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Zecomix: a zero-emissions coal power plant, based on hydro-gasification, CO₂ capture by calcium looping and semi-closed high temperature steam cycle

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Abstract

This paper analyzes various aspects of the Zecomix cycle, a novel coal fired power plant, based on various innovative processes to achieve elevated efficiency and zero-emissions. A coal hydro-gasification process, using recycled steam and hydrogen as gasifying agents, converts carbon to CH_4 , which is then processed by two carbonator reactors where CH_4 , mixed with steam and contacted with calcium oxide, is converted to an H_2/H_2O based syngas while CO_2 is absorbed by the solid sorbent generating $CaCO_3$. The synthetic fuel produced in the chemical island is burned with oxygen in a semi-closed high temperature steam cycle, with a rather complex supercritical heat recovery steam cycle.

The main relevant operating parameters for the chemical island are varied in order to evaluate their effect on plant performance and to optimize the process. In addition, the paper presents a rather detailed analysis of some critical issues, often neglected in previous works from the literature. Net plant efficiency of 44-47% with a virtually complete carbon capture was predicted, a very interesting results with respect to other proposed coal power plants with carbon capture. The high complexity of the chemical island and the importance of a good sorbent performance in the critical conditions typical of this plant should be however taken into account for a fair comparison with other plant concepts.

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Keywords: Zecomix; coal; hydro-gasification; CO2 capture; calcium sorbent

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1. Introduction

In the last years, carbon capture and storage (CCS) technologies, applied to coal power plants, are encountering an increasing popularity. Demonstration plants are now under final planning and development. Being the first-of-a-kind, low-risk capture technologies are adopted, namely: (i) pre-combustion capture, including syngas production from coal or natural gas, water-shift reaction, CO_2 absorption, hydrogen-rich syngas combustion in a gas turbine, (ii) amine capture from flue gases of conventional power plants. A third technology (oxy-combustion) is also considered in many studies, particularly applied to pulverized coal boilers. These processes, even if introducing new milestones in the power industry, are characterized by a relevant loss of efficiency with respect to the reference power plants without capture (e.g. 33-37% vs. 43-45% for coal power plants), yielding to elevated costs of the CO_2 sequestrated.

In a scenario projected to mid-long term applications, a number of alternative processes can be proposed, aimed to improve the performance (efficiency, emissions, economics) of coal power plants with CCS. The process here studied is representative of novel concepts to achieve these goals (coal gasification by steam and hydrogen, CO₂ removal by calcium looping, semi-closed oxy-fuel power cycle). The paper presents a rather detailed analysis of the thermodynamic aspects of the process, with the scope of assessing its potential performance in terms of efficiency and emissions. We will outline the necessity of significant advances, with respect to today's best technology, to make the system viable for industrial application while conserving superior performance, including the development of components not available in today's power industry, provided that they can be developed by resorting to known technologies, but excluding unproven devices whose feasibility cannot be anticipated.

2. Zecomix plant description

ZECOMIX is the acronym of Zero Emission COal MIXed technology, a project leaded by ENEA (an Italian public agency operating in energy, environment and new technologies fields) with the participation of the industrial partner Ansaldo and some Italian universities. This technology promises very high conversion efficiency with zero carbon dioxide emissions. An experimental facility is being built to demonstrate the feasibility of the process at the ENEA Research Centre of Casaccia, near Rome [1].

The schematic of the plant is shown in Fig.1. Four sections can be depicted: (i) the chemical island, where an hydrogen rich syngas is produced, (ii) the power island, made of a high temperature steam cycle obtained via syngas oxy-firing and a lower temperature heat recovery steam cycle, (iii) an air separation unit (ASU) section, where oxygen is produced in a double reboiler distillation process and (iv) the CO_2 compression and liquefaction section.

2.1. Chemical island

The basic idea of the chemical island is the coupling between a hydro-gasifier and a carbonator reactor. The hydro-gasifier is fed by slurry and a hydrogen rich stream, recirculated from the carbonator outlet. It operates at pressures of 30-70 bar and temperatures of $700-1000^{\circ}$ C. Here char is gasified producing mainly methane according to the exothermic hydro-gasification reaction¹ (1):

$$C + 2H_2 \rightarrow CH_4$$
 $\Delta H^\circ_r = -74.6 \text{ kJ/mol}$ (1)

In the hydro-gasifier the following other reactions (2-6), both endothermic and exothermic occur:

¹ Values of heat of reaction are referred to the standard state (25°C, 1 bar) with H₂O as steam.



Figure 1 – Schematic of the Zecomix power plant (HG = hydro-gasifier; Cal = calciner; Carb = carbonator; CEHR = calciner exhausts heat recovery; Ex = syngas expander; SC = steam compressor; HTT = high temperature turbine)

$C + CO_2 \rightarrow 2CO$	$\Delta H^{\circ}_{r} = +172.5 \text{ kJ/mol}$	(2)
$C + H_2O \rightarrow CO + H_2$	$\Delta H^{\circ}_{r} = +131.3 \text{ kJ/mol}$	(3)
$2C + O_2 \rightarrow 2CO$	$\Delta H^{\circ}_{r} = -221.0 \text{ kJ/mol}$	(4)
$CH_4 + H_2O \rightarrow CO + 3H_2$	$\Delta H^{\circ}_{r} = +205.9 \text{ kJ/mol}$	(5)
$\rm CO + H_2O \rightarrow CO_2 + H_2$	$\Delta H^{\circ}_{r} = -41.2 \text{ kJ/mol}$	(6)

The stream exiting the hydro-gasifier enters the first carbonator reactor, where it is contacted with solid CaO and carbon dioxide is captured producing limestone, according to the exothermic reaction (7):

$$CaO + CO_2 \rightarrow CaCO_3$$
 $\Delta H^\circ_r = -179.2 \text{ kJ/mol}$ (7)

The removal of CO_2 from the gaseous phase pushes rightwards the water-gas shift reaction (6) and consequently the steam-methane reforming reaction (5), producing an H₂ rich syngas.

It must be highlighted that the following overall reaction (8) obtained from the single reactions (5), (6) and (7) is very well balanced from a thermal point of view.

$$CH_4 + 2H_2O + CaO \rightarrow 4H_2 + CaCO_3 \quad \Delta H^\circ_r = -14.5 \text{ kJ/mol}$$
(8)

In other words, not only the carbonation reaction facilitates hydrogen production by removing CO_2 from the gaseous phase, but also provides the heat required for the steam reforming reaction, allowing for the use of an adiabatic reactor. A large amount of the syngas exiting this carbonator is partly recirculated to the hydro-gasifier, while the remaining is sent to a second carbonator, where most of the residual carbon in gaseous phase is removed by further steam addition and the final H_2/H_2O -based fuel is produced.

The last reactor in the chemical island is the calciner, where limestone is regenerated to calcium oxide by reversing reaction (7). A CO₂ rich stream ready for heat recovery, compression, liquefaction and final sequestration is produced. Calciner could theoretically operate either at nearly atmospheric pressure or at a pressure similar to the one of carbonators. Atmospheric pressure operation allows for a lower calcination temperature and consequently lower sorbent sintering. In addition, no ash melting problems arise and it is possible to use coal (and unconverted C from hydrogasifier) as heat source for calcination. On the other hand, important problems with solids pressurization will arise and higher consumptions for CO₂ compression would occur. Conversely, pressurized calcination requires higher temperatures which bring about quicker sorbent sintering and the need of an ash free fuel as heat source (melting of ash, certainly occurring at these temperatures, is not acceptable in a fluidized or fixed bed gas-solid reactor like the carbonator). High pressure calcination is assumed for the base case and heat for calcination is provided by refinery residuals (tar) oxy-firing. The usage of a part of the syngas produced as the fuel for the calciner brings about a very large loss of efficiency and it is not analyzed in this study.

2.2. Power island

The high temperature H_2/H_2O based syngas produced in the chemical island is cooled and purified from entrained solids before being expanded in a syngas expander. The cooling temperature is calculated in order to obtain the assumed value of 550°C at the GT combustor inlet (GE already offers a fuel system for fuel gas temperatures up to 538°C [2]). Syngas is then burned with oxygen and expanded in a High Temperature steam Turbine (HTT). In order to limit the combustor outlet temperature (COT) to acceptable values, steam from IP turbine outlet of the heat recovery steam cycle is used as temperature moderator. However, much more dilution steam is needed to limit COT and for blades cooling. Such dilution steam is obtained by recycling part of the steam at nearly atmospheric pressure, available after the HTT expansion and heat recovery. A steam compressor is needed for this task: therefore the power cycle is basically a semi-closed steam Joule cycle. The excess steam, not needed for dilution, is cooled to a proper temperature and expanded in a LP steam turbine. Condensate is pumped to supercritical pressure, superheated and re-heated by recovering heat from the HTT discharge. It is interesting to notice that the reheat steam turbine does not have an IP section because: (i) a large part of IP steam is needed by the hydro-gasification process, (ii) all the remainder is used as dilution steam in the combustor and then expanded in the HTT.

The incondensable species CO_2 , N_2 , Ar, O_2 , contained in the expanding flow, are removed from the condenser and from the deaerator, together with some steam. Rather than venting these gases to the atmosphere, this stream is sent to the calciner after a proper compression (incondensable recovery and compression is a usual practice in geothermal power plants, when using steam with relevant CO_2 content). In such a way, excess oxygen is fully recirculated in the system and used as oxidizer in the calciner, and CO_2 capture is complete, even if some carbonated compounds are present in the syngas. This improves the plant efficiency and virtually no carbon escapes the system.

3. Calculation tools and assumptions

Mass and energy balances and the overall performance have been predicted with GS (Gas-Steam cycles) computer code, developed at Dipartimento di Energia of Politecnico di Milano. The code is a powerful and flexible tool that can be used to accurately predict the performances of a very wide variety of chemical processes and systems for electricity production. GS code was originally designed to calculate gas-steam cycles for power production [3-5] and has been progressively extended and developed to calculate complex systems including gasification processes [6-7], chemical reactors [8-9], fuel cells [10] and essentially any kind of plant for power generation from fossil fuels [11-12]. Ideal behavior is assumed for all the mixtures whose thermodynamic properties are calculated by means of NASA polynomials [13] based on JANAF tables data [14] and on NASA thermodynamic tables [15] for solid calcium based compounds. Pure water/steam is the only compound treated as real fluid according to S.I. tables [16]. Gas composition at reactors outlet is determined by assuming chemical equilibrium, calculated with the model originally developed by Reynolds [17], implemented in the code. For CO₂ compression process, where real gas effect and some other properties like gas solubility which cannot be reproduced by the GS code occur, the commercial tool Aspen Plus[®] Engineering Suite release 2004.1 was used, with Peng-Robinson equation of state.

Low sulfur South African coal is used, similar to that imported in Italy for power generation. Its composition is: 64.4% C, 3.95% H, 7.40% O, 1.49% N, 0.85% S, 9.40% H₂O, 12.67% Ash; 24.62 MJ/kg LHV. The main assumptions used for plant simulation are summarized in Tab.1.

For the cycles here selected, the HTT operates at gas conditions not far from the most advanced gas turbines now present in the industrial market (turbine inlet temperature of 1400-1500°C). However, there are significant differences between combustion gases and steam, as far as the heat transfer properties are concerned. In fact, the higher thermal conductivity and specific heat of steam [18] bring about a higher heat transfer coefficient and, therefore, a higher heat flow (thermal power exchanged per unit surface of cooled blade) at similar temperatures. In order to limit coolant need, the first two stages rows are cooled by means of the high pressure steam flow taken from the compressor outlet and cooled to 250°C. Nearly impulse design was adopted for these stages, to lower the temperature of steam expanding along the rotor blades. The following four stages have a reaction degree increasing linearly (last stage reaction degree is set equal to 0.4) and are cooled by steam taken at intermediate compressor ports at higher temperatures.

The size of the plants was determined setting a fixed gas turbine last row size, with a blade height to mean diameter ratio equal to 0.27, operating at 3000 RPM. This choice was made to properly compare this plant to other solutions based on large industrial gas turbines, thus eliminating the influence of size on turbomachinery efficiency.

In the carbonators, 50% of Ca utilization is assumed, a rather high value for the experienced temperature and blow off fraction (5%) used, if compared with data reported in literature [19]. This optimistic assumption is justified if progresses in sorbent treatment, production and reactivation, compatible with the long term view, are taken into account. However a sensitivity analysis to quantify the effect of CaO utilization was performed.

Hydrogasifier		Other auxiliaries	
Water/solids mass ratio in slurry	0.33	Other chem. island aux., % of fuel LHV	1
Carbon conversion, %	99	Syngas expander	
Gasification temperature, °C	800	Polytropic efficiency, %	88
Pressure loss, %	5	Mechanical efficiency, %	99.7
Heat losses, % of fuel LHV	0.5	Gas turbine	
Carbonators		Compressor inlet pressure, bar	1.02
CaO excess, %	50	Compressor inlet temperature, °C	120
Solids blow-off at exit, %	5	Syngas feeding temperature, °C	550
Pressure loss, %	2	Syngas pressure to GT combustor pressure	1.2
Calciner		Oxygen excess in combustor, %	15
ΔT between calcination and CaCO ₃ decomp. T, °C	25	Heat losses, % of fuel therm. input (LHV)	0.9
Gas-solids ΔT for CaCO ₃ preheating, °C	10	Auxiliaries consumption, % of gross power	0.35
Combustion efficiency, % of fuel LHV	99.5	Compressor/turbine mech. efficiency, %	99.865
Oxygen in oxidant stream, % vol.	50	Electric generator efficiency, %	98.7
Oxygen in combustion products, %	3	Heat recovery steam cycle	
Pressure loss, %	2	HRSG gas side pressure loss, kPa	3
Heat losses, % of fuel LHV	0.5	Heat losses, % of heat transferred	0.7
Hydrogasifier recycle fan		HP live steam pressure, bar	247
Polytropic efficiency, %	85	Maximum SH/RH steam temperature, °C	600/600
Electrical/mechanical efficiency, %	94	Minimum approach point ΔT , °C	20
ASU		Min. gas-liquid ΔT for supercr. level, °C	10
Oxygen purity, % vol.	97	Pressure losses in HP economizer, bar	40
Main air compressor disch. pressure, bar	5.55	Pressure losses in superheaters, %	8
Pressure of O2 delivered by ASU, bar	1.3	Condensation pressure, bar	0.042
Main air compr. polytropic eff., %	89.5	Power for heat rejection, MJe/MJth	0.01
Main air compr. mechanical eff., %	99.7	Hydraulic efficiency of pumps, %	80
Oxygen compressors		Turbine mechanical efficiency, %	99.5
Polytropic efficiency, %	85	Electric generator efficiency, %	98.7
Electrical/mechanical efficiency, %	94	CO_2 compression train	
Max. oxygen feeding temperature, °C	200	# of compressors	1
Other compressors		Compressor isentropic efficiency, %	82
Polytropic efficiency, %	85	Electrical/mechanical efficiency, %	92
Electrical/mechanical efficiency, %	94	Pressure losses at intercoolers, bar	0.1

Table 1 - Main assumptions for the calculation of the Zecomix power plant.

4. Optimization

The steam-to-carbon ratio in carbonators and the fraction of syngas recycled to the hydrogasifier were varied for chemical island optimization. For simplicity, calculations were performed on a simplified scheme using an uncooled turbine, expecting that general trends will not change with the refined power island modeling used later. Hydrogasifier operates at 35 bars and chemical island is coupled with the Joule cycle (there is no syngas expander).

Table 2 – Influence of the recycle fraction and of the steam/carbon ratios on CO_2 captured in the chemical island (i.e. C converted to CaCO₃) and on the efficiency, calculated with a simplified model.

Recycle	H_2O/C	H_2O/C	CO ₂ captured	Net electric
fraction,	in the first	in the	in chemical	efficiency, %
%	carbonator	second	island, %	(simplified
		carbonator		model)
50	2	1.5	83.78	51.23
50	2	2.5	88.25	51.25
50	2.5	1.5	85.18	51.19
50	2.5	2.5	89.05	51.21
66.7	2	1.5	89.49	51.17
66.7	2	2.5	93.43	51.19
66.7	2	3	94.68	51.20
66.7	2.5	1.5	90.74	51.09
66.7	2.5	2.5	93.92	51.10
75	1.5	1.5	88.30	51.17
75	1.5	2.5	93.51	51.21
75	2	1.5	91.33	51.07
75	2	2.5	95.02	51.10
75	2.5	1.5	92.67	50.95
75	2.5	2.5	95.47	50.96

Actual steam-to-carbon ratios in hydro-gasifier and carbonators depend both on the amount of water/steam entering the chemical island and on the fraction of syngas recycled to the hydro-gasifier. In order to keep these values independent, the molar ratio H_2O/C between the water entering the reactors and the carbon from coal is used instead of the actual steam to carbon ratio S/C in the reactors. In addition to steam extracted from IP turbine, water from coal and slurry water are taken into account to calculate H_2O/C ratio for the first carbonator.

The results of the simulation are shown in tab.2. It must be noted that: (i) increasing the recycle fraction, the carbon fraction converted to CH_4 increases, due to the higher concentration of H_2 in the gasifier, (ii) increasing H_2O/C in carbonators, steam reforming and water gas shift reactions are promoted and more H_2 and CO_2 are consequently produced, with a higher fraction of carbon captured as $CaCO_3$ by solid sorbent. Therefore, the increase of recycle fraction and H_2O/C favors carbon conversion to CO_2 and enhance the amount of carbon captured in the chemical island. The syngas produced is mainly made of H_2 and

H₂O, with the H₂O fraction growing with the H₂O/C set in the carbonators. Net plant efficiency is not much influenced by the variation of chemical island parameters, when recycle fraction and $(H_2O/C)_{Carbl}$ are high enough to guarantee a complete carbon conversion in the hydro-gasifier. Steam diverted to the carbonators does not bring about a loss of efficiency, because unconverted steam re-enters the power cycle via the combustor (efficiency loss comparable to the one of steam-injected gas turbine cycles is not present here, there is no air-steam mixing).

Therefore, final operating parameters will be set based on economic and practical considerations rather than on efficiency. High recycles and steam rates will increase size and cost of gasifier and first carbonator, but they improve the carbon conversion (e.g. with 50% or 66.7% recycle and $(H_2O/C)_{Carbl} = 1.5$, carbon conversion is not complete even at equilibrium) and reduce the presence of CO₂ in the condenser, simplifying its design and lowering the cost of the incondensable gases compressor. On the basis of these qualitative considerations, a recycle fraction of 66.7% and H₂O/C in carbonators of 2 and 3 are selected.

An accurate performance prediction was obtained utilizing the cooled turbine model implemented in GS code [20]. According to the optimization of the power island parameters [21], a pressure ratio of 25 and a TIT of 1400°C are set. Syngas expander was also introduced decoupling hydro-gasifier pressure, here set at 70 bars, from Joule cycle pressures. The resulting power balance is reported in Tab.2. A net plant efficiency of 46.69% was calculated for the optimized configuration. About one third of the inlet coal is sent to the calciner for sorbent regeneration, the remaining two thirds being converted to syngas and used in the high efficiency power island. Power generated by low temperature steam turbine is approximately one half of the net power produced by gas turbine. ASU air compressor, oxygen compressors and incondensable gas compressor are the auxiliaries with the highest consumption. CO_2 compression has a low impact on plant performance because of the high calciner pressure and the consequently high pressure of CO_2 at the beginning of compression.

In a second case, calcination is performed at atmospheric pressure. Relevant problems for pressurization of the regenerated hot solids can be anticipated, but calcination can be performed at a lower temperature (920°C), with a lower sorbent degradation and the possibility of using coal to supply the heat required. Ash and unconverted carbon

from hydro-gasifier can also be sent to the calciner increasing overall carbon conversion (depending on kinetic effects not studied here). Results of the calculation, with the same assumptions used for the previous case (therefore not considering extra losses for solids handling), are reported in Tab.2. A penalization of 2.3% points is calculated with respect to the reference case, substantially due to increased CO_2 compression consumption (3.5% points higher) and to a worse quality of the heat recovery from calciner flue gas, where water condenses at a lower temperature (-0.4% points from power produced by LP steam turbine). These effects are not compensated by lower oxygen consumption.

CaO utilization is a key parameter, depending upon the number of carbonation-calcination cycles experienced by the solid particles, also influenced by the presence of sulfur and by the temperature variation experienced during the cycles. There is a lack of experimental data on sorbent properties and on possible reactivation processes, in the

Calciner pressure, bar	64	1.05
Calciner temperature, °C	1250	920
Electric/mechanical power MW		
Gas turbine	566.4	560.3
Steam Turbine	261.5	253.6
Syngas expander	58.3	56.3
ASU air compressor	-102.4	-100.2
O ₂ compressors	-43.9	-29.0
Incondensable gas compressor	-18.2	-9.8
Hydrogasifier recycle compress.	-8.3	-8.2
Chemical island auxiliaries	-14.3	-14.2
CO2 compression/liquefaction	-6.0	-55.6
Other auxiliaries	-25.6	-24.4
Net power output, MW _{el}	667.5	628.8
Total fuel input LHV, MW _{th}	1430	1417
Fuel input to calciner, % of total	32.9%	31.2%
Net LHV efficiency, %	46.69	44.78

Table 3 – Energy balance with cooled HTT

actual plant conditions. A value, probably optimistic, of a 50% utilization of CaO is assumed in this study, for a 5% blow-off. The effect of this parameter was examined and varied between 20% and 66.7%. Keeping a fixed solids blow-off fraction, a decrease of CaO utilization brings about an increase of CaO exiting the plant. Provided that calcium enters the plant as CaCO₃, this means that a significant thermal input is used to produce CaO dispersed in the blow-off fraction. A strong efficiency decrease (-3% pts.) was calculated passing from 66.7% to 20% of CaO utilization, emphasizing the relevance of good sorbent characteristics for plant performance. However, if CaO exiting the plant could be used as plant by-product despite the impurities (for example for cement production), the plant performance could be evaluated taking into account the production of CaO with no CO₂ emission.

5. Technical hurdles

The high complexity of both chemical and power island makes Zecomix plant development particularly crucial. The following criticalities can be summarized:

- Chemical island reactors: design and development of hydro-gasifier and carbonators is required, since these reactors are not used in industrial practice today. Specifically, a design allowing high carbon conversion in the hydro-gasifier, even at its relatively low operating temperature, should be used. Advantages from this point of view would occur with a low calciner pressure, because unconverted carbon, mixed with ash, could be used as fuel in the calciner, making not mandatory a very high carbon conversion in hydro-gasifier.
- Solids handling: a large amount of solids must be circulated in the chemical island. Considering their high temperature, the solids handling system could be particularly complex and expensive. Solutions with a low pressure calciner, requiring a system able to push large quantities of solids at 900-1000°C into a 30-40 bar vessel, are very questionable. The possibility of keeping solids always in the same vessel, switched alternatively between carbonating and calcining conditions, should be investigated.
- Sorbent properties: CO₂ sorption properties of calcium oxide should be assessed in the real plant conditions, under repeated carbonation-calcination cycles.
- Hydrogen recycle compressor, processing gas from first carbonator, operates at very high temperature in the presence of some particulate matter. Recycle gas cooling would bring about serious efficiency penalties.
- Hot gas filtering: syngas exiting the second carbonator must be cleaned of particulate matter before being burned. A nearly zero particulate content should be assured to avoid turbine blades erosion. We assumed to operate the gas filtering at 550°C: syngas cooling at much lower temperatures (e.g. 150-200°C) would allow for efficient and affordable fabric filters, but would bring about important efficiency penalties.
- Development of new turbomachines: high temperature turbine and steam compressor require a new design.
 Steam compressor development does not entail any technological hurdle even if the high outlet temperature

requires the utilization of advanced materials. The blade cooling system of the high temperature steam turbine, even if operating at the same temperatures of advanced gas turbines, will operate at extremely large heat fluxes, due to the enhanced heat transfer properties of steam with respect to air: some relevant technological development and system optimization will be needed.

6. Conclusions

The predicted overall net efficiency (including CO_2 compression and air separation consumptions) is in the range of 45-47%, an impressive value for a coal power station with carbon capture. The system can be proposed as 'zeroemissions' because: (i) 100% CO_2 is captured, by assuming that incondensable gases exiting the LP steam turbine (including pollutants from the combustion chamber) can be recycled to the calciner, (ii) the system does not have a stack. Many formidable technical challenges must be faced to demonstrate the feasibility of the system, as we outlined in chpt.6, but all the processes and the components used in the proposed configuration can be designed and operated by resorting to basically known technologies, not requiring unproven devices and/or exotic materials.

Therefore, this study should be considered as an evaluation of the (large!) potential of the system, useful to outline the key areas for further R&D activities, devoted to a better understanding of the stimulating technical problems brought about by a future generation of zero emissions fossil fuel power plants.

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