes. The assumption about the determining role of electrons in heterogeneous catalysis was put forward by Ostwald [10].

Similar ideas about the role of electrons in catalysis were expressed about 90 years ago by L.V. Pisarzhevsky [11, 12]. The mechanism of catalysis based on the transfer of electrons was pointed out by S. Z. Roginsky [13].

In the field of catalysis, many examples of catalytic reactions have accumulated, where electrons play the role of catalysts [14–18]. We can assume that any factor affecting the ability to donate or accept electrons by reagents can affect the catalytic activity. Therefore, the role of a catalyst can be played directly by the electrons themselves or by the electromagnetic field as a generator of electrons.

3. Electric field-induced catalysis

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*" [19, 20]. 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. 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 [10], Pisarzhevsky [11, 12], Roginsky [13] and Volkenshtein [21] further developed the idea of the electrical mechanism of catalysis.

L. W. Pisarzhevsky 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. Pisarzhevsky 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 [22].

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 [1, 21]. This problem cannot be solved by physics alone.

In recent years, the number of works investigating electric mechanisms of catalysis and electric fields in catalysis has increased dramatically [23 - 66]. It has been discovered that the electric field plays a key role in various types of catalysis [62 - 64]. 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.*" [25, 26, 30, 31]. Some articles refer to the electric field as a class of reagents and call it a "*smart reagent*" [48 - 66].

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* [36, 38, 59, 60]. The immaterial field catalyst has many advantages [28]. 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 [18, 28, 29, 59, 61].

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. 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.

4. Role of spontaneous electric fields in catalysis

In addition to the direct effect of an external field on the reagents and on the catalyst, the effect of internal electric fields on catalytic activity and selectivity was revealed [23, 33]. 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 [33]. 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 [33, 56]. Since spontaneous electric fields are present in all types of catalysis, [33] concluded that *electrostatic phenomenology can be inherent in all types of catalysis*. It is noted in [65] that any heterogeneous process should be considered as having parallels with electrochemical processes.

5. Donor-acceptor mechanism of catalysis

It was shown in [8, 9] that in heterogeneous catalysis, an electron donor–acceptor mechanism for accelerating a chemical reaction is realized. The catalyst substance, when interacting with the reagents, acts as an electron donor and acceptor. The electrons of the catalyst make it possible to reduce the activation energy of the reaction by changing the oxidation state of the reactants. At the same time, the catalyst (substance) itself remains unchanged and participates in the reaction only with its electrons.

The $A + B = AB$ reaction in the presence of a heterogeneous Cat catalyst can be represented by the scheme shown in FIG. 1.

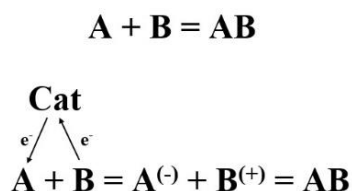


Fig.1. A , B - reagents; AB is the reaction product; $A^{(-)}$, $B^{(+)}$ are ionized reagents; Cat is a heterogeneous catalyst; e^- is an electron.

If the catalyst is an external electromagnetic field, then the donor-acceptor mechanism of catalysis is also realized. Under the field external action, the state of oxidation of the reagents changes due to the transfer of electrons. Field catalysts participate in the reaction with the energy of the field, 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 reagents, and the role of an electron acceptor is performed by another reagent. This makes it possible to reduce the activation energy of the reaction by

changing the oxidation state of the reactants. At the same time, the catalyst itself (the field) remains unchanged and participates in the reaction with its energy and frequency.

The reaction of the form $A + B = AB$ in the presence of a field catalyst can be represented by the scheme shown in Fig. 2.

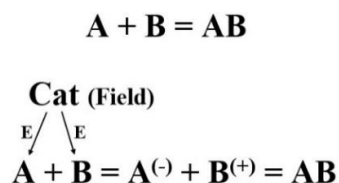


Fig. 2. A, B - reagents; AB is the reaction product; $A^{(-)}$, $B^{(+)}$ are ionized reagents; Cat(Field) - field catalyst; E - field energy.

The main function of catalysts is the generation of elementary particles - protons or electrons and the change with their participation in the oxidation state of reagents. This mechanism is based on the transfer of electric charges to and from the reactants. It is the main mechanism of catalysis. Due to this mechanism, the activation energy of a chemical reaction decreases due to the electric polarization of the reagents.

The donor-acceptor mechanism makes it possible to consider catalytic processes at a deeper level of physical reality. Instead of interaction at the level of molecules and atoms (mechanism of intermediate compounds), catalysis is studied at the level of interaction of elementary particles.

6. The influence of an external electric field on catalysis.

It is well known that the electric field interacts only with electrically charged particles. Such objects are electrons, protons, ionized reagents, and dipoles. In Fig. 3, using examples of field catalysis and combined catalysis, shows the mechanism of action of the electric field in catalysis.

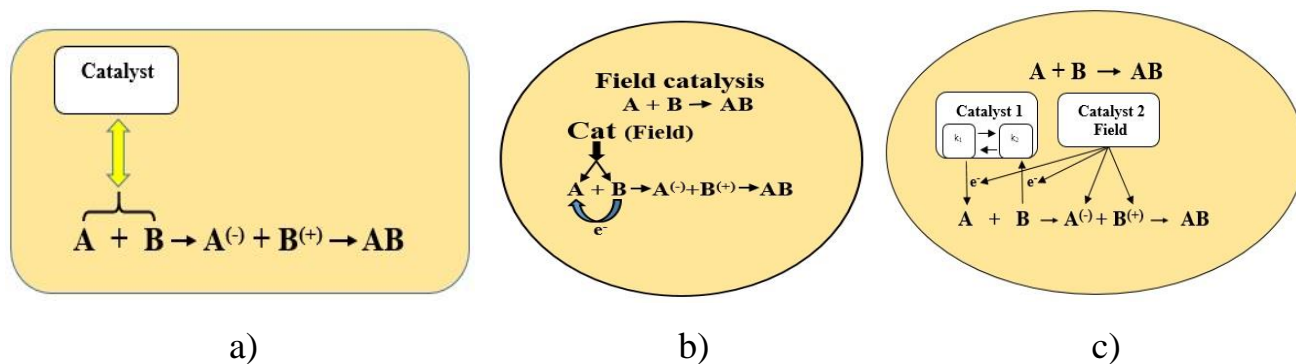


Fig. 3. Mechanism of action of electric field on catalytic reaction. a) - general scheme of mechanism; b) - field catalysis mechanism; c) - mechanism of combined (heterogeneous + field) catalysis; A, B - reactants; AB - reaction product; k_1 - initial oxidation degree of catalyst; k_2 - final oxidation degree of catalyst; $A^{(-)}$ - reactant A in changed oxidation degree; $B^{(+)}$ - reactant B in changed oxidation degree; e^- - electrons.

7. Laws of field catalysis

The field catalysis rate law has the form [68, 71 - 76]:

$$v_{Fcat} = \frac{e \cdot f_e \cdot E_{cat}}{F \cdot m_1 \cdot |z_1 - z_2| \cdot E_i} \quad (1)$$

where: v_{Fcat} - field catalysis rate (mol/s); f_e - is the frequency of the field impact on the reagents; E_{cat} - is the field energy spent on catalysis during one catalysis cycle (J); E_i - is the ionization energy of the reagents (J); F - is Faraday's constant; e - is the electric charge of the electron; z_1 - the degree of oxidation of the reagent in the original product; z_2 - the degree of oxidation of the reagent in the final product; m_1 - is the number of reagent atoms in the final product molecule.

From the field catalysis rate law, the relation for determining the yield of the field catalysis reaction directly follows:

$$n_{Fcat} = \frac{t \cdot e \cdot f_e \cdot E_{cat}}{F \cdot m_1 \cdot |z_1 - z_2| \cdot E_i} \quad (2)$$

where: t - catalysis time (s)

The catalytic balance equation for field catalysis will be obtained by analogy with the 3rd law of heterogeneous catalysis [9]:

$$\frac{2 \cdot E_{cat}}{E_i} = m_1 \cdot N_1^{red} + m_2 \cdot N_2^{ox} \quad (3)$$

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

The relationship for the turnover frequency (TOF) of the catalytic reaction and the turnover number (TON) of the catalytic reaction for field catalysis is:

$$TOF_{Fcat} = \frac{f_e \cdot E_{cat}}{m_1 \cdot |z_1 - z_2| \cdot E_i} \quad (4)$$

$$TON_{Fcat} = \frac{t \cdot f_e \cdot E_{cat}}{m_1 \cdot |z_1 - z_2| \cdot E_i} \quad (5)$$

The relation for redox frequency (ROF) and redox number (RON) [8, 70] is:

$$ROF_{Fcat} = f_e \quad (6)$$

$$RON_{Fcat} = t \cdot f_e \quad (7)$$

The discovery of field catalysis and the addition of a class of material catalysts with an immaterial catalyst - a field, force us to reconsider the established terminology of catalysis and the very definition of a catalyst.

We propose the following definition for the definition of a catalyst: “*a catalyst is a substance or a field that reduces the activation energy of a chemical reaction by changing the oxidation state of the reagents during the donor-acceptor interaction of a material catalyst with the reagents or during the electromagnetic interaction of the field with the reagents.*”

Accordingly, the formulation of field catalysis: “*electric field catalysis is the initiation or acceleration of a chemical reaction under the action of an external field by reducing the activation energy of the reaction by changing the oxidation state of the reagents during the electromagnetic interaction of the field with the reagents.*”

8. Common features in the laws of field catalysis and heterogeneous catalysis

The unified mechanism of heterogeneous and field catalysis indicates that the laws of heterogeneous and field catalysis can also contain common features. To do this, we will analyze the formulas of the laws of heterogeneous and field catalysis. In [8, 9], the formula of the 1st law of heterogeneous catalysis is given and the relation for calculating the yield of the catalytic reaction (n) is obtained:

$$v = \frac{e \cdot n_a \cdot |k_1 - k_2|}{F \cdot (\tau_D + \tau_A) \cdot m_1 \cdot |z_1 - z_2|}; \quad (8)$$

$$n = \frac{e \cdot n_a \cdot t \cdot |k_1 - k_2|}{F \cdot (\tau_D + \tau_A) \cdot m_1 \cdot |z_1 - z_2|}; \quad (9)$$

where: v - is the rate of the catalytic reaction (mol/s); n - is the yield of the catalytic reaction (mol); n_a - the number of active sites of the catalyst involved in the reaction; F - is Faraday's constant; e - is the electric charge of the electron; τ_D - the time of the donor half-cycle of catalysis; τ_A - the time of the acceptor half-cycle of catalysis; k_1 - is the initial oxidation state of the catalyst; k_2 - is the final oxidation state of the catalyst; z_1 - the degree of oxidation of the reagent in the original product; z_2 - the degree of oxidation of the reagent in the final product; m_1 - is the number of reagent atoms in the final product molecule; t - is the time of the catalytic reaction.

In formulas (8) and (9) we see a dimensionless combination of quantities:

$$N_e = \frac{n_a \cdot |k_1 - k_2| \cdot t}{(\tau_D + \tau_A)} \quad (10)$$

This combination of quantities (10) is nothing but the number of electrons N_e obtained by the reactants during the heterogeneous catalysis.

In formula (2) for calculating the yield of a field catalysis reaction, we also see a dimensionless combination of quantities:

$$N_e = \frac{f_e \cdot E_{cat} \cdot t}{E_i} \quad (11)$$

This combination of quantities (11) is nothing but the number of electrons N_e obtained by the reagents during the field catalysis. The number of electrons N_e is a common feature in the formulas of the laws of heterogeneous and field catalysis.

9. Generalized law of field catalysis and heterogeneous catalysis

Thus, for heterogeneous and field catalysis, one can propose a single generalized formula for the catalysis rate law v_{cat} and a single generalized formula for calculating the catalysis reaction yield n_{cat} , which will include the number of electrons N_e as a parameter:

$$v_{cat} = \frac{e \cdot N_e}{F \cdot t \cdot m_1 \cdot |z_1 - z_2|} \quad (12)$$

$$n_{cat} = \frac{e \cdot N_e}{F \cdot m_1 \cdot |z_1 - z_2|} \quad (13)$$

10. Conclusion

With the appearance of a field catalyst in catalysis, a new situation is emerging that does not fit into the traditional understanding of both catalysis and catalysts. This applies both to the terminology of catalysis and to the generally accepted mechanism of catalysis.

The main active factors of field catalysis are field energy and frequency. Under their influence, electrons are generated in the substance. Electrons implement a single mechanism for accelerating chemical reactions in both heterogeneous and field catalysis. This mechanism is the donor-acceptor mechanism of catalysis.

There is a possibility of joint use of two types of catalysts - fields and substances. Exposure of a material catalyst to an external electric field can be the simplest and most efficient way to increase the catalysis rate above the Sabatier maximum [77 - 85].

11. Conclusions

1. The laws and characteristics of field catalysis include the fundamental constants e and F , the characteristics of the electric field, and the oxidation states of the reactants (fig. 4).

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

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

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

Fig. 4. Formulas of the laws of field catalysis and the ratio for calculating the TOF.

2. From the laws of field catalysis it follows that the reaction rate and the amount of the final product directly depend on the number of electrons N_e generated by the field in the substance of the reagents (fig. 5).

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

$$n_{cat} = \frac{e \cdot N_e}{F \cdot m_1 \cdot |z_1 - z_2|}$$

$$N_e = \frac{f_e \cdot E_{cat} \cdot t}{E_i}$$

Fig. 5. Formulas of generalized laws of catalysis and the number of electrons involved in field

3. The change in the oxidation state of reactants in field catalysis is the main factor that leads to a decrease in the activation energy of a chemical reaction (Fig. 6).

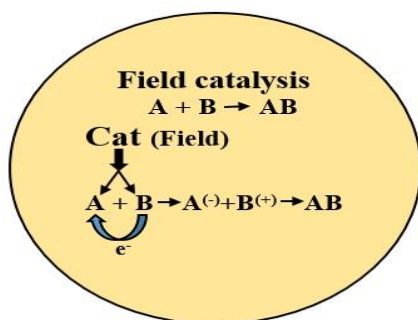


Fig. 6. Mechanism of action of electric field on catalytic reaction.

4. It has been shown that in such different types of catalysis as heterogeneous and field catalysis, the same mechanism of action on reagents by free electrons is realized, the end result of which is a change in the oxidation state of the reagents and, as a result, acceleration of the reaction.

5. It follows from the laws of field catalysis that the field is a more versatile catalyst compared to material catalysts both in terms of its application for a wider range of chemical reactions and in the possibility of controlling the rate and selectivity by changing the characteristics of the field.

6. Based on the laws of field catalysis, a definition of field-induced catalysis is given. A new definition of a catalyst is given, in which the class of material catalysts is extended and supplemented with field catalysts.

7. Both material and field catalysts implement a single donor-acceptor mechanism of catalysis. Material catalysts participate in the reaction with their electrons and act as electron donors and acceptors. Field catalysts participate in the reaction with the energy of the field, which leads to the generation of electrons in the substance of the reactants. In field catalysis, one of the reactants acts as an electron donor, and the second reactant plays the role of an electron acceptor (Fig. 6).

8. The donor-acceptor mechanism claims to be the universal mechanism of catalysis. The limits of applicability of the donor-acceptor mechanism of catalysis go far beyond the scope of heterogeneous and field catalysis. This mechanism is applicable to other types of catalysis.

9. The donor-acceptor mechanism makes it possible to study catalytic processes at a deeper level of physical reality. In it, instead of the interaction of the catalyst and reagents at the level of atoms and molecules, their interaction at the level of elementary particles is studied.

10. A generalized formula for the laws of field and heterogeneous catalysis has been obtained, from which, as partial results, the laws of heterogeneous and field catalysis follow. The main parameter in the generalized formula is the number of electrons involved in changing the oxidation state of the reactants.

11. The change in the oxidation state of the substances involved in the reaction is the main factor that leads to an increase in their reactivity.

12. The table of oxidation states becomes an important tool for selecting catalysts.

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