

INTERPRETATION OF CHEMICAL KINETICS AND CATALYSIS
IN THE TWO-YEAR COLLEGE CURRICULUM
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The main difficulty in teaching chemistry lies in the explanation of chemical phenomena on the basis of *quantum chemistry*: teachers have to explain things that they hardly understand themselves. The widely used quantum chemical explanations include paradigms and principles whose physical bases are not clear. The Transition State Theory employed to explain chemical reaction and catalysis phenomena at the introductory level provides a mechanistic (non-chemical) description and involves and incomprehensible transition state entity. According to J.N.Spencer (JCE No. 3 p. 182 1991) the basics of quantum-chemical interpretations are 'unteachable'. The main difficulties in explaining chemical bonding and chemical reactions are connected with the fact that this problem had never been solved by science in the previous century.

According to S. Lippard (C&EN, Aug. 7, 2000), as a result of the recent revolutionary changes in chemistry, older paradigms are giving way to new principles.

The changes in understanding and explaining chemical reactions in the course of the last chemical revolution took place on the level of changes of the paradigms. It was found that:

1) The interaction of saturated molecules proceeds, as a rule, not along the molecular mechanism, but through an intermediate formation of chemically activated particles (ions, radicals, conences, etc.). Active particles are formed of initial molecules in the course of dissociation that occurs as a result of thermal, photochemical, and other methods of increasing the energy, or via dissolving and dissociating the initial molecules in a solvent.

Previously, in the times of the old paradigm, it was supposed that direct molecular interaction was a common mechanism for chemical reactions, while reactions through intermediate active particles were regarded as special cases.

2) The interaction of active particles with saturated molecules proceeds not along the transition state (TS) situated at the summit of the potential barrier, but along the intermediate compound (IC) situated in the minimum.

3) In the common case (at the bottom of the old paradigm) it was supposed that the interaction of molecules between themselves, and between active particles with molecules, proceed in one stage. According to the new theory (i.e., new paradigm) the interaction of active particles

with molecules proceeds in three stages: 1) association, 2) electronic isomerization, and 3) dissociation.

4) The main driving force of chemical reactions, according to the old paradigm, was the kinetic energy of the transiting initial molecules — Theory of Active Collisions (TAC) and Transition State Theory (TST) — that is spent on the breaking of the old chemical bonds during the reaction. Here it was not clear exactly why most of the chemical bonds break in the course of chemical reactions that proceed at temperatures of 30° – 200°C, when most of the chemical bonds require over 2,000°C for thermal breaking.

According to the new paradigm, during the interaction of an active particle with a saturated molecule proceeding via the above indicated three stages, as a result of the electronic isomerization reaction, the old strong covalent bond turns into a weak Van der Waals bond without any energy expenditure or energy discharge during isomerization. Thermal energy is spent only?? on the formation of extremely small (>0.01 %) active particles.

5) In the framework of the old interpretation, the general explanation of catalysis turned out to be a ‘black hole’, while in the framework of the new interpretation of chemical reaction, the catalysis phenomenon becomes comprehensible and explainable at lessons of Introductory Chemistry relative to both general and special cases of catalytic reactions.

We have developed an alternative approach to describing chemical kinetics and catalysis based on experimental data that became available since the introduction of the Transition State Theory. A comprehensible explanation of the chemical kinetics and catalysis phenomena suitable for the two-year college program is herewith presented [details are provided at <http://www.itchem.com>].

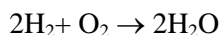
In the course of chemical transformation, the old bonds between atoms break and new ones form. Millions of chemical reactions take place simultaneously in live organisms at temperatures of 20° to 30°C.

As we have seen in the previous section??, we need energy of more than 200 kJ/mol (i.e., more than 2,000°C) to break a chemical bond. Thus, the main question relative to chemical reactions is: *How do chemical reactions proceed at temperatures of 20° to 30 °C if, in order to break a bond, a temperature of more than 2,000 °C is actually required?*

Before we go on to study complex reactions in organisms, let’s take up some simple examples, which can be studied in any school lab. One of the best-known and most instructive experiments is as follows:

The teacher prepares a mixture of two gases: oxygen and hydrogen in a test-tube. He/she demonstrates that these gases will never start a reaction by themselves. Then he lets the gas mixture go into a tin

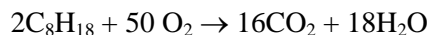
can through an opening at the bottom and he passes an electric current (spark) into the gas. The current causes an explosion, which is the result of a momentary interaction of the hydrogen and oxygen via the reaction:



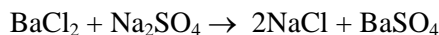
That is, in the process of this reaction, the bonds in molecules H_2 and O_2 were broken, and new bonds were formed between the hydrogen and the oxygen. It is worth mentioning here again that in order to break the bonds in molecules of hydrogen and oxygen a temperature of more than $2,000^\circ\text{C}$ is required.

Of course we can suppose that the spark had caused the momentary heating of the mixture up to this temperature. However, a test made by passing a spark separately into either of the two gases has shown that the temperature of these gases practically does not change.

Indeed, the motor of a car receives air (oxygen) and fumes of gasoline (mixture of hydrocarbons) but the motor will not work because the reaction takes place only when the motor is switched on (i.e., the spark appears). The process that takes place in a motor is well described by the reaction:



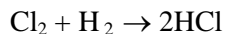
We can also observe in a similar experiment, that if we mix water solutions say, of barium chloride (BaCl_2) and sodium sulfate (Na_2SO_4), we will not get a transparent mixture as with other combinations; we will get some sediment of barium sulfate that is formed via the reaction:



In this case the bonds between barium (Ba) and chloride (Cl), and between sodium (Na^+) and sulfate (SO_4^{2-}) will break, though they actually require a temperature of more than $5,000^\circ\text{C}$ for their rupture.

It is of interest to note that if we dry both these salts at a temperature over 200°C and then mix them, there will be no reaction between them at all.

Other such phenomena are observed in interactions of chloride (Cl_2) and hydrogen (H_2). If we mix these gases in darkness, we will see that there is no reaction between them. If however, we momentarily irradiate the mixture with light, then the following reaction takes place:



What conclusions can we make on the basis of these examples?

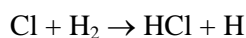
1) Reactions between molecules for which energy of 200 kJ/mol is required for bond breaking (i.e., heating to over 2,000°C) in reality do not proceed at normal temperatures (if the substances are mixed).

2) To proceed with the reaction, we need a momentary energetic stimulant (spark or flash) or the introduction of a third substance (water).

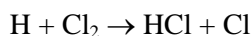
In order to understand the essence of these momentary energetic stimulants, scientists have studied the composition of gas after such coercions. ??? They have found that gas (oxygen + hydrogen) contains separate atoms of oxygen and hydrogen though the initial gases did not contain separate atoms. The concentration of separate atoms amounted to less than 0.01% of the number of molecules in the mixture. Thus, it is not the momentary action of the spark or light that leads to bond breaking in the molecules and to their dissociation into atoms, which then unite to form new molecules.

At the same time we must understand how the presence of separate atoms in tiny concentrations leads to an explosive interaction of the whole mass of hitherto passive molecules.

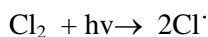
During the development of lab experiments, scientists invariably studied the interaction of separate atoms and ions with molecules. They have found that an atom of chloride rapidly reacts with a hydrogen molecule along the following scheme:



and that an atom of hydrogen also rapidly reacts with a chloride molecule even at room temperature according to the following scheme:



These results allow us to answer the question: *Why and how do reactions lead to the chemical transformation of molecules?* If we take the transformation of the mixture *hydrogen + chloride* the reaction can be described as follows — when this mixture is irradiated with light, a small portion of the chloride molecules dissociates into atoms via the scheme:



This scheme also explains why small concentrations of free atoms cause the transformation of many substances. Every atom performs a large number of cyclic transformations, i.e., this scheme shows how the chemical reaction between molecules proceeds, and the role of short-term energetic action upon reaction mixtures.

However, this scheme does not answer the main question, which arose during the studies of chemical reactions. Indeed, it was found that the interaction of molecules is possible via the chain route where the

carriers of the chain are radicals or ions. The mechanism scheme of the above-cited reaction has two more steps:



In the first of these reactions, all was logical. The chlorine molecule, under the influence of a portion of light (strong, energetic action) broke up into atoms. But in the second reaction (in the presence of the chlorine atom) the bond in the hydrogen molecule dissociated at room temperature without any additional energetic influence. That is, this scheme for chemical transformations allows specifying the main question, which is now formulated as follows:

Why is it that the interactions of:



in the course of which bonds between hydrogen atoms in molecule H_2 break, and bonds between chlorine atoms in molecule Cl_2 (that require heating to more than 2000°C and 700°C respectively) break during the reaction with chlorine atoms and hydrogen atoms at room temperature? ???

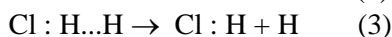
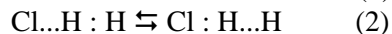
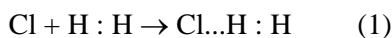
It is of interest to note that the answer to this question was first received theoretically (via discourse) and only then was it proven experimentally. To be more precise, the basis for the theory was a more strictly limited experimental material than the experimental material that appeared after the elaboration of the theory.

Well, what is the answer to the above-mentioned question?

The authors of this theory have suggested that the reaction of the active species (ion, radical, etc.) proceeds not in one step, but in three:

1) association, 2) electronic isomerization, 3) dissociation.

For example, in the case of interaction between a chlorine atom (Cl) and hydrogen (H_2) the reaction mechanism is described thus:



Thereon, the colon (:) represents electrons on the atoms' outermost orbit. Three dots (Cl...)?? indicate Van der Waals bonds (VWB).

How does the above reaction scheme answer the above-mentioned question?

Each of the three steps were studied experimentally, and it was found that each of the reactions proceeds at room temperature, and that the reaction rate of step #3 is close to that of the interaction of a chlorine

atom with hydrogen. It was also found that steps #1 and #2 proceeded with rates by two orders of magnitude higher than that of step #3. That is, the rate of the whole reaction can be defined by the rate of step #3, i.e., step #3 is the *rate-determining step*.

Now let's make out what the *rate determining step* is, or figure out why the slow reaction stage defines the speed of the whole process that proceeds subsequently via several stages. The speed of the whole chemical process is measured by the amount of final product in the given case, by the amount of HCl molecules formed, say, in one minute.

As already indicated, the reaction speeds of #1 and #2 are more than 100 times faster than that of #3. That is, reactions #1 and #2 reach equilibrium. Analogous processes, consisting of successive stages, are found in everyday life.

For example, let's take a poorly organized car assembly line at a factory. The body of a car is moving on a conveyor. During the movement of the conveyor, the body gradually gets its wheels, its motor, its interior, etc. One man is working on each stage of this conveyor.

In order to complete each operation of #1, #2, and #3, it would require, say, 10, 60, and 5 minutes respectively. Less than 5 minutes was spent on all the previous and consequent operations. Such a conveyor will produce one car in 60 minutes. That is, the time required for installing a motor is the time that corresponds to the slowest stage of the sequential process.

Relatively, stage #2 (the motor installation stage) is called *the stage that defines the speed of the whole process*. Since the speed of the slowest stage defines the speed of the whole process, this stage is called *the limiting stage*, since the speed of the whole process (assembly of one car) cannot be less than 60 minutes. That is, this stage limits the amount of cars produced in 60 minutes — to one car.

If we want to increase the production of cars, we should increase the speed of the defining stage, increase the number of workers on the motor-installation stage.

The increase of the number of workers on stages #1 and #3 will not have any effect on the production speed.

Relative to consequential multistage reaction, the defined HCl formation speed of stage #3 (the slowest), in the process is equal to the speed of HCl formation during the process, since stage #3 is the limiting stage.

The influence of parameter change (like the increase of temperature) is obvious (just as the change of the number of workers on the conveyor belt) mainly on stage #3 – the defining stage for increasing the speed of the whole process.

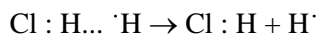
The temperature increase (just as the manpower increase on the conveyor belt) increases the speed of all the stages in the process. But the

temperature increase will cause the speed of the whole process to be close to that of the slowest stage.

We have noted that the speed increase of the fast stages does not affect the whole process. That is, its speed is always defined by the defining stage — the speed of the slowest stage.

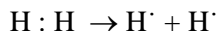
When we study the influence of temperature on the speed of final product formation in a process proceeding via some sequential stages, we define the influence of temperature (temperature coefficient or activation energy) as the very slowest of the sequential reactions.

Likewise, the speed rate of the whole multi-step reaction is defined by the rate of the slowest reaction step. It is this rate that is regarded as the *rate-determining step*, and in the above-mentioned scheme step #3 is the *rate-determining step*:



The VWB, with energy of less than 20 kJ/mol, breaks up at this step.

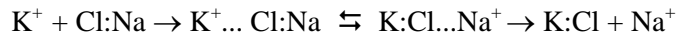
In the case of a thermal break-up of a hydrogen molecule, the reaction proceeds via a single step according to the following scheme:



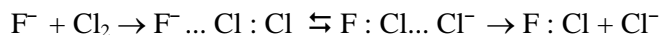
That is, in this case, the covalent bond, whose energy is equal to more than 200 kJ/mol, will break up on the rate-determining step. The ten-fold difference in the energies of bonds, which break on the rate-determining steps, explains why in the presence of radicals the bond breaking reaction proceeds at room temperature, while for breaking such bonds without active species, a temperature of more than 3,000°C is required.

According to the scheme for a chlorine atom interaction with hydrogen, a VWB is formed between the hydrogen atoms as a result of electronic isomerization. As a result of this isomerization (transition of one electron), the covalent bond (H : H) is changed into a VWB.

Positive and negative ions react just like radicals with saturated molecules do. Thus, for example, it has been experimentally confirmed that the interaction of a positively charged ion of potassium (K⁺) with sodium chloride proceeds via the mechanism as follows:



As you see, 2 electrons transit from bond Na:Cl to bond K:Cl. In the case when a saturated molecule is attacked by a negatively charged ion, the following reaction takes place:



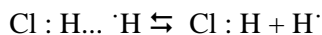
Here, during electronic isomerization, two fluorine electrons sort of substitute for two chlorine electrons.

Thus, the presence of the electronic isomerization step in reaction schemes explains why and how chemical bond breaking reactions take place at room temperature.

The bonding energy in the Van der Waals complex (VWC) is equal to about 15 kJ/mol; that is, this complex is more stable from the thermodynamic viewpoint than the products formed of it.

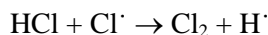
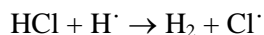
Why doesn't the reaction cease on stage #2 (stage for VWC formation)?

Indeed, the VWC is more thermodynamically stable than the equilibrium in the system



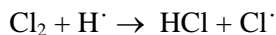
is shifted to the left with a normal temperature (300K). The speeds of the direct and reverse reactions, according to experimental data, are very large even at 300K. The reaction reaches equilibrium in less than one second.

The amount of HCl in the beginning of the reaction is much smaller than the amount of the initial products of Cl₂ and H₂. The interaction of H with HCl leads to the formation of initial H₂ and Cl₂ products along the scheme:



However, this scheme is now accomplished only after producing concentrations of HCl in the system that is commensurable with the concentration of initial substances (H₂ and Cl₂).

Before all this, the formed hydrogen atom reacted with Cl₂ of which there was more than HCl along the scheme:

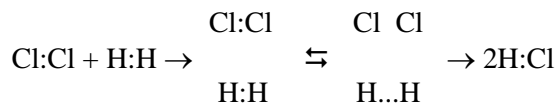


that is, together with the formation of the final product and the Cl atom.

However, some questions still remain:

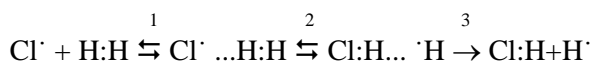
Why are radicals and ions considered to be active species?

Why is it that saturated molecules cannot react between themselves? Why shouldn't chlorine interact with hydrogen molecules according to this scheme?



That is, we have returned to the question given at the beginning of this chapter, only now we can answer it on a higher level.

According to the following chemical reaction scheme:

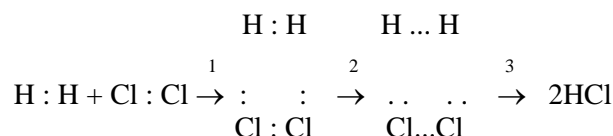


The reaction rate is equal to the rate of the third step which is directly proportional to the concentration of the isomerized associate (IA) (i.e., to the concentration of compound Cl:H...H) and which exponentially depends on the energy of the VWB (H...H) of this compound.

Concentration IA depends on the concentration of the non-isomerized associate and on the rate of the isomerization step. The greater the concentration of the non-isomerized associate (in our example: Cl...H:H) and the greater the isomerization rate — the greater is the concentration of isomerized associate in the compound, and the greater is the rate of the whole reaction. The concentration of non-isomerized associates is defined by the concentration of initial products (in our case, Cl, H₂ and bonding energy Cl...H in compound Cl...H:H).

In order to answer the above offered question: *Why is it that saturated molecules do not react among themselves along the mechanism: **association - electronic isomerization - dissociation**?* let's compare the probable interaction mechanism for saturated molecules with their interaction along the radical mechanism.

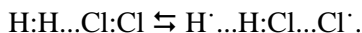
The molecular interaction mechanism for hydrogen (H₂) and chlorine (Cl₂), according to the theory, can be shown as follows:



Now let's compare the rates of each of the steps of the radical and molecular mechanisms. We will begin with step #1.

The origin of the four-member complex, as a result of the association reaction (step #1), is not only a rare phenomenon as compared with the origin of the associate between the radical and saturated molecule, but is hardly possible. The main obstacle in its formation is, first of all, the mutual repulsion of the two electronic circles between the hydrogen and chlorine atoms.

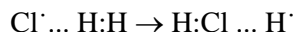
There is a much greater possibility in the formation of a linear complex of the H:H...Cl:Cl type. Out of this complex, as a result of electronic isomerization via the scheme, we get:



An associate is formed containing two VWBs and one covalent bond. The concentration of this associate is much smaller than the initial, non-isomerized one, which has two covalent bonds (H:H...Cl:Cl) and one VWB (H:H...Cl:Cl). The final associate has one covalent bond (H:Cl) and two VWBs, i.e., the electronic energy in this associate is much greater than that in the initial one — over 100 kJ/mol.

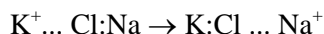
The concentration of the non-isomerized associate is defined by the bonding energy between the molecules. The greater the energy — the greater is the concentration of the associate. It is known that the VWB energy between the saturated molecules comprises less than 5 kJ/mol, while to break the bond of a radical-saturated molecule, we need energy of more than 20 kJ/mol.

When studying electronic isomerization reactions, it was found that the isomerization rate depends on the distance between the atoms and on the number of electrons transiting in the course of isomerization. Thus, for example, isomerization reaction:

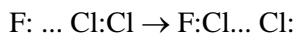


where one electron transits a distance of 1.5Å in the course of the isomerization, the time is equal to 10^{-13} sec.

The transition of two electrons in the isomerization reaction:

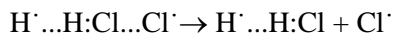


is possible at a time equal to about 10^{-12} and by changing two electrons in the following reaction:



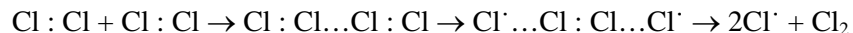
The isomerization rate becomes smaller than that of the association reaction, i.e., it takes place during more than 10^{-11} sec., that is, the isomerization rate in the case of molecular interaction is by two orders of magnitude smaller than in the case of the radical route.

And finally, what is most important, during the breaking of the VWB in an isomerized associate, according to the scheme:



a radical is formed which enters the reaction with saturated molecules, in this case, with hydrogen molecules along the chain mechanism described above. The formation of radicals in the system during the interaction of

hydrogen and chlorine is not due to their interaction, but it is due to the more rapid reaction along the following route:



As a result of this reaction, the final product is not produced. The greater radical formation rate in this route is conditioned by the fact that the bonding energy in Cl_2 is much smaller than in H_2 .

When studying the interaction mechanism for chlorine with hydrogen at high temperatures, it was found that as a result of direct interaction of chlorine molecules and hydrogen, less than 0.001% of the final product is formed. That is, after the formation of about 0.05% of the radicals from Cl_2 , the reaction proceeds along the chain radical route.

Thus the molecules can start an interaction between themselves. This interaction proceeds along the same mechanism:

association - electronic isomerization - dissociation.

However, this interaction proceeds much slower than does the interaction of active species (first of all — radicals and ions) with saturated molecules. This is conditioned by a much smaller energy gain during the formation of the initial associate: *saturated molecule—saturated molecule* as compared with the associate: *radical—saturated molecule*; a great loss of energy as a result of the isomerization reaction of the initial associate and a smaller electronic isomerization speed.

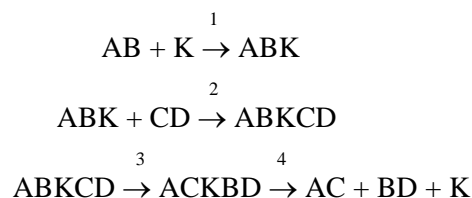
The greater activity of the radicals and ions (as compared with that of saturated molecules) is caused by: the greater bonding energy of the *radical—saturated molecule*, a greater associate isomerization rate, and a greater energy gain during the isomerization reaction (as a result of which the number of covalent and VWBs does not change). The interaction between saturated molecules proceeds along the chain mechanism where radicals or ions play the role of the active intermediate species. There is practically no reaction between saturated molecules at normal conditions ($T = 20^\circ$). The reaction speed between saturated molecules increases abruptly when there are active species in the system like ions, radicals, etc. The concentration of active species is created thermally by radiation, or by electric charges.

Another way of creating the necessary concentration of active species in the system is by introducing available active species like ions, radicals, or substances causing the dissociation of initial substances into active species or substances that dissociate into ions and radicals much easier than the initially saturated molecules. Examples of reaction acceleration at the expense of substances causing the dissociation of the initial substances into active species are reactions in water solutions where the initial substances (like salts) dissociate into ions. In all these cases the mechanism of the reactions does not change. The reaction proceeds along the chain ionic or radical mechanism.

On the other hand, according to TEI, the molecular interaction of substances is not a forbidden route; the above-cited examples are simply much slower than the catalyst reaction. As previously indicated, the main reason for the slow interaction of molecules is the low concentration of the intermediate associates and the much lower electronic isomerization speed. Another way of accelerating the reaction is by introducing substances in the presence of which molecules can react directly.

As indicated above, the low mutual interaction speed of the saturated molecules is conditioned first of all by a low concentration of the initially formed associate and a low concentration of the isomerized associate. Both of these reasons are conditioned by a small bonding energy between the molecules in the associate and by a small electronic isomerization speed. That is, in order to have the molecules react among themselves; we must introduce a substance, which forms strong bonds with both molecules.

A direct interaction of saturated molecules (AB and CD) in the presence of a catalyst (K) can be shown on a scheme as follows:



That is, at the end of the reaction we get the final products and the initial catalyst, i.e., the catalyst does not undergo any chemical changes as a result of this reaction. In order to have substance K as a catalyst, bonds AB–K and ABK–CD should be much stronger than VWB AB–CD. On the other hand, the rate-determining step is step #4. Saturated molecule bonds with catalysts break in the course of this reaction. Thus, in order to accelerate the reaction in the presence of catalyst K it is important that bonds like *catalyst—saturated molecule* are not very strong.

Previously, in chapter *Chemical Bonding* we have described three types of bonds: VWB, DAB, and covalent bonds (including homo-atomic and hetero-atomic covalent bonds). The catalyst forms bonds with saturated molecules, and these bonds are stronger than VWBs and not as strong as covalent bonds. It can be supposed that if there is acceleration of the molecular interaction, the catalyst will form DABs with the reacting molecules.

In the chapter devoted to *chemical bonding and chemical structure*, we have elucidated the fact that DABs are most possible between saturated molecules that include atoms with non-bonding electron pairs and with molecules that include atoms with incompletely filled outermost electronic shells. Such shells have surface atoms of hard

substances and coordinately unsaturated compounds called *conences*. It is these atoms that catalyze molecular interactions, which are accomplished in industrial processes when synthesizing ammonia out of hydrogen and nitrogen, in getting aldehydes out of hydrogen (H₂), carbon oxide (CO), and olefins in the process of hydroformylation, etc. The coordinately unsaturated atoms in the ferment are catalysts of biochemical transformations.

As compared with the first method for accelerating chemical reactions, where the reaction accelerates at the expense of introducing active species (ions, radicals) to the system, the second method (acceleration of the direct molecular interaction) has priority from the viewpoint of selectivity; i.e., as a result of these processes, we generally get individual products.

Conence homogenic catalysis is, from the viewpoint of both methods, a mixed type of catalysis. Here the acceleration is gained at the expense of the conence concentration increase (i.e., active species). The reaction proceeds along the chain mechanism, but during this reaction the initial molecules do not disintegrate, and they do not form VWBs in the associates (which is characteristic of the first method for accelerating reactions). They form DABs.

The acceleration of the catalyst reaction at the expense of introducing active species into the system (radicals, ions, conences, etc.) is described in textbooks as examples of catalyst reactions. The reactions most commonly offered are as follows:

- 1) $\text{RCO}_2\text{R} + \text{H}_2\text{O} \xrightarrow{\text{H}^+} \text{RCO}_2\text{H} + \text{R OH}$
- 2) $\text{Cl}_2 + \text{H}_2 \xrightarrow{\text{Cl}^\cdot} 2\text{H Cl}$
- 3) $\text{SO}_2 + \text{O}_2 \xrightarrow{\text{NO}^\cdot} \text{SO}_3$
- 4) $(\text{CH}_2 = \text{CH}_2)_n \xrightarrow{\text{R}^\cdot} (-\text{CH}_2 - \text{CH}_2-)_n$
- 5) $\text{O}_3 \xrightarrow{\text{Cl}^\cdot} \text{O}_2 + \text{O}^\cdot$
- 6) $\text{RCO}_2\text{R} \xrightarrow{\text{OH}^-} \text{RCO}_2 + \text{ROH}$ RAZVITY???
- 7) $\text{CH}_2 = \text{CH}_2 + \text{CO} + \text{H}_2 \xrightarrow{\text{H}\phi\text{Co}(\text{CO})_3} \text{CH}_2 - \text{CH}_2\text{COH}$
 Were $\text{H}\phi\text{Co}(\text{CO})_3$ conens