

A Reconsideration of the Conventional Rule in Catalysis and the Consequences

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Abstract: The conventional rule that a catalyst increases a reaction rate by lowering the activation energy according to Arrhenius' law is the starting point of this article. However, this rule is incomplete, because the corresponding assignment of the true and the apparent activation energies is missing. The general validity of the rule can be determined by considering the entire reaction route depending on the temperature level. It forms an S-shaped curve, starting from the lowest and going to the highest conversion. In the middle of the curve, there is a turning point, which in catalysis is called the "isokinetic point". This turning point divides the curve into two parts: Below this point, the curve is exponential, and therefore, the Arrhenius equation and even the conventional rule can be applied. This means that the conventional rule does not have a general validity that can be applied to the whole curve. For this reason, an additional rule is introduced for the upper operating state: high activation energy is the condition for very high activity. The further point is the activation energy, which is regarded as an important term in catalysis. According to its definition, the "activation energy" is the "energy barrier" that a reaction must overcome. But this definition does not agree with the roots of this term. In reality, the Arrhenius energy is the temperature coefficient connected with the energy term. The catalyst reduces the temperature of the homogeneous reaction (that means the reaction without the catalyst) to the reaction temperature, and this results in a gain in energy, which will be called "reaction energy" to have a clear distinction with the Arrhenius energy. It is shown that the two energies significantly differ in their magnitudes.



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1. Introduction

Johann Wolfgang Döbereiner (1780–1849) produced, for the first time, a highly dispersed platinum, which was called platinum black [1,2]. This new material was able to ignite hydrogen at room temperature. He used this concept for the catalytic lighter in the year 1823 [3,4]. This lighter was widespread, and influenced Jöns Jacob Berzelius (1779–1848), the founder of modern chemistry, to introduce the new expression "catalysis" in the year 1835/1836 [5,6]. At that time, a catalyst was regarded as a material with a "mysterious quality" [7]. The reason for this assumption is based on the fact that the presence of a particular substance which remains unchanged enables a reaction or enhances the reaction rate. It was not until 1889 that Svante Arrhenius (1859–1927) established an empirical equation for an approximately quantitative description of the temperature dependence of the rates of chemical reactions in which an energetic barrier (activation energy) had to

be overcome at the molecular level [8]. It described a phenomenological connection and applied to many chemical reactions, including catalytic reactions.

Even today, catalysis seems to be a modern form of “Alchemy” according to the responses to two interviews in the year 1990 [9] and in the year 2013 [10]. In these interviews, selected catalysis experts in the relevant fields reported on the progress and even the open questions. The current information on the latest scientific advances in catalysis research is presented in the Roadmap of German Catalysis Research [11].

The characteristic property of a catalyst is its catalytic activity (and selectivity, which is not regarded in this paper), which is the reason for its different applications in the industry; primarily in the chemical and petrochemical industry, because around more than 85% of the products are made through catalytic processes. Even in other fields, catalysts are essential, e.g., electrocatalysis in the context of hydrogen technology or “three-way” catalytic converters, called “cats”, for cars to reduce nitrogen oxides are actual examples. Furthermore, this extends to homogeneous catalysis, organocatalysis, biocatalysis, photocatalysis, and so on. The focus in the following discussion is concerned with heterogeneous catalysis.

The widely used conventional rule explains the effectiveness of a heterogeneous catalyst. The explanation is as follows: if the activation energy is lower, then the effect of catalytic activity is higher [12]. This rule was introduced by Georg-Maria Schwab (1899–1984) in 1931 in his fundamental book concerning the chemical kinetics in catalysis, and which has been cited 559 times in the literature until now [13,14]. This rule has consequently become one of the shibboleths (features) of surface chemistry [14]. For this reason, the rule is regarded as a fundamental declaration.

The conventional rule is very useful to explain the function of a catalyst in a very easy way and is therefore widespread. It is still the subject of modern textbooks on heterogeneous catalysis [15]. In addition, the rule is still used to determine the activity of a catalyst according to the theorem: a low activation energy is an indication of high activity [16]. However, it is increasingly pointed out in the literature that in many cases the well-established rule only partially reflects reality [17–20]. In fact, the rule is incomplete, because it does not distinguish between true and apparent activation energy. In addition, the rule does not have general validity, because the maximum activity is not taken into account. The term “maximum activity” refers to the highest activity or the highest conversion, which is close to the upper operating state. For this reason, highly active catalysts, such as platinum black by Döbereiner, which is able to ignite hydrogen at room temperature, are excluded. This leads to an important message: with a low activation energy, a high activity or a high conversion cannot be achieved. For this reason, some interpretations based on the conventional rule are questionable.

The intention of this article is to analyze and to reconsider the phenomena concerning the influence of the magnitude of the activation energy on the catalytic activity in a critical way. The result shows that although the known doctrines have a very interesting and significant aspect in terms of their meaning, they need to be reinterpreted in their entirety.

2. General-Use Terms, Definitions, and Methods

2.1. The Catalytic Activity

There are different terms and different methods used to determine catalytic activity. A general definition for this activity was introduced by Friedrich Wilhelm Ostwald (1853–1932). His definition states that a catalyst can increase the rate of only those processes that are thermodynamically favourable [21,22]. Furthermore, the thermodynamic equilibrium remains unchanged. But there is no instruction given for the conditions to measure catalytic activity. Also, only the increase in the rate is regarded, but not the highest conversion.

For highly active catalysts, the ignition temperature can be used according to the method of Döbereiner, because a low ignition temperature indicates a high activity. This concept was used to examine the ignition behaviour of an exothermic catalytic reaction and will be discussed later.

And finally, the conventional rule can be applied because a low activation energy indicates a high activity. This concept simplifies the determination of the catalytic activity. Therefore, the activation energy as the Arrhenius activation energy is regarded as an important expression in catalysis, which is based on the following definition: it is the energy barrier that a system must pass on its way from the reactant to the product state [23]. This rule is generally accepted, and a critical review seems to be inopportune. Despite this fact, some questions remain. Does this rule correspond to the scientific standards? This is the main question, which concerns the uniqueness and the general validity of this rule.

2.2. The True and the Apparent Activation Energy

One problem of the rule is the fact that the term “activation energy” is not clearly characterized because a distinction between the true and apparent activation energies is missing [13] (pp. 164–165) [14] (p. 301). Therefore, the rule is incomplete, and the informative value of the rule is reduced.

The mentioned terms are not always clearly presented in the literature. Thus, a short overview will be presented in the context of the corresponding Arrhenius equation. These equations are based on an exponential formula which describes the temperature dependence of the rate of the reaction, v :

$$\ln v = \ln A - E/RT \text{ or } \ln v = \ln A - E_a/RT \quad (1)$$

Equation (1) (with E and E_a , the Arrhenius activation energies) is used if the reaction order is unknown. For a zero-order reaction, we have the rate constant, k ,

$$\ln k = \ln A - E_{\text{true}}/RT \quad (2)$$

and the true activation energy, E_{true} . If there is no zero-order, then the reaction rate, r , and the apparent activation, E_{app} , with the apparent pre-exponential factor, A_{app} , are used:

$$\ln r = \ln A_{\text{app}} - E_{\text{app}}/RT \quad (3)$$

For a zero-order reaction, the Arrhenius terms are not influenced by the educt concentration, but the apparent terms are influenced. More information concerning the kinetics are in the corresponding textbooks.

3. General Validity and the Validity Regions

The general validity can be determined if the whole reaction route is regarded, as seen in Figure 1. This reaction route corresponds with an S-shaped curve (sigmoid curve) and is called the logistic function. This reaction route starts with the lowest conversion and ends with the highest conversion. These two points are called the lower and upper operating states. The most significant is the turning point near the middle of the curve. This point divides the whole reaction route into two different temperature regions. This turning point has many names in catalysis [24]. Only the expression “isokinetic point” or “isokinetic temperature”, T_i , will be regarded in the following discussion.

There are two temperature regions, and each region has its own rule; these rules are in contradiction. In the lower operating state, the curve is exponential. In this region, the Arrhenius equation is applied and the conventional rule is valid. However, there is no

general validity for the entire reaction process, because a high activity is excluded. It must be stated that a catalyst with a low activation energy cannot reach the highest activity or the best conversion, as seen in Figure 1.

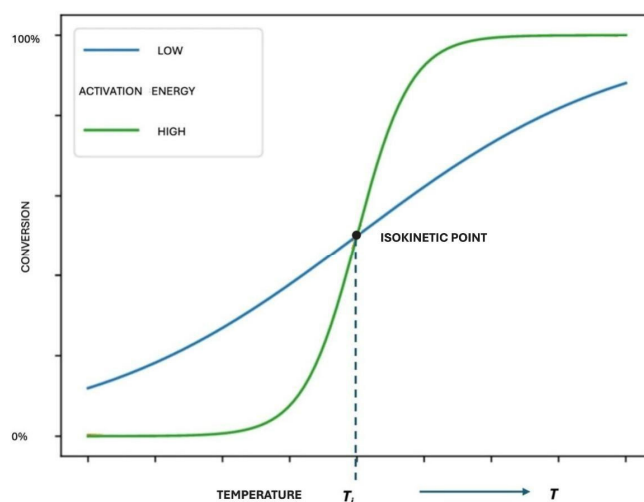


Figure 1. Two sigmoid curves with low and high activation energy. The two rules are easily recognizable. The new rule applies in the upper operating state and is the inverse form of the conventional rule. This means that a high activation energy is the prerequisite for high activity.

3.1. The Temperature Region $T < T_i$

The curve in the temperature region $T < T_i$ has the following advantage: the function is exponential, and the influence of pore diffusion can be neglected. Therefore, the Arrhenius Equation (1) can be applied. Furthermore, the conventional rule is valid in this region, which is preferred in fundamental research. These facts are the advantages of this region. The disadvantage is that the performance (upper operating state) is disregarded.

3.2. The Temperature Region $T > T_i$

This region contains the upper operating state with the highest conversion. A high activity is reached if two conditions are fulfilled: (a) the highest conversion must be reached more or less at (b) the lowest temperature. The first point indicates the performance of the catalyst, and the second point specifies the activity. This definition of the activity is unambiguous in contrast to the conventional terms which were presented in Section 2, with one exception: the ignition temperature.

Very active catalysts are an impressive example, especially catalysts which are able to ignite the educt. They reach the upper operating state by a sudden increase. This behaviour is called “ignition” or “thermal instability”. The condition for this behaviour has many influencing variables: namely, the increase in the values of the enthalpy of the reaction, of the concentration of the educts, and of the surface-specific pre-exponential factor. The key variables have the same influence: a high activation energy and a high surface area of the active component, like platinum black, which was mentioned in Section 1. These conditions have the following consequence: the conventional rule is not valid for very active catalysts. A further consequence is that this rule has no general validity. This region with the upper operating state is important for highly active and/or industrial catalysts. The limitation is caused by pore diffusion and/or by the thermodynamic equilibrium. The influence of the space velocity or residence time on the catalytic activity can be observed and controlled [25,26].

3.3. The Two Rules for the Whole Reaction Route

It must be stated that there are always two rules, and that this is a theoretical dilemma; however, this dilemma can be overcome by ignoring the other case. It is indicated that a low activation energy enhances the activity in the temperature region $T < T_i$ according to the conventional rule, but not in the region $T > T_i$. In this case, a high activation energy enhances the reaction, which is valid for active catalysts. There is a significant difference in the two rules: the conventional rule is valid for catalysts with a low activity, but the contrary is valid for highly active catalysts.

The provisional result of the discussion is that apparent kinetic parameters are responsible for the isokinetic relation. The statement that apparent effects dominate will be used to complete the conventional rule. The reconsidered wording reads as follows: the decrease in apparent activation energy is accompanied by the increase in catalytic activity. In this case, exceptions are disregarded. Furthermore, it must be remarked that the true activation energy is insensitive.

The fact that the apparent activation energy is variable, but the true activation energy is not, raises the following question: can only adsorption effects be observed, but not catalytic effects? This question remains open and must be answered in a separate paper.

4. The Revised Version of the Conventional Rule

4.1. Arrhenius Activation Energy

Schwab [13,14] describes the basis of the activation energy in his fundamental book as follows: the temperature coefficient of the Arrhenius equation is the source for the calculation of the activation energy. But in reality, there is no calculation, because the temperature coefficient is renamed to the activation energy. And consequently, this action leads to a contradiction, because the temperature coefficient is dimensionless.

This contradiction can be solved with the following procedure: the temperature coefficient, α_T , is the real term for the exponent of the Arrhenius equation:

$$\alpha_T/RT \quad (4)$$

and the gas constant, R , must be cancelled to reach conformity with the temperature coefficient, α_T . As a consequence, the Arrhenius equation becomes the reduced form:

$$\ln v = \ln A - \alpha_T/T \quad (5)$$

In this case, α_T is a composed term. It contains the dimensionless temperature coefficient of the reaction and of the adsorption.

The Arrhenius activation energy does not agree with the definition, because it is not the “energy barrier”, but, in reality, the dimensionless temperature coefficient. In other words, the activation energy cannot be determined with the Arrhenius equation. The activation energy is a theoretical construct, because the term does not agree with the definition. Nevertheless, the term is very useful in the conventional theory.

The consequence is that the literature concerning the term Arrhenius activation energy needs to be revised. The term “activation energy” must be transformed into the proposed term—the temperature coefficient—with respect to the new way of looking at it. This procedure will be an exceptional effort, because the literature of the last hundred years must be revised. Furthermore, the corresponding interpretations must be proven, and, if necessary, reformulated.

4.2. The Reaction Energy

The catalytic reaction reduces the reaction temperature, and, as a consequence, the energy requirement. Schwab has already impressively demonstrated that the catalytic reaction reduces the reaction temperature and thus the energy requirement [14] (p. 169). For this purpose, using measurable terms, only the two temperature levels were regarded: the reaction temperature without the catalyst, $T_{\text{no cat.}}$, and the reaction temperature with the catalyst, $T_{\text{cat.}}$. In addition, the term activation energy will not be used in this context, and instead, the term “reaction energy”, E_r , is introduced, which is calculated using the following equation:

$$E_r = (T_{\text{no cat.}} - T_{\text{cat.}}) \cdot R \quad (6)$$

4.3. Comparison of Activation Energy with the Reaction Energy

In the literature, there is no distinction between the activation energy and the reaction energy. Here, we will use a concise example to show that the assumption of the reaction energy to be identical to the Arrhenius activation energy is not justified. The results presented in the work [27] on the total oxidation of ethylene on noble metal catalysts are used as evidence. The catalysts were optimized using a test device based on the DTA principle. The device was designed so that the measurements could be carried out quickly and the ignition point recorded digitally.

Two kinds of Pt and Pd catalysts with a metal content of 1 wt.% and 0.05 wt.%, respectively, were produced by impregnating of the carrier $\alpha\text{-Al}_2\text{O}_3$ ($S = 6 \text{ m}^2/\text{g}$) with Pt or Pd nitrate. The particle size of the metal components was varied by pretreatment at different temperatures: Pt(U) and Pd (U) at 700 K, and Pt(T) at 1200 K for 2 h. The precious metal surface was determined by selective CO adsorption in the proven way as described in [28]. The catalyst Pt(U) has particles with a size of about 1–2 nm and the catalyst Pt(T) from 2 to 7 nm. The Pd catalyst already had a relatively large crystallite diameter of a similar size to Pt(T). The specific surface area and the pore volume of the carrier were not changed by the thermal treatment.

The test reaction used was the total oxidation of 1.5 vol% ethylene in air. The temperature of the gas stream flowing around the catalyst, at which under certain, defined test parameters the reaction ignited or at which the catalyst overheated compared to the gas flow by a specified temperature value (ignition temperature), served as a measure of the catalytic activity. In addition, the activation energy of the reaction was determined in the traditional way by the Arrhenius equation in the lower operating state where the curve was exponential [27].

The catalytic results obtained, which gave rise to us questioning the conventional rule, are shown in Table 1. The ignition temperature was used to determine the activity, because a low ignition temperature is a clear indication of high activity. The activity (low ignition temperature) is influenced by the percentage of the active component, and, as a consequence, the surface area of this component. In general, it can be stated that the higher the surface area, the more active the respective catalyst is. There is an influence of the particle size on the activation energy. The two Pt catalysts (mentioned above) show the following results: For the catalyst Pt(U) with 1% Pt, an activation energy of 90 kJ/mol is determined; on the other hand, for the catalyst Pt(T) with 1% Pt, it is only 71 kJ/mol. Nevertheless, the first catalyst, which has a surface area of the active material of $2.7 \text{ m}^2/\text{g}$, reaches an ignition temperature of 105°C , whereas for the second catalyst with a surface area of the active material of $1.0 \text{ m}^2/\text{g}$, the ignition temperature of 127°C is correspondingly higher.

Table 1. Comparison between activation energy and reaction energy using the example of the total oxidation of ethylene (C_2H_4).

Ignition Temperature Without Catalyst	Ignition Temperature with Catalyst	Surface of the Active Material S (m^2/g)	Temperature Difference	Reaction Energy E_r (kJ/mol)	Activation Energy E (kJ/mol)	Catalyst
~ 425 °C	105 °C	2.7	320 °C	4.9	90	Pt(U)
	160 °C	0.05	265 °C	4.4	90	Pt(U)
	127 °C	1	298 °C	4.7	71	Pt(T)
	185 °C	0.1	240 °C	4.2	71	Pt(T)
	195 °C	0.6	230 °C	4.18	46	Pd(U)
	245 °C	0.1	180 °C	3.76	46	Pd(U)

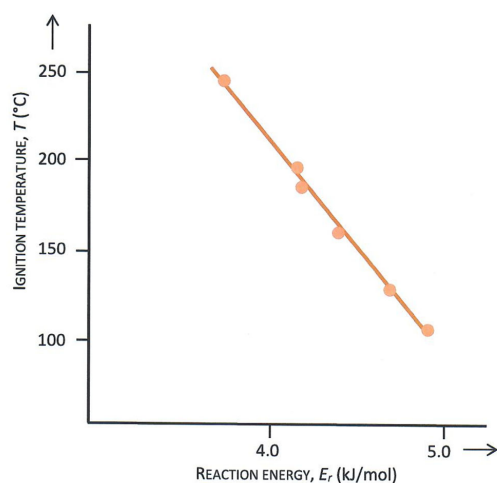
This example shows that a high activation energy correlates with an increase in catalytic activity. The result contradicts with the conventional rule. If the reaction energy is calculated according to Equation (6) and compared with the activation energy, it can be seen that a highly active catalyst has a very low ignition temperature, and consequently, the reaction energy is also high. Regarding the results in Table 1, it can be stated that the following inequality is valid:

$$E_r \neq E \text{ or } E_a \quad (7)$$

There is a large difference between the reaction energy and the Arrhenius activation energy (about one order of magnitude). The consequence of this is that these two terms are not identical. The Pd catalyst has a very low activity and a very low activation energy, which confirms once again that the conventional rule is not valid in the upper reaction state.

The result of the presented data can be expressed in the following way: the conventional rule is not valid for highly active catalysts. The reverse conclusion is that the conventional rule is valid for catalysts with low activity. The Arrhenius activation energy—a central term in catalysis—is not identical with the energy barrier. The consequence is that a variation in the apparent activation energy is caused by the variation in the temperature coefficient of adsorption.

On the other hand, there is a linear relationship between the reaction energy and the ignition temperature (Figure 2). This relationship means that the activity correlates with the reaction energy, or in other words, a low ignition temperature correlates with a high reaction energy. In addition, the reaction energy is a clear activity scale that can indicate the optimal level of activity.

**Figure 2.** Linear relationship between ignition temperature and reaction energy: a low ignition temperature (=high activity) correlates with a high reaction energy.

5. Conclusions

The conventional rule is the basis of theory building because it can explain the effectiveness of a catalyst in an easy way. According to this rule, the activity is enhanced if the activation energy is reduced. However, the rule is not complete, and needs to be supplemented:

1. The distinction between true and apparent activation energy is missing. There are hints that the apparent activation energy is appropriate.
2. The conventional rule has no general validity because the catalysts with high activity or high conversion are not regarded. For this case, a new rule is necessary which is contrary to the old rule: a high activation energy is necessary to reach the highest activity or the highest conversion. Furthermore, with a low activation energy, the highest activity cannot be reached.
3. There are two rules: the conventional rule is valid in the lower operating state and the new rule in the upper operating state. This situation can be described as follows: the first case is valid for catalysts with low activity and the second case for catalysts with high activity.
4. The activation energy is not the “energy barrier” according to the definition. It is—and this is a surprising result—the temperature coefficient connected with the term energy, which is a contradiction. Furthermore, a variation in the “activation energy” is caused by a variation in the temperature coefficient of adsorption.
5. A new term, the “reaction energy” must be introduced to distinguish between “activation energy” and “reaction energy”. The last term is calculated by the following equation: $E_r = (T_{\text{no cat.}} - T_{\text{cat.}}) \cdot R$, with the temperature of the homogeneous reaction $T_{\text{no cat.}}$, which means no catalyst, and the second term, the temperature with the catalyst, $T_{\text{cat.}}$. The term “reaction energy” was introduced to have a clear distinction.
6. A comparison of the activation energy with the reaction energy shows a great difference (more than one order of magnitude).
7. The conventional theory must be reconsidered and supplemented with additional terms and new concepts. This is great challenge, as the time frame concerns the last hundred years.

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