Differentiating the Physical Optimum from the Exergetic Evaluation of a Methane Combustion Process

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Abstract: Combustion processes continue to be essential for the energy supply sector. A reliable energetic evaluation of these processes is crucial, particularly since the pollutants resulting from combustion have a significant impact on global warming. This work evaluates a combustion using the exergetic evaluation and the Physical Optimum (PhO) as it is described in VDI-Guideline 4663. Differences between PhO and exergy are investigated, allowing a distinct differentiation and examining the PhO's added value in combustion analysis. Based on the evaluation of a simulated methane combustion, this paper shows that the PhO-Factor may be used to evaluate combustion processes. However, it shows that the PhO of a combustion process is a simplification of this fuels exergy and does not provide advantages to the exergy evaluation. Nevertheless, an adaption of the PhO is not carried out in the context of this work since the minimal deviation of the simulated energy indicators currently cannot justify an adaptation. In addition, proposed adjustments of the reference value (PhO) could lead to the definition limits of the PhO-Factor being exceeded. The paper introduces the indirect PhO-Factor for a targeted process optimization. It is shown that in this case, the indirect PhO-Factor closely corresponds to the exergy efficiency.

Keywords: fuel; combustion; process evaluation; physical optimum; exergy; exergy efficiency

1. Introduction

In his book *Technische Verbrennung*, Warnatz [1] describes combustion as the oldest technology of mankind [1]. Though it has been used for more than a million years, combustion technology is still of major relevance today. Confirming this statement, Figure 1 shows the composition of Germany's gross electricity production in 2010 and 2019.
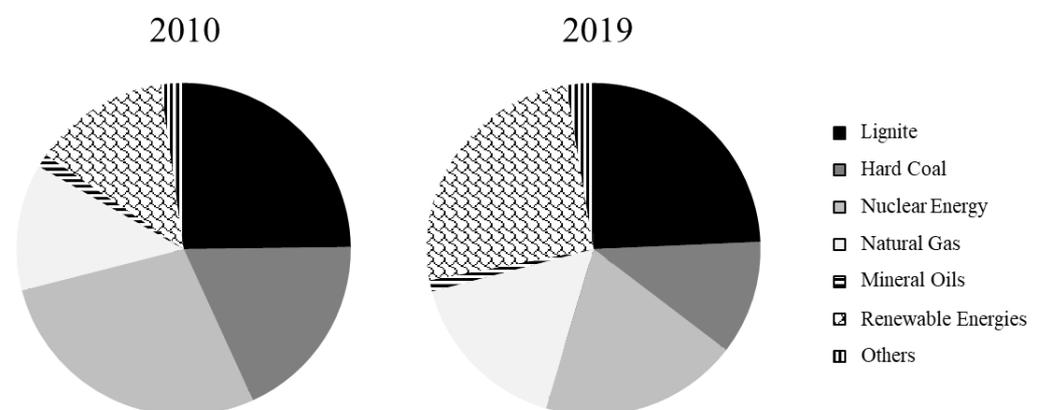
Figure 1. Share of energy sources in Germany's gross electricity production in 2010 and 2019 [2].

While in 2010 only 13% of the gross electricity production was supplied by renewable energy sources, at the end of 2019 25% were generated by primary energy sources such as wind- and hydro power, biomass and photovoltaic. Though there is a recognizable trend towards renewable energies, the current dominance of fossil energy sources is apparent, since the gross electricity production at the end of 2019 was composed as follows: 24% lignite, 9% hard coal, 19% nuclear energy, 16% natural gas and 1% mineral oil [2].

The use of fossil primary energies is based mainly on the combustion process [3]. This releases carbon dioxide, which promotes global warming [4]. In addition, the extraction of fossil energy sources exceeds its new generation, which results in an exhaustion of natural resources [5]. Therefore, an inefficient use of these energy sources must be avoided urgently. Combustion processes are not explicitly used for burning fossil fuels. Sustainable technologies also employ combustion processes to increase efficiency or optimize operation conditions [6–8]. In addition, to achieve sustainable combustion processes, the substitution of fossil fuels by biomass-based products is a trend, which cannot be denied [9]. Hence, the efficiency evaluation of technical combustions will remain relevant in the future.

The Physical Optimum (PhO) is described in VDI-Guideline 4663 [10] whose application to industrial processes was verified within the framework of the WIPANO Project *greoKEMS*, which was funded by the German Federal Ministry for Economic Affairs and Energy. The PhO is the foundation of an energy index (PhO-Factor), which enables a targeted optimization of inefficient processes [11,12]. Therefore, it seems particularly suitable for the evaluation and optimization of combustion processes. This guideline, but also the PhO as a limit value, are based on a total of three dissertations by Volta [13], Keichel [14] and Beckmann [15]. Within the framework of these dissertations, extensive literature research is carried out, which evaluates common methods for the efficiency evaluation of processes. However, a direct comparison between the process evaluation based on the PhO and the exergy is not implemented even though the usefulness and above all the added value of the PhO compared to the exergetic process evaluation is not undisputed and regularly the topic of discussion. This is exemplified by the evaluation of an induction furnace based on exergy and PhO by Wenzel [16]. Since exergy describes the maximum working capacity and thus the quality of energy, there are certain parallels to PhO [17]. It has not yet been studied to what extent the PhO of a combustion differs from the exergy of a fuel. This reflection is essential, since the PhO and VDI Guideline 4663 will only be established as tools for evaluating the efficiency of processes, if they differentiate from exergy and provide advantages compared to the exergy evaluation.

Therefore, the objective of this paper is to compare the exergy and the PhO of a fossil fuel and to analyze differences and similarities. For this purpose, a proven model by Baehr and Kabelac [18] is used to evaluate a fossil fuel's exergy. The definition of the PhO is based on VDI Guideline 4663, respectively on existing models. A comparison of the fossil fuel's exergy with the PhO enables a verification and if necessary, an adjustment. This work also defines the indirect PhO-Factor of a technical combustion, which can be used to realize targeted process optimization.

2. Materials and Methods

The main target of this paper is to distinguish between PhO and the exergy of a fossil fuel. Further models for the efficiency evaluation of processes are not considered for the time being. A complete distinction is only reasonable if the PhO differs significantly from the exergetic evaluation and can generate advantages compared to it. The methods and models used are presented and validated in Sections 2.1–2.3.

2.1. Exergy of a Fossil Fuel

A technical combustion process enables the chemical energy of a fuel to be used as enthalpy or as heat from the exhaust gas. The exergy of a fossil fuel defines the energy portion that is completely convertible into the exergy of other forms of energy in a reversible

process [18]. Thus, the exergy of a fuel can be used to describe the maximum available work of a fuel.

Various models are available to describe the exergy of a fossil fuel. This paper only considers a method of Baehr and Kabelac [18]. Their method allows the determination of fuel exergy even for chemically undefined fuels. Thus, it provides the potential of a general evaluation method. The chemical exergy, which is considered in a fuel's exergy, is always based on an environment model, which must be described exactly. In the ambient state, the exergy will usually be set to zero. To describe the environment, a common model is used in which the air is in thermodynamic equilibrium with liquid water at ambient temperature and ambient pressure. To achieve not only thermomechanical and chemical equilibrium but also material equilibrium at ambient conditions, the model assumes that the air is saturated with water vapor and the water is saturated with air. Thus, a complete thermodynamic equilibrium is achieved at ambient conditions. This allows the calculation of the maximum working capacity of the fuel [19].

If the temperature and pressure of a fuel correspond to the values of the environment, it is in thermal and mechanical equilibrium. However, there may still be a chemical imbalance and thus chemical exergy available. If the fuel is transformed into the surrounding components by a chemical reversible reaction, which are in turn brought into a state without exergy, the chemical exergy can be converted into usable work [18]. In the environmental model needed to determine the exergy, the standard values for temperature and pressure are valid $T_{env} = T_0 = 298.15$ K and $p_{env} = p_0 = 100$ kPa. The composition of the environment is defined based on the molar fractions $x_{N_2,env} = 0.75608$, $x_{O_2,env} = 0.20284$, $x_{H_2O,env} = 0.03171$, $x_{Ar,env} = 0.009096$, and $x_{CO_2,env} = 0.00031$. However, the environmental model used here has deficits in the exergetic evaluation of sulphur-containing fuels. Since sulfur is not considered in the environment, the chemical potential of a sulfur-containing exhaust gas would result in an infinite exergy of the fuel. Adjusting the environment model eliminates this limitation [19].

The procedure for calculating the fuel's exergy according to Baehr and Kabelac is based on an isothermal-isobaric reaction chamber, which is shown in Figure 2 [18]. The figure shows that the reaction products leave the reaction chamber at ambient pressure and temperature. Both the educts and the products are supplied and discharged separately to and from the reaction chamber. This eliminates mixing effects, which would lead to entropy production and thus result in additional exergy losses.
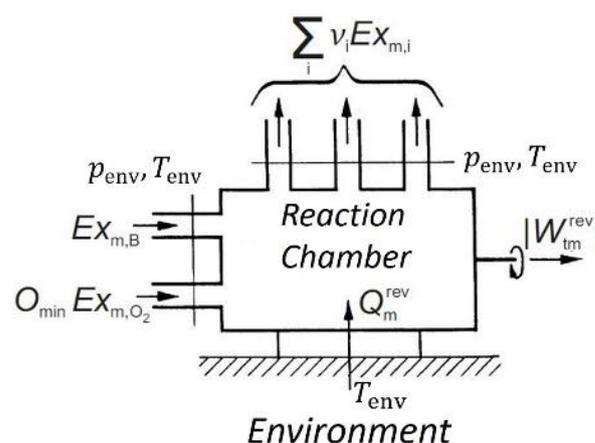
Figure 2. Balance of exergy defining the exergy of a fossil fuel (following [18]).

Heat transfer with the environment only takes place at ambient temperature. Thus, the transferred heat Q_m^{rev} is pure anergy. Therefore, it is not considered in the following exergy balance, which is valid for a reversible reaction. In this reaction, Ex_m describes the

molar exergy of educts and products, O_{min} the minimum oxygen demand, $W_{t,m}^{rev}$ the molar reversible work and ν_i the stoichiometric numbers of the separate products.

$$Ex_{m,B}(T_{env},\ p_{env}) + O_{min}Ex_{m,O_2}(T_{env}, p_{env}) + W_{t,m}^{rev} = \sum \nu_i Ex_{m,i}(T_{env}, p_{env}) \quad (1)$$

$$\Longleftrightarrow Ex_{m,B}(T_{env},\ p_{env}) = \sum \nu_i Ex_{m,i}(T_{env}, p_{env}) - O_{min}Ex_{m,O_2}(T_{env}, p_{env}) - W_{t,m}^{rev} \quad (2)$$

The molar reversible work is described by the difference of the molar Gibbs free energy $\Delta^R G_m$. This in turn is defined by the molar higher heating value $H_{s,m}$ and the molar reaction entropy $\Delta^R S_m$. The following definition applies to the molar reversible work of an isothermal-isobaric combustion [18]:

$$W_{t,m}^{rev} = \Delta^R G_m(T_{env}, p_{env}) = -H_{s,m}(T_{env}) - T_{env}\Delta^R S_m(T_{env}, p_{env}) \quad (3)$$

There are two basic areas of definition. If the Gibbs free enthalpy is $\Delta^R G_m(T, p) < 0$, not only heat but also work can be obtained from the reaction. In this case $W_{t,m}^{rev}$ describes the maximum amount of work or power that can be released from a chemical reaction. The reaction proceeds spontaneously. If the Gibbs free enthalpy is $\Delta^R G_m(T, p) > 0$, then $W_{t,m}^{rev}$ describes the maximum amount of work or power that must be added to the reaction to ensure its proceeding. In this case, a spontaneous proceeding of the reaction is impossible [18]. In conclusion, the exergy of a fuel can be defined:

$$Ex_{m,B}(T_{env},\ p_{env}) = H_{s,m}(T_{env}) + T_{env}\Delta^R S_m(T_{env}, p_{env}) + \sum \nu_i Ex_{m,i}(T_{env}, p_{env}) - O_{min}Ex_{m,O_2}(T_{env}, p_{env}) \quad (4)$$

The equation is confirmed by procedures of Lucas [19] and Schmidt [20]. The exergy of the fuel is primarily determined by the reversible work, which in turn is significantly dependent on the molar higher heating value $H_{s,m}$. The exergy of the molar reaction entropy $T_{env}\Delta^R S_m(T_{env}, p_{env})$ as well as the exergy of the reaction products $\sum \nu_i Ex_{m,i}(T_{env}, p_{env})$ and of the oxygen $O_{min}Ex_{m,O_2}(T_{env}, p_{env})$ usually contribute with a few percent to the fuel's exergy. The choice of the environment only influences the exergy of the reaction products and the oxygen [18]. Therefore, its impact on the overall exergy of the fuel is rather low.

In general, the fuel exergy of gaseous fuels reaches about 95% and that of liquid fuels about 98% of the higher heating value [18]. The deviation of gaseous fuels can be confirmed within the scope of this paper. The calculations performed will show that the fuel exergy of methane amounts to approximately 93% of H_{s,m,CH_4}. However, theoretically the fuel exergy may exceed the higher heating value. For example, the exergy of carbon as a fossil fuel is about 3% above the higher heating value, while the exergy of sulfur as a fossil fuel is as much as 79% above the higher heating value [18]. This is due to the molar reaction entropy $\Delta^R S_m(T_{env}, p_{env})$. If a positive value results from the molar reaction entropy, heat is transferred and the reversible reaction work $W_{t,m}^{rev}$ exceeds $H_{s,m}$. However, this process only takes place if the ambient temperature exceeds the reaction temperature or if additional work is applied to ensure heat transport. In the context of this work, both criteria can be neglected. Thus, the fuel's exergy does not exceed the calorific value of a fuel.

2.2. The Physical Optimum

Volta first defined the PhO in a dissertation [13]. It is described as an optimal reference process that includes all existing scientific laws. By referring a real process to an optimal process, the PhO-Factor is defined as a performance indicator. In his dissertation, Keichel [14] extended the methodology of Volta. Among other things, a consuming and demanding perspective are introduced. Depending on the process, one of the perspectives is chosen. According to Keichel, processes that provide energy are evaluated from the consuming perspective. It compares the real benefit of a process V to the effort under physically optimal conditions V_{PhO}. The effort under physically optimal conditions describes

the maximum available energy in the input of a process. The result of this comparison is an energy index, the PhO-Factor $F_{\text{PhO}}^{\text{V}}$. It is [14]:

$$F_{\text{PhO}}^{\text{V}} = \frac{\text{Benefit}}{\text{Effort(PhO)}} = \frac{V}{V_{\text{PhO}}} \leq 1 \tag{5}$$

One of the major advantages of the PhO-Factor compared to conventional energy indicators is that the PhO is defined by scientific laws and thus, in contrast to a reference to the state of technology, enables the definition of a permanent and time-independent indicator. Furthermore, the PhO-Factor directly indicates the optimization potential of a technical process and can be applied to complex systems, such as Power-to-X processes, where the evaluation using conventional performance indicators shows deficits [11]. To conclude, it is essential that the PhO is determined exactly.

Further on, a combustion process is evaluated based on the PhO. For this purpose, the current definition of the PhO-Factor of a combustion process is specified. Subsequently, it is verified whether specified equations can still be considered as valid, considering previous sections. Primarily the available energy under physically optimal conditions V_{PhO} is of interest. The factor is adjusted if necessary.

2.3. The Physical Optimum of a Technical Combustion Process

According to [14], a combustion is a technical process that provides energy and is therefore evaluated based on the consuming perspective. Within the framework of various student projects, the PhO-Factor of a combustion process was developed at the Hamburg University of Applied Sciences. If the combustion process itself is considered, the factor was consistently defined as shown in Equation (6). Here, Q_{mess} describes the heat in real terms resulting from the combustion, V_B the volume of burned fuel, ρ_B the fuel's density, $v_{RG,tr,min}$ the dry exhaust gas' volume related to the volume of the supplied fuel, and $c_{p,RG,tr,min}$ the exhaust gas' specific heat capacity.

$$F_{\text{PhO}}^{\text{V}} = \frac{V}{V_{\text{PhO}}} = \frac{Q_{mess}}{(V_B\rho_B)\left[H_{\text{s,B}} - v_{\text{RG,tr,min}}c_{\text{p,RG,tr,min}}T_{\text{env}}\right]} \tag{6}$$

The numerator of the PhO-Factor is based on the available heat in real terms resulting from the combustion, which must be defined by using a reference value, such as the environment, since heat is only present due to a temperature difference between two systems. The value Q_{Mess} is real and therefore contains losses. It is defined by the mass flow of the exhaust gas, its specific heat capacity and the temperature difference to the environment. Thus Q_{Mess} is an energy and does not correspond to the exergy of heat. At this point it does not seem necessary to adjust the numerator of the PhO-Factor.

The denominator of the PhO-Factor, which is the PhO, is primarily influenced by the higher heating value of the fuel $H_{s,B}$. However, unavoidable losses must be taken into account, which will always occur during technical combustion, regardless of the combustion process selected. These unavoidable losses reduce the maximum available energy and thus determine the optimization potential of the process. In this case, the unavoidable losses are defined by the energy portion of the dry exhaust gas that cannot be used due to the ambient temperature. Since $v_{RG,tr,min}$ is the dry exhaust gas' volume related to the volume of the supplied fuel, it is a dimensionless value.

In addition, the thermal energy supplied to the combustion chamber by the air and the fuel must be taken into account if the temperatures differ from the ambient temperature. In this case the PhO-Factor is extended according to Equation (7). μ_L describes the mass ratio of combustion air and fuel and $c_{p,L}$ the specific heat capacity of the combustion air.

$$F_{\text{PhO}}^{\text{V}} = \frac{Q_{Mess}}{m_{\text{B}}\left[H_{\text{s,B}} - v_{\text{RG,tr,min}}c_{\text{p,RG,tr,min}}T_{\text{env}} + c_{\text{p,B}}(T_{\text{B}} - T_{\text{env}}) + \mu_{\text{L}}c_{\text{p,L}}(T_{\text{L}} - T_{\text{env}})\right]} \tag{7}$$

3. Results

3.1. Differentiating the Physical Optimum of a Combustion Process from the Exergy of a Fossil Fuel

To allow a differentiation between the PhO and the exergy of a fossil fuel, a Matlab script was developed to simulate an isobaric combustion process of methane. Here methane is burned with dry air. To specify the mole fractions of the air, it is assumed that it consists only of nitrogen and oxygen: $x_{N_2} = 0.79$ and $x_{O_2} = 0.21$.

The air–fuel equivalence ratio λ, which sets the actual air mass in relation to the minimum air mass that is theoretically required for a stoichiometrical combustion, is initially $\lambda = \frac{m_L}{m_{L,stoich}} = 1$. The following reaction equation results:

$$CH_4 + 2O_2 + 2\frac{79}{21}N_2 \rightarrow CO_2 + 2H_2O + 2\frac{79}{21}N_2 \tag{8}$$
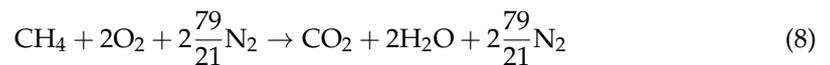
Material values are either calculated from [18] or generated from the NASA text file thermo.inp [21]. The exergetic evaluation was carried out using the method already described by Baehr and Kabelac [18]. The results of the simulation, that determine the maximum available energy of methane, are shown in Figure 3.
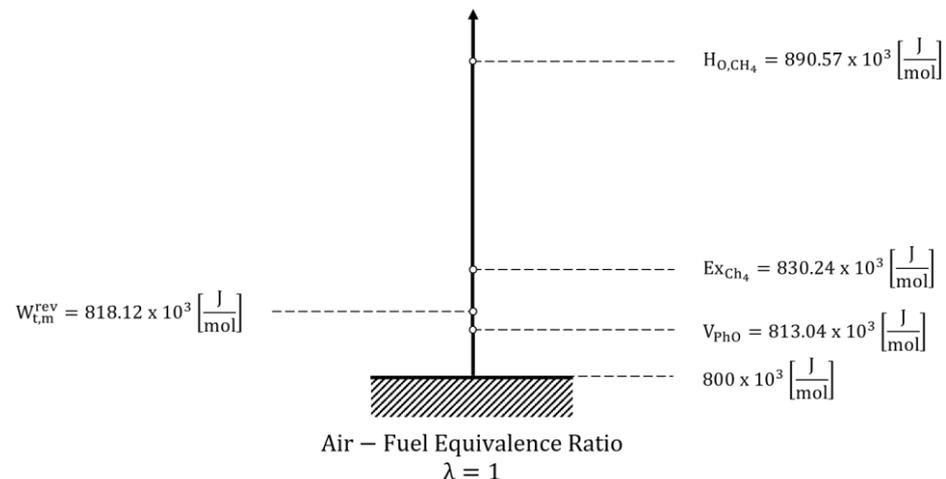
Figure 3. Results of a simulated combustion of methane with dry air and an air–fuel equivalence ratio of $\lambda = 1$.

Figure 3 shows that the higher heating value of a fuel alone cannot be used to define the maximum available energy. The exergy of methane is $Ex_{CH_4} = 830.24 \text{ kJ·mol}^{-1}$ and therefore accounts to only 93.23% of the higher heating value of methane $\left(H_{s,CH_4} = 890.57 \text{ kJ·mol}^{-1} \right)$. If the higher heating value was set as the maximum available energy, the resulting PhO-Factor would indicate a greater potential for optimization than actually viable. Particularly in case of energetically advanced plants, this deviation could lead to high investment costs, which will not improve the process. The influence will alternate depending on the type of fuel. In case of methane, an unrealizable optimization potential of 6.77% would be present.

Comparing the fuel's exergy with the PhO $\left(V_{PhO} = 813.04 \text{ kJ·mol}^{-1} \right)$ shows that they deviate only by 2.07%. Due to the minimal deviation of the values, parallels between the fuel's exergy and the PhO must be analyzed. Since both procedures aim to define the maximum available energy of the fuel, the cause of the deviation of the values needs to be examined and evaluated.

Both in the PhO and in the exergetic evaluation, the higher heating value of the fuel is of great significance. The PhO only considers the higher heating value of the fuel and the thermal energy of the exhaust gas. The combustion air and the fuel are supplied at ambient temperature and therefore do not add usable energy to the system. The maximum

of the PhO is initially defined by the energy available due to the higher heating value of the fuel. The dry exhaust gas resulting from the combustion is cooled down to the ambient temperature. Thus, the available energy of the exhaust gas is limited by ambient conditions (see Equation (6)).

In addition, in the definition of the exergy of the fuel a relation to the ambient temperature is established, since all components are both supplied and discharged at ambient pressure and temperature (see Equation (4)).

The exergy of the fuel is significantly limited by the reaction entropy, which results from the chemical conversion of the fuel. If only the higher heating value and the reaction entropy are considered in the exergetic evaluation, this value corresponds to the molar reversible work of the fuel $W_{t,m}^{rev}$. It describes the available energy only in terms of the higher heating value, the chemical conversion and the release of the products at ambient conditions. Thus, the same effects are considered in the molar reversible work as for the PhO. A comparison between the PhO and the reversible work $\left(W_{t,m}^{rev} = 818.12 \text{ kJ·mol}^{-1} \right)$ confirms this assumption and shows that the quantities are approximately equal. They only differ by 0.62%. Therefore, it is possible to validate the slight deviation of the PhO from the exergetic evaluation and to recognize parallels between the meanings of these values. In both cases, a fuel is supplied with a higher heating value, which initially describes the maximum energy of the process. In both cases, this energy is reduced by discharging the products (exhaust gas) at ambient conditions.

The deviations between the exergy of a fuel and the PhO can be verified with the remaining terms of Equation (4). First, it is noticeable that in the exergetic evaluation a consideration of the term $\sum \nu_i Ex_{m,i}(T_{env}, p_{env})$ leads to an increase of the total available energy. This is explained by the chemical potential of the individual flows of the exhaust gas to the environment. The chemical potential can be used energetically and must be considered in this balance of exergy (see Figure 2).

Analogously, the chemical exergy of the supplied oxygen $O_{min} Ex_{m,O_2}(T_{env}, p_{env})$ reduces the available energy. The exergy of the nitrogen of the combustion air has no influence since it does not actively participate in the reaction and thus has the same chemical potential as educt and product. From these results different conclusions can be drawn. It should be explicitly pointed out that the following statements are currently based solely on the simulation of the combustion of methane.

Initially, it should be noted that an adjustment of the definition of the PhO-Factor for a combustion process is not mandatory at present. The deviation of only 2.07% between the PhO and the exergy of this fossil fuel cannot be regarded as critical and therefore does not justify an adjustment due to high deviation. This also applies to a comparison with the reversible work. In both cases, it cannot be assumed that a technical combustion process will be optimized up to this extent. In addition, an adjustment of the ratio will not be necessary due to a changing state of the technology.

Nevertheless, based on the previous comparison of the PhO with the exergy of methane, it must be noted that the current definition of the PhO does not define the total available energy of a combustion thermodynamically exactly. Due to the neglect of the available chemical energy, the PhO is a simplification and accordingly has no direct advantages over the fuel's exergy. Using the exergy, the available energy of a fuel is completely described and thus offers the possibility to directly define an optimization potential. Even with an air–fuel equivalence ratio $\lambda \neq 1$, the PhO cannot provide an advantage to the exergy of a fossil fuel, since the exergy is always related to the minimum required air as well. In order to make a general statement about whether the deviations between PhO and the fossil fuel exergy are always negligible, further simulations of the combustion of different fuels must be conducted. If there are significant deviations, the PhO cannot be used to define the maximum available energy of a fuel.

A possible approach in adjusting the PhO-Factor might be to replace the PhO with the exergy of the fuel. The PhO would be a value that results from the system balance and thus cannot be determined subjectively. Furthermore, the energy available in the fuel would be

described completely and thermodynamically exactly. However, such an adjustment is not possible, since with a ratio of the heat available in the exhaust gas as the numerator and the fuel exergy as the denominator, the definition limits of the PhO-Factor (cf. Equation (5)) are not necessarily met. This can be clarified by means of an independent example, which is based on a calculation to evaluate the exergetic efficiency of a heat pump [20].

In Figure 4, a countercurrent heat transfer device is shown in which air transfers heat to another mass flow. It is assumed that the specific heat capacity $c_{p,L} = 1004 \text{ J·kg}^{-1}\text{·K}^{-1}$, the specific gas constant of air $R_L = 287 \text{ J·kg}^{-1}\text{·K}^{-1}$ and the air mass flow $\dot{m}_L = 14.1 \text{ kg·s}^{-1}$ are constant values. The ambient temperature is $T_{env} = 273.15 \text{ K}$.
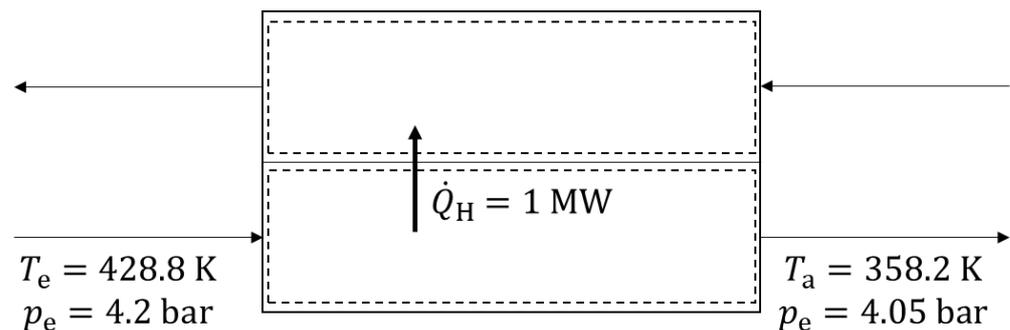
Figure 4. Schematic diagram of a heat exchanger and its operating parameters.

The transferred heat flux can be calculated by applying the first law of thermodynamics. Potential and kinetic energy changes are neglected.

$$\dot{Q}_H = \dot{m}_L c_{p,\,L}(T_e - T_a) \approx 1 \text{ MW} \tag{9}$$

In addition, the exergy flow $\dot{Ex}_H$ shall be calculated, which is emitted by the air mass flow. This exergy flow can be determined by the change in exergy between the inlet and outlet states, where changes in potential and kinetic energy are also negligible:

$$\dot{Ex}_H = \dot{m}_L(ex_e - ex_a) \tag{10}$$

$$\dot{Ex}_H = \dot{m}_L\left[c_{p,L}(T_e - T_a) - T_{env}\left[c_{p,L}ln\left(\frac{T_e}{T_a}\right) - Rln\left(\frac{p_e}{p_a}\right)\right]\right] = 344.17 \text{ kW} \tag{11}$$

Since the transmitted exergy flow $\dot{Ex}_H$ in contrast to the transmitted heat flow $\dot{Q}_{HH}$ takes irreversibilities into account, it only amounts to about 34% of the heat flow $\dot{Q}_{HH}$. Comparing the exergy and the energy is therefore not appropriate and cannot provide a valid statement in this case. This is transferable to the combustion process. An adjustment of the PhO-Factor, in which the PhO is described based on the fuel exergy, could lead to an exceeding of the definition limits, which additionally does not show any direct optimization potential:

$$F_{PhO}^V = \frac{Q_{Mess}}{Ex_B} \underset{>}{\overset{<}{=}} 1 \tag{12}$$

For compliance with the definition limits, a simultaneous adjustment of the denominator would be necessary. This would take the exhaust gas' exergy into account instead of Q_{Mess}. In this case, the PhO-Factor would be equal to the exergy efficiency.

3.2. The Indirect PhO-Factor of a Combustion Process

The PhO-Factor defined by Volta [13] and Keichel [14] shows the maximum optimization potential of a process. However, this factor does not reveal how losses occur in a process and how much energy is required to compensate these losses. The indirect PhO-Factor should, in analogy to the indirect efficiency, determine the share of individual

process losses in the total effort of energy [11,22]. This enables and significantly simplifies a targeted process optimization. The indirect PhO-Factor is developed based on the correlation between effort, benefit and losses [11]. This is the general approach:

$$\text{Effort} = \text{Benefit} + \text{Losses} \tag{13}$$

According to Equation (5), the PhO-Factor of the consuming perspective is defined by using the ratio of benefit and effort. If Equation (13) is solved according to the benefit and used in Equation (5), the indirect PhO-Factor of the consuming perspective results in [11].

$$F_{\text{PhO,ind}} = 1 - \frac{\sum \text{Losses}}{\text{Effort(PhO)}} = 1 - \frac{\sum L}{V_{\text{PhO}}} \leq 1 \tag{14}$$

The primary goal of the indirect PhO-Factor is the targeted process optimization. It may also be used to verify the assumptions and simplifications made.

In the following, the indirect PhO-Factor of a combustion process is determined. According to [18], the losses of adiabatic combustion can be determined by two essential factors: losses from adiabatic combustion and losses due to cooling of the combustion gas. The losses resulting from adiabatic combustion must be considered in any case. They result from the selected process of converting the primary energy. However, no further losses need to be taken into account in this analysis, as only the combustion itself is analyzed. Here, air and fuel are supplied to a reaction chamber. Exhaust gas leaves the reaction chamber with the adiabatic combustion temperature. Further process steps are not considered. For this purpose, the system boundary needs to be extended and additional components would be considered. For example, the heat of the exhaust gas could be used in a heat transfer device. The exhaust gas would emit heat; cool down and entropy would be generated. This would result in additional losses. The exhaust gas could also be released to the environment at a temperature higher than the ambient temperature. This would result in additional losses due to the unused thermal potential.

It must be noted that the losses of the indirect PhO-Factor are real values determined by measurement technology. Therefore, when calculating the indirect PhO-Factor, it is necessary to check whether the individual losses can be determined using measured values. If this is not possible, alternative methods for determining the losses must be found. In the following, the loss of adiabatic combustion is defined.

3.3. The Loss of Exergy in an Adiabatic Combustion

If the chemical energy of a fuel is converted into internal energy or heat by means of a technical combustion process, this process results in significant energy losses. However, it is not possible to measure the losses resulting from adiabatic combustion directly or to determine them only based on measured values. Therefore, a theoretical model must be used to describe the losses in order to consider them in the indirect PhO-Factor.

The calculation of the exergy loss from adiabatic combustion is based on an entropy balance, which is shown in Figure 5.

Considering an adiabatic combustion, the internal entropy $\dot{S}_{\text{irr}}$ is determined by the difference between exhaust gas entropy $\dot{S}_{\text{RG}}$ and fuel- $\dot{S}_{\text{B}}$ as well as air entropy $\dot{S}_{\text{L}}$. They may also be described using the molar flow rates $\dot{n}_{\text{i}}$ and the molar entropy $S_{\text{m},i}$.

$$\dot{S}_{\text{irr}} = \dot{S}_{\text{RG}} - \left(\dot{S}_{\text{B}} + \dot{S}_{L} \right) \tag{15}$$

$$\dot{S}_{\text{irr}} = \sum \dot{n}_{\text{RG},i} S_{\text{m,RG},i} - \left(\sum \dot{n}_{L,\,i} S_{\text{m,L},i} + \dot{n}_{\text{B}} S_{\text{m,B}} \right) \tag{16}$$
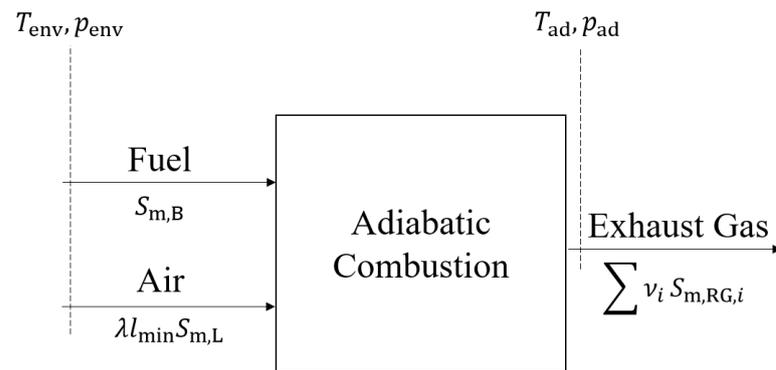
Figure 5. Entropy balancing to calculate the exergy loss of an adiabatic combustion.

Below, the internal entropy generation is divided and therefore referenced to the fuel's mass flow:

$$\dot{S}_{\mathrm{m,irr}} = \frac{\dot{S}_{\mathrm{irr}}}{\dot{n}_{\mathrm{B}}} = \Delta S_{\mathrm{m}} = \sum \frac{\dot{n}_{\mathrm{RG},i}}{\dot{n}_{\mathrm{B}}} S_{\mathrm{m,RG},i} - \left(\sum \frac{\dot{n}_{\mathrm{L},i}}{\dot{n}_{\mathrm{B}}} S_{\mathrm{m,L},i} + S_{\mathrm{m,B}} \right) \tag{17}$$

According to [19], the molar reaction entropy of a chemical reaction is determined by the partial molar entropies of the individual components:

$$\Delta S_{\mathrm{m}} = \sum_{i} \nu_i S_{\mathrm{m},i} \tag{18}$$

Here the stoichiometric numbers of the products are positive, while the stoichiometric numbers of the educts are negative due to the chosen balance. The partial molar entropy of the components is defined as follows [18].

$$S_{\mathrm{m},i} = S_{\mathrm{m,0},i}(T, p_0) - R ln \left(\frac{p_i}{p_0} \right) \tag{19}$$

$S_{\mathrm{m,0},i}(T, p_0)$ describes the entropy of the component at an arbitrary temperature and a reference pressure p_0. The term $R ln \left(\frac{p_i}{p_0} \right)$ is called mixture entropy, is always negative and results from the partial pressure of the component, which differs from the pressure of the mixture. The molar exergy loss related to the fuel can finally be calculated from the ambient temperature T_{env} and the molar irreversible entropy production $Ex_{\mathrm{m,v}}$ resulting from irreversibilities [23]:

$$Ex_{\mathrm{m,v}} = T_{\mathrm{env}} S_{\mathrm{m,irr}} \tag{20}$$

Using these equations, the exergy loss of the already described simulated combustion of methane was determined. A variable air–fuel equivalence ratio λ was assumed, which varies exemplarily from $\lambda = 1$ to $\lambda = 1.4$. The calculation of the exergetic losses requires the calculation of the corresponding adiabatic flame temperature. Figure 6 shows the significant reduction of the adiabatic flame temperature of methane with an increasing air–fuel equivalence ratio. Excess oxygen and nitrogen do not actively participate in the reaction, thus reducing the adiabatic flame temperature. The results are in accordance with expectations and have been confirmed by various literary sources [18,19,24].
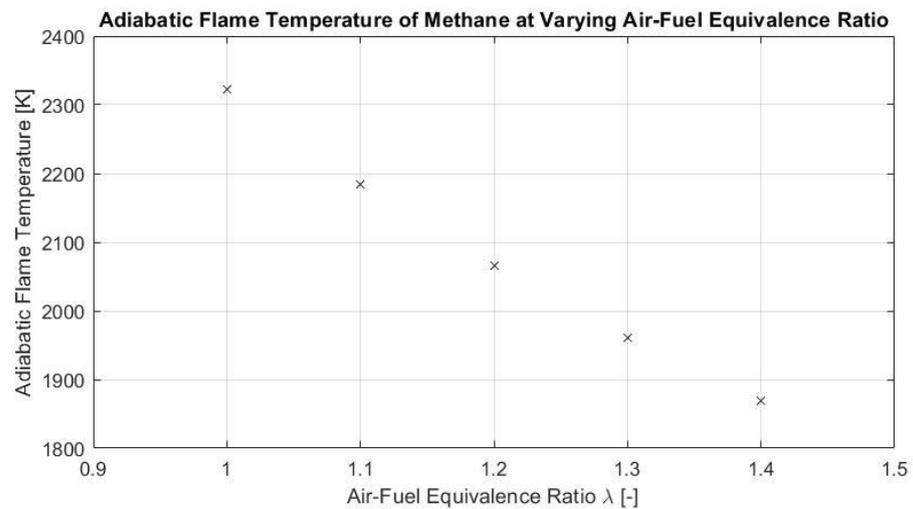
Figure 6. Effect of the air–fuel equivalence ratio λ on the adiabatic flame temperature of isobaric CH_4 combustion.

The partial molar entropy of the exhaust gas, which is required to calculate the exergy loss, was determined at the corresponding adiabatic flame temperature. Building on this, the exergy loss of a methane combustion was determined simulative. The results are shown in Figure 7. The available energy of the adiabatic combustion can be determined from the difference of the fuel's exergy and the exergy loss.
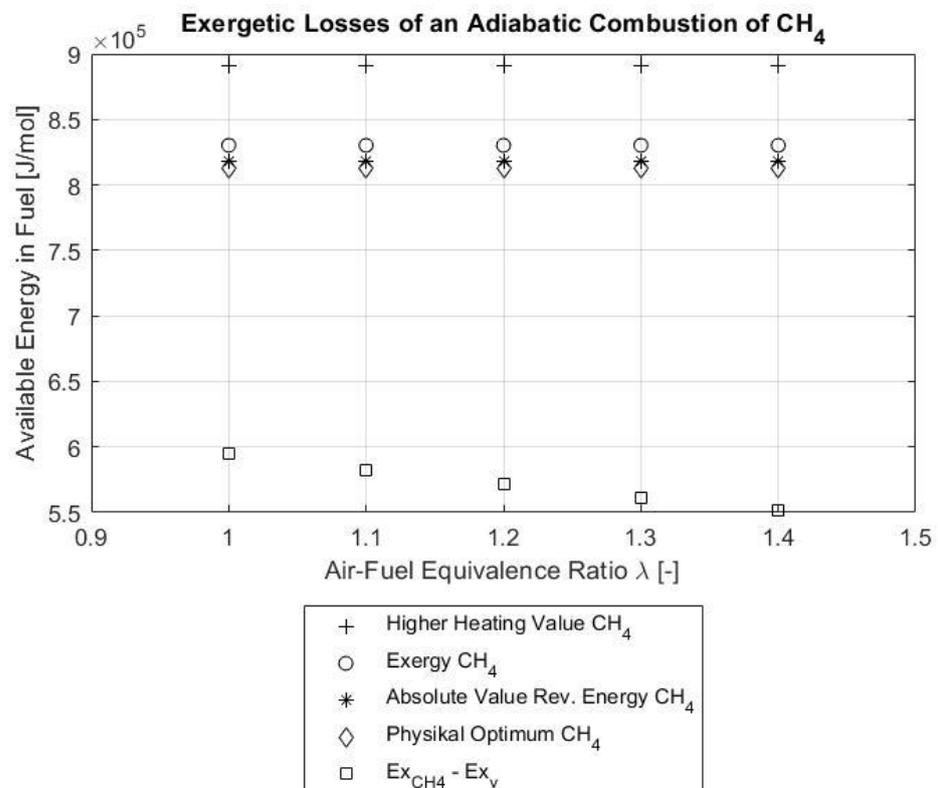
Figure 7. Exergy loss of a simulated combustion of CH_4 with dry air and a varying air–fuel equivalence ratio λ.

Figure 7 shows the significant impact of the exergy loss on the energy that can be provided by this process. Because of the higher heating value, the reversible work, the fuel's

exergy and the PhO are based on an air–fuel equivalence ratio $\lambda = 1$; they are constant parameters.

While the fuel's exergy of the stoichiometric combustion is $Ex_{CH_4} = 830.24$ kJ·mol^{-1}, the available energy taking into account the exergy losses is only $Ex_{CH_4,real} = 594.60$ kJ·mol^{-1}. This corresponds to a reduction of about 28.4%. Figure 7 also shows the influence of excess combustion air on the fuel's available energy, which is calculated from the difference of the fuel's exergy and the exergy loss:

$$Ex_{CH_4,real} = Ex_{CH_4} - Ex_v \tag{21}$$

As the air–fuel equivalence ratio increases, excess oxygen and nitrogen are added to the reaction chamber. These components do not actively take part in the reaction but are merely heated by the combustion. Equation (17) clearly shows that this will result in an increase of the entropy production $S_{m,irr}$. Since entropy production has a significant impact on exergy loss (cf. Equation (20)), the energy provided by this theoretical combustion will decrease. Thus, taking into account the exergy losses at an air–fuel equivalence ratio of $\lambda = 1.4$, the combustion process can only provide $Ex_{CH_4,real} = 551.21$ kJ·mol^{-1}. This corresponds to a reduction of the fuel's exergy of about 33.6%. Raising the combustion air intake from $\lambda = 1$ to $\lambda = 1.4$ results in a difference in available energy of 5.2%. A reference to the PhO will, of course, yield analogous results with only minor variations due to the neglect of chemical exergy. It is obvious that great efficiency losses are to be expected, especially in processes that are operated with a high excess of combustion air.

Based on the exergetic losses and the fuel exergy, the exergy efficiency ζ is determined according to Equation (22).

$$\zeta = 1 - \frac{Ex_v}{Ex_B} \tag{22}$$

In contrast to thermal efficiency, exergy efficiency considers that conversions are always influenced by limits that depend on the environment and are not actively variable. Therefore, the exergy efficiency can be considered as a measure that defines the quality of an energy conversion [25]. The effect of the air–fuel equivalence ratio on the exergy efficiency, and thus the energetic use of the chemical energy of the fuel, is shown in Figure 8.
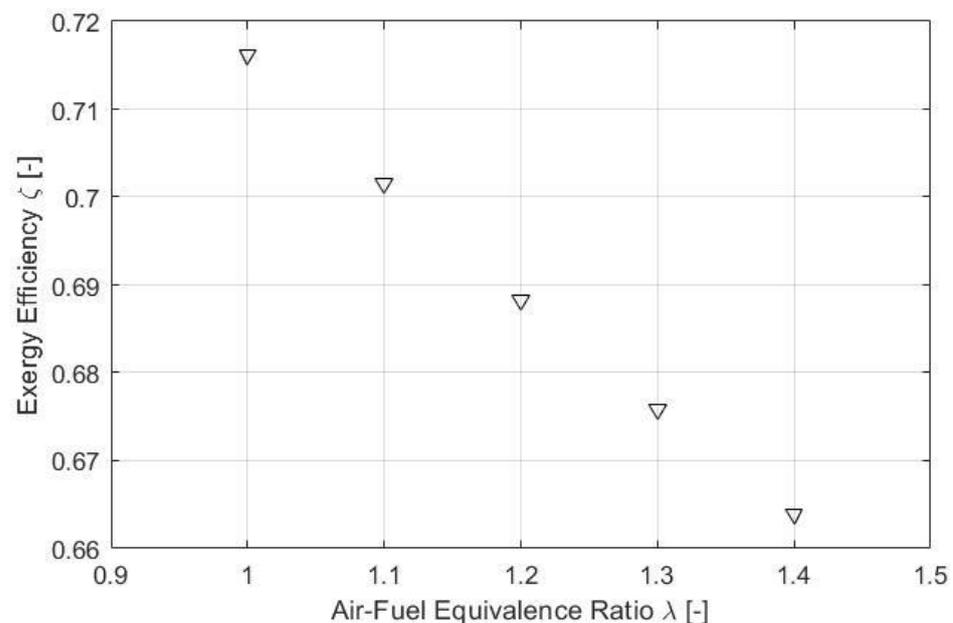
Figure 8. Effect of the air–fuel equivalence ratio λ on the exergy efficiency of isobaric CH_4 combustion.

Figure 8 shows that the exergetic efficiency decreases with an increasing air–fuel equivalence ratio. While this is $\zeta = 71.62\%$ for stoichiometric combustion, an exergetic

efficiency of only $\zeta = 66.39\%$ is achieved at an air–fuel equivalence ratio of $\lambda = 1.4$. This effect is plausible as it reflects the direct link between exergy losses (cf. Figure 7) and exergy efficiency, as it is defined in Equation (22). Thus, the supply of excess combustion air shows analogous effects on exergy loss and exergetic efficiency. The aforementioned expectations are confirmed: it can be shown that a combustion with high excess air is more efficient than combustion processes operated almost stoichiometrically.

The indirect PhO-Factor of adiabatic isobaric combustion is defined below. Since air and fuel are supplied at ambient temperature, they are not considered in Equation (23):

$$F_{\text{PhO,ind}} = 1 - \frac{Ex_{\text{v}}}{(V_{\text{B}}\rho_{\text{B}})\left[H_{\text{s,B}} - v_{\text{RG,tr,min}}c_{\text{p,RG,tr,min}}T_{\text{env}}\right]} \tag{23}$$

In this case, the reference of an exergy to an energy can be considered as noncritical, since the energy (the PhO) describes the maximum working capacity of the fuel. This is confirmed by the fact that the PhO deviates only minimally from the fuel exergy. Since in this case the PhO deviates from the fuel exergy by approximately 2%, it is expected that the value of the indirect PhO-Factor is quite similar to that of the exergy efficiency. This is confirmed by a comparison of the values. At an air–fuel equivalence ratio of $\lambda = 1$ an exergy efficiency of $\zeta_{\lambda=1} = 0.7102$ and an indirect PhO-Factor of $F_{\text{PhO,ind},\lambda=1} = 0.7162$ are achieved. This corresponds to a deviation of only 0.84%. The simulation shows that with an increasing air–fuel equivalence ratio the deviation between the exergy efficiency and the indirect PhO-Factor increases slightly. Thus, at $\lambda = 1.1$ a deviation of approximately 0.9% is achieved, while the deviation at $\lambda = 1.4$ is approximately 1.1%. However, for methane a relevant deviation of the ratios is not expected in a technically relevant range of the air–fuel equivalence ratio.

It is concluded that when evaluating a combustion process, the indirect PhO-Factor cannot provide advantages over exergetic process evaluation. The exergy loss must be determined in both cases. Only the reference to the maximum available energy of the fuel leads to a, as already discussed, small deviation of the factors. In addition, the effort to determine the key figures is comparable.

4. Conclusions and Outlook

Combustion processes are still highly present in energy technology and their use of fossil fuels continues to have a significant impact on greenhouse gas emissions [26,27]. However, sustainable energy conversion processes may include combustion processes, making them relevant in the future as well [6–8,28]. Considering upcoming regulations, the evaluation and optimization of the efficiency of these processes is essential. In order to evaluate the efficiency of a technical combustion and to define its optimization potential, the exergy of a fuel and the PhO are determined within the scope of this work. A comparison of these methods will determine whether the PhO, as a new evaluation method of the VDI Guideline 4663, can generate advantages over the conventional exergetic process evaluation.

For this purpose, an exergetic process evaluation, which is based on a method of Baehr and Kabelac [18] and an environment model of Lucas [19], as well as a process evaluation based on the PhO, are initially performed to define the maximum energy available by methane combustion. Using a Matlab script, a specific value of $Ex_{\text{CH}_4} = 830.24 \text{ kJ·mol}^{-1}$ is determined. The PhO of the methane combustion process is $V_{\text{PhO}} = 813.04 \text{ kJ·mol}^{-1}$. This corresponds to a deviation of only 2.07%. Due to this small deviation, parallels between the exergetic process evaluation and the PhO are studied. For this purpose, the reversible work, which is $W_{\text{t,m}}^{\text{rev}} = 818.12 \text{ kJ·mol}^{-1}$, is initially compared with the PhO. It can be shown that the reversible work and the PhO are almost identical and only differ by 0.62%. An evaluation of the presented equations shows that this deviation is based on the neglect of the chemical exergy in the evaluation based on the PhO. In the exergetic process evaluation, the term $\sum v_i Ex_{\text{m,i}}(T_{\text{env}}, p_{\text{env}})$ describes the chemical potential that the products have to the environment. Since this potential is exergy, it can be converted into

usable work and must be considered when evaluating the maximum available work of the fuel. In this process, neither the liquid water nor the nitrogen have any impact on the chemical exergy. While the chemical exergy of the water is negligible, which is confirmed by scientific literature [19], the nitrogen does not actively participate in the reaction and thus has the same potential as educt as well as product.

Based on this result, it can be concluded that the PhO is in this case a simplification of the fuel's exergy and cannot generate any advantages compared to the exergetic process evaluation. Neglecting the chemical exergy leads to an incomplete evaluation of the maximum available energy. At the same time, it should be noted that neglecting the chemical exergy does not significantly reduce the amount of work required to theoretically define the PhO compared to the fuel's exergy. However, the deviation of only 2% between PhO and exergy cannot justify an adjustment of PhO at present since combustion processes are not expected to be optimized to this extent. Within the scope of this work, it has also been demonstrated that if the fuel's exergy is considered in the PhO-Factor, the result is a performance indicator that does not necessarily comply with the intended definition limits of Keichel [14].

To enable targeted process optimization based on the PhO, the indirect PhO-Factor is introduced. This factor relates the individual losses of a process to the physically optimal effort and, in case of this particular combustion process, can be reduced to the losses that result from adiabatic combustion. It is shown that the air–fuel equivalence ratio has a significant effect on exergy losses and therefore process efficiency. Thus, taking adiabatic combustion losses into account reduces the 830.24 kJ·mol^{-1} contained in the fuel's exergy in adiabatic combustion to only $Ex_{CH_4,real} = 594.60$ kJ·mol^{-1}, a reduction of nearly 30%. A comparison of the exergetic efficiency of stoichiometric combustion $\zeta_{\lambda=1} = 0.7102$ with the indirect PhO-Factor of stoichiometric combustion $F_{PhO,ind,\lambda=1} = 0.7162$ shows that they nearly concur, differing by only 0.84%. This is due to the fact that exergetic losses are included in the definition of the indirect PhO-Factor as well as in exergy efficiency. The paper shows that the deviation of the ratios grows with an increase of excess air. Thus, at $\lambda = 1.4$, the factors deviate approximately 1.1% from one another. However, for methane a relevant deviation of the ratios is not expected in a technically relevant range of the air–fuel equivalence ratio. In conclusion, the indirect PhO-Factor cannot generate any advantages over the exergetic efficiency.

The calculations performed here are based on exact thermodynamic formulas. Nevertheless, the Matlab script can be improved for the calculation of the fuel's exergy and the PhO. A current limitation of the model is the assumption of dry combustion air. The script should also be further developed to allow an analysis of sulfur-containing fuels. In order to check if the deviation between the exergy of a fossil fuel and the current definition of the PhO is generally small, the indicators must be calculated for different fuels. If there are large deviations for some fuels, the PhO-Factor is not suitable as a reference for the maximum available energy of a fuel. In addition, evaluating other fuels can increase the scope of application. In addition, an evaluation of complete energy conversion processes based on PhO and exergy must be performed. In this context, processes are to be analyzed that are based on technical combustion but combine a variety of components for conversion (e.g., steam turbine or internal combustion engine). Again, differences and parallels in evaluation need to be found in order to register an added value of PhO compared to conventional evaluation methods. Another point that should be considered in the future is the evaluation of combustion processes using the PhO based on legal limits.

Author Contributions: Conceptualization, L.K., A.S. and B.S.; methodology, L.K., B.S. and A.S.; validation, L.K., A.S. and B.S.; formal analysis, L.K.; investigation, L.K.; resources, L.K.; writing—original draft preparation, L.K.; writing—review and editing, L.K., B.S. and A.S.; visualization, L.K. All authors have read and agreed to the published version of the manuscript.

Institutional Review Board Statement: Not applicable.

Nomenclature

Roman Symbols Symbol	Unit	Description
c_{P}	$\frac{\mathrm{J}}{\mathrm{kgK}}$	Specific heat capacity
Ex	J	Exergy
F_{PhO}	−	PhO-Factor
G	J	Gibbs free energy
H	J	Enthalpy
H_{s}	$\frac{\mathrm{J}}{\mathrm{mol}}$	Higher heating value
L	J	Loss
$l_{\min}$	−	Min. mass-specific air intake
m	kg	Mass
N	−	Quantity
n	mol	Number of mols
P	W	Power
p	Pa	Pressure
Q	J	Heat
R	$\frac{\mathrm{J}}{\mathrm{molK}}$	Individual gas constant
S	$\frac{\mathrm{J}}{\mathrm{K}}$	Entropy
T	K	Temperature
V	J, m^3	Effort, Volume
v	$\frac{m^3}{\mathrm{kg}}$	Specific volume
W	J	Work
x	−	Molar fraction

Sub- and Superscript Symbol	Description
0	Reference state
a	Outlet
ad	Adiabatic
B	Fuel
e	Inlet
env	Environment
i	Consecutive number
ind	Indirect
irr	Irreversible
L	Air
m	Molar
Mess	Measured value
min	Minimal
PhO	Physically optimal
R	Reaction parameter
rev	Reversible

RG	Exhaust gas	
stoich	Stoichiometric	
t	Technical	
tr	Dry	
v	loss	
Greek Symbols		
ζ	—	Exergy efficiency
λ	—	Air–fuel equivalence ratio
μ	—	Mass ratio combustion air and fuel
ν	—	Stoichiometric number
ρ	$\frac{kg}{m^3}$	Density

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