International Journal of Thermodynamics (IJoT)
ISSN 1301-9724 / e-ISSN 2146-1511
https://dergipark.org.tr/en/pub/ijot

Vol. 28 (No. 2), pp. 051-058, 2025
doi: 10.5541/ijot.1642883
Published online: June 1, 2025

Research Article

Exergetic Efficiency of Coal Gasification Using High-Temperature Mixtures of O_2/N_2 and O_2/CO_2

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Received 19 February 2025, Revised 04 April 2025, Accepted 10 April 2025

Abstract

A search for ways to improve the efficiency of energy technologies requires the selection of appropriate efficiency criteria and a study of the influence of different factors on them. This paper focuses on coal gasification, including hard coals and brown coals, in air and steam media, considering different efficiency criteria for gasification processes: cold gas efficiency, energy efficiency that takes into account the heat of the gasification agent, and exergy efficiency that considers the chemical, thermal, and mechanical energy of reactants and products. The dependence of these efficiency criteria on stoichiometric ratios and air temperature is demonstrated, and the applicability of these criteria in power plant analysis is discussed.

Keywords: *Thermochemical conversion; chemical thermodynamics; optimization; fuel utilization.*

1. Introduction

Solid fuel gasification can be used for various purposes: waste recycling, production of feedstock for chemical syntheses (primarily hydrogen), or fuel gases for gas engines or electrochemical converters, etc. Depending on the purpose, different criteria may be used to evaluate the efficiency of gasification processes [1]. The most frequently used criteria are given in Table 1.

Table 1. Efficiency criteria for gasification processes.

Criteria	Formula	Notation
Cold gas efficiency (chemical efficiency)	$\eta_{chem} = \frac{\sum_i q_i G_i}{QB}$	q_i – heat of combustion of i-th component; G_i – mass flowrate of *i*-th component; Q – heating value of fuel; B – fuel flowrate
Thermal efficiency	$\eta_{therm} = \frac{\sum_i q_i G_i}{QB + q_{in}}$	q_{in} – energy used for gasification agent heating
Exergetic efficiency	$\eta_{ex} = \frac{\sum_i Ex_i G_i}{Ex_f B + Ex_g G_g}$	Ex_i – specific exergy of *i*-th component; G_g – gasification agent mass flowrate
Carbon conversion efficiency	$\eta_C = \frac{\sum_i y_i^C G_i}{y_f^C B}$	y_C – carbon content
Component yield	$\eta_i = \frac{G_i}{B}$	

Gasification efficiency metrics in the present study are cold gas efficiency, which is a fraction of solid fuel heating value converted into gas heating value; thermal efficiency, which takes into account the physical heat energy of the gasification agent; and exergy efficiency, which weighs input and output exergy flows. It should be noted that chemical exergy is not equal to heating value but is usually very close. Cold gas efficiency does not include the sensible heat of gas and its expansion work.

Gasification technologies allow for the production of multiple products, so it is necessary to have a method for comparing the different outputs of chemicals, heat, and electricity, depending on the conditions. Considering this, exergy analysis can be considered as such a method.

The main difference between energy and exergy analyses lies in the choice of the reference state. For technical thermodynamics, the reference state is absolute zero temperature and vacuum. In exergy analysis, however, the environment serves as the reference state.

Accordingly, a system at rest with the conditions of the environment has zero useful energy, as it cannot serve as a source or sink for heat or matter in any engine, due to the second law of thermodynamics. Thermal, chemical, and mechanical energy stored within a system cannot be completely utilized. These forms of energy, when weighted by their availability, are referred to as exergy.

Following standard definitions [2], we can express the exergy value of a system as follows:

$$Ex = U + P^{env}V - T^{env}S - \sum_j^{N_s} n_i \mu_i^{env} \qquad (1)$$

Here Ex is system exergy, U is internal energy of the system, index *env* refers to the environment, and V is system volume. Using enthalpy instead of internal energy, we can write the expression for exergy as:

$$Ex = H - (P - P^{env})V - T^{env}S - \sum_j^{N_s} n_i \mu_i^{env} \qquad (2)$$

In this form, exergy can be calculated using tables of thermodynamic properties of substances. Standard exergy values for individual substances are tabulated in corresponding reference datasets. For example, data from references [3] and [4] are used for calculations in this paper. Calculating exergy for solid fuels involves ill-defined estimates of specific entropy. However, there are reference books and recommended formulas available (for example, [5] for materials and [6] for fuels) to assist with this process.

The full exergy of a gasification-based power plant may also include slag, sulfur, and high-purity nitrogen, among other possible products. However, we will not consider these potential outputs in this discussion, as we focus on the raw producer gas leaving the gasifier unit.

In the case of high-temperature gasification, the producer gas contains both chemical energy that can be used for combustion and thermal energy that can be used, for example, for generating steam. Therefore, the presentation of gasification efficiency in terms of cold gas efficiency (Table 1) is not sufficient to evaluate the overall useful energy potential. It should also be noted that integrating a gas cleaning system into the thermal circuits for water-steam heating can significantly improve the efficiency of the entire power plant.

Many works focused on the analysis of exergy flows in energo-technological plants (combined heat and power plants [7], integrated gasification combined cycles [8], Rankine cycles [9], co-gasification of coal and biomass [10], multifuel plants [11]), including multi-product coal-fired plants (hydrogen [12], methanol [13], ammonia [14]) and CO_2 capture systems (including flue gas recycle [15] and cooling-compressing unit [16]). Some studies compare different capture schemes (for example, advantages of the water shift stage are considered in [17] and the choice of absorbent is considered in [18]) and the energy/exergy efficiency of the plant or specific units under different conditions. Of interest is the relationship between the exergy efficiency of gasification units and process parameters, such as the flow rate and temperature of the gasification agent.

Exergy balances for industrial coal gasification processes were developed in [19] for Hygas and in [20] for Lurgi. The main exergy losses are related to oxidation processes, although these are lower than those for traditional combustion processes [21]. The exergy balance for plants that produce liquid fuels from gasification products is discussed in [22], and the reduction of exergy losses through the use of indirect combustion methods for coal, including gasification, is examined from a general perspective in [23]. Similarly, schemes involving heat recirculation between stages of coal conversion are explored in [24] for steam-oxygen gasification and in [25] for air gasification.

The current study aims to compare the energy and exergy efficiency criteria as applied to some coal gasification processes. In relation to the scope of this paper, we are also interested in works related to the study of the spatial distribution of specific exergy in solid fuel conversion reactors. These works include one-dimensional approximations [26] and 2D/3D formulations (for multiphase flows [27] and gaseous fuels [28]). Data obtained through measurements and mathematical modeling allow us to control the destruction of exergy directly in the reaction zone. However, such detailed models usually do not allow full-scale parametric optimization due to high computation costs.

Let us consider the process of coal gasification with the hot air. Usually, high-temperature heating improves the conditions for gasification, increasing the conversion of fuel and improving the quality of producer gas. Thermodynamic analyses show that reducing the temperature difference between the input and output material flows may increase the efficiency of the fuel conversion process (see, for example, works on general theoretical consideration [29] and, specifically, carbon conversion [30]). However, we must consider the heat input required to obtain high-temperature air. Chemical and thermal energy have different qualities: chemical energy can be converted directly into useful products, while thermal energy can be converted into useful work with significant thermodynamic losses only. Therefore, we need the exergy approach to evaluate the efficiency and to compare different approaches.

A feature of the thermodynamics of solid fuel gasification processes in oxygen-containing environments (compared to combustion processes) is the existence of a certain threshold value of the stoichiometric ratio above which a complete conversion of fuel matter to gaseous products occurs (these thresholds were determined for solid carbon in [31] and for biomass in [32]). This threshold value is typically much lower than that corresponding to complete combustion (ranging from 0.25 for biofuels to 0.5 for pure carbon), and the addition of additional gasifying agents such as steam and carbon dioxide can reduce this value even further. However, a purely stoichiometric approach is not sufficient without considering the thermodynamics and kinetics of heterogeneous reactions. In this study, we examine the kinetic constraints on achieving equilibrium in a reactive system, which allow us to identify temperature ranges for thermodynamically efficient coal gasification.

The novelty of the study is the estimation of exergy efficiency for oxyfuel gasification processes. These processes were proposed in several works for both one-stage [33] and two-stage [34] reactors. The advantage of oxyfuel gasification is the lower cost of carbon dioxide capture, although there are crucial changes in heat transfer and produced gas properties. The papers [34, 35] give estimates for the net efficiency of oxy-fuel IGCC plants of about 40-45%, taking into account carbon capture based on available experimental data. Further investigation may lead to an even lower carbon capture penalty. Traditional and promising coal power plants with carbon capture have net efficiency of 30-35% [36]. The advantage of IGCC is very important in the climate change context, when thermal power plants become subject to strict environmental restrictions, including carbon emissions. The use of cheap coal fuel based on new clean technologies will not only improve the environmental situation but also achieve the specified requirements with acceptable economic efficiency.

It is important to evaluate the thermodynamic efficiency of new promising gasification processes with respect to exergy destruction and to compare them with traditional processes. Moreover, instead of considering a single set of parameters, we obtain exergy efficiency in a wide range of stoichiometric ratios and input temperatures, which allows us to optimize coal gasification conditions.

2. Mathematical Model

The calculation of coal gasification processes is carried out using a mathematical model, which is described previously in detail in the paper [37]. These works use a spatially one-dimensional model of the coal dust gasification process in an oxidizer flow, similar to models [38] and [39].

In formulating the mathematical model, the following assumptions are made. The drying rate is limited by external mass exchange with the ambient air, the pyrolysis rate is proportional to the volatile content in the particle and depends on temperature according to the Arrhenius law, the gasification rate of individual particles is described by the diffusion-kinetic theory of carbon combustion, reactor heat losses can be neglected, and the effects of gas recirculation and slag film are not taken into account.

The chemical kinetics of reactions in the gas phase are not taken into account. It is assumed that substances entering the gas phase immediately reach a state of thermodynamic equilibrium. Therefore, chemical transformations are described using a thermodynamic model with macroscopic constraints on the rate of heterogeneous reactions (for example, lumped models [40] and reduced-order models developed for power plant calculations [41]). This approach is suitable for high-temperature processes where the rate of gas-phase reactions is much faster than that of heterogeneous reactions. Validation of this assumption by means of comparison of calculated gas composition with experimental data can be found in previous works [37]. The possible effects of interaction between organic and mineral components of fuels (due to thermal effects or slagging conditions [42]) are not considered. Therefore, gasification efficiency is mainly determined by stoichiometry and fuel heating value.

One-dimensional approximation does not allow capturing all relevant phenomena in high-temperature gasification processes. For example, feeding systems design and gas-wall interaction, including jet formation and destruction, are important topics in gasification technology. However, this approximation allows estimating reaction zone length and output gas composition, which gives the main technical parameters of the gasifier at the level of a power or chemical plant. The reduction of computations from 3D to 1D is crucial, and additional simplifications (such as the equilibrium gas model) allow for full-scale optimization. Some models include several 1D pieces allowing to reproduce mixing and recirculation effects [38, 43]. We do not take into account these features, modeling the flow in the gasifier as uniform.

The model allows estimating the influence of heterogeneous reactions on exergy efficiency. A similar analysis was presented in [44] for the oxygen-fed coal gasification process (Texaco), where authors obtained an exergy efficiency of 87% with 50% of all losses due to heterogeneous reactions. Temperature range was, however, very narrow. In the present work, we consider a wider range of gasification processes.

In calculations, we varied gasification conditions, namely stoichiometric ratio, fuel composition, gasification agent composition, and its temperature. Depending on carbon content, the optimum stoichiometric ratio varies from 0.25 to 0.5 [45]. In our calculations, we used a range from 0.15 to 0.7 to localize the optimum. Brown and hard coals were considered as a fuel (see Table 2). Brown coals contain less carbon, so their heating value is lower, but a higher content of elemental hydrogen and oxygen (including moisture) allows for decreased specific air consumption and obtaining syngas with a higher content of molecular hydrogen. Hard coals have a higher heating value, which allows for a higher conversion temperature but requires the addition of steam to convert this heat to combustible products. In this regard, carbon dioxide may be considered as an alternative to steam.

The properties of a gasification agent are usually the main control parameter in the conversion process. We use a range of temperatures from 300 K to 1400 K. The latter value is quite high and non-typical for industrial processes. Nevertheless, we consider this range in order to investigate the limits of coal gasification caused by physico-chemical features, which are discussed in Sec. 6.

Table 2. Coals' characteristics.

Coal source		Berezovskiy	Mugunskiy	Urtuysky	Kuznetsky
Coal composition, % mass.	W^r	33	22	29.5	2
	A^r	4.7	15.6	8.8	15.4
	S^r	0.2	0.9	0.3	0.9
	C^r	44.2	46	46.9	68.1
	H^r	3.1	3.6	3	4.2
	N^r	0.4	0.9	0.5	1.6
	O^r	14.4	11	11	7.8

3. Air Gasification of Brown Coals without Heating

Let us consider the gasification process in a cylindrical reactor with a diameter of 3 m and a length of 9 m; the working pressure is about 15 atm [46]. The fuel consumption is about 180 t/h, and the average particle size is 0.1 mm. The gasifying agent is a mixture of air and water vapor (temperature 655 K). The variable parameters are the specific air consumption (1-6 kg/kg of fuel) and the specific steam consumption (0-0.1 kg/kg of fuel). The fuel composition is given in Table 2 [47].

Figure 1 shows a comparison of the exergy and cold gas efficiencies of air-steam gasification of brown coals. From a practical point of view, the most interesting region is the vicinity of the maximum efficiency. Before reaching the extremum, both criteria are practically the same: the cold gas efficiency closely characterizes the exergy of the producer gas. However, as the specific consumption of oxidizer increases, the cold gas efficiency decreases at a sharper rate. At high values of stoichiometric ratio, fuel carbon is almost absent, and the excessive oxygen consumes combustible components of the producer gas. Cold gas efficiency is more sensitive to these losses. Exergy efficiency remains at the level of 30-40% even after complete combustion (stoichiometric ratio of 1), due to the high thermal exergy of combustion products. Interestingly, the dependence is similar for different compositions of coal.

Three presented coals are quite similar in composition: Mugunsky coal contains 2-3 times more ash and less moisture; Urtuisky coal contains more carbon in the organic mass and therefore has a higher heating value. The stoichiometric amount of air required for complete combustion varies for the coals in the range of 5.5-6.3 kg/kg. Urtuysky coal is characterized by the highest cold gas efficiency (68.3%), due to its higher heating value. It is followed by Berezovsky coal (67.5%) and, finally, Mugunsky coal (67.3%). The last two coal compositions, however, differ little, and within the accepted assumptions, the characteristics of their gasification can be considered almost equal. Exergy efficiencies are very close to cold gas efficiencies (68.3%, 67% and 67.3% correspondingly).

4. Coal Gasification with Air Heating

Now, let us consider the gasification process in a reactor with a fuel capacity of 100 t/h with an operating pressure of 30 atmospheres [48]. The geometric dimensions of the reactor are as follows. The length of the reaction zone is 15 meters; the internal diameter is 3 meters. The temperature of the fuel entering the reactor is 300 K; the steam temperature is 600 K, and the air temperature varies from 400 to 1300 K (with increments of 100 degrees). The steam consumption ranges from 0 to 0.2 moles per mole of carbon, and the stoichiometric ratio ranges from 0.1 to 0.7 (with increments of 0.05). Kuznetsky coal with characteristics from Table 2 is used as the fuel.

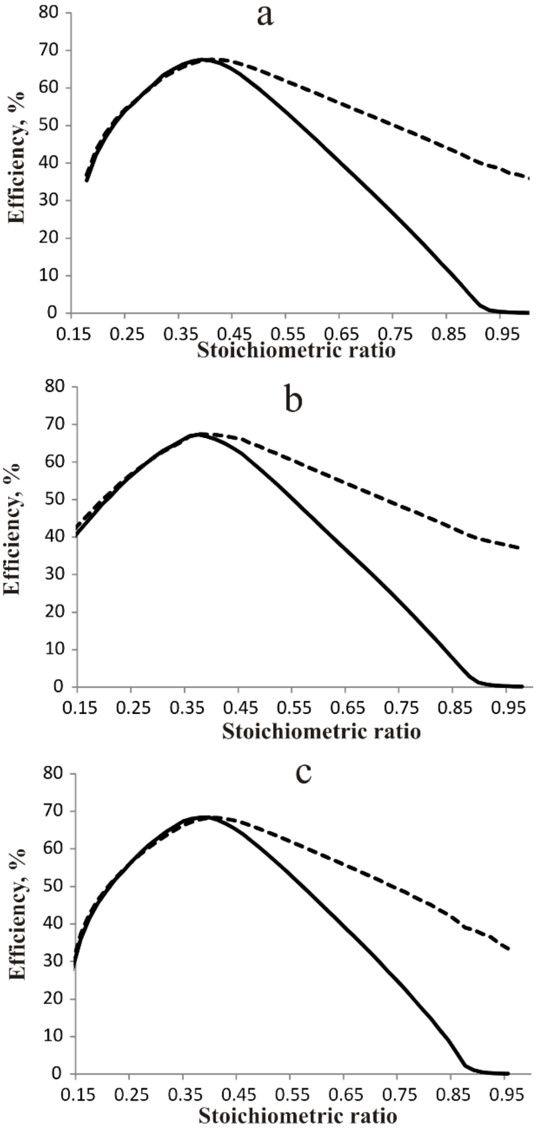

Figure 1. Comparison of cold gas efficiency (solid line) and exergy efficiency (dashed line) of brown coal gasification: Berezovsky (a), Urtuysky (b), Mugunsky (c).

For each parameter set, output gas flow rate and composition were calculated using the mathematical model. Then, criteria presented in Table 1 were estimated. The results are presented below.

A comparison of cold gas, thermal, and exergy efficiencies is presented in Figure 2. The x-axis in each subfigure is the stoichiometric ratio, and the y-axis is the input air temperature. Isolines show surfaces of efficiency criteria dependence on gasification agent parameters for the fixed fuel composition and flow rate. The observed dependences are typical for solid fuel conversion. There is a range of stoichiometric ratios where efficiency achieves its maximum. Qualitatively, these surfaces are similar. However, the optimal conditions for each criterion differ: cold gas efficiency has a relatively flat maximum, with the value above 78% extending along the temperature axis. In contrast, thermal and exergy efficiencies exhibit a more distinct extremum, corresponding to lower temperatures.

Thermal efficiency indicates that high-temperature air heating does not justify an increase in combustible gas yield. Exergy efficiency maximum corresponds to air heating up to 600 K. Cold gas efficiency does not change significantly with increasing temperature, and the thermal exergy of input air included increases. Although the thermal exergy of the gasification products also increases, there is an optimum, marking the point after which the exergy efficiency decreases more slowly than the cold gas efficiency (similarly to the previous section).

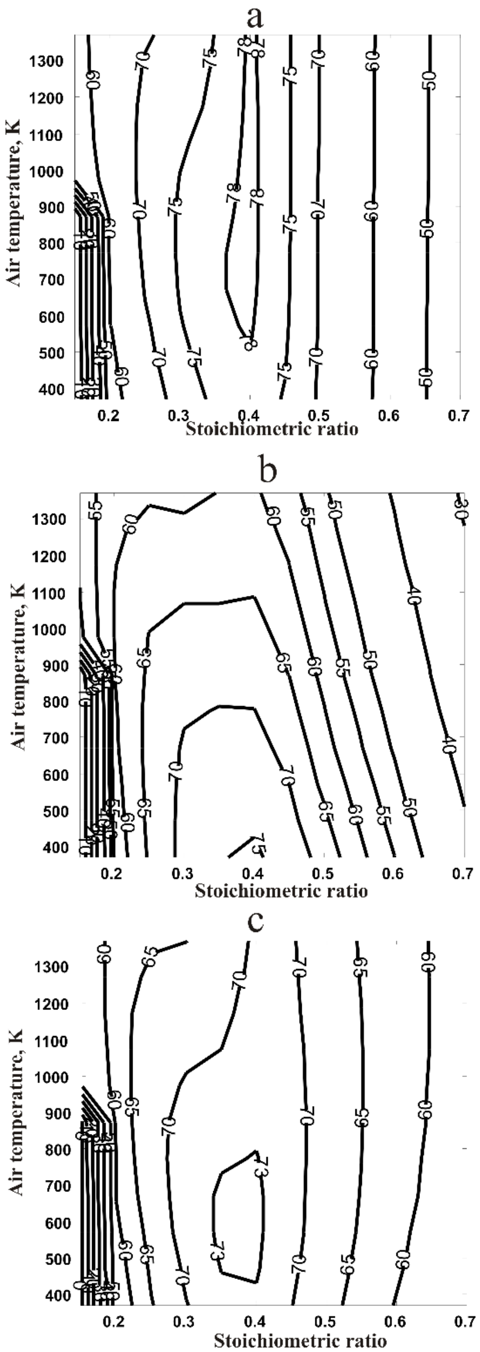

Figure 2. Coal gasification efficiency (%) at a specific steam consumption of 0.1 mol/mol carbon: cold gas efficiency (a), thermal efficiency (b) and exergy efficiency (c).

Kuznetsky coal has a higher heating value compared to brown coals, which results in higher cold gas efficiency. Higher gasification temperature improves conditions for endothermal reactions leading to CO and H_2 formation. The exergy efficiency is lower, although still higher than for brown coals.

5. Coal Gasification with High-Temperature O_2/CO_2 Mixtures

Then let us consider the gasification process under oxy-fuel conditions, when the gasifying agent is a mixture of oxygen with combustion products. By excluding nitrogen, highly concentrated CO_2-H_2O mixtures are obtained after combustion [49]. The extraction of CO_2 for further transportation and disposal is simplified compared to using air.

The higher heat capacity of CO_2 inhibits fuel ignition at low temperatures while also contributing to thermal stability and improved gas quality when using heating [50]. The reactor dimensions and fuel characteristics remain the same as in the previous section. Detailed calculations can be found in [48].

When switching to O_2-CO_2 (ratio is 20/80), increasing the temperature allows for several benefits. Firstly, it expands the range of stable conditions. Secondly, it can result in an increase in cold gas efficiency by up to 20% per 1000 K. The optimal value of the stoichiometric ratio decreases as the gasification agent temperature increases, as CO_2 acts as the gasifying agent in this case.

Figure 3 compares the cold gas, thermal, and exergy efficiency surfaces in the space of stoichiometric ratio and gasification agent temperature as in the previous section. In the vicinity of optimal parameters, all three criteria behave similarly: the maximum efficiency is observed with increasing temperature within a narrow range of stoichiometric ratios. However, the maximum efficiency decreases from 95% for the cold gas efficiency to 88% for the thermal efficiency and 84% for the exergy efficiency.

As the stoichiometric ratio increases, the cold gas efficiency becomes independent of temperature and is only determined by stoichiometric conditions. When taking into account the air heating, the efficiency naturally decreases as temperature increases. In contrast, the exergy efficiency increases with temperature due to the increase in the thermal component of the exergy of the gasification products.

A significant difference between fuel gasification in air and in O_2/CO_2 mixtures is the initial exergy of the gasifying agent: atmospheric air has, by definition, zero chemical exergy; O_2/CO_2 mixtures (even at an oxygen concentration equal to atmospheric) have significant chemical exergy due to the high CO_2 content. In this case, CO_2 is not an inert diluent but a gasification agent, the presence of which improves the conditions of coal conversion and, in some cases, increases the heating value of the producer gas. Therefore, the transition from unstable (low-temperature) gasification conditions to effective high-temperature conversion occurs with a larger jump compared to air gasification.

Authors of [51] noted that the 1D model of the oxy-fuel coal gasification process gives similar results as the equilibrium model. In this regard, our results may be of interest with respect to gasification process characteristics. Cold gas efficiency of 76% was achieved in a laboratory reactor in [35], which is close to the optimum point at the lower temperature boundary (see Figure 3a). The estimated power production efficiency, taking into account carbon capture, is about 40-45%. The presented results show that gasification agent heating can enhance gasification efficiency, allowing to achieve exergy efficiency of 80% at lower stoichiometric ratios. In this regard, the promising power plant needs less oxygen, which is compensated by the CO_2 heating up to 700 K and higher. Thermal energy costs of the heater in some cases are less than power production efficiency increase [52].

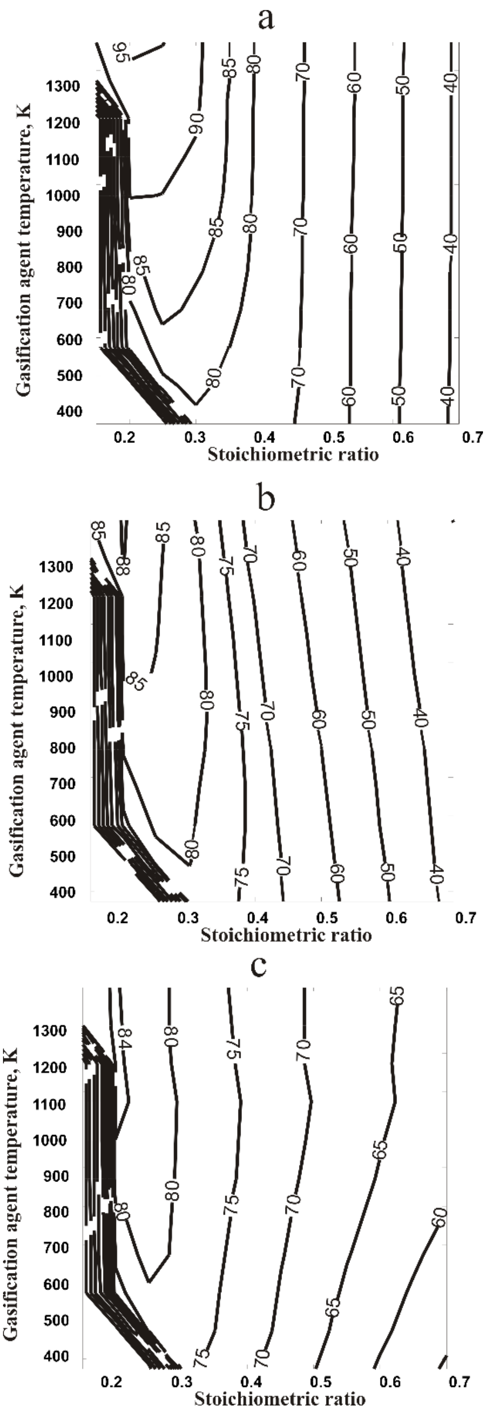

Figure 3. Comparison of coal oxyfuel gasification efficiency criteria (%): cold gas efficiency (a), thermal efficiency (b) and exergy efficiency (c).

6. Discussion

The cold gas and exergy efficiencies of the gasification process are almost equal in the region of low stoichiometric ratios, where the chemical exergy greatly exceeds the thermal exergy. The divergence is observed when the stoichiometric ratio exceeds a value corresponding to the full coal conversion. At this point, oxidation leads to the conversion of the chemical energy of gasification products into thermal energy. The positions of the maximum efficiency are equal for both criteria. The gasification products have significant thermal exergy value after the threshold [31]. In this regard, the conclusion of paper [53], proposing the optimal conditions at stoichiometric ratios slightly above a full conversion threshold, is correct.

Gasification in O_2/CO_2 mixtures allows for higher efficiency due to the utilization of carbon dioxide as a gasification agent in the reaction zone. Full conversion threshold shifts towards lower stoichiometric ratios (which are calculated based on molecular oxygen). In order to maintain high temperatures, internal heating is required (due to the higher heat capacity of CO_2). Nevertheless, the exergy efficiency value for oxyfuel gasification is lower than other efficiency criteria. This is due the higher specific exergy of concentrated CO_2, which increases exergy input in oxyfuel systems but allows simpler carbon capture.

It should also be noted that the efficient use of producer gas thermal exergy is limited due to the requirements for temperature and purity of fuel gas. Harmful impurities, such as sulfur compounds and solid particles, must be removed. A common solution to this is water spray cooling, which reduces the heating value of the producer gas through a water shift reaction and dilution. In some cases, the water shift stage is introduced to enhance the H_2/CO ratio or to capture CO_2 before combustion. The exergy loss during the gas conditioning is estimated to be up to 80% of total exergy losses according to studies [54, 55]. Reducing these exergy losses is possible through hot gas cleaning methods, such as hot filters and sorbent materials [56, 57]. This approach has the potential to improve thermodynamic efficiency, although it may require more expensive materials and more complex equipment to implement.

7. Conclusion

The paper examines the influence of the stoichiometric and heating factors on the efficiency of coal gasification processes. The exergy efficiency of gasification is shown to be similar to the cold gas efficiency when the stoichiometric ratio is below the optimum point, but it exceeds the cold gas efficiency at higher values of the stoichiometric ratio due to the thermal energy of the gas products. When switching to oxyfuel gasification, the initial energy of the gasification agent is higher due to the high CO_2 content, while the higher heat capacity of the mixture can worsen the thermal stability at lower initial temperatures but improve the conditions for high-temperature gasification.

Optimum exergy efficiency is about 73% for air gasification heated to 600-700 K and about 80% for oxyfuel gasification at the same gasification agent temperature. Even higher exergy efficiency is possible at temperatures of O_2/CO_2 mixtures of 1100 K and stoichiometric ratios of about 0.2, but it would be difficult to maintain a stable gasification process in these conditions.

Acknowledgements

The research was carried out under the State Assignment Project (no. FWEU-2021-0005) of the Fundamental Research Program of the Russian Federation 2021-2030 using the resources of the High-Temperature Circuit Multi-Access Research Center.

Nomenclature

Ex – exergy
$IGCC$ – integrated gasification combined cycle
U – internal energy
S – entropy
P – pressure
T – temperature
n – molar quantity
μ – chemical potential
η – efficiency
G – gas mass flowrate
q – gas component heating value
Q – coal heating value
B – coal mass flowrate
y^C – carbon content
W^r – moisture content
A^r – ash content
C^r – carbon content (y^C)
H^r – hydrogen content
O^r – oxygen content
S^r – sulfur content
N^r – nitrogen content
$chem$ – chemical
$therm$ – thermal
eq – equilibrium
in – input
env – environmental

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