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Presentation of an Innovative Zero-Emission Cycle for Mitigating the Global Climate Change

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Abstract

In the spectrum of possible options to cope with the global climate change, a novel technology based on the zero CO_2 emission MATIANT cycle (contraction of the names of the 2 designers : MATHIEU and IANTOVSKI) is presented here. This latter is basically a regenerative gas cycle operating on CO_2 as the working fluid and using O_2 as the fuel oxidiser in the combustion chambers. The cycle uses the highest temperatures and pressures compatible with the most advanced materials in the steam and gas turbines. In addition, reheat and staged compression with intercooling are used. Therefore the optimized cycle efficiency rises up to around 45% when operating on natural gas. A big asset of the system is its ability to remove totally the CO_2 produced in the combustion process in liquid or supercritical state and at high pressure, making it ready for transportation, for reuse or for final storage. It avoids the cost in performance (decrease of efficiency and power output) and in money of the CO_2 capture by a MEA scrubber. The assets and drawbacks of the cycle are mentioned. The technical issues for the design of a prototype plant are examined.

Key words: CO_2 emissions mitigation, CO_2/O_2 power cycles, zero emission power systems, CO_2 removal and storage.

1. Background

After the Kyoto global climate change summit in December 1997, a protocol involving economy against ecology was issued. According to this protocol by 2012 the developed countries would have to cut emissions of equivalent CO_2, namely CO_2, CH_4, N_2O, SF_6 and 2 fluorocarbons, by an average of 5.2% below their 1990 levels. Looking at the battery of available weapons to cope with the greenhouse effect, a sequence of successive stages of CO_2 mitigation methods can be adopted as follows:

1. Implementation of low cost technologies leading to a 'non-regret policy": the efficiency increase of fuel saving and energy supply and end-use. In this category, we find: cogeneration, increase of the share of renewables, switching from coal to gas as the fuel.

2. It is generally believed that the major drawback of the previous measures is their limited reduction potential. Therefore, more costly and more efficient CO_2 mitigation technologies have to be considered, namely CO_2 capture from the flue gas of conventional power plants and CO_2 sequestration, the possible revival of nuclear programmes with new reactor types, a large-scale energy production based on renewable energy (hydroelectricity, photovoltaic cells, biomass).

3. The CO_2 capture is feasible when it is carried out by end-of-pipe techniques, like MEA scrubbers installed in the flue gas. These techniques are very penalising on both the performance and cost of generated electricity. They ask for serious reductions in both costs and performance penalties (like for example, scrubbers with membranes as developed by STATOIL).

4. However, the nuclear alternative is met with public non acceptance whilst the renewables still cost too much and, therefore, CO_2 capture and storage remain the option to develop. In a very comprehensive analysis (Riemer et. al.,

1993) it was emphasized that storage is 4 to 5 times less costly than capture. This shows that the designer of a low emission CO_2 cycle has to concentrate his know-how and expertise on the cost in performance and money of CO_2 capture and, if it is possible, to get rid of this cost. In order to provide an idea of the interest to avoid this capture of CO_2, realistic Figures are given here for a 500MW$_e$ pulverized coal-fired steam plant with a 65% annual capacity factor (Herzog et al. 1997 and Herzog 1998). Without CO_2 capture, the generating cost is typically 4.6 cent/kWh$_e$. When using a MEA absorber/stripper with 90% capture efficiency, the power output is reduced by 20% (400MW$_e$ instead of 500). If the captured CO_2 is compressed up to 100 bar for transportation and storage, the increase in cost of generated electricity is 2.84 cent/kWh$_e$ (or 38%) or

2.84 cent/kWh$_e$/(0.828-0.104) kg CO_2/kWh$_e$
=39 US$/ton CO_2 **avoided**

where, 0.828 kg/kWh$_e$ is the CO_2 specific emission with coal for 500 MW$_e$ power output and 0.104 is 10% of that value for 400 MW$_e$ power output ($0.1 \times 0.828 \times 500/400$). For comparison, the current CO_2 tax in Norway of 50US$/ton CO_2 is avoided.

We present here one possible option meeting the requirement of avoiding the CO_2 capture: the zero-emission MATIANT cycle. It is of course not the only possible zero-emission cycle and additionally it can still be improved a lot. However, it has a major asset compared to others since it combines power generation and CO_2 storage.

As a consequence of the different Global Climate Change Summits, the CO_2 removal could be the near-future challenge for the energy world. Should it be considered as a basic requirement in the design of an energy system, then novel technologies burning fossil fuels (no disruption in fuel supply and use) might be more suitable than end-of-pipe techniques used in existing or new installations.

The MATIANT cycle presented here only claims to show that the challenge can be met. Very low emission cycles can be designed and this one is not only an example amongst others but also the starting point of more efficient environment-friendly systems, and hopefully at a cost comparable to that of a conventional plant burning the same fossil fuel. In this context, the attractiveness of a novel technology can be assessed for the four main decision factors:

- estimated costs (of electricity generation and of CO_2 emission reduction)
- confidence in its operation (feasibility, maturity of technology, complexity, novelty of process and materials)
- acceptance (safety, environment, social)
- applicability (CO_2 removal, CO_2 storage, capacity available)

2. Scope and Objective

Many approaches to the CO_2 emissions mitigation have been extensively investigated in the scientific literature (Blok et al., 1992, Riemer, 1993, Riemer et al, 1993, Riemer and Smith, 1995 and Manfrida, 1998). An original concept of zero-release of pollutants in the atmosphere is proposed here as a competitor with systems using MEA scrubbing or membrane techniques and cutting the CO_2 releases by 80 to 90%. The removed CO_2 is then disposed of in proper sites, like in aquifers, in deep oceans or in depleted oil and gas fields. Of course, zero-emission is an ideal and desirable target, however leakage is unavoidable. This is discussed here.

In this paper, two concepts are introduced:

i. The stack downwards. The common features to zero-emission cycles are the use of CO_2 as the working fluid and O_2 as the fuel oxidizer. In order not to release CO_2 in the atmosphere, we make a knot in the stack or better, we turn the power plant upside down (see *Figure.1*), so the emissions are avoided and the combustion products are dealt with as effluents, possibly contaminated with NO_X, SO_X, particles, and toxics. The objective is to keep the effluents under total control. Such zero-emission cycles all require an air separation unit (ASU) (Iantovski et al., 1997 a,b,c).

Let us mention that a Rankine cycle operating on CO_2 instead of H_2O with external combustion was proposed for the first time by Hochstein (1940). An improvement of this concept with *internal* combustion was proposed much later by Lorentzen and Pettersen (1990). The MATIANT cycle combines the both.

ii. Grave-into-the-cradle. We take the fossil fuels from the bowels of the earth, so we must also send back the products of their combustion (Marchetti, 1979).

In the MATIANT concept, the removed CO_2

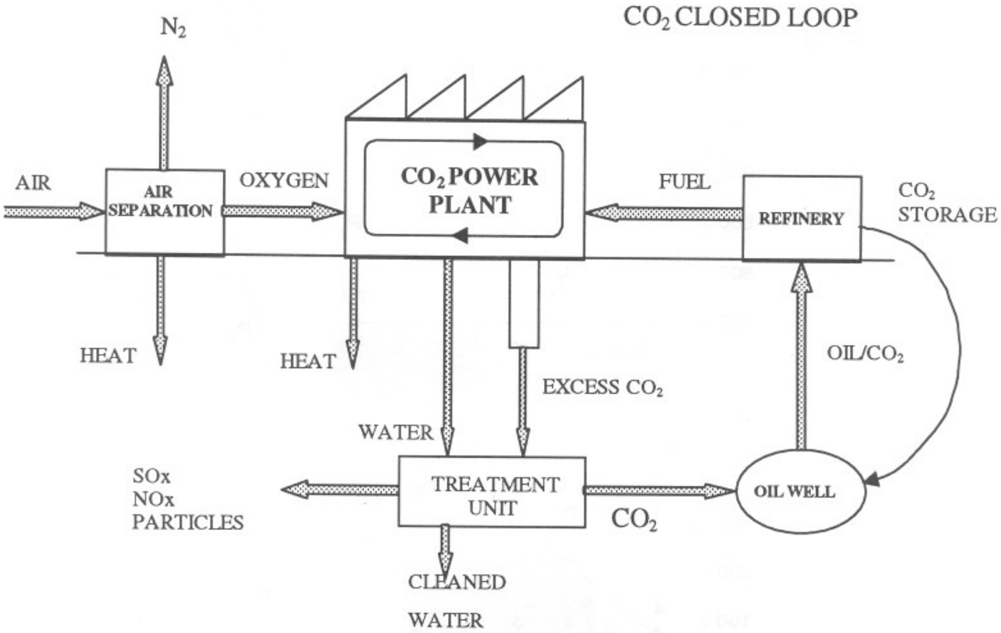


Figure 1: stack downwards and grave-into-the-cradle concepts

flow is liquid and at high pressure. We can consequently use it in applications where pressurized liquid CO_2 is required, as is the case in extraction of oil. CO_2 is used to increase the oil recovery efficiency. The higher the CO_2 injection pressure, the higher this efficiency. The oil well is then more depleted and the CO_2, separated from the extracted mixture CO_2/oil mixture, can be reinjected in the depleted well. Hence the waste (CO_2) in its final storage is replacing the fuel which produced it in the fuel site. So a closed fuel/CO_2 cycle is set up (*Figure.1*). This has to be considered as a concept, the technical feasibility and the associated technical issues are the subject of a separate study.

3. Cycle Diagram and Layout of the Plant

The MATIANT gas cycle is shown on the T-s diagram in *Figure 2* and the layout of the corresponding plant is in *Figure.3* (Iantovski and Mathieu, 1996, Iantovski et al. 1996, Iantovski et al. 1997a). It comprises a supercritical part (2-3-4-5-6) combined with a regenerative CO_2 Brayton cycle with reheat (6-7-8-9-10-11-12-1-2).

The combustion takes place along the isobar 7-8. The fuel is injected at the proper pressure P_2 in the burners as well as the mixture of the working fluid CO_2 and O_2 at 7. At exit 8 of the combustion chamber (1300°C), the mixture CO_2/H_2O is expanded along 8-9 and then reheated up to the temperature 10 in a second combustion chamber where other fractions of fuel and O_2 are injected at P_3. At 10, the fluid contains the combustion products in stoichiometric proportions, namely 8% CO_2 and 6% H_2O in addition to the 100% CO_2 circulating along the total cycle. This fluid is expanded along 10-11. These two expanders produce electricity. The fluid is then cooled from 11 to 12 in a recuperator where it gives up its heat to 2. CO_2 gas flows first at the higher cycle pressure P_1 and after expansion in 5-6 at the pressure P_2 of the combustion chamber. CO_2 is expanded along 5-6, in a steam-like turbine and generates electricity. As to O_2, its total throughput is 10% of the CO_2 mass flow rate which is around 7% and is injected at 7 and another 3% is injected at 9 for the reheat process. The O_2 can be reheated in the recuperator from its temperature at the ASU outlet up to the temperature at the inlet of the burners 7 and 9 of the combustion chambers.

At the recuperator outlet, the water is condensed in a cooler and extracted in a CO_2/H_2O separator. A very small amount of CO_2 (around 0,5% of the total flow) is dissolved in the removed water, (Rasmussen, 1998).

The remaining gas is then CO_2 and it is compressed in a 4 staged compressor with intercooling along 1-4 above the CO_2 saturation line in order to carry out a compression as close as possible to isothermal as possible.

At 4, the excess CO_2 (about 8% of the recirculated CO_2 mass flow rate) is removed in

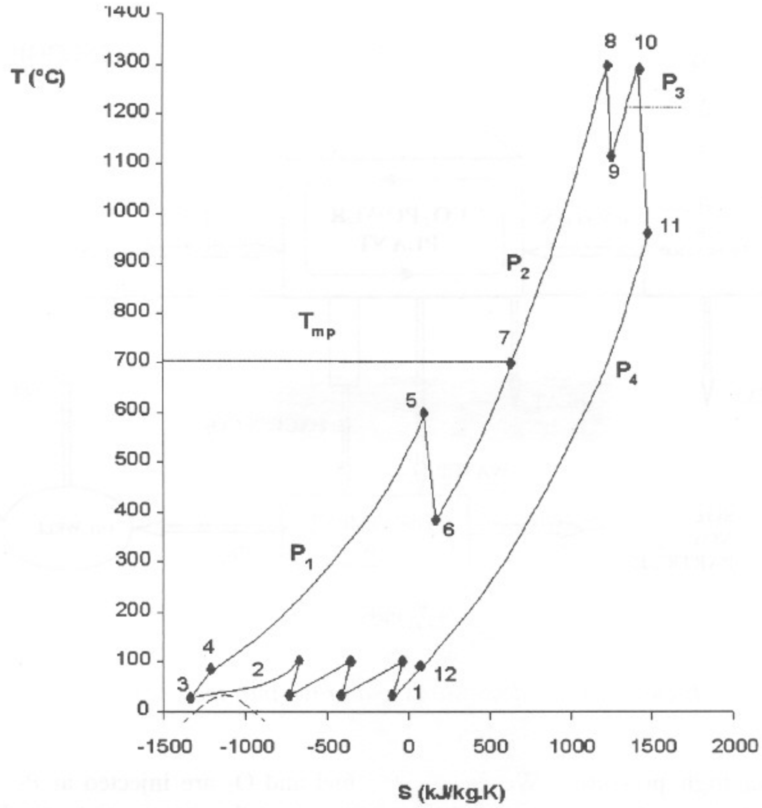
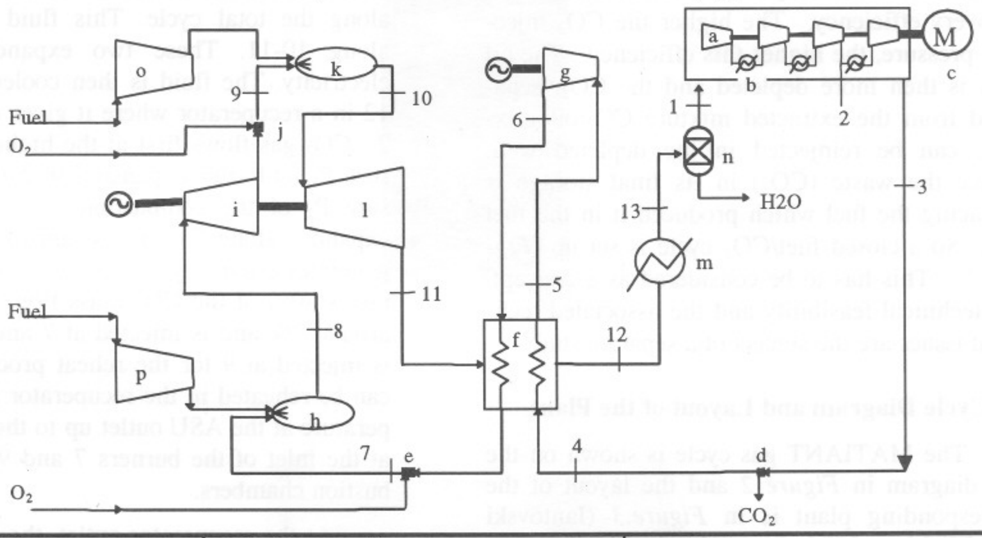


Figure 2 : Cycle representation in (T-s)
The CO_2 saturation curve is at the bottom (left)



a : CO_2 compressor	b : intercooling	c : driving motor
d : splitter	e : mixer	f : recuperator
g : uncooled HP expander	h : cooled combustion chamber 1	i : MP expander with internal cooling
j : mixer	k : cooled combustion chamber 2	l : LP expander with internal cooling
m : cooler	n : water separator unit	o and p : fuel compressors

Figure 3: Layout of the plant

liquid or supercritical state through a valve, without any energy consuming and costly system as it is the case in a CO_2 scrubber or membranes installed in the flue gas.

Basically, the effluents, possibly contaminated, remain under control. At this stage of the modeling, the cooling of the hot parts of the cycle is not included yet but in this analysis, a cycle efficiency penalty of 2.5 percentage points to take it into account is adopted similar to an air gas turbine, (Bolland et Saether 1992 and 1993).

Of course, the fluid in the recuperator 11-12 contains small quantities of Ar and N_2 coming from the ASU and of O_2, not used in the combustion process because an excess of oxygen is necessary in practice to make the combustion complete (no CO in the flue gas). They are present in the cycle and have an impact on the efficiency. However, the used ASU delivers O_2 at a purity of 99.5% with a specific electricity consumption of 0.28 kWh/Kg O_2 at 5 bar. In the natural gas, the major part of non-condensable gases are N_2 and CO_2, depending on its composition. The adiabatic exponent γ of N_2 and Ar is much higher than that of CO_2, and so it is for the resulting mixture. The compression requires more electricity than with pure CO_2 but more power is provided in the high temperature expanders 8-9 and 10-11. The net resulting effect on the cycle efficiency can be neglected because the accumulation of Ar in the cycle remains lower than about 1% of the total mass flow rate and hence does not change significantly the physical properties (C_p,γ, molar mass) of the working fluid.

Improvement

In a previous configuration (Iantovski and Mathieu 1996, Iantovski et al. 1996 and Iantovski et al. 1997), a 3-stage intercooled compressor was used to bring the working fluid from P_4 up to the CO_2 saturation pressure (70 bar) at 30°C. Then CO_2 was condensed at 70 bar and pumped in a liquid state up to P_1 making a CO_2 Rankine-like cycle. In order to avoid an accumulation of the non-condensable gases in the cycle, they are vented out of the condenser from a cold zone. However, the CO_2 vapour is partly carried away with the extracted gases and its amount depends on the partial pressures, the gaseous mixture composition, the solubilities of the gases in liquid CO_2 and the temperature of the subcooled condensate. With CO_2, the entrained amount is higher than in a conventional steam/water condenser with air extraction. Consequently, in order to achieve zero emission, the CO_2 has to be recycled or recovered using devices like scrubbers or membranes already used for CO_2 removal from flue gases at large scale. To avoid the large associated additional costs, we decided to get rid of the condenser and to use a 4-stage intercooled compressor, the working fluid being cooled in the last intercooler along a supercritical isobar (80 bar) that runs quasi horizontally above the critical point (73 bar, 31°C) and down to the cold source temperature (point 3 in *Figure 2*). All in all, the mean integrated temperature of the heat sink is hardly different than that obtained with a condenser. The improvement here is twofold: -the compression from P_4 to P_1 without penetrating into the CO_2 saturation line and -economy of an additional component, the condenser, with two separate phases.

4. Optimal Design Point

Assumptions and data used for the calculation are described below :

The data are summarised in TABLE I.The fuel is natural gas. It is not pure methane but a mixture of CH_4, ethane, propane, CO_2 and other gas traces, whose Low Heating Value, in Europe anyway, is around 42 MJ/kg.

In order to comply with the admissible mechanical stress criteria for the advanced materials used here, the high pressure level is limited to 300 bar and the temperature of this high pressure gas flow at the recuperator outlet is limited to 600°C (point 5 in *Figure 2*). The higher the pressure, the lower this temperature limit.

Similarly, as the pressure P_2 is varied between 40 and 100 bar, the temperature T_{mp} at the recuperator outlet (point 7 in *Figure. 2*) has to be limited to 700°C but should the materials be able to resist, this latter could be further increased. However, attention must be paid that, at this temperature, there is no self-inflammation of the mixture CH_4/O_2 (Rasmussen, 1998).

Due to the limitation of the cooling techniques and of the materials resistance, the temperature inlet of the MP and LP expanders is limited to 1300°C.

Typical values of the pressure losses in the heat exchangers are taken as 5% of the inlet pressure and 3% in the combustion chambers. Pressure losses in mixers, splitters et cetera are also taken into account. Having not included a model of the cooling yet, it is assumed that the cycle efficiency penalty due to the cooling of the hot parts of the system is equal to 2.5 percentage points in every case (Bolland and Saether, 1992).

The calculations are carried out using the commercial ASPEN PLUS code.

Results of the optimisation:

Figure 4 shows 2 curves, the upper one corresponding to an isentropic effectiveness of

TABLE I. Data for the calculations.

Upper intermediate pressure (P_2)	40 bar	High pressure of the cycle (P_1)	300 bar
ΔT pinch at the recuperator outlet	20 °C	Low pressure of the cycle (P_4)	1 bar
HP expander inlet temperature	600 °C	$\varepsilon_{S,E}$ of the 3 expanders	0.87
LP & MP expander inlet temperature	1300 °C	$\varepsilon_{S,C}$ of the O_2 compressors	0.75
Cycle low temperature	30 °C	$\varepsilon_{S,C}$ of the fuel compressors	0.75
$\varepsilon_{S,C}$ of the three first stages of the CO_2 compressor: 0.85 (axial compressor.) $\varepsilon_{S,C}$ of the last stage of the CO_2 compressor: 0.8			

0.9 for the compression without taking into account the cooling of the hot parts whilst the lower one corresponds to an isentropic effectiveness of 0.75, the cooling being taken into account.

The optimisation process provides a P_3 of 9.7 bar. The CO_2 multi-stage compressor isentropic effectiveness is taken as 0.85 for the three first stages and 0.8 for the last one.

In this modelling, the fuel is considered to feed the burners at ambient temperature (17°C) and at 3 bar. The sensitivity of the cycle η is very small to both of these latter quantities. In reality, the delivery pressure of the natural gas depends on the network and in Europe in particular, this pressure can be as high as 40 or 60 bar. Consequently, depending on P_2, the fuel compression can be low or even zero. If the combustion chamber is at a pressure lower than that of the network, a recovery in an expander is possible. In this case however, a preheat of the fuel in the recuperator is required before its injection in the burners.

On the cold side, our calculations are made with a low cycle temperature of 30°C. However, in very cold countries like Denmark, Norway, Sweden and Finland, the heat sink is more favourable (less than 10°C in the sea water).

For 1 kg/s of CO_2 recirculated, the fuel consumption is equal to 0.0328 kg/s, the O_2 consumption is equal to 0.1106 kg/s, and the mass flow rates of CO_2 and water removed from the cycle are 0.0815 kg/s and 0.0618 kg/s, respectively.

The energy consumption and production in kJ/kg of recirculated CO_2 are given in TABLE II.

Figure 5 shows the share of the electricity consumptions that are equal to 47% of the total production. The compressor appears to be the biggest exergy consumer and, hence, especially requires a careful design.

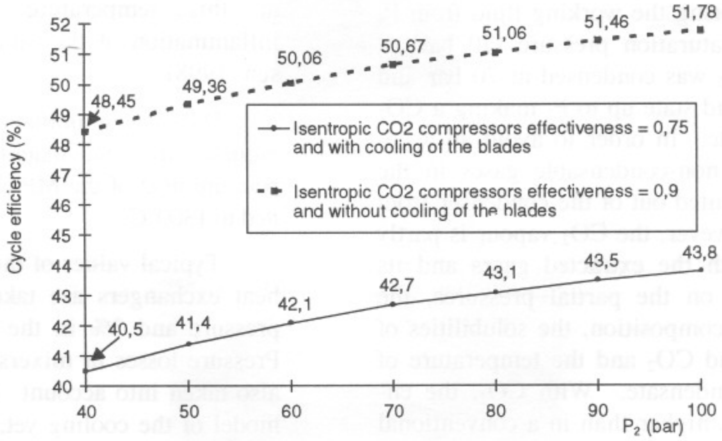


Figure 4: Cycle efficiency versus P_2 for 2 values of $\varepsilon_{s,C}$

TABLE II. (kJ/kg of recirculated CO_2)

Fuel compressors consumption	15.9	HP expander production	244.5
O_2 compressors consumption	34.8	MP expander production	362.1
ASU consumption (O2 at 5bar, 99.5%)	112.3	LP expander production	611.7
CO_2 intercooled compressor consumption	412.5		
Total consumption	**575.5**	**Total production**	**1218**

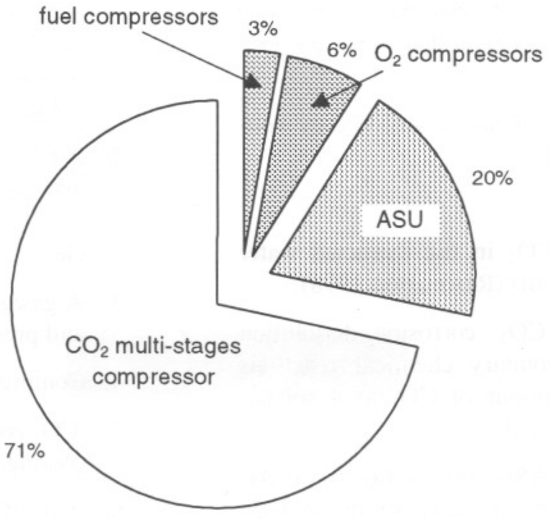


Figure 5: Share of the total electricity consumption (47% of the total production)

The net cycle efficiency is obtained by

$$\frac{W_{prod.} - W_{cons.}}{m_{fuel}\ LHV_{fuel}} - (\text{cooling penalty})$$
$$= \frac{1218 - 575.5}{0.0328 \times 42000} - 2.5 = 44.2\ \%$$

when the cooling demand induces an efficiency penalty of 2.5 percentage points and the ASU consumption, taken from ASU manufacturers' data, is 0.28 kWh/kg O_2 at a purity of 99.5% and at 5 bar.

The compression of the gaseous O_2 at 5 bar up to the pressure of the second combustion chamber (P_3=9.7 bar) adds an electricity consumption of 0.03 kWh/kg O_2 and of 0.12 kWh/kg O_2 up to 40 bar. So taking into account the respective mass flows of oxygen at P_2 and P_3, the total consumption due to the production and the compression of the O_2 injected in the combustion chambers is 0.37kWh/kg O_2.

Let us mention that in the low temperature range (0-300°C) and at pressures near the CO_2 saturation line (around 60 bar), the compressor consumption is reduced with respect to that of an ideal gas and that non-ideality behaviour nearly offsets the ASU consumption at the design point.

5. Technical Advantages and Drawbacks

We have modelled CC plants (reference case) using CO_2 scrubbers for CO_2 removal from the flue gas without and with flue gas recirculation. CO_2 removal from the flue gas by an absorber/stripper system using MEA is considered currently as the state-of-the-art technology.

In addition to be very costly, this chemical plant installed at the exhaust of the plant leads to an efficiency penalty typically of the order of 6 to 8 percentage points for a natural gas fueled CC plant. As far as zero-emission is concerned, we have studied CO_2 semi-closed CC cycles (Mathieu and Deruyck 1993). It is shown (Bolland and Mathieu, 1997) that a CC with recirculation has better performance and lower cost for CO_2 scrubbing than a semi-closed CO_2 cycle of the same type. The MATIANT cycle has been designed as a zero-emission system, like the semi-closed CO_2 CC, but without using a heat recovery boiler and a Rankine cycle.

The assets and drawbacks of the MATIANT cycle are notably the following ones

Assets :

1) good net cycle efficiency (between 40 and 45% when burning natural gas).

2) negligible release of pollutants in the atmosphere.

3) avoidance of a CO_2 scrubber and removal of a highly pressurized liquid CO_2 flow and possible reuse or final storage without further compression It is the biggest difference with a semi-closed CO_2 CC.

4) no use of an industrial gas turbine as such; the compressor and expanders are sepa-

rated from each others, so there is no danger of surge of the compressor, especially when the LHV of the fuel is lower than that of natural gas, like that of a syngas.

5) no need of expensive and energy consuming CO_2 scrubber.

Drawbacks :

1) leakage of CO_2 in the extracted water (solubility is very small) (Rasmussen 1998).

2) chemistry of CO_2 : corrosion, dissolution in other fluids, secondary chemical reactions with materials; behaviour of CO_2 as a solvent have to be well modelled.

3) requires an ASU (99.5% O_2; 0.5% Ar, traces of N_2); electrical consumption of 0.25 kWh/kg O_2 at 1 bar.

4) net cycle efficiency very sensitive to inefficiencies, in particular compressor effectiveness.

6. Technical Issues and Design of a Prototype Plant

The ASU has to be integrated in the cycle and in particular the flow of N_2 or maybe the rejected heat. A new design or a redesign of known industrial components is required when they operate on CO_2 instead of air or water.

Components already available or needing adaptations:

1. A compressor whose design is very important. (Indeed, as shown in *Figure 6*. 6, the cycle efficiency is very sensitive to the compressor effectiveness. As an illustration, a decrease of this latter from 0.9 to 0.75 provokes an efficiency drop of around 5.5 percentage points.)
2. A CO_2 expander without cooling at 600°C, 300 bar, similar to the high pressure section of an UltraSuperCritical Rankine cycle,
3. A gas/gas recuperator at high temperatures and pressures.
4. Compression of a supercritical fluid.
5. CO_2 condenser and intercoolers with water cooling circuits.
6. Water/CO_2 cooler and separator.

Components requiring a new design:

1. Combustion chamber using O_2/CO_2 instead of air.
2. Temperature expanders (1300°C) with internal cooling of the blades and vanes. (The design of gas turbines has previously been investigated, (Mathieu 1994 a and b, Mathieu, 1995), with a very simplified one dimensional model. It is a major issue that can affect the performance. Further development is absolutely needed here.)

We are currently integrating the cooling of the hot parts of the cycle in the model. When natural gas is replaced by a liquid or a solid fuel, an integration of a gasification unit and a clean-

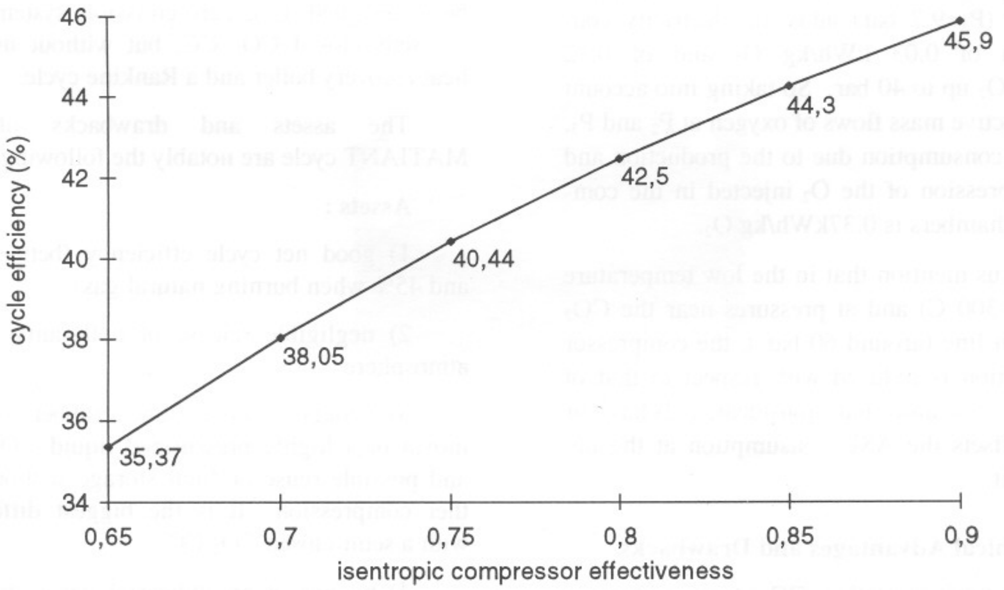


Figure. 6 :Cycle efficiency versus the average compressor effectiveness up system of the syngas in the MATIANT plant is required. This is the following development of the present study and this will modify the design of the cycle discussed here above.

7. Conclusions

The MATIANT concept leads to a power plant design targeting high performance and no releases of pollutants in the atmosphere.

Target 1: Cycle efficiency is very sensitive to inefficiencies and may vary from some 48-49% down to 40-41% depending on the values of the parameters. An efficiency of around 50% is obtained when the fuel is pure methane, when the effectivenesses of the turbomachines are close to 0.9, when the pressure losses are 2 to 3% of the inlet pressure through the heat exchangers and when the cooling of the hot parts is not taken into account. Compressions of oxygen and of the fuel as well as the ASU consumption play also a significant role. *The target is 45%.*

Target 2 : Zero-Emission corresponds to an ideal system, without leakage. With real fluids and components, some leakage is unavoidable. Leaks have to be minimised and recovered, in particular, where fluids are extracted. This is the case in the cooler/separator, with the CO_2 dissolved in the extracted water (around 0,5% of the total throughput). To achieve zero-emission, this gaseous water has to be treated or sent to the storage with CO_2.

Target 3 : Compression of CO_2 from around 80 bar to 300 bar in liquid and supercritical state.

Target 4 : System simulation has to be made with real fluids in real conditions.

Target 5 : A prototype (with zero power and zero efficiency) to demonstrate the technical feasibility of a zero-emission cycle and evaluate the costs has to be built.

Several applications are obtained by combination with other systems like a gasification unit, an MHD unit, heat pumps, chemical recuperators, oil recovery installations. The concept is also used with technologies like various regimes), of cost, of environmental impact ?

There are at least two good reasons to go to zero-emission plants on top of those mentioned above:

1. It is easier to remove the CO_2 in centralized production power plants than in disperse sources like cars. Now the transport sector can use vehicles consuming electricity or hydrogen in fuel cells without releasing the CO_2 associated with the combustion, even very efficiently, of fossil or bio-fuels.
2. After Kyoto, the implementation of a market of pollution permits was proposed. Each time you release less CO_2 than your assigned limit, you can get money on a CO_2 market.

Therefore, if an efficient zero-emission plant is a requirement for complying with the total CO_2 emissions allowed for a country, it is time to design a pilot plant now and this is an appeal towards the potential financial supporters.

Nomenclature

ASU	Air separation unit
CC	Combined cycle
GT	Gas turbine
HP or P_1	High pressure (bar)
LHV	Low heating value (MJ/kg)
LP or P_4	Low pressure (bar)
MEA	Mono ethanol amine
MP orP_2	Medium pressure (bar)
P_3	Reheat pressure (bar)
TIT	Turbine inlet temperature (°C)
T_{mp}	Outlet recuperator temperature at P_2 (°C)
$\varepsilon_{s,c}$	Isentropic effectiveness of the compressor
$\varepsilon_{s,E}$	Isentropic effectiveness of the expander

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