

Review Article

Opinion — On a New Mechanistic Model Toward the Catalytic Reactions: From Hydrogen Combustion to Fischer-Tropsch Reaction

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Abstract

Mechanism research in catalytic chemistry is both fascinating and confusing, particularly when it comes to solid-state catalysts, the nature of catalytic behaviours has been unidentified so far. For a mechanistic model to be acceptable, it should have an ability to explain all unique aspects of a given catalytic reaction and provide an illuminating explanation to a widely range of catalytic reaction. In our recent reports, a new mechanistic model was suggested for catalytic CO₂ reduction reaction on Cu metal and hydrogen evolution reaction on various transition metals, which provides a reasonable interpretation to both catalytic reactions (from the diversity of product distribution and catalytic behaviour of various metals). Here, it is expected to extend this new mechanistic model to a wider range of catalytic reactions over various catalysts. Such as hydrogen combustion with Cu metal adding, oxidation of SO₂ by O₂ to give SO₃ with NO adding, conversion of CO and NO into CO₂ and N₂ with Ru metal adding, and hydrogenation of propylene with Pt metal adding. Importantly, this model seems also to pertain to the mechanism of the Fischer-Tropsch (T-F) reaction, i.e. the conversion of CO and H₂ to hydrocarbons, principally a mixture of linear alkanes (including methane) and alkenes, by passage over various heterogeneous transition-metal catalysts (Fe, Co, Ni et.al.).

Keywords

Mechanistic Model, Fischer-Tropsch Reaction, Catalytic Reaction Mechanism

1. Introduction

Catalysts play an important role in modern chemical industry. More than 90% of the commercially produced chemical products proceeds with the aid of a solid metal catalyst at some stage. [1] Catalysts can increase the rate of a chemical reaction and are not consumed by the reaction, [2] how catalysts achieve this function in chemical reactions? This problem has been studied for more than a century but still remains in debate. [3] Until today, catalytic chemistry is usually regarded to be mysterious

and charismatic for researchers.

In our recent publications [4, 5] it is assumed that the metal atom with unpaired electrons on valent orbitals is a radical centre, which can convert spin-paired reactant into reactive radical state by radical abstraction or addition starting the chemical reaction or can bond two reactants by radical dimerization supporting the formation of new chemical bond as shown in Figure 1. This assumption can provide an illumi-

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nating explanation to the catalytic property of copper metal for conversion CO_2 into widely range of products and various transition metals for hydrogen evolution. In the present work we wish to continually show the line of thought and several examples which have led us to this point of view, hoping that this approach may be useful to some researchers in area of chemistry.

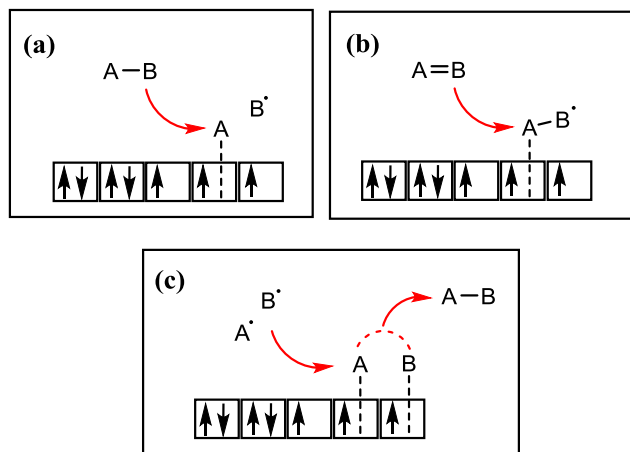


Figure 1. The process of catalytic reaction on metal atom with unpaired electrons: (a) a spin-paired reactant is converted into free radical and bonded species via radical abstraction; (b) a spin-paired reactant with double bond is converted into absorbed radical via radical addition; (c) two active radicals are bonded on the surface of metal atom via radical dimerization to support the formation of new chemical bond.

2. Discussion

2.1. H_2 Oxidation into H_2O

The discussion initiates with the consideration of H_2 oxidation to produce H_2O . The combustion of mixtures of H_2 with O_2 usually does not take place at room temperature, and its starting needs to be heated to at least 500°C by a spark or flame. [6] The reactants (H_2 and O_2) are spin-paired; thus, it here is assumed that the initial elementary reaction step in this reaction requires activation energy to homolysis of H-H σ bond or O-O π bond into H^\cdot or $\cdot\text{O}-\text{O}^\cdot$ reactive radicals, respectively. Namely H_2 molecule or O_2 molecules by absorbing energy is broken into H^\cdot or $\cdot\text{O}-\text{O}^\cdot$ reactive radicals, which then react with O_2 or H_2 molecules by addition forming HOO^\cdot radicals (Figure 2a). Following by stepwise radical reactions, H^\cdot or $\cdot\text{O}-\text{O}^\cdot$ radicals react with O_2 or H_2 molecules producing H_2O with energy releasing, the remaining H_2 or O_2 molecules are gradually converted into reactive H^\cdot or $\cdot\text{O}-\text{O}^\cdot$ radicals by absorbing energy released from previous reactions, the mixtures of H_2 with O_2 are gradually starting combustion under this condition

It is known that metal catalysts (Cu or Pt) can affect the combustion of mixtures of H_2 with O_2 , making it occurring at

room temperature. [7] Cu or Pt atoms have unpaired electrons, It seems evident that directly converting H_2 molecules into reactive H^\cdot radicals by homolysis of H-H σ bond or O-O π bond at high temperature is avoided if it is assumed that the atoms of metal surface with unpaired electrons can react with H_2 or O_2 by abstraction forming a free H^\cdot or bonded $\cdot\text{O}-\text{O}^\cdot$ radical (Figure 2b). In this condition, the global reaction does not require energy to convert spin-paired H_2 or O_2 molecule into reactive H^\cdot or $\cdot\text{O}-\text{O}^\cdot$ radicals, which can be produced by radical abstraction by surface atoms of metal catalysts. With further radical reactions occurring, energy is gradually released from radical reactions, the combustion of mixtures of H_2 with O_2 will be started.

2.2. SO_2 Oxidation into SO_3

In the following, it is discussed that the oxidation of SO_2 to SO_3 ($2\text{SO}_2 + \text{O}_2 \rightarrow 2\text{SO}_3$) catalyzed by adding nitric oxide (NO), this reaction can take place at high temperature (without NO adding). [2] By analyzing its elementary reaction steps firstly (Figure 2c), the initial step may be homolysis of π bond of $\text{O}=\text{O}$ molecules forming $\text{O}^\cdot-\text{O}^\cdot$ radicals, energy input (activation energy) is required in this step. As O-centred radicals ($\text{O}^\cdot-\text{O}^\cdot$ radicals) are extremely reactive, both radical centers of $\text{O}^\cdot-\text{O}^\cdot$ radicals can react with spin-paired SO_2 molecules forming $\cdot\text{O}-\text{S}(\text{O})-\text{OO}-\text{S}(\text{O})-\text{O}^\cdot$ intermediate by addition and this intermediate could then decompose into two SO_3 molecules by adjusting electron structure.

Thus, it is our opinion that the step of directly homolyzing π bond of $\text{O}=\text{O}$ molecules for O-centred radicals might be replaced by adding NO. As there is single unpaired electron on the N atom of NO, the initial step of the oxidation of SO_2 by NO may be that ON^\cdot radicals react with spin-paired O_2 molecules forming a O-centred radical ($\text{ONO}-\text{O}^\cdot$) by addition (Figure 2d), $\text{ONO}-\text{O}^\cdot$ radicals then react with SO_2 by addition, following by stepwise radical reaction two SO_2 molecules are finally converted into two SO_3 molecules and NO is regenerated in final step. Thus, the step of homolyzing π bond of $\text{O}=\text{O}$ molecules forming $\text{O}^\cdot-\text{O}^\cdot$ radicals by extra energy input is avoided from global reaction by adding NO into reaction system, and thereby the rate of oxidation of SO_2 is speeded up.

2.3. CO Reacts with NO to Give CO_2 and N_2

In industrial, the exhaust from the burning of fossil fuels usually contains some of harmful byproducts (such as CO and NO). In theory, CO can react with NO to give CO_2 and N_2 ($2\text{CO} + 2\text{NO} \rightarrow 2\text{CO}_2 + \text{N}_2$). Although the reactant of NO has a single unpaired electron on N atom, this reaction strongly relies on the metal catalysts (such as Pt and Rh). [8] By analyzing this redox reaction, the C atom of CO is oxidized by O of NO into high valence state and N atom of NO is reduced by C of CO into zero valence state. Thus, it seems that the N-centred radical (NO) will not directly react with C or O atoms of CO molecule by abstraction, NO radicals are

more trend to dimerize to give a spin-paired ON-NO. Here, we assume that nitric oxide reactants in form of ON-NO involve in reaction with CO on metal catalysts to give CO₂ and N₂. In metal catalytic process, the initial reaction might be the conversion of CO into reactive radicals. As illustrated in Figure 2e, CO molecules might undergo radical addition by

surface atoms of metal catalysts forming C-centred radicals, which might then attack ON-NO by a further addition reaction to give N-centred radicals, then followed by stepwise elementary reactions, the reactants (CO and ON-NO) are converted into CO₂ and N₂ in final step, surface atoms of metal catalysts are regenerated.

2.4. Hydrogenation of C=C Double Bonds

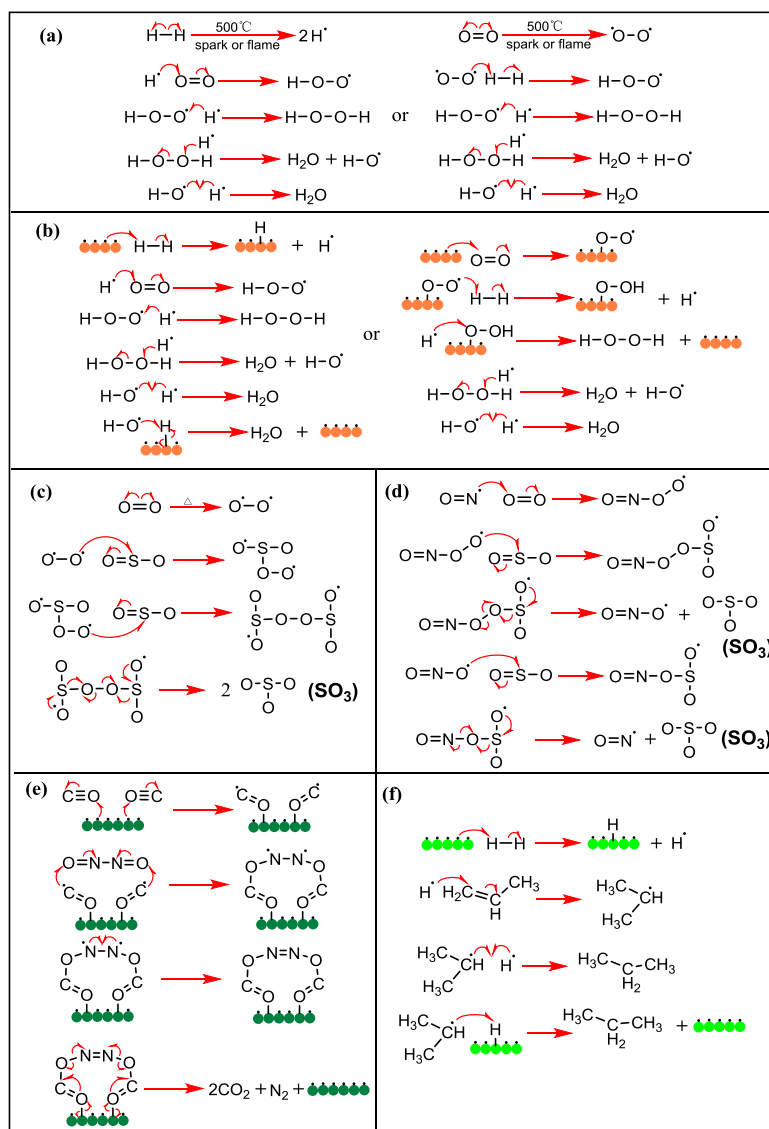


Figure 2. Elementary reaction mechanism steps of various chemical reactions: (a) the combustion of mixture of H₂ and O₂ initiated by spark or flame, (b) the combustion of mixture of H₂ and O₂ is motivated on metal surface, (c) oxidation of SO₂ by O₂ to give SO₃ without NO adding, (d) oxidation of SO₂ by O₂ to give SO₃ with NO adding as catalyst, (e) the mixture of CO and NO is converted into CO₂ and N₂ on metal surface, (f) propylene is hydrogenated by H₂ on metal surface. Origin solid balls represent Cu metal atoms, green solid balls represent Ru metal atoms, and light green solid balls represent Pt metal atoms.

The C=C double bonds can be reduced by using H₂ to produce fully saturated alkanes and this type of reaction also strongly relies on the metal catalysts adding (usually Ni or Pt). [9] Similar to catalytic H₂ reacts with O₂ to give H₂O, the initial step of homolysis of H-H bond require energy input, we

consider the atoms of metal surface with unpaired electrons can react with H₂ by abstraction forming a free H[•] radical and a bonded H[•] radical (Figure 2f). Taking propylene hydrogenation as example, the free H radicals produced then may attack the C atoms of C=C double bonds to give a C-centred

radicals, which could react with another free H^\bullet radicals by dimerization or react with H^\bullet radical bonded on catalyst sur-

face by abstraction to achieve totally addition reaction and releasing the metal surface site.

2.5. Fischer-Tropsch Reaction

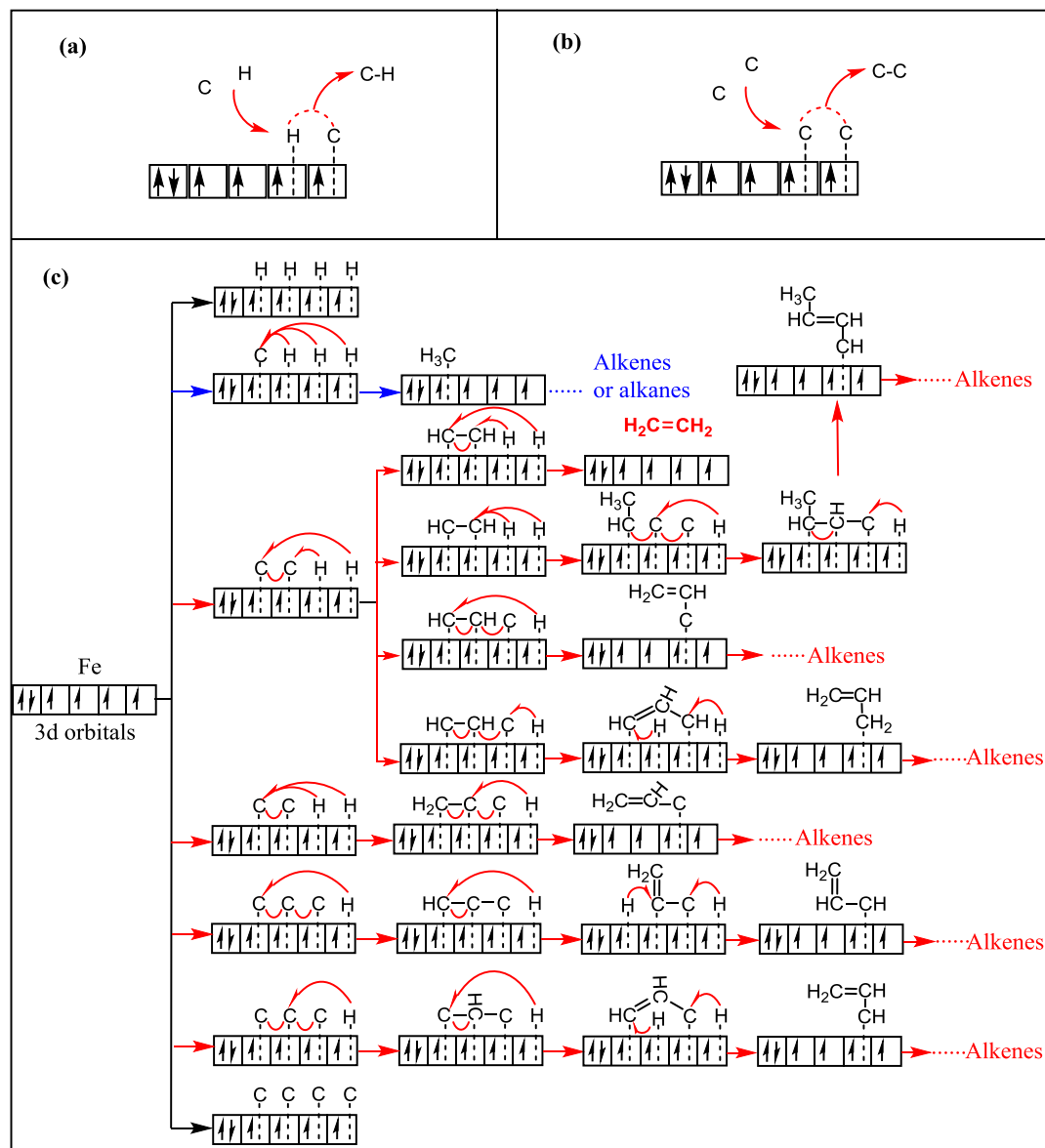


Figure 3. The process of catalytic reaction on transition-metal atom with HFOs: (a) the metal site bonds C-atom and H-atom on its surface via orbital interaction supporting the formation of C-H, (b) the metal site bonds two C-atoms via orbital interaction supporting the formation of C-C bond, (c) the possible process of F-T reaction on the surface of Fe atom to form the dominated products (alkenes). Red line routes represent a high possibility for F-T reaction following with this line, blue line route represents a low possibility for the occurrence of F-T reaction, and black line routes represents that it is impossible for F-T reaction occurring.

Since the discovery of the T-F reaction, various separate proposals for the mechanism of the reaction have been suggested, most of them focus on the formation of C-C bonds of the products. In 1926, Fischer and Tropsch [10] suggested that the C-C bonds of the product are produced through polymerization of CH_2 fragments on the metal surface. Later, Anderson and Emmett and their co-workers [11] suggested that C-C

bonds are made via a condensation reaction involving less of H_2O between hydroxy methylene groups on the surface. Subsequently, Pichler and Schultz [10, 12] suggested that the C-C bond analogous to the insertion of CO into metal-alkyl bonds in homogeneous systems. However, until today the distribution of products over various transition-metal catalysts and the formation of various chemical bonds of products are

still in debate. To understand the product diversity and distribution over various catalysts and design more effective and selective catalysts for desired product, there is a continuing requirement for studying the mechanism that various products are formed on the surface of catalysts. And for a mechanistic model to be acceptable, it should explain all the unique aspects of the F-T products.

It is claimed that iron ($[\text{Ar}]3d^64s^2$)-based F-T catalysts have the advantage of higher selectivity to alkenes, the remainder being alkanes. [13-15] The synthetic on cobalt($[\text{Ar}]3d^74s^2$)-based catalysts is rich in *n*-alkanes with a wide carbon number distribution. [16, 17] The lighter product fraction also contains some alkenes. However, the synthetic on nickel($[\text{Ar}]3d^84s^2$)-based catalysts is dominated by methane. [18] Those three metals lie in the same period in the periodic table which means most of the properties are similar. The major difference is the number of half-filled valence sub-orbitals (HFO); Iron has four HFOs on the 3d-orbital,

Cobalt has three HFOs on 3d-orbital and Nickel has two HFOs on the 3d-orbital. Therefore, there seem to be a certain correlation between the distribution of product and the VEC of metals, which is similar to our recent observation of catalytic property of various transition-metal for hydrogen evolution reaction. [5] Thus, it is assumed that surface atoms of metals with multiple HFOs have a potential to simultaneously bond reactive C and H atoms on its surface to support the formation of C-H and/or C-C bonds in the process of F-T reaction as showed in Figure 3a and 3b.

In 1976, Araki and Poncet observed that CO dissociation into C atoms via Equation 1 is a prerequisite for F-T reaction. [19] thus, the discussion of the formation of various hydrocarbons on catalyst initiates with the atomic C and H in present paper.

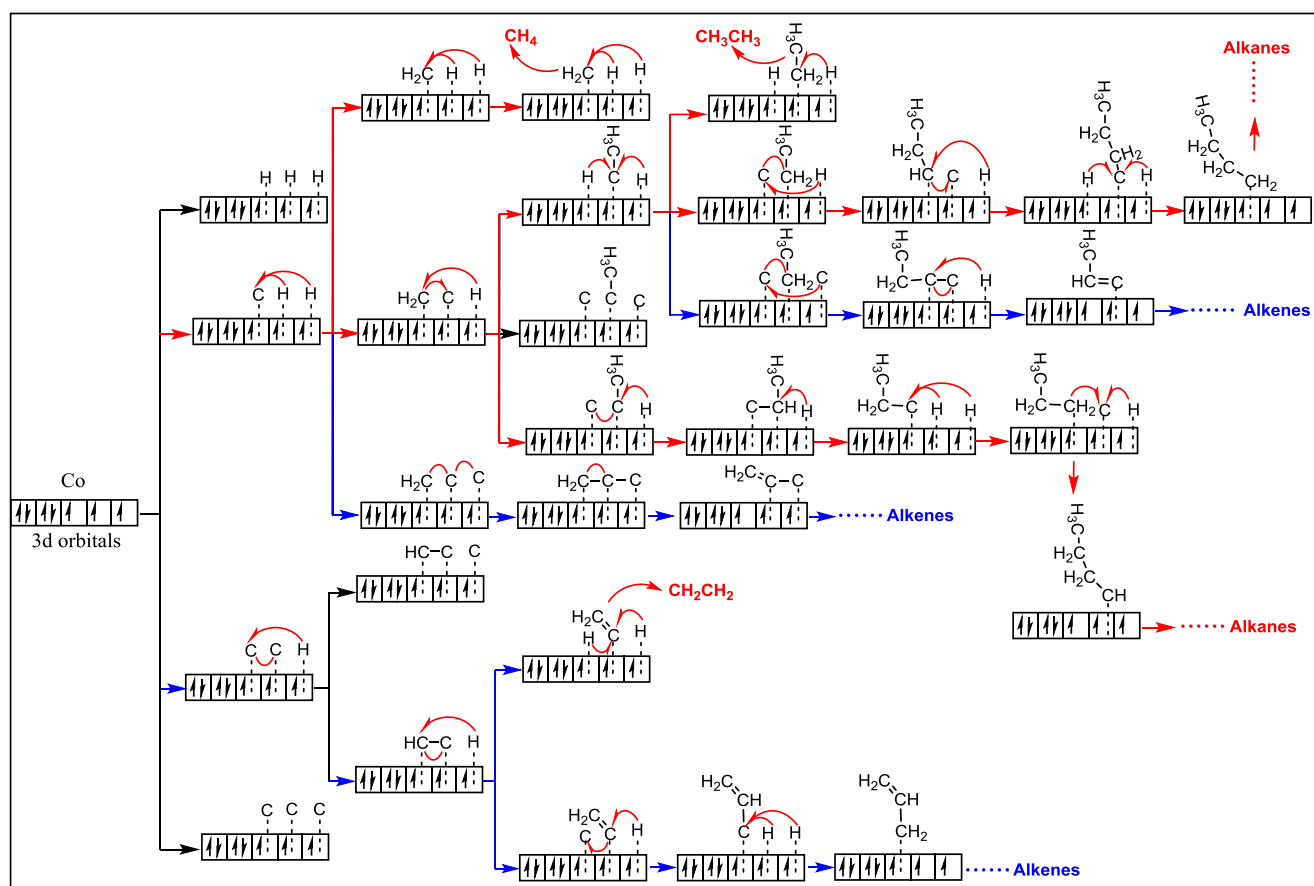
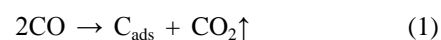


Figure 4. The possible pathways of F-T reaction on the surface of Co atom for the formation of the alkanes and alkenes. Red line routes represent a high possibility for F-T reaction following with this line to form alkanes, blue line route represents a low possibility for the occurrence of F-T reaction to form alkenes, and black line routes represents that it is impossible for F-T reaction occurring.

In the first place we will report on the treatment of the Fe-based F-T catalysts for higher selectivity to alkenes. One Fe atom have four HFOs, it means that based previously proposed mechanistic model, there are five types of possibility about the

bonding atomic C and H on the surface of Fe atom via orbital interaction: four H-atoms, one C-atom and three H-atoms, two C-atoms and two H-atoms, three C-atoms and one H-atom, and four C-atoms. We believe that it is impossible for the occurrence

of F-T reaction when Fe atom links with four atomic H or C via orbital interaction because the generation of hydrocarbon requires the formation both H-C and C-C bonds. Due to Iron element with low EN (1.8), [20] it is our opinion that there is a low possibility that Fe atom bond one C-atom and three H-atoms. Therefore, we assume that the F-T reaction occurring on Fe atom mainly starts with the bonding of two C-atoms and two H-atoms or three C-atoms and one H-atom via orbital interaction to support the formation of initial C-C and C-H bonds and producing the initial intermediates, such as HC-CH, H₂C-C, HC-C-C or C-CH-C as illustrated in Figure 3c. Those formed intermediates will link with Fe atom via C atom again and the left HFOs on Fe atom will bond with other C-atom and/or H-atoms via orbital interaction supporting the further formation of C=C and/or other C-H and/or C-C bonds on the base of initial intermediates. With subsequent stepwise bonding with further C-atom and/or H-atoms on the Fe atom and the formation of C=C and/or other C-H and/or C-C bonds based on the last intermediates, a widely range of alkenes products might be made. It is suggested, furthermore, that for the Fe atom, a high possibility of bonding one intermediates via C-atoms and two C-atoms is benefit to the formation of C=C bond, which might be the main reason that the Fe-based catalysts have an advantage of higher selectivity to alkenes.

In the case of Co-based catalysts there are three HFOs on a single Co atom, it is impossible to initiates the F-T reaction when three HFOs are occupied by three H-atoms or three C-atoms. Thus, it is assumed that the occurrence of F-T reaction on Co atom initiates with the linkage of one H-atom and two C-atoms or one C-atom and two H-atoms via orbital interaction to support the formation of C-H bond producing the initial intermediate (CH₂ or CH-C species) as illustrated in Figure 4. Due to a higher EN (1.9) of Co element compared with Fe (1.8), it has a relatively higher affinity with H-atom. Therefore, [20] we assume that there is a high possibility to simultaneously bond the C-based intermediates and two H-atoms or bond the C-based intermediates, one C-atom and one H-atom supporting the formation of C-H or C-C bonds, and those types of bonding might alternately occur and contribute to dominate the chain propagation and result in the formation of alkanes. It should be noted that in some case, three HFOs of Co site might be occupied by C-based intermediates and two C-atoms, this may contribute to the formation of C=C bond resulting the production of alkenes, but with low distribution in products due to high affinity of Co with H-atom. In addition, when three HFOs of Co atom might be occupied by two C-atoms and one H-atom, this may result in formation of alkenes, which might be the main reason that light alkenes

are usually observed in this case.

There is only two HFOs on a Ni atom, it is impossible for the occurrence of F-T reaction if both HFOs are occupied by two H-atoms or C-atoms. Thus, the catalytic reaction on Ni atom starts with the linkage of one H-atom and one C-atom via orbital interaction supporting the formation of C-H bond producing the CH species as showed in Figure 5. Ni with a high EN (1.9), [20] identical to Co element, have a high affinity with H-atom, therefore it is our opinion that there is a high possibility to bond CH_x intermediate and one H-atom on the surface of Ni atom via orbital interaction supporting the further formation of C-H bond, and a low possibility to bond CH_x intermediate and one C-atom supporting the formation of C-C, which might be the main reason the methane is main product on Ni-based catalysts.

Identical to the hydrogen evolution reaction, the catalysts (Fe, Co or Ni) require to be made in fine powder deposited on chemical-stable carrier when applied to drive the catalytic F-T reaction. Our proposal might provide a reasonable interpretation for this behavior. As we have suggested in our previous publication, [5] on the iso-surface of the atom, due to the orbital orientations in space, a part of the orbitals on the outermost of an atom might be blocked by neighbouring atoms in crystal and that those totally blocked sub-orbitals are inaccessible to involve in orbital interaction with reactants during the catalytic process. Only when considering atoms on the surface, a portion of the valence sub-orbitals on a particular atom is accessible with the portion of electron density for surface interactions. When an atom is lying on the inner basal plane, one-third of sub-orbitals have part region of electron density exposed, two-thirds of sub-orbitals have a region of its electron density exposed when an atom is lying on the edge site, and all sub-orbitals have region exposed when an atom is situated on the corner of a crystal. The 3d-orbital has five sub-orbitals, each of which exposes a part of the electron density while the atoms are lying in the corner. Each atom on the edges has two sub-orbitals that reveals a part of the electron density region. Atoms on the centre of crystal have just one sub-orbital with a visible zone of electron density. For driving the F-T reaction, it is required to bond more C-based species and H-atoms via orbital interaction to support the formation of C-H, C-C and C=C bonds. This means that only corner or edge atoms of metal are active during the catalytic reaction, explaining why reduced Fe, Co or Ni metals on the nanoscale could achieve the catalytic activity to drive F-T reaction. As a result, the highest surface area is also a fundamental prerequisite for building efficient solid-state catalysts to drive F-T reaction.

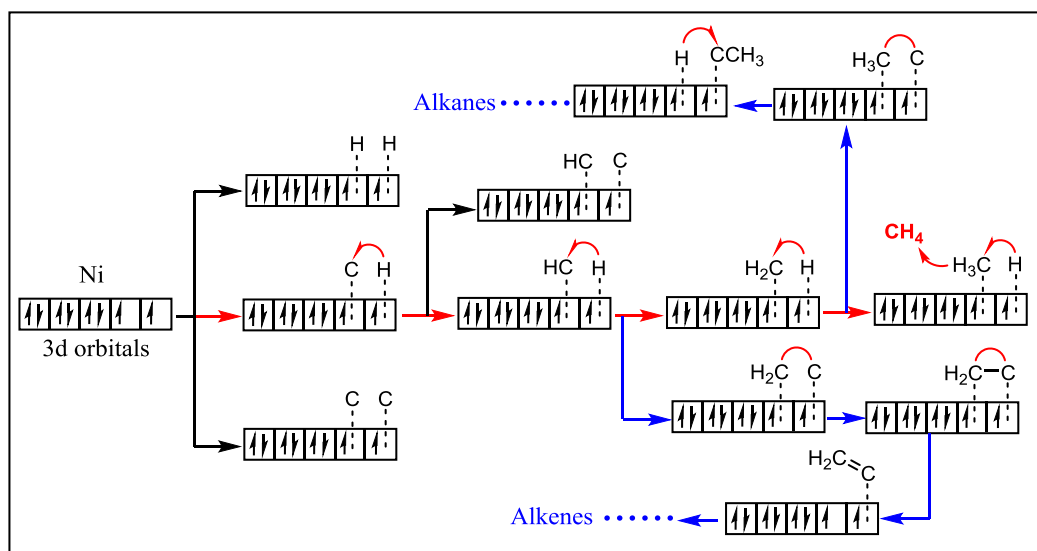


Figure 5. The possible process of F-T reaction on the surface of Ni atom to form the dominated products (methane). Red line routes represent a high possibility for F-T reaction following with this line, blue line route represents a low possibility for the occurrence of F-T reaction, and black line routes represents that it is impossible for F-T reaction occurring.

3. Conclusions and Future Work

In our treatments so far disclosed the part catalytic reactions occurring on metal catalysts, many of catalytic reactions are left untouched. If this mechanistic model is correct, it must be valid for all reactions catalyzed by metal catalysts. We shall try to gradually discuss them in our future publications.

The assumption as discussed above may be considered as corresponding to the interpretation of different metal catalytic reactions. A correlation between them needs to be confirmed by further investigations.

The opinion that the metal atoms with single or multiple unpaired electrons considered as radical centers could convert reactants into reactive radicals, as well as that an active site may also have an ability to bond two or more reactive radicals on its surface for formation of new chemical bond involved in the above discussion, is hoped to find applications more widely to problems on the catalytic activity of metal catalysts systems.

Abbreviations

F-T	Fischer-Tropsch
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Author Contributions

Youyi Sun is the sole author. The author read and approved the final manuscript.

Conflicts of Interest

The authors declare no conflicts of interest.

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Biography



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Research Field

Youyi Sun: catalytic reaction mechanism, electrochemical reaction, renewable energy storage, electrochemical reaction mechanism, catalytic ammonia synthesis, Fischer-Tropsch reaction.