

Research Article

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Does it occur or not? – A structured approach to support students in determining the spontaneity of chemical reactions

<https://doi.org/10.1515/cti-2022-0046>

Received November 3, 2022; accepted August 17, 2023; published online November 3, 2023

Abstract: Thermodynamics is a central topic of chemistry as it holds a crucial role in understanding chemical processes. Due to its complexity, level of abstraction and amount of “mathematics”, chemical thermodynamics is one of the most challenging subjects resulting in a huge number of inappropriate conceptions. To determine whether a chemical reaction is spontaneous in a particular direction, students need to apply a huge amount of chemical knowledge when completing a complex, multi-step thinking process. In a Design-Based Research project, we developed “unmathematical” instructional material that aims at supporting students in accomplishing this demanding task. By providing a step-by-step instruction and incorporating scaffolding measures, students are guided through four successive tasks. The material was implemented in more than ten upper secondary chemistry classes and four teacher education courses and was proved to be proper for independent work, supporting students in connecting thermodynamics to basic chemical knowledge. Working on the material enables students to overcome barriers in the field of chemical thermodynamics and to successfully complete tasks in the area of thermodynamics and physical chemistry. Teachers can easily adapt the material to specific contents, levels, and teaching requirements. Further, the supportive material can be applied to offer differentiated and individualised learning paths.

Keywords: chemical thermodynamics; material development; secondary chemistry education; design-based research

1 Introduction

Thermodynamics provides a framework that is essential to understanding chemical processes and thus plays a central role in chemistry. It gives information about energy changes that occur during chemical reactions and allows chemists to predict whether a particular reaction is spontaneous (in the suggested direction) or not. In this way, chemical thermodynamics can be used to explain natural phenomena and industrial processes (e.g., Brown et al., 2022; Engel & Reid, 2021) and is thus an obligatory part of upper secondary chemistry curricula.

However, students experience chemical thermodynamics as a complex, abstract and mathematic-loaden topic and consider it to be one of the most challenging subjects across the chemistry curriculum (e.g., Nilsson & Niedderer, 2014; Tsaparlis, 2007). Researchers have found that chemical thermodynamics is mainly focused on

By publishing the material under an open license, we want to invite chemistry teachers and chemistry education researchers to translate and adapt the material according to their own needs and implement it in teaching practice and research. We welcome any form of feedback, suggestions for improvement, and ideas for joint research projects.

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concepts such as energy, work and heat (Goedhart & Kaper, 2002) and that students are confronted with a “mess” of different terms and concepts across the science disciplines (Finkenstaedt-Quinn et al., 2020; Jewett, 2008). Cooper et al. (2014) also complain that the concept of energy is often introduced from three different perspectives (macroscopic, atomic-molecular, quantum-mechanical) that are not connected to each other, what prevents students from developing a comprehensive understanding. Beyond that, lessons about chemical thermodynamics are characterised by mathematical formula and calculations rather than by qualitative (chemical) descriptions (Carson & Watson, 2002; Nilsson & Niedderer, 2014). Therefore, it is hardly surprising that students struggle to connect thermodynamics to their prior chemical knowledge, resulting in a variety of inappropriate conceptions (e.g., Bain & Towns, 2018; Barker & Millar, 2000; Kind, 2004; Nilsson & Niedderer, 2014; Saricayir et al., 2016; Sözbilir, 2004; Tsaparlis, 2007). As a result, students’ self-efficacy beliefs related to physical chemistry are consistently low, even though positive beliefs would be important for successful learning (Nicoll & Francisco, 2001; Sözbilir, 2004).

To determine whether a chemical reaction occurs spontaneously in a particular direction, students need to apply a huge amount of basic chemical knowledge, e.g., constitution of compounds and state of matter, bonding models and bonding energy, chemical reactions and reaction equations (e.g., Boo, 1998; Kind, 2004; Sözbilir et al., 2010). The issues thermodynamics deals with are mainly related to changes that are represented in the form of graphs, diagrams or mathematical equations. Mastering tasks in the field of chemical thermodynamics thus requires students to switch between different levels of representation and description, making use of mathematics in chemical contexts (Cooper et al., 2014; Goedhart & Kaper, 2002; Nilsson & Niedderer, 2014). Beyond that, determining the spontaneity of chemical reactions is a complex, multi-step thinking process including a number of inverse relations to consider.

To support students in accomplishing this challenging task, it is for one thing necessary to link thermodynamics tasks to prior chemical knowledge (Cooper et al., 2014) and for another thing to reduce cognitive load. We therefore aimed at developing “unmathematical” instructional material for upper secondary chemistry education that supports students in applying the basic analysis steps of chemical thermodynamics. Guided by the material, students should apply their prior chemical knowledge to make qualitative predictions about changes in enthalpy and entropy, and finally draw conclusions about the spontaneity of chemical reactions. Additional supportive material addresses students’ different prior knowledge and various needs, hence enabling them to work on the tasks independently and individually and experience themselves as competent to successfully complete tasks in the field of chemical thermodynamics.

2 Design & method

To develop the material in a systematic and structured way, a design project following the Design-Based Research approach (DBR Collective, 2003) was conducted by two researchers who are also secondary chemistry teachers. The DBR approach was selected as it is considered suitable for designing innovative solutions for practical teaching problems whilst at the same time producing theory-generating results (Oppl et al., 2022).

The process of material development followed a five-step DBR-cycle suggested by Fraefel (2014; see Figure 1). In the first step (*Problem Analysis*), we explored the theoretical background of chemical thermodynamics and examined students’ conceptions and challenges in this field. Based on these findings and including practical experience from former teaching, we derived implications for the design of the instructional material and developed an initial version (*Design*). In the next step (*Implementation*), we proved the developed material in both upper secondary chemistry classes and chemistry teacher education and examined its appropriateness and applicability. The experiences made during the implementation were then exchanged and the material was discussed with regard to beneficial aspects and aspects in need of improvement (*Reflection*). After having revised and adapted the material accordingly (*Re-design*), we implemented it again (see Figure 1). Overall, this iterative cycle was processed several times including implementation more than ten times.

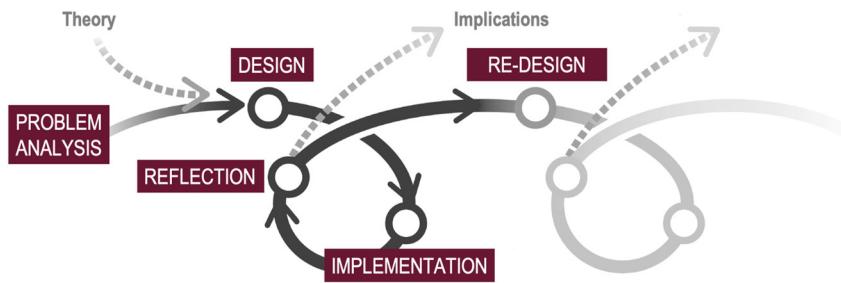


Figure 1: The iterative Design-Based Research cycle (adapted from Fraefel, 2014, p. 9).

2.1 Problem analysis

Chemical reactions are processes in which substances are transformed into new substances with different properties (e.g., Brown et al., 2022; Chang & Goldsby, 2013; Tro, 2018). These transformation processes can proceed at different speed and occur either on its own without any external intervention or be forced to, e.g., by supplying energy or manipulating pressure-conditions (e.g., Brown et al., 2022; Lucas, 2008; Mortimer, 2008). The “voluntary” chemical processes are called spontaneous, although spontaneity refers to only one direction (“natural direction”). The reverse chemical reaction is nonspontaneous and must be forced, if requested. Whether a chemical reaction does or does not occur spontaneously is mainly driven by changes in two thermodynamic quantities, enthalpy and entropy (e.g., Atkins et al., 2019; Brown et al., 2022; Engel & Reid, 2021; Frenking & Shaik, 2014; Klotz & Rosenberg, 2008; Mortimer, 2008).

Reviewing the existing body of literature, two factors are broadly found to influence students’ success in thermodynamics and physical chemistry in general: mathematical proficiency and problem-solving abilities (Bain et al., 2014; Becker & Towns, 2012; Derrick & Derrick, 2002; Hahn & Polik, 2004; Nicoll & Francisco, 2001; Tsaparlis, 2007). This is not surprising as thermodynamics lessons are mostly mathematic-loaden and characterised by physical concepts, even in chemistry education (Goedhart & Kaper, 2002; Nilsson & Niedderer, 2014). However, even if thermodynamics lessons are designed more “chemical”, the topic is still highly demanding for students who need to apply a huge amount of basic chemical knowledge. Analysing chemical processes from the perspective of thermodynamics requires students to link observations at the macroscopic level with descriptions at the particle level mostly including mathematical formula and other symbolic representations (Anderson et al., 2005; Cooper et al., 2014; Goedhart & Kaper, 2002; Nilsson & Niedderer, 2014). According to Cooper et al. (2014), chemical thermodynamics and especially the concept of energy is discussed by referring to three different perspectives: macroscopic, atomic-molecular, and quantum-mechanical. However, there is often little to no connection between these perspectives, hindering students from developing a comprehensive understanding of energy changes associated with chemical processes (Cooper et al., 2014).

In the following sections, we present the chemical background the developed material relies on by integrating all perspectives, macroscopic, atomic-molecular, and quantum-mechanical. Completing the problem analysis, we present an overview of students’ conceptions and challenges in the field of chemical thermodynamics.

2.1.1 Energy changes in chemical reactions

During chemical reactions, particles are rearranged by breaking existing bonds and forming new bonds, which is accompanied by changes in energy (e.g., Brown et al., 2022; Chang & Goldsby, 2013; Tro, 2018). These energy changes are mainly associated with changes in potential energy arising from electrostatic interactions at the particle level. Each type of chemical bonding results from electrostatic attraction between oppositely charged particles, e.g., cations and anions (ionic bonding), nuclei and “electron gas” (metallic bond), or nuclei and shared pair(s) of electrons (covalent bonding). Thus, breaking chemical bonds means to increase the distance between the bonding partners by overcoming the forces of attraction. The greater the magnitude of charge and the stronger the attraction between the bonding partners, the stronger the resulting bond and the more energy is

required to break it. The reverse is true when bonds are formed (e.g., Brown et al., 2022; Frenking & Shaik, 2014; Mortimer, 2008). Summarised, breaking/forming strong bonds (e.g., ionic bonds) requires/releases more energy than breaking or forming rather weak bonds (e.g., nonpolar covalent bonds).

The energy changes associated with rearrangements at the particle level are always accompanied by energy transfer from the system (substances involved in the reaction) to its surroundings (reaction vessels and everything beyond) and vice versa (e.g., Atkins et al., 2019; Brown et al., 2022; Klotz & Rosenberg, 2008). The chemical energy of the reactants represents the internal energy of the system in its initial state ($E_{initial}$) and the chemical energy of the products represents the internal energy of the system in its final state (E_{final}). The change in internal energy (ΔE) is defined as difference between E_{final} and $E_{initial}$ as represented in equation (1) (e.g., Atkins et al., 2019; Jenkins, 2008; Keszei, 2012; Mortimer, 2008).

$$\Delta E = E_{final} - E_{initial} \quad (1)$$

When the amount of internal energy of a system decreases in the course of a chemical reaction, i.e., the reactants are higher in energy than the products, ΔE is negative and the energy released by the system is absorbed by the surroundings. If the products are higher in energy than the reactants (ΔE is positive), the amount of internal energy of the system increases by gaining energy from its surroundings (e.g., Atkins et al., 2019; Keszei, 2012; Klotz & Rosenberg, 2008; Mortimer, 2008; Smith, 2008). This absorbed energy is then “stored” as chemical energy in the bonds (e.g., Brown et al., 2022; Dauer et al., 2014; Lucas, 2008).

2.1.2 Enthalpy changes in chemical reactions

In most cases, chemical reactions (especially those outside of the laboratory) occur in open systems and under constant pressure (e.g., atmospheric pressure). In these reactions, energy can be exchanged between the system and its surroundings in the form of heat and work. This energy transfer is described by the thermodynamic quantity called enthalpy. Enthalpy (H) is defined as the sum of the internal energy of a system (E) and the product of the pressure (p) and the volume (V) of the system. The change in enthalpy (ΔH) is then given by the relationship presented in equation (2) (e.g., Atkins et al., 2019; Keszei, 2012; Klotz & Rosenberg, 2008; Mortimer, 2008).

$$\Delta H = \Delta E + p \cdot \Delta V \text{ (constant pressure)} \quad (2)$$

If the considered reactions are additionally (to conditions at constant pressure) limited to processes doing no other work than volume change work, the change in enthalpy (ΔH) equals the amount of heat the system gains from or releases to its surroundings. If the change in enthalpy is negative ($\Delta H < 0$), i.e., the amount of enthalpy decreases in the course of a chemical reaction, then heat is released from the system and absorbed by its surroundings. Such reactions are called exothermic and are characterised by an increase in temperature. Conversely, ΔH is positive ($\Delta H > 0$) for endothermic reactions in which heat is gained by the system from its surroundings (decrease in temperature) (e.g., Atkins et al., 2019; Keszei, 2012; Klotz & Rosenberg, 2008).

Enthalpy is a state function, which means that the change in enthalpy depends only on the enthalpy of the system in its initial state (enthalpy of reactants; $H_{reactants}$) and on the enthalpy of the system in its final state (enthalpy of products; $H_{products}$), but not on the specific reaction path (e.g., Atkins et al., 2019; Keszei, 2012; Klotz & Rosenberg, 2008; Mortimer, 2008). Thus, the enthalpy change of a chemical reaction (ΔH_{rxn}) is given by the relationship in equation (3).

$$\Delta H_{rxn} = H_{products} - H_{reactants} \quad (3)$$

As the change in volume is small for the majority of chemical reactions, the difference between ΔE and ΔH is small for most chemical reactions as well (e.g., Atkins et al., 2019; Brown et al., 2022; Keszei, 2012). Thus, enthalpy changes in chemical reactions can be qualitatively estimated by considering the internal energy of reactants and products associated with their bonding properties (see Section 2.1.1). Considering that energy must be provided for each bond to be broken and energy is released for each bond that is formed, the enthalpy change of the reaction is equal to the difference between the total energy to be provided for breaking bonds and the total energy released by forming bonds (Atkins et al., 2019; Klotz & Rosenberg, 2008; Smith, 2008). If the bonds in the reactants

are stronger than the bonds in the products, more energy is needed for breaking bonds than energy is released by forming new bonds, $\Delta H > 0$, and the reaction is endothermic. In the opposite case (bonds in the products are stronger than bonds in the reactants), $\Delta H < 0$, and the reaction is exothermic.

Applied to chemical processes, this means that reactions in which ionic compounds (great in bond strength) are formed from covalent ones and metals (smaller in bond strength) are therefore very likely to be exothermic. Conversely, reactions in which polar covalent bonds are present in the reactants and weaker, nonpolar covalent bonds are present in the products (e.g., electrolysis of water) tend to be endothermic.

In general, a decrease in enthalpy, as it occurs in exothermic reactions, fosters the spontaneity of a chemical reaction. However, energy changes in chemical reactions are not limited to breaking and forming bonds, but also include effects of dispersing particles. Thus, changes in entropy must also be taken into account in order to determine whether a chemical process does or does not occur spontaneously (e.g., Atkins et al., 2019; Brown et al., 2022; Keszei, 2012; Lucas, 2008; Smith, 2008).

2.1.3 Entropy changes in chemical reactions

Entropy (S) is a thermodynamic quantity that reflects the degree of randomness in a system. From the perspective of statistical thermodynamics, entropy reflects the number of possible microstates; the number of different ways particles can be arranged (e.g., Alzeer, 2022; Atkins et al., 2019; Brown et al., 2022; Jenkins, 2008; Lucas, 2008; Vogt, 2017). The higher the number of possible microstates (“arrangement possibilities”), the more possible ways the system has to distribute its energy and the more likely it is to reach a state of low(er) internal energy. An increase in entropy is therefore energetically favourable and thus promotes the spontaneity of a chemical reaction (e.g., Alzeer, 2022; Atkins et al., 2019; Lucas, 2008; Smith, 2008; Vogt, 2017).

Since entropy is a state function, like enthalpy, changes in entropy can be estimated by comparing the entropy of the system in its initial state and its final state (equation (4)).

$$\Delta S_{rxn} = S_{final} - S_{initial} \quad (4)$$

The number of possible microstates of a system can be estimated by the degree of dispersion of particles and their freedom to move independently. Consequently, considering the states of matter and the number of particles of reactants and products can be used to make qualitative predictions about the entropy changes in chemical processes (Lambert, 2002b; Lauth & Kowalczyk, 2022; see also e.g., Atkins et al., 2019; Brown et al., 2022; Smith, 2008).

In solids, particles are arranged in lattice structures (e.g., crystalline structures or metal lattices) and are located to nearly fixed positions. Particle motion is restricted to vibration only what means a quite small number of possible microstates. In the liquid phase, particles are still relatively close to each other, however, their motion is no longer limited to vibration. The particles are additionally free to rotate and move around one another. This crucially increases the number of possible microstates and thus the entropy compared to solids. The largest number of microstates is possible in gases. Here, particles are completely free to move and may spread independently from each other. Consequently, the entropy of gases is higher than of liquids, which in turn is higher than of solids ($S_{gas} > S_{liquid} > S_{solid}$) (e.g., Atkins et al., 2019; Brown et al., 2022; Klotz & Rosenberg, 2008; Lauth & Kowalczyk, 2022; Mortimer, 2008; Smith, 2008).

The situation is similar for reactions in which the number of particles changes (mostly prevalent in reactions including molecular reactants and products). A decrease of the number of molecules is associated with an increase in molecular size. Larger molecules are less free to move (in particular in the gaseous phase) and hence the dispersion in the system is reduced. Consequently, the number of possible microstates and thus the entropy in the system decrease (e.g., Brown et al., 2022; Jenkins, 2008; Lambert, 2002b; Lauth & Kowalczyk, 2022; Mortimer, 2008; Smith, 2008).

Summarised, there are two “categories” of reactions that are very likely to be associated with an increase in the entropy of the system, what in turn fosters the spontaneity of these reactions: (1) reactions in which lattice structures “disappear” and/or gases arise from either solids or liquids, and (2) reactions in which the number of molecules of gaseous products is higher than the number of molecules of gaseous reactants.

However, to make a definitive statement about the spontaneity of a chemical reaction, entropy changes need to be linked to enthalpy changes and the temperature of the reaction.

2.1.4 Changes in Gibbs free energy and the spontaneity of chemical reactions

Gibbs free energy is a state function that relates to both thermodynamic concepts, enthalpy and entropy and also considers the absolute temperature of a chemical reaction (in Kelvin). Assuming reaction conditions at constant temperature and pressure, the change in Gibbs free energy (ΔG) is defined by the difference of change in enthalpy (ΔH) and the product of absolute temperature and change in entropy ($T \cdot \Delta S$). This relationship is shown in the Gibbs-Helmholtz-Equation (equation (5)) (Atkins et al., 2019; Jenkins, 2008; Klotz & Rosenberg, 2008; Mortimer, 2008; Smith, 2008).

$$\Delta G_{rxn} = \Delta H_{rxn} - T \cdot \Delta S_{rxn} \quad (5)$$

The sign of ΔG provides valuable information about the spontaneity of chemical reactions. For constant temperature and pressure, ΔG of a spontaneous (exergonic) chemical reaction is negative ($\Delta G < 0$) what means that the reaction occurs in the forward direction. For reactions that are nonspontaneous (endergonic; reactions need to be forced by external intervention), ΔG is positive ($\Delta G > 0$). If $\Delta G = 0$, a reaction is at equilibrium and does not occur spontaneously in any direction (Atkins et al., 2019; Jenkins, 2008; Klotz & Rosenberg, 2008; Lucas, 2008; Mortimer, 2008; Smith, 2008).

Whether a chemical reaction is spontaneous or not (in a given direction) depends on both the sign and magnitude of the terms ΔH and $T \cdot \Delta S$. Exothermic reactions ($\Delta H < 0$) that are accompanied by an increase in entropy ($\Delta S > 0$) result in a negative value for ΔG , regardless of temperature, and are thus spontaneous in any case. Conversely, endothermic reactions ($\Delta H > 0$) that are associated with a decrease in entropy ($\Delta S < 0$) always result in positive values for ΔG , what indicates that they are nonspontaneous at any temperature. If both ΔH and ΔS are like in sign, it depends on the temperature whether the reaction is spontaneous or not. Exothermic reactions with $\Delta S < 0$ tend to be rather spontaneous at lower temperatures and endothermic reactions with $\Delta S > 0$ tend to be spontaneous rather at higher temperatures (Atkins et al., 2019; Brown et al., 2022; Jenkins, 2008; Keszei, 2012; Klotz & Rosenberg, 2008; Lucas, 2008; Mortimer, 2008; Smith, 2008).

It must be remembered, however, that the situation becomes much more complicated if both reactants and products are present in a system in its initial state or if solutions or mixtures are involved in the reaction. If both reactants and products are present in a system (in nonequilibrium state), the reaction will spontaneously proceed in the direction necessary to reach equilibrium. Similar applies for reactions including solutions or mixtures. For these reactions, ΔG depends on the concentration and proportion of components in the mixture, respectively. In these cases, it is necessary to refer to the equilibrium constant (K) to determine the spontaneity of a chemical reaction. In addition, it must be noted that for reactions with gases (unlike for solids and liquids) pressure essentially influences ΔG , so that the Van't Hoff equation should be relied on to determine the spontaneity of these reactions more accurately (Atkins et al., 2019; Keszei, 2012; Klotz & Rosenberg, 2008; Mortimer, 2008; Smith, 2008).

2.1.5 Alternative conceptions in the field of chemical thermodynamics

Analysing chemical reactions from the perspective of thermodynamics is a complex and multi-step thinking process that requires students to grasp abstract concepts and connect them to their prior chemical knowledge. As stated in the previous sections, it is possible to make qualitative predictions about the spontaneity of chemical reactions by analysing and comparing the properties of reactants and products with regard to the concepts of enthalpy and entropy. In so doing, it is necessary to be familiar with the principles of chemical bonding (to make qualitative statements about enthalpy changes; see Sections 2.1.1 and 2.1.2), with the meaning of possible microstates regarding the structure of matter (to make qualitative statements about entropy changes; see Section 2.1.3), and with the concept of Gibbs free energy connecting the concepts of enthalpy and entropy (to determine

the spontaneity of chemical reactions; see Section 2.1.4). Accordingly, respective alternative conceptions are of great importance as they prevent students from both developing an appropriate understanding and completing tasks in the field successfully.

It has been shown in several studies that many students struggle with correctly understanding the principles of chemical bonding (e.g., Kind, 2004; Levy Nahum, 2007; Özmen, 2004; Taber et al., 2012). Chemistry education research is aware of a number of relevant alternative conceptions, such as that forming bonds would require energy while breaking bonds would release energy (e.g., Bain & Towns, 2018; Bain et al., 2014; Barker & Millar, 2000; Cooper & Klymkowsky, 2013; Galley, 2004; Kind, 2004; Levy Nahum et al., 2010; Saricayir et al., 2016; Sözbilir, 2001) or that both bond formation and breaking would be energy consuming processes (Boo, 1998; Nimmermark et al., 2016; Wren & Barbera, 2013). These conceptions are devastating for understanding energy changes in chemical reactions as they imply inverse relations and take the concept of spontaneity to absurdity (see Sections 2.1.1 to 2.1.4).

Concerning the concept of entropy, analogies with the term “disorder” are particularly problematic as they foster alternative conceptions preventing students from developing an appropriate understanding of entropy and its role in chemical processes (Bucy et al., 2007; Christensen et al., 2009; Meltzer, 2007; Bennett & Sözbilir, 2007). Abell and Bretz (2019) overall conclude that “many chemistry students do not understand the nature of entropy nor the connection between entropy and energy” (p. 1810).

In general, changes in enthalpy and entropy are often considered isolated from each other, neglecting the relationship represented in the Gibbs-Helmholtz-Equation. Resulting from this, students hold the conception that all exothermic reactions would be spontaneous while all endothermic would be nonspontaneous (Sözbilir, 2001, 2004; Sözbilir et al., 2010). Similar ideas exist for entropy changes, where an increase in entropy is equated with spontaneity (Bucy et al., 2007; Bennett & Sözbilir, 2007; Smith et al., 2009).

Beyond that, students tend to confound the concepts of thermodynamics and kinetics, e.g., by conflating spontaneity with equilibrium (e.g., Thomas & Schwenz, 1998; Sözbilir, 2002; Turányi & Tóth, 2013) or enthalpy with activation energy (Boo, 1998).

2.2 Design of the material

Taking up these issues, we aimed at developing “unmathematical” instructional material (as suggested by Carson & Watson, 2002) that supports students in determining the spontaneity of chemical reactions. The material should be suitable for student-centred lessons at the upper secondary level and meet the following requirements:

- connecting thermodynamics to basic chemical knowledge,
- relying on qualitative descriptions rather than on mathematical formulas,
- being thoroughly structured and including supportive measures.

Considering these requirements, we expected the material to elicit students’ prior knowledge and to support them in applying it. In this way, we aimed at fostering students’ conceptual understanding and enable them to experience themselves as competent to complete tasks in the field of thermodynamics and physical chemistry (as usually mathematic loaden topics) in general. For this purpose, the instructional material was planned to consist of several, sequential parts that follow the order of the analytical process for determining the spontaneity of chemical reactions based on the analysis of the properties on reactants and products. Accordingly, after an introductory part (Task 1), the bonding properties are to be analysed, from which qualitative predictions about the enthalpy change can be made (Task 2). By analysing the particle structure of reactants and products, predictions of the entropy changes can be made (Task 3), which are to be combined with the results of Task 2 and finally be applied to the Gibbs-Helmholtz equation to determine the spontaneity of the respective chemical reaction (Task 4).

To develop material that fosters students’ conceptual understanding about chemical thermodynamics on the one hand and that is manageable at the upper secondary level at the other hand (e.g., Le Maréchal & El Bilani, 2008; NRC, 2012), the underlying chemical background needed to be reduced and simplified according

the approach of educational reconstruction (Duit et al., 2012). Moreover, care was taken to incorporate all perspectives (macroscopic, atomic-molecular, and quantum-mechanical) in the material as suggested by Cooper et al. (2014). For this purpose, the three main types of chemical bonding were considered being separable, neglecting that no definite bounds exist between them.

Since qualitative estimations were aimed at, number and position of bonds broken (in molecular compounds) were ignored. Instead, the stability of the entire molecules was determined. Other disregarded aspects were intermolecular forces, physical side effects, such as solubility effects or enthalpies of solution or vaporisation, and the distinction between water formed as liquid or gaseous product. Furthermore, we did not address that Gibbs free energy is only a limited criterion for the spontaneity of chemical reactions including solutions and mixtures (due to effects of concentration/ratio), gaseous reactants (due to effects of pressure) and reactions in which both reactants and products are present in the initial state of the reaction (cf. Section 2.1.4). Concerning the concept of entropy, we decided to refrain from using the term “disorder” as this analogy has been found to cause inappropriate conceptions (Bucy et al., 2007; Bennett & Sözbilir, 2007; Lambert, 2002a). Instead, we consistently used “possible number of microstates” to characterize entropy (cf. Section 2.1.3).

The overall instructional goal for the material was set to help students understand that they can make thermodynamic predictions by analysing reactants and products of the reaction. Since several studies have identified tasks in the field of chemical thermodynamics as complex and overchallenging to many students (e.g., Nilsson & Niedderer, 2014; Tsaparlis, 2007), it was decided to provide instruction as concrete and precise as possible.

First of all, the multi-step thinking process was broken down into individual analysis steps that are formulated as several tasks and respective sub-items. To prevent students from cognitive overload, the tasks are formulated in the form of explicit instruction supported by scaffolding measures (e.g., Hammond & Gibbons, 2005; Hmelo-Silver et al., 2007; Quintana et al., 2004). Determining the spontaneity of chemical reactions by analysing the properties of reactants and products requires students to apply a huge amount of (already known) chemical concepts. Therefore, conceptual scaffolds providing essential knowledge (e.g., definitions, terminology, or relations) were developed to support students to recall their prior knowledge and strategies and to connect them with the concepts and procedures addressed in the task (Quintana et al., 2004). When implementing these scaffolding measures both implicitly (structure and instruction of the task) and explicitly (supportive material), the developed material is considered a “well-structured problem” (Jonassen, 2000) that aims at familiarising students with a possible procedure – an analysis scheme – they can then apply independently to a wide variety of reactions.

According to empirical findings, worked-out examples are appropriate to support students in working on well-structured problems that address demanding contents and complex analysis processes (Atkinson et al., 2000; Jonassen, 2011; Renkl & Atkinson, 2010). Thus, selected worked-out examples that enable students to reconstruct the problem-solving process were added to the task. Furthermore, the worked-out examples were formatted similar to reaction equations in order to provide a consistent structure throughout the entire task. In this way, the information required (e.g., type of bonding, stability of bond) can be directly assigned to reactants and products, which is expected to facilitate determining the changes in the course of the reaction.

From the perspective of practicing teachers, it was also important to us that the material is flexible for adaption and allows for differentiation. For this reason, the reactions were not directly integrated into the task, but were placed separately at the beginning so that they can be easily replaced without having to change the entire task. Varying both the number and type of reactions to be worked on and providing or withholding the scaffolds and worked-out examples allows teachers to adapt the task according to content focus and level of difficulty (*differentiation by content*; Taylor, 2015).

2.3 Implementation

To prove the developed material regarding its appropriateness and applicability, we implemented it for several times, as designated in DBR approaches (DBR Collective, 2003). Overall, students from more than ten upper secondary chemistry classes (grades 10/11) and four chemistry teacher education courses (6th semester of Bachelor programme) worked on the material.

During the material implementation, we collected information for revising the material in the form of lesson observation primarily (Henning et al., 2009; Knoblauch, 2019). In doing so, we documented and collected our experiences for the joint reflection and re-design by paying particular attention to the following aspects (Knoblauch, 2019):

- *Comprehensibility and clarity of the instruction*: At what points in the task do students not know what to do? At what points in the task are students asking for clarification? Which terms and instructional phrases result in inquiries?
- *Application of chemical concepts and practices*: Which concepts and practices cause difficulties in application? For which concepts do students need to refer to their textbook, individual notes, supportive materials or instruction by the teacher? What chemistry-specific practices are students not familiar with?
- *Progress in problem-solving*: At what points in the task do students get stuck and for what reasons? At what points, for what purpose, and how often do students use the supportive material or ask for support by the teacher?
- *Suitability for students' independent work*: Do students work (collaboratively) in a task-oriented manner? How long does it take students to complete the task? Do students use the worked-out examples to assess their results and in what ways?

Complementing the lesson observation, we held informal discussions with students and integrated tasks similar to the instructional material as part of the usual exams and/or assessments. In so doing, we strived for a kind of summative assessment (Dixson & Worrell, 2016) about students' abilities to make qualitative predictions about the spontaneity of a selected chemical reaction after having worked with the developed instructional material in both chemistry lessons and exam preparation. The outcomes of these tasks were corrected and analysed regarding common errors and problems.

The experiences gained from implementation are summarised in the following two sections, with Section 2.4 providing a general overview, before illustrating some exemplary challenges and how they were addressed when revising the material in Section 2.5.

2.4 Reflection

Having implemented the developed material for several times resulted in an increased amount of practical experience regarding the challenges of the task and the content addressed. We could identify aspects that were especially demanding for students and needed to be supported by intensive scaffolding both material-based and in person.

First of all, implementing the material revealed major differences in students' chemical knowledge and skills they were asked to apply when working on the task. This was confirmed in each implementation cycle, regardless of whether students were in their first year of secondary chemistry education or nearly at the end of their Bachelor programme. In one upper secondary chemistry class, for example, there were some students who successfully completed tasks 1 and 2 without any further support, others were already struggling with generating the chemical formulas and determining the main types of chemical bonding. This again confirmed that immense differences in knowledge and skills can arise even after a few months in chemistry class. Thus, scaffolding measures are essential to enable students to work in their zone of proximal development (Vygotsky, 1978). To address students' individual needs and promote their learning in the best possible way, the material-based scaffolding has shown to be indispensable. Only in this way, the teacher can allow for individual learning paths

and handle students' diverse capabilities and needs. As shown in the implementation, students were able to work independently on the material if they knew how to purposefully use supportive material, such as information cards or worked-out examples. One of the secondary chemistry students stated: *"It was very difficult at the beginning. (...) But the worked-out examples helped me a lot."*

In general, working on the material was perceived as a demanding but "fruitful" experience. Many of the students – even the student teachers – were basically familiar with the required concepts, however, they did not have all the concepts ready at the same time. One student mentioned that *"each task in itself was actually not that difficult"* as similar tasks had been practiced in former chemistry lessons. However, it was found *"challenging to remember so many different concepts and apply it all at the same time"*. Another one emphasised the sequential structure of chemical subject-matter and concluded that the material *"requires you to apply everything that you have learned before"*. In this regard, the students gave the feedback that after a first, exhausting run, the *"hardest part had been overcome"* and that proceeding the tasks would become easier with each further chemical reaction to be worked on. They went on to say that *"it [the material] shows you exactly what you can and cannot do of all this"*, which was seen as *"quite disillusioning"*. However, it was also concluded that working on the material would make one not only to learn about chemical thermodynamics, but also to recall numerous basic chemical concepts. One student even stated that they were *"a bit proud"* when having completed the task successfully, because *"then you know that you understand a large part of chemistry"*.

Also, many of the student teachers gave the feedback that they would not have dealt with chemical thermodynamics in such a fundamental way until then. This was highlighted by the following statement made by one student teacher in their 6th semester of Bachelor degree: *"Until just now, I didn't know that you can really derive it [enthalpy change] from the properties of reactants and products."*

2.5 Re-design

In the course of several iterative DBR-cycles, a variety of revisions was made. While the basic structure of the material (four tasks addressing introduction, enthalpy changes, entropy changes, and changes in Gibbs free energy) essentially remained the same throughout the entire project, a number of changes were made in the structure, wording, and formatting of the tasks as well as the information included in the supportive material. In addition, the worked-out examples were even more closely aligned with the wording of the instructional sheet. In the following, we provide an insight into the reflection and re-design processes by going into some selected revisions.

It became apparent in several occasions that students had particular difficulty with tasks including a varying "number of possible answers", e.g., gradations in bond polarity and bond stability, or energy content of reactants and products (Task 2). Therefore, in initial revisions it was explicitly stated where to differentiate into gradations. In the further course, we included the graduated options in the supportive material. Finally, the task was formulated in a way that there is one "correct answer", which is additionally concretised by answer options in some parts. These options are directly integrated into the instruction and are visually highlighted (formatting in boxes) to be recognisable to the students (see Figure 2 in Section 3.1). Furthermore, the entire material was revised

Task 2: Making Qualitative Predictions about the Change in Enthalpy (ΔH_{rxn})

- 2a) Determine the **type of chemical bonding** in the reactants and products.
For covalent bonding, also state whether the bonds are **nonpolar**, **slightly polar**, or **highly polar**.
- 2b) Make a statement about the **stability of the bonds** in the reactants and products. Consider the predominant bonding properties (including bond polarities of covalent bonds) and make gradations between **little stable**, **moderately stable**, **stable**, **very stable**, and **extremely stable**.
- 2c) Make a statement about the **energy content** of the reactants and products. Consider the stability of bonds and make gradations between **low energy content**, **moderate energy content**, and **high energy content**.

Figure 2: Excerpt from the step-by-step instruction of task 2.

to make the wording more “relative” (e.g., the change in entropy rather promotes the spontaneity of the reaction) so that it becomes clear that the analyses provide trends and qualitative statements and do not allow any definitive statements or concrete values.

Concerning the chemical concepts and practices, students’ needs and difficulties were greatly diverse – also caused by absences in former lessons. However, some problems occurred frequently, e.g., with generating the chemical formula of element molecules (e.g., N and Cl instead of N_2 and Cl_2). For this reason, we added an information card (listing those elements usually occurring as diatomic molecules) to the supportive material.

Another problem arose with determining the polarities of covalent bonds. Several students conflated the polarity of single bonds with the polarity of the entire molecule. This was addressed in one hint added to the supportive material. Further, students struggled with the inverse relationship between stability and energy, which might be caused by alternative conceptions that are well known from the literature (e.g., Bain et al., 2014; see Section 2.1.5). For this reason, analysing this challenging relationship was split into three separate subtasks (bonding type, stability, energy content). Moreover, we included both information and hint cards that explicitly address this inverse relationship, e.g., by providing guiding principles, such as “The more stable the bond, the lower the energy content of the compound”.

Overall, it turned out that applying many chemical concepts and practices at the same time was completely overwhelming for some students. Since the material aims at determining the spontaneity of chemical reactions (and not, e.g., at generating chemical formulas or formulating and balancing reaction equations), providing not only conceptual, but also procedural and strategic scaffolds (e.g., Belland et al., 2017; Hmelo-Silver et al., 2007; van de Pol et al., 2010) has proven to be indispensable. For this purpose, we added references to the textbook (e.g., for chemical formulas) and an online tool (“online balancer”) for balancing chemical equations. Additionally, the supportive material was assigned to the respective tasks and categorised into two groups: information (including definitions, concepts, relationships) and hints (addressing problem solving strategies or further measures of support). After the material had become increasingly extensive, differentiating between information and hints made it easier for both students and teachers to orientate themselves through the supportive material.

3 Resulting instructional material

The instructional material developed in the framework of the DBR-project consists of the instruction sheet, two worked-out examples and additional supportive material including information cards and hints. The material in its final form is presented in overview herein after and attached in full as Supplementary Material.

3.1 Task & instruction sheet

The instructional material is structured in such a way that there is a general work order consisting of four tasks, which then have to be processed for different chemical reactions (e.g., reaction of sodium and chlorine to sodium chloride, oxyhydrogen reaction, decomposition of silver(I) oxide, burning of ethanol).

Task 1 constitutes the **introduction** to the respective chemical reaction. Students are asked to reflect on their general knowledge on the specific reaction and where it appears, write down names and formula of reactants and products and formulate the stoichiometrically balanced chemical equation. If students have no prior knowledge on the respective reaction, they can still proceed with the task.

In task 2, students make qualitative predictions about the **enthalpy change** that occurs during the reaction. For this purpose, they follow a step-by-step instruction and process the following aspects successively (see Figure 2): (a) determining bond types, (b) estimating stability of the bonds, (c) estimating the energy content of the compound, (d) applying the values to conclude ΔH_{rxn} , (e) deciding whether the reaction is exothermic, endothermic or no statement is possible and (f) hypothesising whether the change in enthalpy would rather promote or hinder the spontaneity of the reaction.

Task 3 deals with **entropy change** and is designed analogously to task 2. In the first step, students are asked to analyse in what state of matter the reactants and products exist under normal temperature (25 °C/298,15 K) and pressure (1 atm) and complete the chemical equation by adding the indices for the states of matter (s, l, g). Then, they analyse the reactants and products regarding the following two aspects: changes in lattice structures and gaseous compounds (3b) and number of gaseous molecules (3c). Considering results from these analyses, students decide whether the entropy is increasing or decreasing throughout the reaction and qualitatively estimate the value of ΔS_{rxn} . Analogous to task 2, students finally make a statement concerning the consequence of ΔS_{rxn} for the spontaneity of the chemical reaction.

In task 4, students have to combine their results from tasks 2 and 3 and consider the relationship described by the Gibbs-Helmholtz-Equation. For this purpose, they are supposed to recollect how both enthalpy change and entropy change affect the spontaneity of the reaction. Substituting the estimated values for ΔH_{rxn} and ΔS_{rxn} into the Gibbs-Helmholtz-Equation, students are asked to make a qualitative prediction about the **change in Gibbs free energy** (ΔG_{rxn}) and determine the potential impact of the temperature. Finally, it has to be concluded whether the analysed reaction would be spontaneous or nonspontaneous, or if the spontaneity is influenced significantly by the temperature.

3.2 Worked-out examples & supportive material

Each task is supported by information cards and hints for processing the task. The information cards address aspects that should have already been covered in previous lessons, such as types of chemical bonding or the definition of exothermic and endothermic reactions. Students who do not remember this information can use the cards to recall it. The hint cards include concrete solution strategies to proceed with some of the tasks or indicate further measures of support. Figure 3 shows an example of an information and hint card each.

In addition, worked-out examples for each reaction are provided to the students. For one thing, these examples can be used to check the results and for another thing, they can serve as guide for working on the tasks. An excerpt from the worked-out example for reaction 3, the decomposition of silver(I) oxide, can be found in Figure 4, two complete worked-out examples (R3 – thermal decomposition of silver(I) oxide and R4 – combustion of ethanol) can be found in the Supplementary Material.

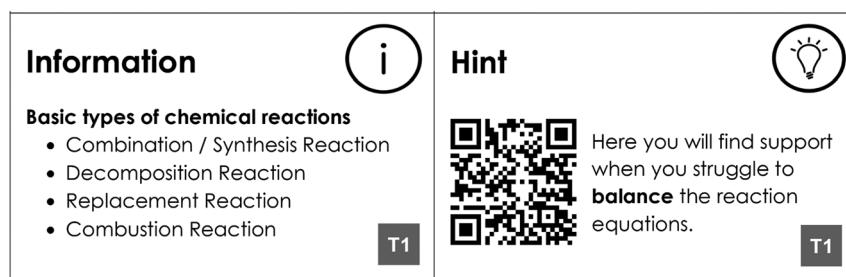


Figure 3: Examples for information and hint cards.

1c	$2 \text{Ag}_2\text{O}^{2-}$	\rightarrow	4 Ag	+	O_2
2a	ionic bonding		metallic bonding		covalent bonding nonpolar bonds
2b	extremely stable		little stable		moderately stable
2c	low energy content		high energy content		moderate energy content
2d	The stability of bonds decreases during the reaction. The energy content of compounds increases during the reaction. The enthalpy increases during the reaction; $\Delta H_{rxn} > 0$.				

Figure 4: Excerpt from the worked-out example of R3 – decomposition of silver(I) oxide.

4 Discussion

In the framework of a DBR project (DBR Collective, 2003), we developed “unmathematical” instructional material that supports students in determining the spontaneity of chemical reactions. Following a step-by-step problem-solving process, changes in enthalpy and entropy can be estimated by analysing the properties of reactants and products. Interpreting these with regard to changes in Gibbs free energy allows students finally to make qualitative predictions about the spontaneity of chemical reactions. The material was developed according to the DBR cycle starting from a problem analysis. After having implemented and revised the material for several times, material consisting of an instruction sheet (with four tasks), two worked-out examples and supportive material including cards with information and hints resulted.

In order to generate material that is adequate for upper secondary chemistry education whilst at the same time allowing for statements regarding the spontaneity of chemical reactions, various didactical reductions and simplifications were necessary (see Section 2.2). For this reason, the material is limited concerning the chemical reactions it is applicable or adequate for (e.g., there are limitations for reactions in which other than volume change work is done or reactions in which both reactants and products are present in the initial state of the reaction) and the results – that are even qualitative statements and tendencies.

Implementing the material in more than ten upper secondary chemistry classes and four chemistry teacher education courses proved it to be both appropriate and applicable. The students worked in a self-responsible and task-oriented manner using the instruction sheet, the worked-out examples as well as the supportive material. Not only the lesson observations, but also the use of similar tasks in assessments showed that students were able to make qualitative statements about the spontaneity of chemical reactions after having worked with the developed material. However, it must be noted that even material of high quality is at risk of not being effective in practice as teachers are not aware of its purpose, structure and didactical approaches (Heck et al., 2012; Roth McDuffie et al., 2018). We strived to counteract this by describing not only the chemical concepts (Sections 2.1.1 to 2.1.4) and underlying didactical considerations (Sections 2.1.5 and 2.2) this material is based on, but also by providing insights into experience gained from implementation (Section 2.4).

The experiences we made in the course of this DBR project did not only contribute to the further development of the material, but also influenced our teaching of chemical thermodynamics in general. Observing students when they were working on the tasks provided us with feedback on aspects on basic chemistry we should emphasise more or could put less attention on in our future teaching. Implementing the material, it became apparent that especially knowledge about the state of matter and the bonding properties are crucial for students’ understanding of chemical thermodynamics. In particular, it turned out that the inverse relations (e.g., energy vs. stability) cannot be discussed often enough.

Despite several iterative cycles of implementation and revision, it must be admitted that the material has not yet undergone a systematic empirical evaluation in the sense of a pre-post-test investigation, which can provide information about students’ conceptual understanding and growth. However, the material has proven itself in teaching practice in that the students were able to work with the material independently and according to their individual prior knowledge and needs. It was shown that the material allows for differentiation and adaption and enables students to successfully complete tasks in the field of chemical thermodynamics.

4.1 Recommendations for implementation

To allow students to work through all tasks for at least one of the listed reactions, a minimum of two lessons should be assessed. Depending on content focus, the material can be processed in various ways. One possibility is to complete tasks 1 and 2 for several chemical reactions and then work on tasks 3 and 4. In so doing, emphasis is put on the basic principle of enthalpy change and exothermic and endothermic reactions (see Sections 2.1.1 and 2.1.2). Changes in entropy and Gibbs free enthalpy (see Sections 2.1.1 and 2.1.2) can then be seen as optional elaboration for the purpose of content deepening or extension. Another approach is to work through all tasks in

one go, explicating the connection between the change of Gibbs free energy and the changes of enthalpy and entropy. Of course, students can work on the tasks also in different sequences depending on their prior knowledge, skills and needs and the content level they aim for. However, to understand how a statement about the spontaneity of a chemical reaction can be made from analysing properties of reactants and products, students should work through task 1 to task 4 at least once. Alternatively, they can reflect on the worked-out examples and comprehend the applied strategies.

To enable students to work successfully on the material, the underlying theoretical concepts (chemical reactions, reaction equations, models of chemical bonding, energy of bonds, bond polarity etc.) need to be addressed in preceding chemistry lessons. For one thing, students must be basically familiar with these concepts and for another thing, they also need to be used to applying their knowledge and skills independently. Only then is it possible for the teacher to support students by providing scaffolding adapted to students' individual needs. Depending on prior experience with scaffolds, students may rely on these scaffolds independently or the teacher may provide them as needed. Some of the scaffolds (in particular hints for tasks 1 and 4) can also be applied in the sense of incremental scaffolds (Schmidt-Weigand et al., 2009).

Beyond that, it is also recommended to make clear what simplifications and limitations have been made in the material and to discuss what impact these simplifications and limitations have on the validity of the results. In any case, it must be clear that the results are to be understood only as tendencies and qualitative predictions rather than as exact results.

5 Conclusions

In this article, we presented instructional material that relates on basic chemical knowledge and guides students step-by-step through the complex process of determining the spontaneity of chemical reactions. By analysing the properties of reactants and products, students can draw conclusions about the compounds' stability, energy content, and number of microstates. Comparing the reactants and products, qualitative statements about the spontaneity of a chemical reaction can be made without performing mathematical calculations or referring to physical concepts, such as work. To address students' diverse prior knowledge, skills and needs and to foster their independent work, additional supportive material (information and hint cards, worked-out examples) is provided.

The material was developed in the course of a Design-Based Research project and was implemented for several times in both upper secondary chemistry classes and courses of chemistry teacher education. The "unmathematical" material has proved to be proper for students' independent work and individual working pace. It was evident from both lesson observations and integrated assessment tasks that students were, for the most part, able to determine the spontaneity of chemical reactions after having worked with the material. Students' informal feedback also indicated that the material supported them in connecting thermodynamics to their prior chemical knowledge. Further, the step-by-step instruction and supportive material enabled students to successfully complete the tasks and experience themselves as competent to complete tasks in the field of chemical thermodynamics. However, the material is yet to be proved systematically on a larger scale, evaluating its efficacy by assessing students' conceptual understanding and problem solving strategies.

Acknowledgments: We are thankful to the students who worked on the material and provided feedback for its improvement.

Research ethics: Not applicable.

Author contributions: The authors have accepted responsibility for the entire content of this manuscript and approved its submission.

Competing interests: The authors state no conflict of interest.

Research funding: None declared.

Data availability: Not applicable.

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Supplementary Material: This article contains supplementary material (<https://doi.org/10.1515/cti-2022-0046>).