

SIMULATION OF METHANE COMBUSTION IN THE ALLAM CYCLE

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ABSTRACT

The Allam cycle is an oxy-fuel cycle incorporating carbon capture technology, able to play a role in the frame of the energy transition in support to a full switch to renewables. In the case of LNG, combustion takes place in a CO_2/O_2 environment so that the products are in theory only H_2O and CO_2 . The former can be condensed and separated, and the latter can be extracted as a liquid to be directed to sequestration. An issue which is not adequately covered in the scientific literature is the possible presence of secondary species in the combustion products. This presence may invalidate the performance of the condensation and sequestration of the low-temperature section and can also affect the performance of components like the expanders, as well as impact that of the combustors themselves. Based on data from cycle simulations, a series of CH_4 -Oxygen equilibrium calculations in CO_2 enriched environment, used both in the primary zone and as a coolant, was set up using Cantera. The approach pursued allowed to identify the potential presence of secondary species which resulted considerably affected by the recirculation rate of CO_2 . Tuning of this parameter can allow a very low concentration of secondary species, which are limited to CO .

The consequence of dissociation over the thermodynamic performance was evaluated developing a model for the combustion process and comparing the results of complete combustion and equilibrium composition. This analysis demonstrates that the reactants regenerative preheating arrangement of the Allam cycle is very effective in decreasing the combustion exergy destruction. The differences determined by equilibrium conditions are marginal.

1 INTRODUCTION

The Allam or NET cycle (Allam et al., 2017) is an innovative solution in support of energy transition (IEA, 2020). The energy transition requires a switch to Renewable Energy Sources (RES). The energy transition necessitates the development of technologies capable to harnessing renewable energy sources (RES) for reaching carbon neutrality (IEA, 2020). Among these, solar photovoltaics and wind power are the most significant in terms of market potential and are classified as Variable Renewable Energies (VRE). However, VREs require support from baseload power plants or extensive energy storage deployment. Geothermal and biomass energy can support VREs to some extent, but extensive storage presents challenges regarding economic profitability and sustainability, such as the life cycle impact of batteries. In the near future, clean fossil fuels like Liquefied Natural Gas (LNG) will play a crucial role in supporting the transition to RES.

The advantage of oxy-combustion cycles, which involve extensive recycling of combustion products, is that water can be separated through condensation, leaving CO_2 as the primary combustion product. This CO_2 can then be directed to storage and sequestration, thereby substantially reducing the carbon footprint. A notable feature of the Allam Cycle is the extensive and effective preheating of reactants, achieved through a complex regenerative heat transfer network (Scaccabarozzi et al., 2016).

One issue that is not extensively covered in the scientific literature is the possible presence of secondary species in the combustion products. The assumption that the working fluid consists solely of a mixture of CO_2 and H_2O requires validation as the Allam Cycle is further developed. The consequences of this assumption range from combustor performance and efficiency to the realistic modelling of expansion and the final challenge of separating carbon compounds. This is the specific focus of the present research.

2 ALLAM CYCLE

A schematic of the Allam cycle is included in Figure 1. Supercritical CO_2 is compressed by a series of intercooled compressors when it is in the gas phase and pumps when it is in the liquid phase. It is then mixed with oxygen, reaching up to 99.5% purity, supplied by the Air Separation Unit (ASU). The working fluid passes through the regenerator, where it is preheated before being directed to the combustor. Following expansion in the turbine, the heat of the flow is recovered in the regenerator, a complex multi-flow heat exchanger. The mixture is subsequently cooled in a condenser, which also separates water. Finally, the remaining CO_2 is partially stored and partially compressed again to initiate a new cycle.

Currently, the primary focus in the development of the Allam Cycle is associated with the utilization of LNG, which can be considered as pure methane for practical purposes. The efficiency of the natural gas-fired Allam cycle can reach almost 60%, with nearly 100% carbon capture (Allam et al., 2017; Chan et al., 2023). It is acknowledged as one of the most appealing solutions for oxy-fuel cycles and related CCS applications.

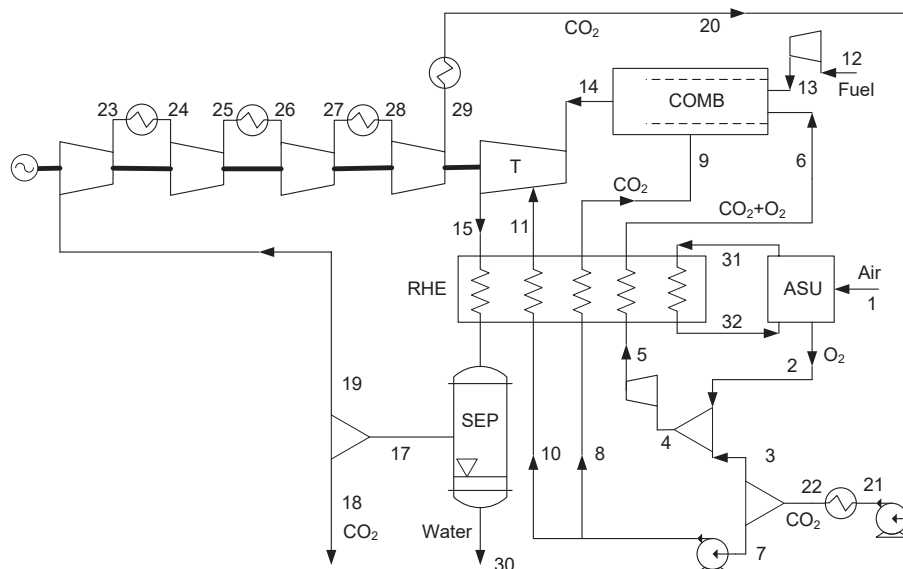


Figure 1: Simplified Allam cycle layout

2.1. Oxy-Fuel Combustion processes

Oxy-fuel combustion has emerged as a leading technology for implementing carbon capture and sequestration across various applications in the power plant industry, encompassing atmospheric boilers, fluidized beds, and gas turbines (Stanger et al., 2015).

Conventional gas turbines operate with a high excess of air to maintain turbine inlet temperatures within allowable limits for structural integrity. This excess air provides the oxygen necessary for combustion. In order to mitigate nitrogen oxides (NO_x), which originate from both nitrogen in the air and from fuel-

bound sources, conventional gas turbines employ premixed combustion. However, in oxy-fuel operation, combustion occurs at nearly stoichiometric conditions without the presence of nitrogen in the reactants. Thus, the need for a premixed flame is eliminated, as the fuel and oxidizer can be supplied separately. While NO_x emissions may arise due to nitrogen in the fuel, the working fluid derived from LNG contains very low concentrations of nitrogen, rendering NO_x emissions non-critical.

The scientific literature currently lacks extensive research on the oxy-fuel combustion process, particularly in conjunction with the Allam cycle. In the Allam cycle, combustion occurs under extreme conditions primarily due to the exceptionally high pressures involved. Compared to conventional turbines and combustors, the pressures in the Allam cycle are significantly elevated, ranging from 250 to 350 bar as opposed to the typical interval between 17 and 40 bar found in present gas turbine cycles. Uysal (2021) demonstrated that for the Allam cycle, the optimal pressure value is 300 bar to maximize net power output.

Komarov et al. (2020) conducted a simulation of combustion at a pressure of 300 bar for a supercritical carbon dioxide gas turbine. They presented the results of chemical kinetics, comparing them with a nitrogen-based environment. Their study demonstrated that in a CO₂ environment, the normal flame propagation velocity decreases, leading to a slower combustion process. This deceleration occurs because CO₂ hampers chemical reactions more than nitrogen does. The flame velocity can drop to as low as values between 0.05 and 0.10 m/s, and a crucial design factor to enhance it to levels between 0.2 and 0.3 m/s is operating combustion at nearly stoichiometric conditions ($\Phi = 1$) in the primary zone, alongside using a low dilution factor in the combustor $\gamma = \text{CO}_2/(\text{CO}_2 + \text{O}_2)$. An essential parameter influencing flame velocity is the combustor inlet gas temperature: values exceeding 750°C are recommended. Therefore, gas preheating emerges as a pivotal issue to achieve both high efficiencies and stable combustion.

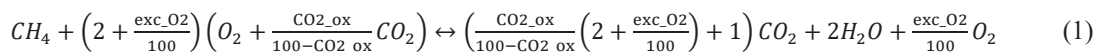
3 COMBUSTION MODELS

3.1 Complete Combustion Model

Previous cycle simulations conducted by Mariani et al. (2021) have established the operating conditions for the combustion chamber, specifically a pressure of 30 MPa and a preheating temperature of 1000 K for the reactants. The model employed assumes complete combustion without dissociation reactions, resulting in CO₂, H₂O, and excess O₂ as the sole products.

The objective of this model is to generate preliminary results that can guide more advanced investigations. Developed in Engineering Equation Solver (EES), the model is referred to as E-CCM. Key assumptions include adiabatic combustion, which allows for the calculation of the adiabatic flame temperature, an open system, and no work transfer.

The fuel considered in the model is pure methane (CH₄), and the oxidizer is a mixture of O₂ and CO₂. All species are treated as ideal gases. The reaction is represented by Equation (1).



The variable exc_O_2 represents the percentage excess of O₂ compared to stoichiometric conditions, while $\text{CO}_2\text{_{ox}}$ denotes the molar fraction of CO₂ in the oxidant mixture comprising O₂ and CO₂. This parameter, in molar terms, corresponds to the γ proposed by Komarov. The coefficient multiplying CO₂ ensures that the proportions in the oxidant mixture remain correct when varying exc_O_2 . In this model, exc_O_2 is initially set to zero to maintain stoichiometric conditions, minimizing the use of O₂, as its separation from air is both energy-intensive and costly. It is crucial to employ CO₂ as a diluent; otherwise, using pure oxygen would lead to extremely high temperatures.

An exergy analysis was associated with the chemical reaction of Equation (1) with the purpose of evaluating the exergy efficiency through Equation (2):

$$\eta_x = \frac{Ex_p}{Ex_{in}} \quad (2)$$

The exergy of products, of the reactants and the exergy destruction are computed using the relations from Equation (3) to Equation (5):

$$Ex_p = H_{p, fis} - T_0 S_{r, fis} \quad (3)$$

$$Ex_{in} = \varepsilon - Ex_r \quad (4)$$

$$Ex_d = T_0 (S_p - S_r) \quad (5)$$

The chemical and physical exergy of the reactants are defined by Equation (6) and Equation (7):

$$\varepsilon = -\Delta G_{react} \quad (6)$$

$$Ex_r = H_{r, ph} - T_0 S_{r, ph} \quad (7)$$

3.2 Incomplete Combustion Models

An incomplete combustion model for CH₄ in oxygen with CO₂, used both in the primary zone and as a coolant, was developed using Cantera. This model assumes equilibrium conditions and employs the same operating parameters as those for complete combustion. The setup allowed for the identification of potential secondary species such as CO, which were significantly influenced by the CO₂ recirculation rate. By tuning this parameter, the model can achieve very low concentrations of secondary species, primarily CO. This model treats combustion as incomplete, facilitating a more advanced analysis that includes dissociation and endothermic reactions, and considers a broader range of products. A general model, referred to as P-ICM (Python Incomplete Combustion Model), was created using Python and the Cantera library, which is designed for thermochemical analysis. The chemical reaction in this context is expressed by Equation (8):



The variable *Products* stands for all the 53 species considered by the Cantera library.

The conceptual model is designed to replicate the processes occurring within a real combustor, despite not accounting for its actual geometry as it applies a zero-dimensional approach. The combustor is ideally divided in four main zones, as it can be seen in Figure 2. In the Pre-mixing zone (ZONE 1), the streams of CH₄, O₂ and CO₂ enter separately and create the reactants mixture. In the Primary zone (ZONE 2), the combustion starts and ends. In the Cooling zone (ZONE 2.5), a stream of CO₂ is used to cool the liner of the combustor and it takes a part of the heat of the combustion products. In the Dilution zone (ZONE 3), the CO₂ stream is utilized to cool the combustion chamber *CO₂_cool2* mixes with the products stream exiting the primary zone, before finally entering the turbine after dilution. The combustor as a system is considered adiabatic, but an adiabatic efficiency ($\eta_{cc}=0.85$) is introduced for the percentage of heat subtracted from the products exiting the Primary zone and removed by the CO₂ coolant in the Cooling zone.

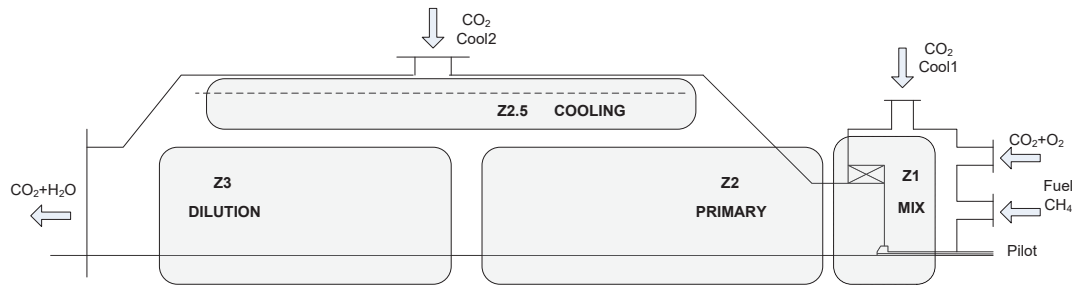


Figure 2: Conceptual layout of the combustor

The model allows performing a four-variable parametric analysis varying the following quantities: excess of CO_2 in the premixing (CO_2_{ox}), quantity of CO_2 dilution relatively to combustion products mass (CO_2_{cool}), T_1 and p_1 .

Regarding exergy analysis, the model calculates all the necessary exergy streams and transfers: exergy of reactants, exergy of CO_2 used as coolant, exergy of products, chemical exergy, exergy entering the system, exergy destructions of each section of the combustor, efficiencies in direct and indirect form. These quantities are computed by applying the formulations from Equation (11) to Equation (23):

$$Ex_{CH_4} = H_{CH_4, fis} - T_0 S_{CH_4, fis} \quad (11)$$

$$Ex_{CO_{2ox}} = H_{CO_{2ox}, fis} - T_0 S_{CO_{2ox}, fis} \quad (12)$$

$$Ex_{O_2} = H_{O_2, fis} - T_0 S_{O_2, fis} \quad (13)$$

$$Ex_{CO_{2cool2}} = H_{CO_{2cool2}, fis} - T_0 S_{CO_{2cool2}, fis} \quad (14)$$

$$Ex_{gas2} = H_{gas2, fis} - T_0 S_{gas2, fis} \quad (15)$$

$$\varepsilon = -\Delta G_{react} \quad (16)$$

$$Ex_{in} = Ex_{CH_4} + Ex_{CO_{2ox}} + Ex_{CO_{2cool}} + Ex_{O_2} + \varepsilon \quad (17)$$

$$Ex_{d, mix1} = T_0 (S_r - S_{CH_4} - S_{CO_{2ox}} - S_{O_2}) \quad (18)$$

$$Ex_{d, comb} = T_0 (S_p - S_r) \quad (19)$$

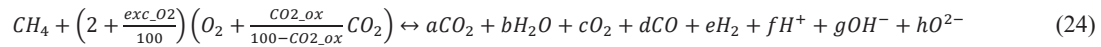
$$Ex_{d, HE} = T_0 (S_{CO_{2cool2}} - S_{CO_{2cool1}} - \frac{Q_{loss}}{T_p}) \quad (20)$$

$$Ex_{d, mix2} = T_0 (S_{gas2} - S_p - S_{CO_{2cool2}} - S_{O_2}) \quad (21)$$

$$\eta_{dir} = \frac{Ex_{gas2}}{Ex_{in}} \quad (22)$$

$$\eta_{ind} = 1 - \frac{Ex_{d, tot}}{Ex_{in}} \quad (23)$$

A smaller number of species in the products was included in the EES model (E-ICM, EES Incomplete Combustion Model), using input data from the results of Python. The reaction considered is defined by Equation (24):



The dissociated species included in Equation (24) are only those calculated in the P-ICM with a molar fraction in products higher than 0.1% under the $\text{CO}_2_{\text{ox}}=0$ conditions, which is the case with the highest dissociation effects. The stoichiometric coefficients of the products are calculated from the molar fractions of P-ICM, for each value of CO_2_{ox} . NASA libraries are used in E-ICM for a correct thermodynamic calculation of the dissociated species.

4 RESULTS

3.1 Results - Complete Combustion Model

A sensitivity analysis for E-CCM was performed varying T_1 and CO_2_{ox} , as shown in Figure 3. This last shows that T_p is decreased as CO_2_{ox} increases, and the same does the efficiency. This occurs because, with the same amount of fuel and oxygen, part of the heat generated is used to heat the inert CO_2 . Additionally, it can be observed that both quantities increase with T_1 . This is due to the fact that higher-temperature products are produced using higher-temperature reactants, as the heat released by the reaction is not influenced by pre-heating.

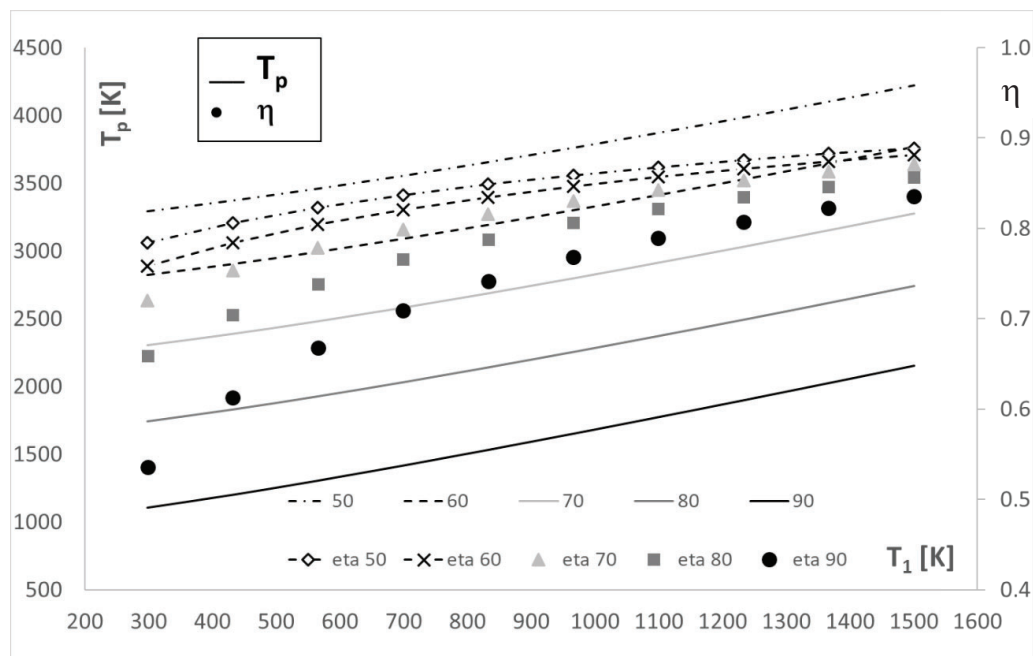


Figure 3: T_p and η varying CO_2_{ox} and T_1

3.2 EES and Python Incomplete Combustion Models

The results of the incomplete combustion models are presented below, allowing for a comparison with those of the complete combustion model explained in Subsection 3.1.

It is essential to examine the composition of the products, considering that the exit stream from the combustor represents the working fluid that expands in the turbine. After expansion, the mixture passes through the water separator. A portion of the dry working fluid, now only CO_2 , is then withdrawn and directed to sequestration. Therefore, understanding its composition is crucial for the operability of the plant. The first notable point is that there are more species among the products, not just CO_2 , H_2O , and any excess O_2 as in the E-CCM. For example, with $p_1=30$ MPa, $T_1=1000$ K, $\text{exc}_{\text{O}_2}=0$ and $\text{CO}_2_{\text{ox}}=50$, the main species predicted by the P-ICM approach are reported in Table 1.

Table 1: Main species analysis for the mixture of combustion products
($p_I=30$ MPa, $T_I=1000$ K, $exc_O_2=0$ and $CO_2_ox=50$)

Species	Molar fraction
H_2	0.88377
H^+	0.16194
O^{2-}	0.27558
O_2	4.31301
OH^-	2.51622
H_2O	35.39272
CO	9.21954
CO_2	47.21926

As expected, H_2O and CO_2 remain the predominant substances. The other substances present are CO , O_2 , OH^- , H_2 , O^{2-} and H^+ . Figure 4 shows the trend of the mixture composition with varying CO_2_ox . In Figure 5, the difference between the temperatures of the products calculated by the different models is illustrated. The incomplete combustion models (P-ICM and E-ICM) predict similar temperatures of the combustion products across the entire range of CO_2_ox . This demonstrates that a limited set of species (E-ICM) can be considered instead of the extensive list included in P-ICM. The complete and incomplete combustion models converge at high CO_2_ox levels which are above 80% as visible in Figure 5. If the chemical composition of the products (calculated in P-ICM) is analyzed, for CO_2_ox values higher than 80%, the molar concentration of all species other than CO_2 and H_2O is nearly zero, as confirmed in Figure 4. Regarding temperature prediction and energy balance, the complete combustion model is appropriate as long as CO_2_ox exceeds 80%. These conditions are applied in the Allam cycle to maintain the combustor temperature at levels comparable to those in modern gas turbine (GT) combustors.

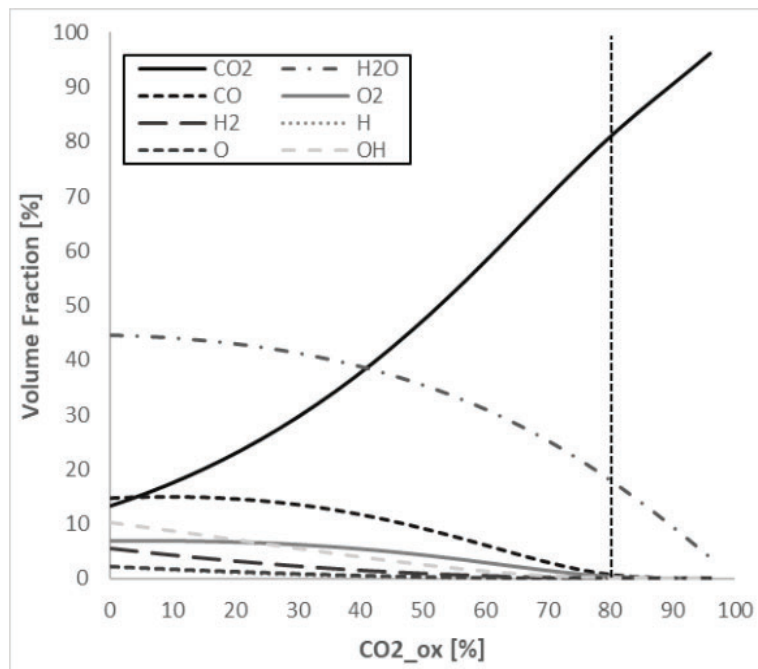


Figure 4: Chemical composition of the products (P-ICM)

In addition to the flame temperature, Figure 6 presents the trend of exergy efficiency with varying CO_2_{ox} for the incomplete combustion models P-ICM and E-ICM. The results indicate that both models agree within the design range for the Allam Cycle with CO_2_{ox} greater than 80% of total. However, for lower CO_2 recirculation rates, the simplified model E-ICM (which considers only a limited set of species) becomes inaccurate when compared to the P-ICM.

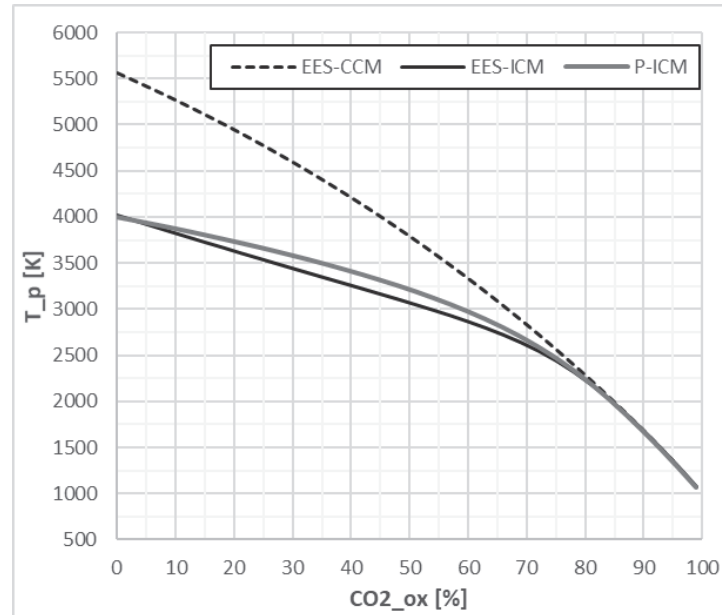


Figure 5: Trend of the product flame temperature in models E-CCM, P-ICM and E-ICM

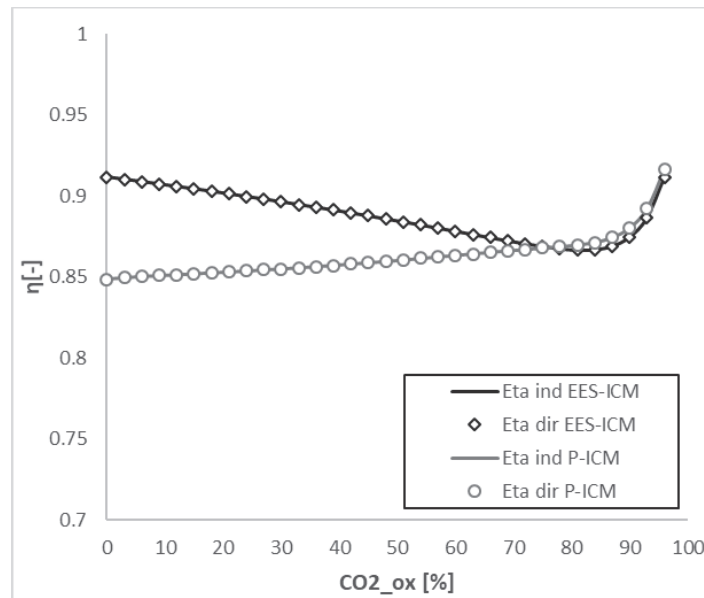


Figure 6: Comparison of exergy efficiencies in E-ICM and P-ICM. Direct and indirect modes of calculation are shown.

3.3 Combustor Design Chart (incomplete combustion)

Using the model described in Subsection 3.2, it is possible to investigate the potential ranges of the most important parameters for the initial design of the combustor in the Allam cycle.

The "degrees of freedom" of this model include the quantity of CO_2 that can be introduced in ZONE 1 (CO_2_{ox}) or ZONE 2.5 (CO_2_{cool2}), the excess oxygen (exc_{O_2}), the combustor pressure (p_1), and the preheating temperature (T_1) of the CO_2 and O_2 streams. The CH_4 stream temperature is maintained at 418 K, resulting from compressing the fuel from ambient pressure up to the combustor pressure.

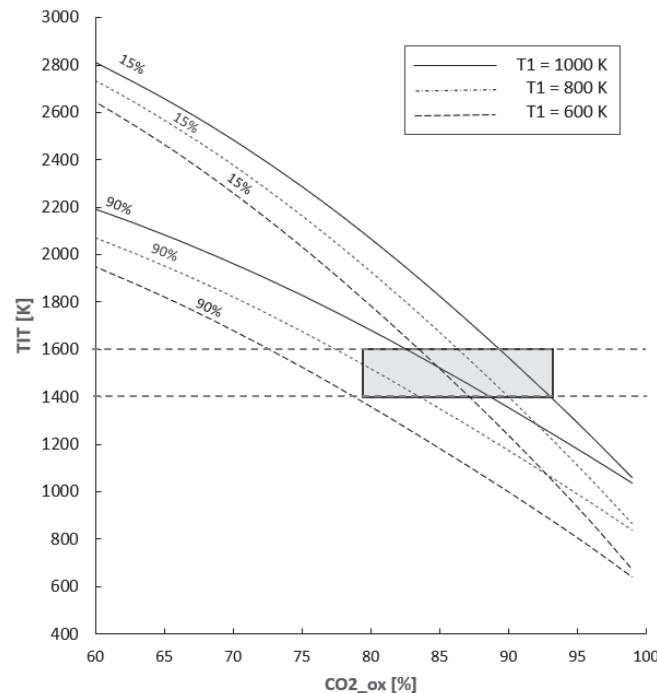


Figure 7: Trends of TIT as a function of CO_2_{ox} , CO_2_{cool} and T_1 . For each value of T_1 , the upper line is the case with $\text{CO}_2_{cool} = 15\%$, the lower one is the case with $\text{CO}_2_{cool} = 90\%$ ($p_1 = 300$ bar).

To narrow the scope of analysis, the first consideration is the turbine inlet temperature (TIT), for which Chan et al. (2023) identify a range between 1400 and 1600 K. The TIT must not be too high due to mechanical constraints of the turbine operability, but it also must not be too low, as the temperature of the hot gases entering the regenerator would be insufficient to preheat the other streams. The target range for the exit temperature, which can be evaluated as a function of CO_2_{cool} and T_1 in Figure 4, should always ensure that CO_2_{ox} is above 80% to achieve complete combustion. In Figure 7, the curves for three different reactant temperatures, T_1 equal to 1000 K, 800 K and 600 K are presented. For each temperature, an upper curve is drawn with CO_2_{cool} equal to 15%, and a lower one with CO_2_{cool} equal to 90%. Several operating conditions can be adjusted within the range between 15% and 90% for CO_2_{cool} . As anticipated, the higher the CO_2_{cool} , the lower the TIT, due to the increased injection of coolant into the combustion chamber. The shaded rectangle delineates the operational range of the

combustor: depending on T_i , various suitable combinations of CO_2_{ox} and CO_2_{cool} can be identified. To determine the optimal T_i , one must consider the exergy efficiency of the combustor, as depicted in Figure 8. As anticipated, higher values of T_i correspond to higher efficiencies. Additionally, it is preferable to operate at elevated levels of CO_2_{ox} , to increase efficiency.

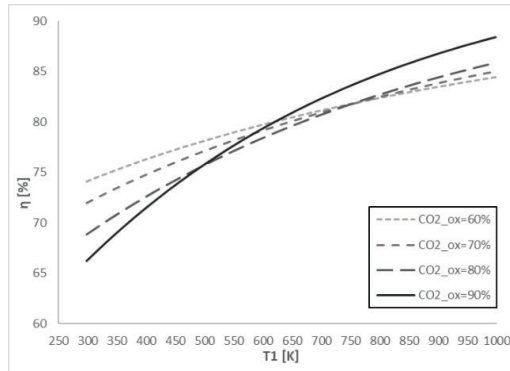


Figure 8: Trends of exergy efficiency versus T_i and CO_2_{ox} ($CO_2_{cool}=50\%$, $p_i=300$ bar).

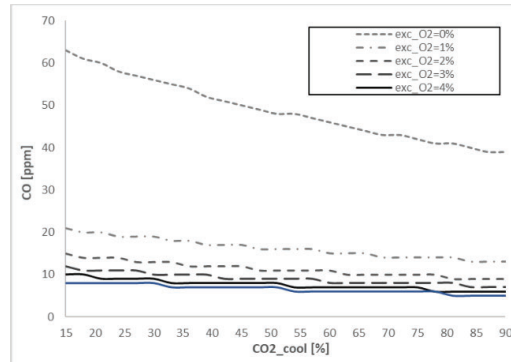


Figure 9: Trends of CO emissions as a function of CO_2_{cool} and exc_{O_2} ($CO_2_{cool}=90\%$, $T_i=1000K$, $p_i=300$ bar).

Given these considerations, the optimal design conditions would be at temperature T_i of 1000 K and CO_2_{ox} of 90%. This choice aligns with the recommended preheating range suggested by Chan et al. at a temperature between 973 and 1023 K. However, it is worth noting that the operability range narrows at high values of T_i according to Figure 7 with CO_2_{ox} between 84% and 92% with corresponding adjustments in the value of CO_2_{cool} . These operating conditions could represent a crucial aspect if the combustor operates in off-design functioning.

Analyzing the chemical composition of the hot gases exiting the combustor reveals a mixture predominantly composed of CO_2 and H_2O . However, it is crucial to regulate the presence of CO, which is the first dissociated species predicted by equilibrium models. The NO_x emissions are absent as the oxidant is pure oxygen. In conventional gas turbine power plants, CO emissions exceeding 10 ppm at the stack are rare. In the Allam cycle, where the CO_2 stream is either recirculated or captured for sequestration, CO levels should ideally be minimized, as the working fluid ideally comprises only CO_2 and H_2O . In Figure 9, predicted equilibrium values are analyzed as a function of CO_2_{cool} and exc_{O_2} . With 0% excess O_2 , the 10 ppm threshold is substantially exceeded with values over 40 ppm. Although it is not recommended to conduct combustion under stoichiometric conditions in the Allam cycle, a reasonable value could be 3%, as suggested in the literature and confirmed by the equilibrium model.

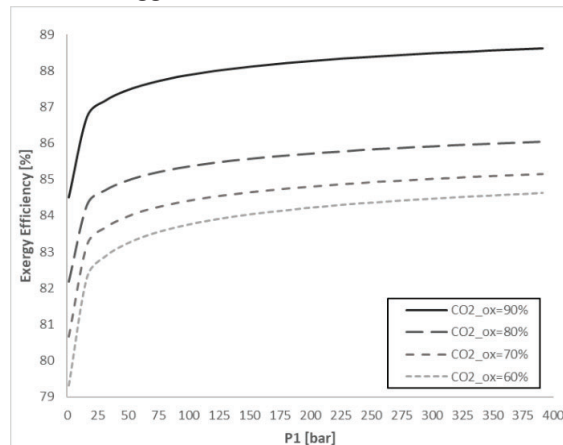


Figure 7: Trends of exergy efficiency as a function of p_i and CO_2_{ox} ($CO_2_{cool}=50\%$, $T_i=1000K$)

Lastly, all the results mentioned above are calculated with a reference pressure of the combustor of 300 bar. Figure 10 illustrates the trend of the exergy efficiency concerning p_1 and CO_2_{ox} . The model validates the appropriateness of selecting p_1 , as η_x decreases significantly below 200 bar. The potential advantage of operating at pressures higher than 300 bar seems to be marginal in comparison to technical constraints.

CONCLUSIONS

The consequences of dissociation on the thermodynamic performance were assessed by developing a model for the combustion process and comparing the results of complete combustion with equilibrium compositions. The findings confirm that the regenerative preheating arrangement of the Allam cycle for reactants is highly effective in reducing combustion exergy destruction. The disparities determined by equilibrium conditions are marginal, provided that the combustor is operated correctly at high CO_2 recycle rates with CO_2_{ox} greater than 80%.

The main conclusions are listed in the following:

- The results of the exergy analysis indicate that the highest efficiencies are attained when the operating pressure in the combustor is approximately 300 bar. Further increases result in technological complications and yield negligible gains.
- According to the parametric analysis, a design point was identified: compliance with TIT constraints and the attainment of optimal efficiencies necessitate operating with a preheating temperature of 1000 K with CO_2_{ox} equal to 90% in the oxidant mixture, and an appropriate value of CO_2_{cool} in the CO_2 stream used as a coolant, enabling precise adjustment of the TIT.
- Considering carbon capture and storage, it is advisable to minimize CO impurities by operating with a slight excess of oxygen. A reasonable value for this excess is 3%.

NOMENCLATURE

Acronyms

0D	Zero-dimensional
ASU	Air Separation Unit
CCS	Carbon Capture and Storage
CFD	Computational Fluid Dynamics
EES	Engineering Equation Solver
E-CCM	EES Complete Combustion Model
E-ICM	EES Incompl. Combustion Model
FOAK	First-of-a-kind
HHV	Higher Heating Value
LHV	Lower Heating Value
MAC	Main Air Compressor of ASU
MHE	Main Heat Exchanger of ASU
P-ICM	Python Incompl. Comb. Model
SNG	Substitute Natural Gas
TIT	Turbine Inlet Temperature

Subscripts

0, std	Standard Conditions
1	Preheating Conditions
ad	Adiabatic
cc	Combustion Chamber
d	Destruction
dir	Direct
fis	Physical
gas2	Combustor Outlet
in	Inlet
ind	Indirect
r	Reactants
p	Products

Symbols

CO ₂ _cool	Quantity of CO ₂ dilution relatively to combustion products mass [%]	Q	Heat [kJ]
CO ₂ _ox	Molar fraction of CO ₂ in the O ₂ -CO ₂ mixture [%]	R	Universal Gas Constant [kJ/kmol*K]
DG	Gibbs Free Energy [kJ]	S	Entropy [kJ/K]
Ex	Exergy [kJ]	T	Temperature [K]
exc_O ₂	Excess of oxygen [%]	X	Molar Fraction [-]
H	Enthalpy [kJ]	Y	Mass Fraction [-]
m	Mass [kg]	ε	Chemical Exergy [kJ]
MM	Molar Mass [kg/kmol]	η	Efficiency [-]
n	Number of Moles [kmol]	τ_{pf}	Residence time [s]
p	Pressure [kPa]	ϕ	Equivalence ratio [-]

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