

Catalysis Mechanism and Application of Carbon Gasification Reaction-A Comparison of Two Heterogeneous Catalysis Mechanisms

Jia Min Jin

Correspondence: Jia Min Jin, Shanghai Research Institute of Materials, Shanghai, China.

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Abstract

This article is a brief summary article of research. The results of the three times experiments are reviewed. two heterogeneous catalysis mechanisms are introduced, namely: Chemical Reaction Mode Cyclic Catalysis Mechanism-CRM and Electron Cyclic Donate-Accept Catalysis Mechanism-ECDAM or Electron Orbital Deformation-Recovery Cyclic Catalysis Mechanism -EODRM. Some difficulties encountered by CRM are listed. The author clearly points out that the CRM is not credible. This false theory has misled us for more than 100 years.

About ECDAM, the article also gives a brief description. The main point of ECDAM is that the catalysis phenomenon are physical rather than chemical phenomenon. The catalysts do not participate in chemical reactions. It's just contact, electron cyclic donate-accept or electron orbital deformation-recovery cycle. The theory contains three viewpoints:

1. There is a boundary between the catalyst and the poison.
2. The active of the catalyst or the degree of toxicity of the poison is closely related to the electronegative value of the catalyst or poison.
3. The active of catalyst is closely related to the chemical state of the catalyst

The selectivity of catalyst is also related to electronegative or energy level

According to ECDAM, the author considers that there are several problems worth studying in production and scientific research. such as: alumina is a poison in the Fe ammonia synthesis catalyst. The Cordierite ($2\text{MgO}\cdot 2\text{Al}_2\text{O}_3\cdot 5\text{SiO}_2$) ceramic honeycomb support is also a poison in automotive exhaust purification catalyst. The Cordierite ceramic honeycomb is retardant in wall flow filter for diesel vehicles. Activated carbon is a poison in the Ruthenium catalyst for ammonia synthesis. Alumina and activated carbon all are a poison to noble metal catalysts, and so on.

Keywords: heterogeneous catalysis, catalysis Mechanism, CRM, EODRM

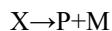
1. Introduction

Because the chemical and physical properties of solid catalysts affect the catalytic active of catalysts at the same time. The result is that the catalysis phenomena are very complicated and were regarded as mysterious and not amenable to rational interpretation. For a long time, the selection study of catalysts is always in the skill stage of the laboratory. The author thinks that the basic reason is lack of correct theoretical guidance.

At present, it can be seen from literature that there are two heterogeneous catalysis mechanisms. One is the CRM widely adopted, and it is raised to the height of the "principle" by the scholars, this CRM is also instilled in high school textbooks. It can be said that CRM is deeply rooted in catalytic academia. Another catalysis mechanism is the EODRM or ECDAM proposed by author. The EODRM is completely different from CRM, which consider that catalytic phenomena are physical rather than chemical phenomena, and that catalysts do not participate in chemical reactions. It's just contact, electron cyclic donate-accept, electron orbital cyclic deformation-recovery. The electrocatalytic, photocatalytic, microwave catalysis, laser catalysis are all physical phenomena, only different energy levels.

2. About the CRM

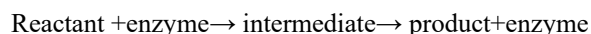
In the Catalyst Handbook (Catalyst Handbook 1982), the expression for the CRM is as follows;



Where R is reactant, P is product, M is catalyst, X is intermediate. The catalyst M takes part in a chemical reaction and regenerates the same catalyst as original after reaction. From reactant to product, $\text{R}\rightarrow\text{P}$, it does not generate, Instead, the

intermediate is generated by catalyst with reactant, and then the product is generated by the intermediate. The reason is that such a reaction way requires lower energy.

In the high school textbooks in China on the catalytic mechanism of biocatalyst enzyme, it is also based on CRM.



Enzymes take part in chemical reactions, produce intermediate compounds, and finally release the same enzymes as original. The Enzymes are constantly decomposed and generated.

About the CRM, the most obvious way to put it should be “Sabatier principle” (in 1902) and “Boudart principle”(in 1992).

In 2009, Deutschmann, O, et al had wrote a book, [*Heterogeneous catalysis and solid catalysts*] (Deutschmann.,O,2009). In this book, two “principles” advocated by the catalysis academia are repeatedly mentioned. One is the “Sabatier principle”, and the other is the” Boudart principle”, which author call as the “S-B principle”.

The Sabatier principle proposes the existence of an unstable intermediate compound formed between the catalyst surface and at least one of the reactants. The intermediate must be stable enough to be formed in sufficient quantities and labile enough to decompose to yield the final product or products.

The Boudart principle is that the most fundamental principle in catalysis is that of the catalytic cycle, which may be based on a redefinition of a catalyst 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”. “The activity of the catalyst is defined by the number of cycles per unit time or turnovers or turnover frequency(TOF;unit:S⁻¹),The life of the catalyst is defined by the number of cycles before it dies.”

1981 year, Mckee. D.W. (Mckee., D,W,1981) has studied the catalysis of the alkali metal oxides and their salts, alkaline earth metal oxides and their salts, transition group metals and their oxides, noble metals and other catalysts on C+O₂, C+CO₂, C+ H₂O and C+H₂ reactions with advanced instrument(CAEM-Controlled Atmosphere Electron Microscope) . He has done a lot of experimental studies, wrote more than 100 pages of lengthy articles. Finally, he considers that on balance “specific oxidation-reduction cycles” have been conspicuously successful in interpreting the effects of these compounds in the various types of carbon gasification reactions. But the writer feels that the "specific" word is nowhere to be seen. Although the author shows that the complex catalysis remains unclear and needs to be better studied afterward.

A senior American scholar has studied the catalysis of the noble metal Pt on the C+CO₂=2CO and C+H₂O=H₂+CO reaction, they also adopted CRM to account for the catalysis mechanism. They believed that Pt is continuously reduced and oxidized by H₂,CO and H₂O,CO₂ in the catalyze process. This is also Oxygen Transfer Mechanism-OTM. Even though the paper says " proposed mechanism".

In 2016, Chen ping, a young scientist from Chinese Academy of Sciences, and other researchers published a high-level paper in the international top academic Journal (Nature Chemistry). They have proposed the design theory of "dual-activation center" ammonia synthesis catalyst. Two activation centers such as TM (transition group metal) and LiH are continuously decomposed and regenerated in the catalyzing process of ammonia synthesis. In fact, they also follow the “S-B principle” or the traditional ancient CRM principle. It can be seen that the younger generation also inherited the idea of CRM.

Based on the above, CRM has been widely adopted in the catalytic community for more than 100 years and it is deeply rooted in catalytic field.

3. Three Times Experimental Studies

The most main ideal of the CRM is that the catalyst must take part in the chemical reaction, and it has to be a cyclic reaction. In order to explore whether CRM can be trusted, the author has conducted three times experimental studies.

3.1 BaCO₃ Catalysis in Carburizing Box (the 1960s)

The BaCO₃ is commonly used as catalyst in solid carburizing agent of surface of steel parts. At the beginning, the mechanism of carbonate decomposition to release CO₂ which reacts with carbon to produce carbon monoxide and thus increasing the rate of carburization was used to account for the reason for accelerating carburizing rate. Later, it was found that CaCO₃, which can release a large amount of carbon dioxide, but it has no catalytic activity, while barium carbonate, which does not decompose at the carburizing temperature, has carburizing activity. Therefore, the mechanism of carbon dioxide release was abandoned and the Oxygen Transfer Mechanism, namely OTM, was adopted.

Namely:





The OTM is also CRM. This is a typical CRM. BaO is an intermediate compound, and the BaCO₃ is constantly decomposed and generated in the carburizing process.

The mechanism was first proposed by R.A. Ragaz & L.O. Kowalke at 1931 (R.A. Ragaz & L.O. Kowalke 1931).

The equilibrium pressure of reaction (1) was measured by author (Jin, J.M., 1963). At the carburizing temperature 946°C, the equilibrium pressure is 0.226 atm., which is much lower than the 1 atm. of carbon monoxide pressure in the carburizing box. It indicates that the BaCO₃ cannot be decomposed by carbon, that is, the BaCO₃ does not take part in chemical reaction and the cyclic reaction of "decomposition-generation" cannot at all occur.

3.2 The catalysis of Fe, Co, Ni, Cu, Ag and SiO₂, S on the Reducton Reaction Rate (Jin, J.M. 1982) (the 1970s)

The authors have done a few times verification experiments refer to the CRM and EODRM. These have included the active of catalyst and poisonousness of poison, such as Fe, Co, Ni, Cu, Ag, SiO₂, S. The experimental results show that Fe etc. appear catalytic active on carbon gasification reaction. Iron appears the greatest catalytic active. Their relative activities are Fe > Cu, Fe > Ni, Cu > Ni, Ag > Cu and Co > Ni. The SiO₂ and S are poison. The poisonousness of SiO₂ appears to be more poisonousness than S. The experimental results agree well with EODRM. At the same time, it also strongly shows that the CRM simply cannot account for the results of the experiment. Because they can't take part in a chemical reaction in the reaction box. Because there's a lot of solid carbon in the reaction box, while the carbon is a powerful reducing agent at 1000°C. Thermochemical data can justify perfectly that Fe, Co, Ni, Cu and Ag can only take the form of metallic state. The oxidation-reduction cyclic reaction cannot at all occur in the reduction reaction box.

3.3 The Catalysis of Alloy Elements and the Mechanism of Carbon Dissolving Into Fe in Iron-Graphite Powder Metallurgy Compact During Sintering (T. Hong 1986). (the 1980s)

Sintered steel parts are made of iron powder, graphite powder, alloy powder after mixing and pressing sintering. How graphite is dissolved into iron and the effect of alloying elements are naturally the subject of research

The experiment result shows that:

1. Temperature, compacts density, atmosphere, alloy elements, vacuum degree all affect the rate of carbon dissolute into Fe
2. The dissoluting rate of carbon into iron depends entirely on the carbon gasification reaction. The solid-solid reaction between Fe-C is almost zero. Carbon gasification reaction is the control step of carbon dissoluting rate,
3. Alloying elements W and K₂CO₃ appear catalytic active to accelerate carbon dissoluting rate, and the SiO₂ and S is poison. In particular great significance, the experimental result shows that the hydrogen appears catalytic active on the carbon gasification. Because the hydrogen is less electronegative than carbon. But it is a poisoner on the Fe catalyst. Because the its electronegativity is higher than Fe.

The results of the above three times experiments for long 30 years are in complete agreement with the ECDAM judgment. But the CRM was unable to explain the results of three times experiment study.

The CRM can neither illustrates that a catalyst, such as Fe, appears catalytic active in a wide temperature range, such as 600⁰-1000⁰c, on the FeO reduction, nor that many elements or compounds all appear catalytic active at the same temperature, such as 1000⁰c. Because the cyclic generation-decomposition or oxidation-reduction chemical reaction equilibrium requires a strict reaction condition and the reaction equilibrium condition is all different.

If according to CRM, the catalyst must participate in chemical reactions, the result will appear many chemical reactions and intermediate compounds, and each said his own, it is dazzling, the research of catalysis mechanism will be leaded to bottomless abyss.

A familiar example is that housewives know that salt (NaCl) appears catalyze active for coal combustion, and CRM cannot explains the reason at all, because it is impossible for NaCl to decompose- generate repeatedly cyclic reactions in coal furnaces. Owing to NaCl is a very stable compound.

The author can cite many examples to illustrate that CRM is uncredible Because it's too cumbersome, Thus, I don't want to say any more.

Therefore, the author considers that the catalytic academia to improve CRM to "principle" is incredible. Using "TOF" to define catalyst activity and service life is obviously out of thin air. It is to fool people.

4. The ECDAM or EODRM's Viewpoint

Back in the 1960s, based on the experimental results and theoretical analysis, the author points out that the CRM is not credible. Unfortunately, it didn't get attention.

The ECDAM were proposed in the 1970s. The ECDAM or EODRM contains three viewpoint.

4.1 There Are a Boundary Between the Catalyst and Poison

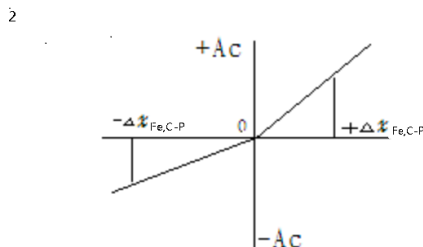


Fig. 1. Relationship between electronegativity differences and catalytic activities.

In figure 1, +Ac shows catalytic active, -Ac shows poisonousness, $+\Delta\chi_{Fe,C-P}$ shows the electronegativity difference, Footnotes Fe,C shows iron or carbon, P shows the catalyst, promoter, poison or support . For example; $+\Delta\chi_{Fe-P}$ Shows the difference between Fe-promoter, such as Al_2O_3, LiH etc.,. $+\Delta\chi_{C-P}$ shows the difference between C and catalyst, such as $BaCO_3, Fe, Ni$ etc. catalyst.

From Fig.1, the coordinate "0" point of the $\Delta\chi=0$ is a demarcation between the catalyst and the poison or inhibitor. For the carbon gasification reaction, this demarcation is the electronegativity value of the carbon, $\chi_c=2.55$. Any elements or compounds with electronegativities value greater than 2.55 should be a poison, such as P, S, Cl. Any elements or compounds less than 2.55 should be a catalyst, such as Fe, Co, Ni, Cu, Pt etc.. For iron - based ammonia synthesis catalyst, the demarcation is the electronegativity value of iron, $\chi_{Fe}=1.83$. Any elements or compounds with electronegativities greater than 1.83 should be a poison, such as C, N, P, S, Cl, Ni, Cu, etc .and any elements or compounds less than 1.83 should be a catalyst, such as alkali metal, alkaline earth metal. For the noble metal catalysts, the demarcation is the electronegativity value of noble metal, $\chi_{Pt}=2.28$.

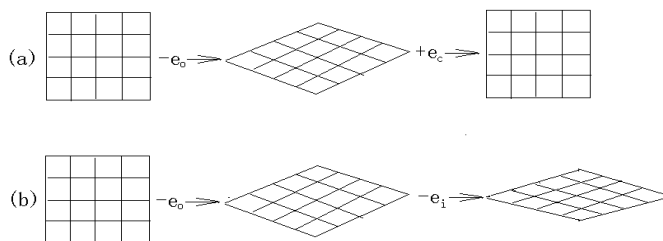
On the element periodic table, for carbon gasification reaction, these elements at the right of carbon ,such as Alkali metal, Alkaline earth metal ,Transition group metal and Noble metal should be catalyst and those elements at the left of carbon ,such as P, S, Cl, F should be poison,

For iron - based ammonia synthesis catalyst, these elements at the right of Fe, such as Alkali metal, Alkaline earth metal should be catalyst and those elements at the left of Fe, such as, Cu, Ni, C, P, S, Cl, F should be poison,

$SiO_2, Al_2O_3, cordierite(2MgO \cdot 2Al_2O_3 \cdot 5SiO_2)$ are all acidic materials, their electronegativity is greater than carbon, iron and Pt, Pd, Ru. Therefore, they are poison to iron Ni, noble metal catalyst and carbon.

A very interesting is that the Cu is a catalyst for carbon gasification reaction, but it becomes a poison in iron based ammonia synthesis catalyst, which had been proved to be true in production practice. Because the Cu is between iron and carbon on the element periodic table, its electronegative is less than carbon and more than iron. This result strongest proofs that ECDAM or EODRM is credible.

4.2 The Active Size of Catalyst Is Closely Related to the Electronegativity Value of Catalyst



Scheme 1. The imaginative catalyzing and poisoning model.
 (a). catalyzing. (b). poisoning.
 $-e_o, -e_i$ ---electron is seized by oxygen or inhibitor.
 $+e_c$ ---electron is donated by catalyst or promotor.

The scheme 1 (a) is a imagine figure of catalyzing .The scheme (b) is a imagine figure of poisoning.

From scheme 1(a), because the oxygen is more electronegative than carbon, $\chi_o > \chi_c$, the suspend electrons on the surface of the carbon must be snatched by the adsorbed oxygen, there was a contest for electrons between oxygen and carbon. the result is a deformation of electron orbitals in the carbon crystal or matrix, the crystal structure changes from square to diamond. If you are using chemical or physical methods to donate electrons to the carbon matrix, the contest electron situation is eased. The carbon bond breaking energy is reduced, CO is easy to generate. Therefore, the function of a catalyst, promotor or accelerator is to donate electrons to carbon or iron matrix, to revert a deformed electron orbital or crystal structure from a diamond to a square. Thus, catalysis is only an electron donate-accept cycle or electron orbital deformation-recovery cycle, without chemical reactions, let alone intermediate compounds. Therefore, any element or compound that can give electrons to carbon(or Fe) has catalyze active; otherwise, it is poison. the catalyze phenomenon is a physical phenomenon, not a chemical one, the catalyst does not participate in the chemical reaction, it is just contact, electron donate-accept, or electron orbital deformation- recovery.

From scheme 1 (b), these elements or compounds that are higher electronegative than carbon will further take away the electrons of carbon, the electron orbits in carbon matrix is further deformation, the result is that the carbon bond breaking energy is elevated, the CO desorption is more difficult and therefore the reaction rate is slow down.

From Fig.1, The $+\Delta\chi$ value higher is, the higher the catalytic active is. The $-\Delta\chi$ value higher is, the higher the poisonousness is. Therefore, whether carbon gasification reaction, iron based ammonia synthesis reaction or noble metal catalytic reaction, alkali metals and their salts are always the most active catalysts or promoters, while P, S and C are always poisons. Long-term production practice has proved this rule beyond doubt.

4.3 The Catalytic Actives of the Catalysts Are Closely Related to the Chemical States of the Catalyst

The catalytic activity of a catalyst is closely related to its chemical state. When the chemical states have changed, the electronegativity is different, and the catalytic activity is different.

The most compelling example is iron. The Iron has three chemical states, namely Fe, FeO and Fe₂O₃. For carbon gasification reaction, the experimental study proves that the Fe has catalytic active, but the FeO has no catalytic active, while the Fe₂O₃ becomes a poison at high temperature.

For salt catalysts, the catalytic activity varies with the cation and anion in salt.

When the anion is constant in the salt, the molecular electronegativity of salt increases with the increase of the electronegativity of cations in salt. Similarly, when the cation is constant, the molecular electronegativity of salts increases as the electronegativity of anions increases its catalytic activity changes with the increase of the electronegativity of the anion.

For carbon gasification reaction, for alkali metal carbonates, their relative catalytic active is Cs₂CO₃>K₂CO₃>Na₂CO₃. For alkaline earth metal carbonates, the their relation catalytic active is BaCO₃>SrCO₃>CaCO₃>MgCO₃. For sodium ealts catalyst, the catalytic activity of Na₂CO₃ is greater than NaNO₃.Na₂SO₄, NaCl, Na₃PO₄. All these have been confirmed by experiments and production practice.

The research results of Chen Ping's team found that using LiH as a promotor for iron-based ammonia synthesis catalyst can greatly improve the catalytic activity of iron catalyst. According to EODRM, this is inevitable Because LiH is less electronegative than carbonate.

Different chemical states, there are different electronegativity, so there are different catalytic activity.

The core idea of EODRM is that the catalyst does not participate in chemical reactions, it's just "contact", electron donate-accept cycle or electron orbital deformation- recovery cycle, there is no intermediate. There are no generation-decomposition and oxidation-reduction cycles. Adsorption of the reactants results in the electron movement and distortion of electron orbitals. Desorption of the product results in electron orbital recovery. The electronegativity difference is what drives electrons to move. A continuous cycle of adsorption and desorption completes the production proces. The activity and selectivity of the catalyst depend on the energy level of the catalyst.

In modern times, electrocatalysis, photocatalysis, microwave catalysis and laser catalysis all are physical phenomena, which coincide with EODRM.

5. Using the EODRM to Judge Some Probrems Existing in Industrial Production

According to EODRM, it is found that there are many problems worthy of study in the field of research and production.

5.1 Alumina Poisoning in the Iron-Based Ammonia Synthesis Catalyst

Iron-based ammonia synthesis catalyst is considered by the academic community to be a classic catalyst. It is also the most studied catalyst. It has been used for more than 100 years. The chemical composition of the catalysts produced in

different countries is almost the same. It contains 2,5-3,0% Al_2O_3 which is called structural promoter. However, the experimental study of Aika.K (Aika, K., 1985) and H. G. Du (H, G, Du., 2002) have proved that alumina appears poison to carbon gasification reaction. The electronegativity of alumina is greater than that of carbon and iron, and it is poison to both carbon and iron catalysts. The production practice shows that C, P and S are poison to iron - based ammonia synthesis catalyst. The greater the poison, the lower the permissible content. The permissible content of sulfur in ferro-based catalysts is $<0.003\%$. The content of alumina is 1000 times that of sulfur, which is seriously poisons the catalytic activity of iron. The author considers that there are three high situations, such as high temperature, high pressure and high energy consumption, in synthetic ammonia production, which may be caused by improper use of alumina.

The physical and chemical properties of the catalyst simultaneously affect the catalytic activity of the catalyst, lead to the catalytic phenomenon is very complex. The advantages of alumina are high melting point, high strength, and especially high specific surface area, The favourable side of the physical properties covered up the unfavorable side of the chemical properties. It disturbs people's sight. In terms of chemistry property, Alumina should be a poison. Both adsorption and desorption increase energy consumption. Therefore, author considers that it is not appropriate to use alumina as a promoter for the synthesis of iron - based ammonia.

5.2 Carbon Support Poisoning in the Ruthenium Ammonia Synthesis Catalyst

The Ruthenium-based ammonia synthesis catalyst has been developed by British-American two companies cooperative study after for more than 10 years. The support material of the catalyst Ruthenium is the activated carbon after with high temperature graphitization.

According to EODRM, $\Delta\chi_{\text{Ru-C}}=2.2-2.55=-0.35$, The active carbon is poison to Ruthenium catalyst. The use of this support, must use more noble metals, the result is that the catalyst is very expensive.

5.3 Catalyst Support for Automobile Exhaust Purification

The most frequent type of catalytic converter in automobiles is the three-way catalyst (TWC) for stoichiometrically operated gasoline engines with an annual production of over 60×10^6 units. The TWC systems contain Pt, Pd and Rh. The catalytic components are supported by a cordierite honeycomb monolith coated with high surface area $\gamma\text{-Al}_2\text{O}_3$. According to EODRM, the cordierite ($2\text{MgO} \cdot 2\text{Al}_2\text{O}_3 \cdot 5\text{SiO}_2$) is acid material, it is poison to noble metal catalyst. In order to meet emission standards, more catalysts must be used.

In order to meet higher emission standards, The Germans have developed the Fe-Cr-Al metal coil honeycomb support. Compared with cordierite ceramic honeycomb carriers, At the same emission standard, noble metals loaded with metal coil honeycomb support are approximately 1/2 of the ceramic support. This clear indication that inappropriate support materials can waste a lot of noble metals catalyst. It is estimated that about 100 tons of noble metals are wasted each year.

The second support, i.e. $\gamma\text{-Al}_2\text{O}_3$ plating layer is also poison to noble metal catalyst. Fortunately, rare earth oxides are gradually replacing alumina.

The automobile catalyst is developing in a reasonable direction, which is in perfect agreement with EODRM's expectation.

The author believes that the powder metallurgy porous metal honeycomb support should be the most rational support. It's much better than the metal coil honeycomb. Large specific surface area and the plating layer is not easy to peel off, we did it and got good experimental results. Due to the lack of funds and other reasons, it is give up at halfway.

5.4 Diesel Exhaust Wall Flow Soot Filter

Diesel exhaust soot filter, at present, it still use the cordierite ceramic honeycomb wall flow filter. The cordierite is a poison to carbon gasification. The filter is too slow to regenerate, it cannot be used continuously. As a result, the americans used two filters, one job and the other to regenerate, with the result that prices are too expensive to promote.

The author suggests that the powder metallurgy honeycomb filter should be used in diesel soot filter, because the catalytic active of iron on carbon gasification reaction is much greater than that of noble metal.

5.5 Support Materials

Alumina and active carbon are mass produced as noble metal catalyst support in many factories around the world. High strength, high melting point, high specific surface area, high chemical stability, abundant resources and so on are the advantages of alumina, active carbon. However, according to EODRM, both are poison to Fe, Ni, Pt, Pd, Ru and so on catalysts, Unreasonable support will consume more noble metals. As far as automotive TWC catalysts are concerned, because of the use of unreasonable support materials. About 100 tons of noble metals are wasted every year.

From the internet, the support of fuel cell Platinum catalyst is made by carbon black or graphite at early stage. Later, the carbon nanotube is developed. The catalyst support of denitrification in power plant has adopted the cordierite ceramic honeycomb or stainless steel mesh. The author thinks that all are unreasonable.

6. Conclusion

1. Experimental and theoretical analysis show that the decomposition - generation, oxidation - reduction cyclic reaction does not exist, so it is unthinkable to improve CRM to "principle", using "TOF" to represent catalyst active and service life is obviously out of nothing.
2. Catalysis is a physical phenomenon rather than a chemical one. The catalyst does not participate in the chemical reaction. It is only contact, electron cyclic donate-accept and electron orbital cyclic deformation- recovery. EODRM or ECDAM's three viewpoints are confirmed by experiment and production practice. The active and selectivity of the catalyst are closely related to the electronegativity or energy level of the catalyst.
3. According to EODRM, author considers that there are several unreasonable measures in industrial production. It is lead to serious economic losses.

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