AVOIDING FIRST-YEAR UNIVERSITY CHEMISTRY TEXTBOOKS' MISREPRESENTATIONS IN THE TEACHING OF SPONTANEOUS REACTIONS

Educação

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This paper summarizes the misrepresentations related to Gibbs energy in general chemistry textbooks. These misrepresentations arise from a problem in the terminology textbooks use. Thus, after reviewing the proper definition of each of the terms analyzed, we present two problems to exemplify the correct treatment of the quantities involved, which may help in the discussion and clarification of the misleading conventions and assumptions reported in this study.

Keywords: first-year chemistry; textbooks; spontaneity.

INTRODUCTION

Research on learning difficulties associated with thermodynamics is well documented.[1] Educational studies[2,3] suggest that one of the sources of the students' learning difficulties in physical chemistry lies in how textbooks and teachers deal with key chemistry concepts. For example, several authors[4-11] have made an inventory of university students' misconceptions due to a poor understanding of the spontaneity concept. Some of those misunderstandings may have their origin in the way this concept is taught.

Misrepresentation of Gibbs energy

Misrepresentations in Gibbs energy (G) terminology can be found in many general chemistry textbooks. A complete discussion on this issue has been reported recently.[12,13] It was found that there are Gibbs energy changes that refer to different processes and that frequently they are not properly defined. For example, some authors discuss the sign of ΔG^o in order to stablish the spontaneous direction of a reaction mixture. Hence, these presentations often lead to the assumption that $\Delta G^o < 0$ corresponds to a general condition for spontaneity; conversely, it is assumed that if $\Delta G^o > 0$ the forward reaction is forbidden.

Those two referred studies concluded that most of the textbook confusions arise due to the overuse of the symbol 'Δ' in teaching thermodynamics in introductory university chemistry courses. That is, it is usually assumed that ΔG^o plays the role of $\Delta_r G^o$; similarly, $\Delta_r G$ is normally misrepresented as ΔG. For example, Q-K inequalities are normally employed to decide the direction of a disturbed equilibrium system, but this discussion is usually based on the following equation

$$\Delta G = RT \ln \frac{Q}{K} \qquad (1)$$

(instead of $\Delta_r G = RT \ln \frac{Q}{K^0}$).

Spontaneity and equilibrium

In order to avoid the teaching of misrepresentations similar to the ones stated above, in this section the aim is to consider practical situations exemplifying accurate calculations for the discussion of spontaneous reactions. Therefore, we will focus on discussing the meaning of spontaneity, stressing that this concept refers both to determining whether a reaction is product- or reactant-favored and to predicting the direction in which a reacting system shifts in response to a disturbance. Examples 1 and 2 are presented for this purpose. These problems focus on both the meaning of the sign of $\Delta_r G$ and its units. Eventually, Table 1 provides a glossary of Gibbs energy terminology in order to summarize the discussion that follows.

Before presenting those examples, it may be useful to review briefly the meaning of $\Delta_r G$, which also serves to stablish the different meanings of the three remaining quantities examined in this study, ΔG, $\Delta_r G^o$ and ΔG^o. An extended discussion of the basis of the following treatment can be found in several papers and advanced textbooks as it has been reviewed previously.[12-14]

$\Delta_r G = \sum_i \nu_i \mu_i$ is the so-called free energy of reaction, and represents the rate of change of G with respect to the extent of reaction (ξ), at constant T and P, and also the rate of change of A with respect to the extent of reaction, at constant T and V,

$$\Delta_r G = \left(\frac{\partial G}{\partial \xi} \right)_{T,P} \qquad (2)$$

$$\Delta_r G = \left(\frac{\partial A}{\partial \xi} \right)_{T,V} \qquad (3)$$

It must be stressed now the meaning of these equations. $\Delta_r G$ is a derivative and not an ordinary difference despite the use of "Δ", as signaled by the sub-r feature.

A general equation for the spontaneous direction of reaction in a specified reaction mixture, at a specified temperature, is the following[12,13]

$$\Delta_r G d\xi < 0 \qquad (4)$$

That is, for a spontaneous reaction from reactants to products [a A(g) + b B(g) → r R(g) + s S(g)], since $d\xi > 0$, then $\Delta_r G < 0$. For the reaction to reverse spontaneously [a A(g) + b B(g) ← r R(g) + s S(g)], since $d\xi < 0$, then $\Delta_r G > 0$.

Similarly, the general equilibrium condition can be written as follows[12,13]

$$\Delta_r G d\xi = 0 \qquad (5)$$

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Thus, the sign of $\Delta_r G$ allows us to predict the direction of the spontaneous chemical reaction. Moreover, if $\Delta_r G = 0$, the equilibrium has been attained.

ΔG is a finite difference in the Gibbs energy between two states. That is,

$$\Delta G = G_{final} - G_{initial} \tag{6}$$

These final and initial states can even be the equilibrium situation or reactants or products. For example,

$$\Delta G = G_{eq} - G_{reactants} \text{ and } \Delta G = G_{eq} - G_{products} \tag{7}$$

Notice that in both cases $\Delta G \neq 0$.

ΔG^o is also a finite difference, but now ΔG^o is the difference in the Gibbs energies of the products and reactants when they are unmixed and each is in its standard state,

$$\Delta G^o = G^0_{products} - G^0_{reactants} \tag{8}$$

It should be noticed that $\Delta_r G \neq \Delta G$. Not only is there a conceptual distinction between these two quantities, but there is also a differentiation in the units used to measure each quantity. That is, one should realise that ΔG is an extensive quantity; it is expressed in energy units only, kJ. Conversely, $\Delta_r G$ is not a finite difference: it is an instantaneous rate of change of G with respect to the extent of reaction. It is an intensive quantity and is normally reported in units of kJ mol^{-1}. At equilibrium the rate of Gibbs energy change is zero, $\Delta_r G = 0$. But, the value of ΔG is indeed a finite diference between a given initial situation and a final one that can be the equilibrium state. That is, as the process goes from the initial mixture to equilibrium a change in Gibbs energy occurs, thus $\Delta G \neq 0$. However, it is true that some authors may state their readers that the initial situation is already equilibrium. Hence, then, obviously, $\Delta G = 0$, since the process will have done nothing to get to equilibrium.

The standard free energy of reaction, $\Delta_r G^o$, is the rate of change of standard Gibbs energy, viz.

$$\Delta_r G^0 = \left(\frac{\partial G^0}{\partial \xi}\right)_T \tag{9}$$

It is a constant quantity,

$$\Delta_r G^0 = \frac{\Delta G^o (kJ)}{\Delta \xi \ (mol)} = \frac{G^0_{products} - G^0_{reactants}(kJ)}{(1-0)(mol)} \tag{10}$$

[In eq. (10) we have considered that initially we have $n_0(A) = a$ mol and $n_0(B) = b$ mol; thus, $\xi_{initial} = 0$ and $\xi_{max} = 1$]

Therefore, $\Delta_r G^o$ is an intensive quantity and is expressed in kJ mol^{-1}.

Textbook discussions concerning the meaning of the value of this last quantity are usually made with incorrect analyses. That is, neither can the negative value of $\Delta_r G^o$ be used as the general condition for spontaneity, nor is it true that a positive value of $\Delta_r G^o$ means that a chemical reaction will not proceed. It is the sign of $\Delta_r G$ that should always be considered for that purpose. In example 1 we analyse this situation. In this case, despite $\Delta_r G^o > 0$, the forward reaction is spontaneous ($\Delta_r G < 0$). We will make an enlarged point of clarification on this issue after discussing the meaning of equation (14).

A proper calculation of $\Delta_r G$ makes use of the following equation

$$\Delta_r G = RT \ln \frac{Q}{K^o} \tag{11}$$

where, K^o is the thermodynamic equilibrium constant. For a given equilibrium reaction such as: a A(g) + b B(g) $\rightleftharpoons$ r R(g) + s S(g), it is expressed as follows,

$$K^o = \frac{\left(\frac{p(R)_{eq}}{p^o}\right)^r \left(\frac{p(S)_{eq}}{p^o}\right)^s}{\left(\frac{p(A)_{eq}}{p^o}\right)^a \left(\frac{p(B)_{eq}}{p^o}\right)^b}; p^o = 1 \text{ bar} \tag{12}$$

and Q is the reaction quotient, which has the form of the equilibrium constant, K^o, but it is not equal to the equilibrium constant [observe that when $\Delta_r G = 0$ (equilibrium), then $Q = K^o$],

$$Q = \frac{\left(\frac{p(R)}{p^o}\right)^r \left(\frac{p(S)}{p^o}\right)^s}{\left(\frac{p(A)}{p^o}\right)^a \left(\frac{p(B)}{p^o}\right)^b} \tag{13}$$

K^o and Q are here defined for homogeneous gas phase reactions. Notice that equation (13) contains non-equilibrium specification of partial pressures, while the thermodynamic equilibrium constant, K^o, defined in equation (12) contains equilibrium specifications.

As $\Delta_r G^o = -RT \ln K^o$, equation (11) can be written as follows

$$\Delta_r G = \Delta_r G^0 + RT \ln Q \tag{14}$$

Equation (14) allows us to calculate the value of $\Delta_r G$ and, therefore, to discuss the direction of the spontaneous reaction. At this point the reader is reminded that the forward reaction is spontaneous when $\Delta_r G < 0$ and for the reaction to reverse spontaneously $\Delta_r G > 0$. This has been the approach performed in the two examples below.

But, one does not need to calculate the value of $\Delta_r G$ in order to know the direction of the spontaneous reaction. Q-K inequalities can be used as a basic criteria for spontaneity in isothermal conditions. From equation (11) we can state that if $Q < K^o$ ($\Delta_r G < 0$), the forward reaction is spontaneus; conversely, if $Q > K^o$ ($\Delta_r G > 0$), the reaction is spontaneus in the backward direction. Eventually, if $Q = K^o$ ($\Delta_r G = 0$), there is an equilibrium mixture. In example 1 we have calculated the value of $\Delta_r G$ corresponding to an initial non-equilibrium situation. As $\Delta_r G < 0$, we have concluded that the reaction will proceed in the forward direction. We could have reached the same conclusion stating that in that initial conditions, $Q < K^o$. And in example 2, the equilibrium disturbance has caused that $\Delta_r G > 0$, concluding that the reaction will proceed in the backward direction. Similarly, we could have reached the same conclusion stating that in that disturbed equilibrium conditions, $Q > K^o$.

Finally, it should be stressed that only when $Q = 1$ can the sign of $\Delta_r G^o$ serve to predict the direction of the spontaneous chemical reaction, as in this situation $\Delta_r G = \Delta_r G^o$.

Table 1 summarizes the Gibbs energy (G) terminology associated with the four quantities ($\Delta_r G$, ΔG, $\Delta_r G^o$ and ΔG^o) that have been discussed previously. It should be stressed that $\Delta_r G \neq \Delta G$. ΔG is an extensive quantity; its value is indeed a finite diference between a given initial situation and a final one. Conversely, $\Delta_r G$ is not a finite difference: it is an instantaneous rate of change of G with respect to the extent of reaction. It is an intensive quantity and can be calculated using an equation that has two terms: one is the value of $\Delta_r G^o$, which is constant, and the second one depends on the value of the reaction quocient, Q. At this point, it should also be emphasised that the value of $\Delta_r G$ changes as the composition of the reaction mixture varies, which means that the value of Q is modified. But the value

Table 1. Glossary of Gibbs energy (G) terminology for a chemical reaction represented as: a A(g) + b B(g) $\rightleftharpoons$ r R(g) + s S(g)

Free energy of reaction (intensive quantity; kJ mol^{-1} units):

$$\Delta_r G = \left(\frac{\partial G}{\partial \xi}\right)_{T,p} = \sum_i \nu_i \mu_i, \text{ where } \mu_i \text{ are the chemical potentials}$$

$$\Delta_r G = \Delta_r G^0 + RT \ln Q = RT \ln \frac{Q}{K^0}$$

Standard free energy of reaction (intensive quantity; kJ mol^{-1} units):

$$\Delta_r G^0 = \left(\frac{\partial G^0}{\partial \xi}\right)_T = \sum_i \nu_i \mu_i^0, \text{ where } \mu_i^0 \text{ are the chemical standard potentials}$$

$$\Delta_r G^o = -RT \ln K^o$$

Change in the Gibbs energy (extensive quantity; kJ units):

$$\Delta G = G_{final} - G_{initial} \text{ (for example: } \Delta G = G_{eq} - G_{reactants} \text{ or } \Delta G = G_{eq} - G_{products})$$

Change in the standard Gibbs energy (extensive quantity; kJ units):

$$\Delta G^o = G^0_{products} - G^0_{reactants}$$

of $\Delta_r G^o$ is constant and thus it can only play the role of $\Delta_r G$ when $Q = 1$. Hence, the sign of $\Delta_r G^o$ cannot be used as a general criterion for spontaneous reactions.

At this stage, in order to make a point of extended clarification, it may be useful to stress the previous discussion on the meanings of the signs of both $\Delta_r G$ and $\Delta_r G^o$. It will be done with the help of example 1.

In this case, it should be noticed that although $\Delta_r G^o > 0$, the forward reaction is spontaneous. That is, the relationship $\Delta_r G^o > 0$ means that if a reaction mixture in which $Q = 1$ at the temperature specified (600 K), then the backward reaction is the spontaneous direction of reaction. However, this particular case (ie. $Q = 1$) is not the one dealt with in example 1 and of course it is not the condition for most chemical reactions. Rather, the sign of $\Delta_r G$ accurately establishes the direction of the spontaneous reaction whatever the initial composition of the reaction mixture may be. In our example, the relationship $\Delta_r G < 0$ means that in the definite reaction mixture with the specified amounts of substances, at 600 K, the forward reaction is spontaneous.

Another application of the above discussion is the prediction in the evolution of a disturbed chemical equilibrium system when a reactant is added to an equilibrium mixture. This case was found to be misrepresented in many general chemistry texbooks as well as in official chemistry exams[15] as most of the cases studied, as a rule, set forth qualitative questions whose statements left out, for the most part, the variables which remain constant when the equilibrium is disturbed. It was ascertained that teachers wanted their students to apply the Le Châtelier´s principle as an infallible rule to solve the problem. In example 2 it is discussed this case on a problem involving the addition of one of the reactants at constant temperature and pressure.

Indeed, the backward reaction is not expected when the application of Le Châtelier's qualitative principle is intended for this particular situation.[16-23] Adding a reactant at constant pressure changes the concentrations of all gaseous components of the raction mixture (as the volume of the reactor increases). The Le Châtelier's principle is limited to make a prediction on the direction of the subsequent reaction, although it has been generally assumed that adding a reactant always shifts the perturbed equilibrium mixture to the direction of the forward reaction. That is, in cases of mass perturbations, it is

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Example 1

At 600 K, $K^o = 1.24 \times 10^{-4}$ for the equilibrium corresponding to the methanol synthesis

$$CO\,(g) + 2\,H_2\,(g) \rightleftharpoons CH_3OH\,(g)$$
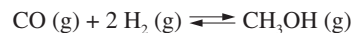
In the study of this reaction, the total pressure of the gas mixture is 500 bar at 600 K, and the amounts of each substance are: $n(CO) = 1.64$ mol, $n(H_2) = 0.82$ mol and $n(CH_3OH) = 0.95$ mol. State the direction of the spontaneous reaction.

Solution

If $\Delta_r G < 0$, the forward reaction [CO(g) + 2H$_2$(g) $\rightarrow$ CH$_3$OH(g)] is spontaneous. Conversely, if $\Delta_r G > 0$, the backward reaction [CH$_3$OH(g) $\rightarrow$ CO(g) + 2 H$_2$(g)] is then spontaneous. The value of $\Delta_r G$ can be determined as follows

$$\Delta_r G = \Delta_r G^0 + RT \ln Q$$

Firstly, we can calculate the value of $\Delta_r G^o$
$\Delta_r G^o = -RT \ln K^o = -(8.3145\ \text{J K}^{-1}\ \text{mol}^{-1})\,(600\ \text{K})\,(\ln 1.24 \times 10^{-4})$
$= 44.9\ \text{kJ mol}^{-1}$

The reaction quotient, Q, is

$$Q = \frac{p(CH_3OH)/p^o}{\{p(CO)/p^o\}\{p(H_2)/p^o\}^2}\ (p^o = 1\ \text{bar})$$

where

$$p(CH_3OH) = y_{CH_3OH}\ p = \frac{0.95}{3.41} \times 500 = 140\ \text{bar}$$

$$p(H_2) = y_{H_2}\ p = \frac{0.82}{3.41} \times 500 = 120\ \text{bar}$$

$$p(CO) = y_{CO}\ p = \frac{1.64}{3.41} \times 500 = 240\ \text{bar}$$

Thus, we have

$$Q = \frac{(140)}{(240)(120)^2} = 4.05 \times 10^{-5}$$

Finally,
$\Delta_r G = \Delta_r G^0 + RT \ln Q$
$\Delta_r G = 44.9 \times 10^3\ \text{J mol}^{-1} + (8.3145\ \text{J K}^{-1}\ \text{mol}^{-1})(600\ \text{K})(\ln 4.05 \times 10^{-5})$
$= -5.49\ \text{kJ mol}^{-1}$.
As $\Delta_r G < 0$ (observe also that $Q < K^o$), the forward reaction [CO (g) + 2 H$_2$(g) $\rightarrow$ CH$_3$OH (g)] will proceed till an equilibrium state is reached. (Notice that although $\Delta_r G^o > 0$, the forward reaction is spontaneous).

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doubly incorrect to assert (and teach) that Le Châtelier's principle predicts that an increase in the amount of one component shifts the equilibrium in the direction that decreases the mass of that component, because such prediction is neither universally true nor Le Chatelier's, having been disproved and disowned by Le Châtelier himself.[24-26] This case has been a source of misconceptions among teachers and first-year university students.[2,27-33] For example, many students and teachers assume that the forward reaction will always take place after one of the reactants has been added to an equilibrum mixture.

Example 2

At 700 K, $\Delta_r G^o = 23.4$ kJ/mol, for the equilibrium corresponding to the ammonia synthesis:

$$1/2\ N_2\ (g) + 3/2\ H_2\ (g) \rightleftharpoons NH_3\ (g)$$
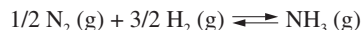
In the study of this reaction, the total pressure of the gas mixture is 100 bar at 700 K, and the equilibrium amounts of each substance are: $n(N_2) = 0.513$ mol, $n(H_2) = 0.204$ mol and $n(NH_3) = 0.139$ mol. If it is added, at constant temperature and pressure, 0.120 mol of $N_2(g)$, which will be the direction of the spontaneous reaction?

Solution

Although this is a case of a disturbed equilibrium, its treatment is similar to the example 1. The aim is to calculate $\Delta_r G$ after disturbing the initial equilibrium. If $\Delta_r G < 0$, the forward reaction [$1/2\ N_2\ (g) + 3/2\ H_2\ (g) \rightarrow NH_3\ (g)$] is spontaneous. Conversely, if $\Delta_r G > 0$, the backward reaction [$NH_3\ (g) \rightarrow 1/2\ N_2\ (g) + 3/2\ H_2\ (g)$] is then spontaneous.

The value of $\Delta_r G$ can be determined as follows

$$\Delta_r G = \Delta_r G^0 + RT \ln Q$$

As we know the value of $\Delta_r G^o$, we need to calculate the value of Q.

The reaction quotient, Q, is

$$Q = \frac{p(NH_3)/p^o}{\{p(N_2)/p^o\}^{1/2}\{p(H_2)/p^o\}^{3/2}} \quad (p^o = 1 \text{ bar})$$

where

$$p(NH_3) = y_{NH_3}\ p = \frac{0.139}{0.976} \times 100 = 14.2 \text{ bar}$$

$$p(H_2) = y_{H_2}\ p = \frac{0.204}{0.976} \times 100 = 20.9 \text{ bar}$$

$$p(N_2) = y_{N_2}\ p = \frac{0.633}{0.976} \times 100 = 64.9 \text{ bar}$$

Thus, we have

$$Q = \frac{(14.2)}{(64.9)^{0.5}(20.9)^{1.5}} = 1.84 \times 10^{-2}$$

Finally,

$\Delta_r G = \Delta_r G^0 + RT \ln Q$

$\Delta_r G = 23.4 \times 10^3$ J mol^{-1} + (8.3145 J K^{-1} mol^{-1})(700 K)(ln 1.84×10^{-2})

= 146 J mol^{-1}.

As $\Delta_r G > 0$, the the backward reaction [$NH_3\ (g) \rightarrow 1/2\ N_2\ (g) + 3/2\ H_2\ (g)$] will proceed till a new equilibrium state is reached. Notice that for this reaction we can calculate the value of the equilibrium constant, K^o. In this case, we have $K^o = 1.79 \times 10^{-2}$. That is, $Q > K^o$, which is in agreement with the positive value of $\Delta_r G$, and hence with the prediction of reaction shift made.

These assumptions can be overcome if Le Châtelier's principle is avoided and then the essential part of the argumentation is founded on a sound thermodynamic basis, as it has been stated by several authors.[2,12,13,17,27,34-45] Once again, Q-K^o inequalities may play an accurate

basic role. That is, one merely needs to distinguish between the ratio of partial pressures or concentrations at the disturbed equilibrium situation (Q) and the value when the system is at equilibrium (K). In example 2, as $Q > K^o$ the backward reaction is then spontaneous.

Although similar exercises as examples 1 and 2 can be found in some first-year chemistry textbooks, in most of the cases studied their authors have not paid attention to the basis of the above discussion, which has caused several misrepresentations and misleading assumptions. In summary, the sign of $\Delta_r G^o$ must not be used as a general condition in order to predict the direction of a spontaneous reaction; it is the sign of $\Delta_r G$ that should always be interpreted in order to accurately predict the direction of the spontaneous reaction when giving a situation involving some initial conditions. The same is true when studying the evolution of a disturbed chemical equilibrium system. That is, Le Châtelier's principle is a limited rule that can be overcome analysing the meaning Q-K inequalities, which are grounded on the meaning of the sign of $\Delta_r G$. Textbooks usually concentrate on practical situations where the addition of one of the reactants is assumed at constant volume. Example 2 studies the evolution of a disturbed chemical equilibrium in which a reactant has been added at constant presssure. The presentation of new possible perturbed situations in which the addition of one of the reactants is made at constant pressure may contribute in overcoming some student (and also teacher) misconceptions connected with this topic.

CONCLUSIONS

The presentation of Gibbs energy accomplished in this paper assists in establishing the fundamentals to make alternative accurate approaches to the current misrepresentations found in general chemistry textbooks.[12,13] This paper has focused on the appropriate calculation of both $\Delta_r G$ and $\Delta_r G^o$ and on the meaning of their sign. To accomplish this purpose, the examples examined in this study were designed on the proper use of free energy of reaction in the analysis of spontaneous reactions. It is stressed that the calculation (and then the sign) of $\Delta_r G$ serves to establish the direction of a chemical reaction. Clarification in the meaning of the different terms involved seems essential, which provides sound approaches when dealing with spontaneous reactions. Two problematic cases have been analysed on the basis of current misrepresentations and misconceptions. It has been emphasized that: a) the sign of $\Delta_r G^o$ must not be used as a general condition in order to predict the direction of a spontaneous reaction and b) adding a reactant to a chemical equilibrium mixture at contant pressure is a case in which the Le Châtelier's principle is a limited rule that can be overcome analysing the meaning of Q-K inequalities (which are grounded on the meaning of the sign of $\Delta_r G$) in order to make accurate predictions.

Hence, similar problems to the ones discussed in this study can be presented to students when dealing with spontaneous reactions. This may help instructors in avoiding both current misrepresentations of Gibbs energy and the misuse of Le Châtelier's principle.

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