ADVANCED COGENERATION CYCLES FOR MINIMIZING CO_2 EMISSIONS

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Abstract. *Cogeneration is an important item of energy conservation techniques and also an adequate method of reducing environmental impacts when the centralized energy structure of a country is based on thermal power generation (especially if coal is the preferred fuel). Traditional thermal cycles burning natural gas are nowadays recommended to several applications, and in particular combined gas/steam cycles because of their high thermal efficiencies, which is nowadays in the range of 55 to 60%. A good idea may be, however, improved – and that is the way in which advanced cogeneration cycles are proposed. Several technologies are under development for defining a "(near) zero-emission" of CO_2 niche, i.e. concepts and schemes that integrate power generating systems to technologies for reducing or eliminating CO_2. In this paper, a discussion about the taxonomies proposed for classifying zero-emission technologies is presented, as well as the most representative schemes available in the literature in each category is also analyzed.*

Keywords*: cogeneration, CO_2 emissions, advanced thermal cycles, zero-emission technologies.*

1. INTRODUCTION

Several articles present the capture and storage of CO_2 by describing a great profusion of technologies - most of them not usual technologies - and they do not allow to the non-initiated readers to be appropriately faced to the great diversity and variability of the denominations. In this article, low carbon emission technologies are defined, and the taxonomy of the different separation alternatives and storage of CO_2 is also described. In this context, several proposals of solutions are presented, both the ones that are commercially available and the ones that are devised as potentially appropriate to be available in medium and long term.

2. ANALISYS OF ADVANCED TECHNOLOGIES FOR CO_2 EMISSION REDUCTION

2.1. Initial concepts

The search for advanced technologies that reduce in a significant way the emissions of CO_2 in the atmosphere has been the object of intense evaluations on universities and thermal cycle manufacturer's research centers. Manufacturers not only see in that theme significant commercial opportunities for their activities in the medium and long-term but also their survival condition in a competitive market that is more and more limited for environmental constraints.

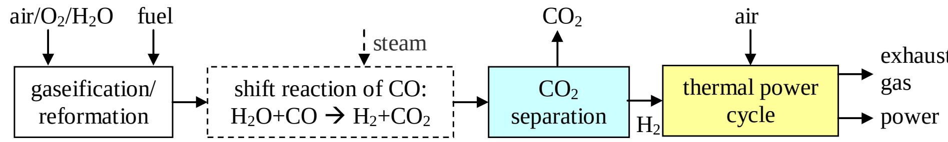
Advanced technologies are defined as the ones that are revealed to be better than the conventional ones in terms of energetic and/or exergetic efficiency, smaller emissions of chemical species and/or energy, smaller investment costs, smaller costs operational and/or specializations requested for their operation, and with a larger reliability (LIOR, 1997). In that context, zero emission technologies (ZET) or near-zero emission technologies (NZET) concepts are introduced.

Grübler, Nakićenović and Victor (1999) divide technological development in six stages: invention, innovation, market/commercialization niche, diffusion, saturation and obsolescence. For them, the technological stages must be associated to radical, incremental or mature technologies. Radical technologies introduce new concepts that stand out significantly of the previous practices, while the incremental technologies refine and improve existent processes; the incremental technologies explore the potential of established projects (mature technologies) and frequently reinforce the domain of market of the companies that own the technology for their development and implantation.

2.2. Advanced technologies taxonomy

According to the state-of-the-art review, the division of technologies proposed by Göttlicher and Pruschek (1997) is accepted as the more general; it is then presented and adopted as the basis to include the concepts and new thermal cycles described in another references. In agreement with such authors, five families of technologies can be identified:

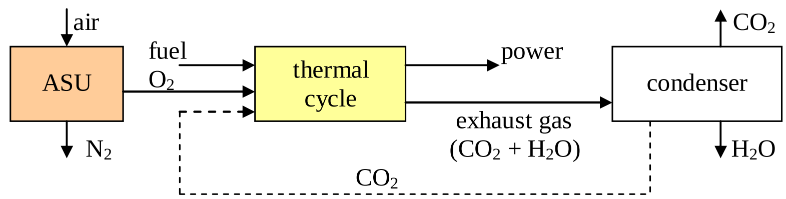
Family 1: processes in which CO_2 is captured from the synthesis gas produced by steam reforming process, partial oxidation of natural gas or gas coal gasification (Damen et al., 2006); the fuel is rich in hydrogen and it is burned with air after the separation of H_2 and CO_2. The separation of H_2 and CO_2 with or without catalytic shift reaction is also possible. For Feron (2006), the techniques of this technological family are appropriate to be used when a new thermal power station is to be constructed. Fig. 1 illustrates Family 1 structure.



Source: Lyngfelt and Leckner (1999)

Fig. 1 – Family 1 structure (pre-combustion)

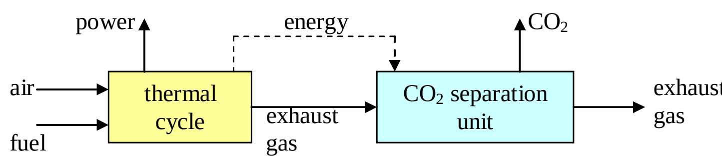
Family 2: processes in which fuel is burned in an atmosphere of mixed oxygen and recycled CO_2 and/or steam, resulting in a concentrated flow of CO_2 and steam ($H_2O_{(v)}$) that can be separate for condensation. Instead of separating CO_2 from the exhaust gases, basically nitrogen in that case, N_2 is removed from the combustion air by an air separation unit (ASU); in the energy generation process, the thermal cycle burns the fuel with the oxygen obtained in ASU. Eide et al. (2005) refer to that technological route as oxy-fuel technology; Lyngfelt and Leckner (1999) refer as O_2/CO_2 firing; Feron (2006) calls it denitrogenation. Fig. 2 illustrates the Family 2 structure.



Source: Lyngfelt and Leckner (1999)

Fig. 2 – Family 2 structure (denitrogenation, oxy-fuel, as O_2/CO_2 firing)

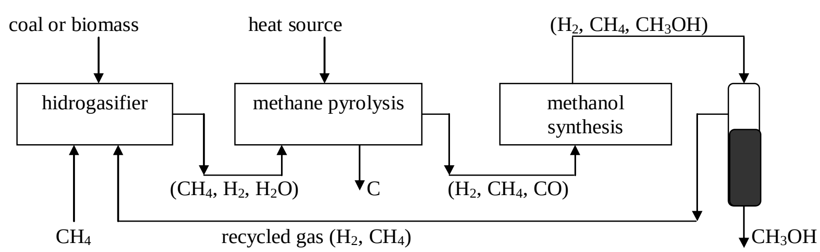
Family 3: these processes include the forms of power generation based in the burning of fossil fuels in which CO_2 is removed of the exhaust gases after the combustion (post-combustion). This technique is recommended for existent power plants; for Lyngfelt and Leckner (1999), as the energy for the CO_2 removal is obtained from the thermal cycle as low pressure steam, the net efficiency is reduced (Fig. 3).



Source: Lyngfelt and Leckner (1999)

Fig. 3 –Family 3 structure (post-combustion)

Family 4: this technological route refers to the process Hydrocarb, in which carbon is separate from the fuel before the combustion. Developed by Brookhaven National Laboratory, it was conceived to produce carbon and methanol starting from coal with excess of hydrogen; hydrogen is obtained from a methane rich gas, which is thermally decomposed in a reactor (Lane and Spath, 2001). For Steinberg (1997), the process Hydrocarb is based on three reactions - the hidrogasification of the coal, the decomposition of the methane and the synthesis of the methanol - whose complete reaction is $CH_{1,4}O_{o,7}$+0,34 CH_4 → 0,66 C+CH_3OH (Fig. 4).



Source: Steinberg (1997)

Fig. 4 – Family 4 structure – Hydrocarb process

Family 5: this technological route includes CO_2 separation processes with fuel cells adapted for burning fossil fuels derived gases. Eide et al. (2005) report fuel cells and chemical looping as un-mixed technologies that can be included in this technological route.

2.3. Air and CO_2 separation processes

Smith e Klosek (2001) state the set of air separation available technologies as:

- **Criogenic process:** cryogenics is nowadays the most efficient way and with better cost/benefit ratio among existing technologies to produce large amounts of nitrogen, oxygen and argon as liquid or gaseous products. This technology is based on low temperature distillation to separate atmospheric air into its three main components, being specially recommended when 100 ton per day or more of oxygen is needed; design pressures in the range 0.44-0.72 MPa are common, although the products of this process are disposed at near-atmospheric pressures (Wimer et al., 2006). The higher the pressure, the more compact will be the equipment, and this reduces the ASU investment cost; another potential advantage is the possibility of injecting nitrogen into gas turbines, a way of justifying the economic feasibility of air separation process.
- **Non-cryogenic processes:** some of these technologies are under development but here presented to state their potential for a future association to energetic systems:
    - **Adsorption**: is the phenomenon for which, contacting a solid and a mixture of fluids, one of them is retained by the solid, resulting in an enrichment of the non adsorbed fluid; it is based on the capacity of some natural and synthetic materials of preferably adsorb nitrogen. In the case of zeolites, non-uniform electric fields present in the material's empty spaces are responsible for preferential adsorption of molecules which are more polarizable as those that have greater electrostatic quadrapolar moments (Smith and Klosek, 2001). In the air separation process, molecules of nitrogen are more strongly adsorbed than molecules of oxygen and argon; when the air is passed at a zeolite bed, nitrogen is retained and a rich flow in oxygen leaves the bed.
    - **Polymeric membranes**: such process is based on the diffusion rates difference between the oxygen and the nitrogen through the membrane that separates high and low pressure flows. Flow and selectivity (permeability rate of gases to be separated) are properties that determine the economic attractiveness of membrane systems.
    - **Ion transportation membranes**: these are inorganic solid oxide ceramic materials that produce oxygen by passing ions through a ceramic crystal structure. Generally operating in temperatures greater than 590ºC, the molecules of oxygen are turned into ions of oxygen in the membrane surface and transported in the membrane by the application of an electric voltage or by difference of oxygen partial pressure. This process could be integrated with energy generating systems that request oxygen for combustion or gasification processes.

According to Gambini e Vellini (2003), CO_2 separation can be done by means of physical and chemical absorption methods. After CO_2 separation from exhaust gases, it is necessary to compress, liquefy and dehydration of CO_2 so that it can be transported, stored or employed in a process without future energetic consumption. An ultimate CO_2 drying is also necessary because its combination with water, the carbonic acid (H_2CO_3), is corrosive.

Physical absorption consists of dissolving CO_2 through a solvent at high pressures and small temperatures because the solubility of CO_2 increases with such conditions. After the physical absorption, CO_2 is removed of the solvent by a slow process of reduction of the pressure. As the exhaust gases of thermal cycles are with near-atmospheric pressures, this flow should be compressed to an appropriate value of pressure for which the retreat of CO_2 is possible, and this represents an energy consumption in the compression process; because of this, Gambini and Vellini (2003) affirm that the physical absorption is not recommended for the treatment of exhaust gases in the post-combustion processes (Family 3); however, the same can be convenient for processes of pre-treatment of gases.

Chemical absorption is the most appropriate process to the CO_2 separation of the gases when it presents low concentration (5 to 15% in volume) in the gas flow at atmospheric pressure. The chemical absorption of CO_2 consists of two stages: the absorption of CO_2 by chemical solvents at low temperature (between 40ºC and 65ºC), and the recovery of CO_2 from the chemical solvents with low temperature heat (between 100ºC and 150ºC), usually available from the thermal cycle. Aqueous solutions of amines absorb CO_2 by means of complex chemical reactions that depend on several variables, as the pressure of the absorber, the solvent flow and concentration, and the number of plates in the absorption column. Different solvents can be considered, as monoethanolamine (MEA), diethanolamine (DEA), diisopropanolamine (DIPA), diglycolamine (DGA), methyldiethanolamine (MDEA) and triethanolamine (TEA).

3. ANALYSIS OF ADVANCED CO_2-FREE THERMAL CYCLES UNDER DEVELOPMENT

Energy generating thermal cycles were considered in this paper considering the innovations presented for meeting the objectives of increasing thermal efficiency, potential of burning alternative fuels and/or reduction of atmospheric emissions. Göttlicher and Pruschek (1997) proposed the taxonomy here adopted for analyzing CO_2-reduced thermal cycles, and Damen et al. (2006) established other parameters for comparing the same technologies; the classification divergences will not be discussed in this paper. Moreover, for the space limitation, it will just be presented the most representative schemes of each technological route; family 4 will not be presented given the limitation of its current development.

3.1. Technological Family 1 (pre-combustion)

According to Otter (2004), pre-treatment processes are positively distinguished from post-treatment ones (Family 3) by the lower volume content of CO_2 in the exhaust gas; this reduces the gas separation unit capacity, and consequently its investment cost. Furthermore, less selectivity techniques can be recommended, whit lower energy consumption, because of the higher CO_2 concentration. The gasification of coal, biomass and refinery wastes produces a synthetic gas (*syngas*) and as some of the advanced technologies are designed for burning such syngas, it is necessary to state some information about the conversion of thermal machines to the new fuel. Wimer et al. (2006) present important results about this; the difference in composition and heating value of this exhaust gases impose two challenges for adapting gas turbines to the new fuel, and both can be partially solved by integrating gas turbine to the air separation unit:

- **different combustion characteristics**: recently, the use of low-NOx burners for natural gas combustion in gas turbines became a reality. Because of syngas combustion characteristics, a one-stage diffusion combustion chamber is recommended to avoid hot spots and to control NOx formation (according to these authors, it is more difficult to avoid hot spots because of the high adiabatic flame temperature of hydrogen, 2047ºC, and of CO, 2100ºC, compared to 1875ºC from methane). Turbine inlet temperature (TIT) is reduced by introducing diluents such as N_2 produced in the ASU, low-pressure steam or even water.
- **higher (mass and volumetric) fuel and exhaust gas flow in the combustion chamber and gas turbine**: gas turbine net power is dependant on mass flow; because of mechanical limits, such torque and surge line, there is a maximum mass flow (and as a consequence, a maximum power) in which gas turbine can be safely operated. A determined amount of fuel heat power must be produced for an air mass flow entering combustion chamber, whichever fuel is to be burnt; using syngas, whose heating value is lower than methane's one, higher mass (5 to 6 times) and volumetric flow (4 to 5 times) of syngas will be necessary to produce the same fuel heat value.

The two proposals of configurations presented in the sequence are classified in this technological route; the structure presented in Lozza and Chiesa (2002a) presents a combined cycle based on partial oxidation (Fig. 5), with absorption of CO_2 for physical or chemical process. The scheme presented in Lozza and Chiesa (2002b) involves combined cycle with reform of methane (Fig. 6); the configuration with partial oxidation of natural gas and chemical absorption of CO_2 was the one that was revealed more promising, with efficiency of 48.5%. The system based on steam reforming presents lower generated power and efficiency, but produces hydrogen with 95% of purity, while the partial oxidation generates fuel with nitrogen dilution.

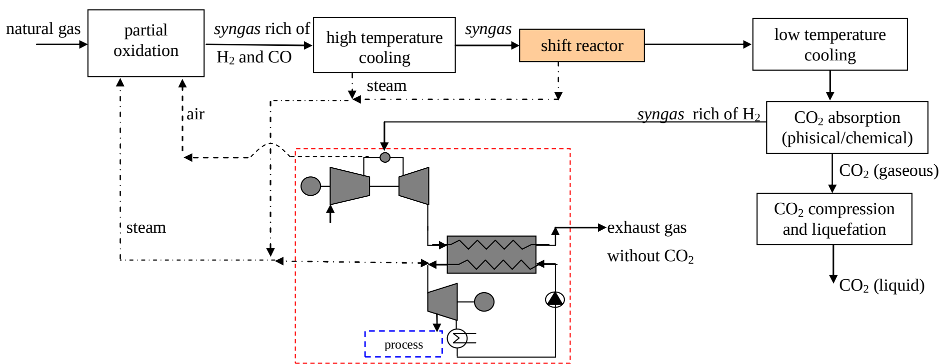


Figure 5 – combined cycle with partial oxidation, by Lozza and Chiesa (2002a)

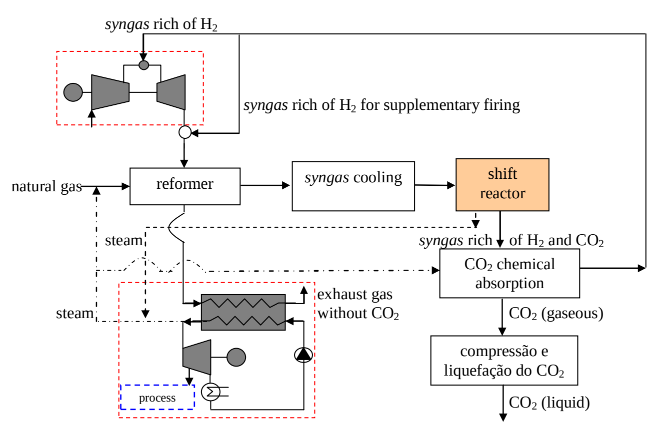


Figure 6– combined cycle with steam reforming, by Lozza and Chiesa (2002b)

3.2. Technological Family 2 (denitrogenation or *oxy-fuel*)

The employment of nitrogen for dilution can increase the useful life of the components of gas turbines and to reduce the formation of thermal NOx for eliminating hot spots in the combustion chamber. The dilution of nitrogen can especially be beneficial for gas turbines that operate in high temperature atmospheres and that suffer with the low density of the ambient air (WIMER et al., 2006); besides, the injection of nitrogen represents an increase of net power (between 20 and 25%) in the gas turbine since it increases the mass circulating flow (GTW, 2007a/2007b). The integration between ASU and the extraction of air of the gas turbine compressor is recommended, once it represents a process of energy improvement for the auxiliary power consumption reduction, it reduces the costs of an extra air compressor and improves the global air efficiency of the installation. It is recommended that the extraction of compressor be in the range 30% to 40% (GTW, 2007b).

The proposition of cycles based on CO_2 separation and its use as energetic fluid is being proposed in this technological route; Shao et al. (1995) present a proposal that is based on a fuel burnt in an atmosphere of oxygen and recycled CO_2 and/or steam, resulting in a concentrated flow of CO_2 and separate steam for condensation, with oxygen supplied by ASU; this scheme was named P2C2 (power plant with CO_2 captures). Some cycles are more commented in the literature, as the MATIANT (Mathieu and Nihart, 1999), AZEP and Graz (DAMEN et al., 2006) cycles; Fig. 7 present the MATIANT cycle scheme and Fig. 8 illustrate the AZEP cycle. AZEP (advanced zero emission power plants) consists of a combined cycle in which the combustion chamber is substituted by a reactor of mixed conducting membrane (MCM) which includes a combustion chamber, a “low” temperature heat exchanger, a MCM membrane and a high temperature heat exchanger (EIDE et al., 2005).

3.3. Technological Family 3 (post-combustion)

Gabrielli and Singh (2003) present three schemes that are framed in the group of the combustion technologies with pure oxygen, whit the water/steam mixture as working fluid. From the operational point of view, the thermal cycle uses pure oxygen; the gas natural pass through a steam reformer, producing synthesis gas (basically CO, H_2, CO_2, CH_4 and steam) that is burned in the combustion chamber of the gas turbine in the presence of water steam.

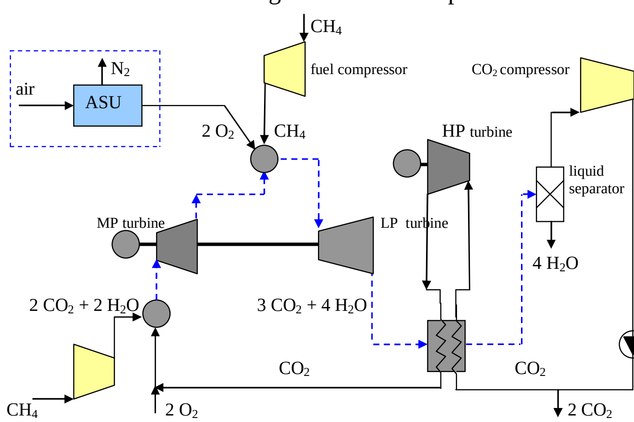


Figure 7 – MATIANT cycle, by Mathieu and Nihart (1999)

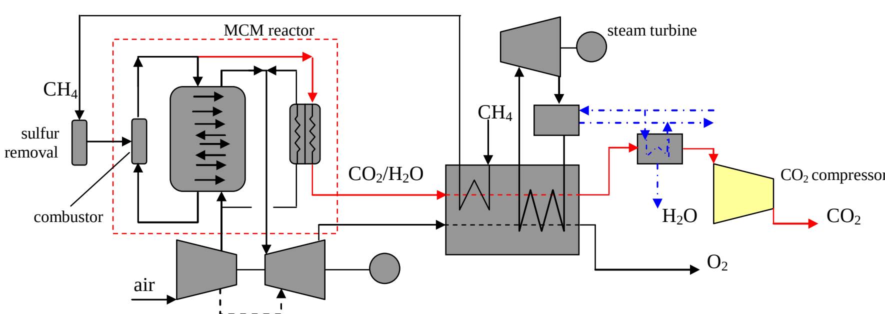
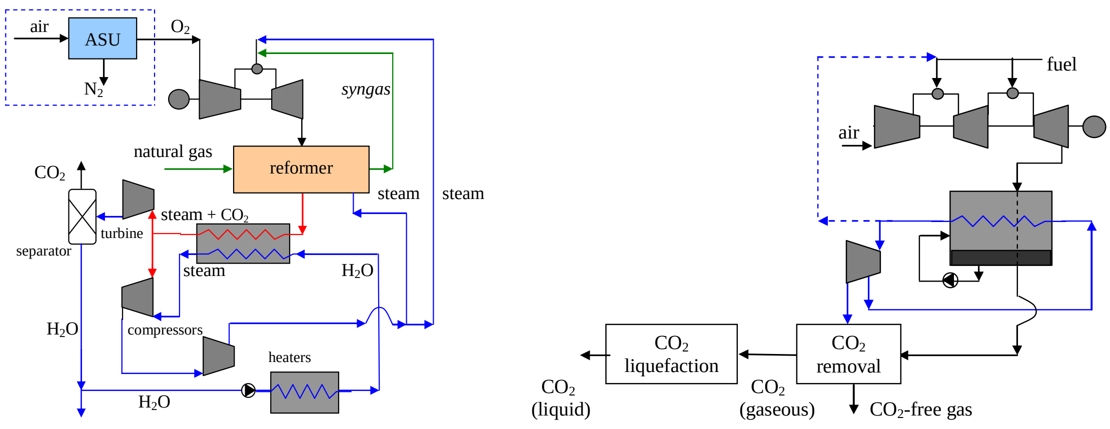


Figure 8 – AZEP cycle, by Eide et al. (2005)

The exhausted gases contain 90% in volume of steam and CO_2 that expand in a steam/CO_2 turbine and furnish heat to the steam reformer and later to the recovery steam generator. A portion of these gases is mixed to the steam of the recovery steam generator, being in the sequence transported by a battery of compressors before entering the reformer and combustion chamber; the non-recirculated part is condensed and sent to a CO_2/water separator. Fig. 9(a) presents the first of the proposed schemes, once the others differ slightly for some pressure levels, presence or absence of heat exchangers and intermediate cooling.

Gambini and Vellini (2003) present conventional thermal cycles and an advanced mixed cycle (AMC) with post-combustion recovery of CO_2; the AMC cycle presented in Fig. 9(b) consists of a gas/steam combined cycle in which the steam constitutes a closed cycle. An initial heating of the steam is observed, followed by a regenerative process in which the same is partially sent to a separator, and partially mixed with exhausted gases resulting from natural gas combustion. In this cycle, the separation of water and the recovery are necessary to operate the steam cycle in the closed mode; the removal of CO_2 is done by chemical absorption.



(a) (b)

Figure 9 – (a) thermal cycle by Gabrielli and Singh (2003); (b) AMC cicle, by Gambini and Vellini (2003)

3.4. IGCC – integrated gasification combined cycle

It was decided to include IGCC after the presentation of the first three Families because this technology contains elements of all of them. IGCC cycles were described by Damen et al. (2006) as composed by the three technological families, being considered as pre-combustion (Family 1) the employment of shift reactor after the section of cleaning of the synthesis gas, as denitrogenation (Family 2) the employment of ASU providing O_2 for the combustion of synthesis gas or new gas turbines, projected for operating with CO_2/H_20 as work fluid, and as post-combustion (Family 3) the use of chemical absorption after the gas turbine, without shift reactor or gas turbine modifications.

From the point of view of the development stage, IGCC can be considered a radical technology in the demonstration phase - although it counts with more than 30 years of developments and tests, once the Lünen unit, in Germany, is from 1972 (GTW, 2007a). The reasons for the delay in its use for the larger coal power plants refer to the low operational reliability demonstrated (in the last years, however, IGCC have already reached reliability indexes of over 90%) and to costs (between 10 and 20% higher than pulverize coal power plants). Figure 10 present a possible IGCC scheme.

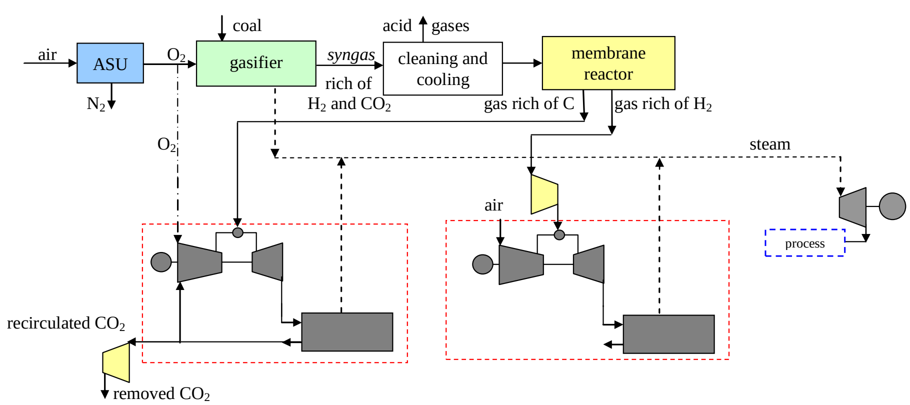


Figure 10 – IGCC with dual cycle (gas turbine and CO_2 recovery) by Duan et al., 2004

3.5. Technological Family 5 (fuel cells and chemical looping cycle)

Family 5 presents processes of separation of CO_2 with fuel cells and chemical looping technology. Fuel cells nowadays constitute an interesting niche of technological development because of their highly favorable efficiency and emissions characteristics, while the processes of chemical looping is best defined in the context of the energy generation associated to industrial transformation processes (specially chemistry and petrochemical industries). Figures 11 and 12 illustrate concepts of combined cycles integrated, respectively, to fuel cells and chemical looping reactors.

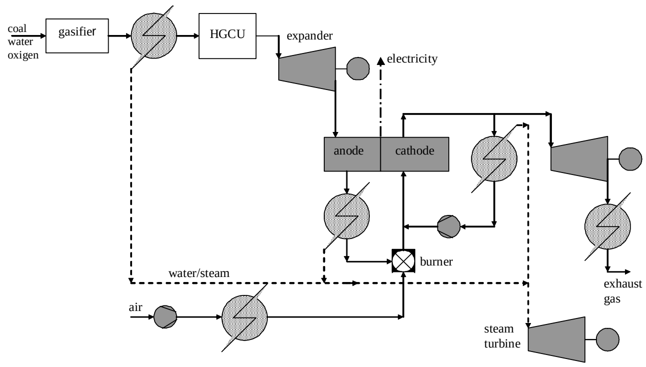
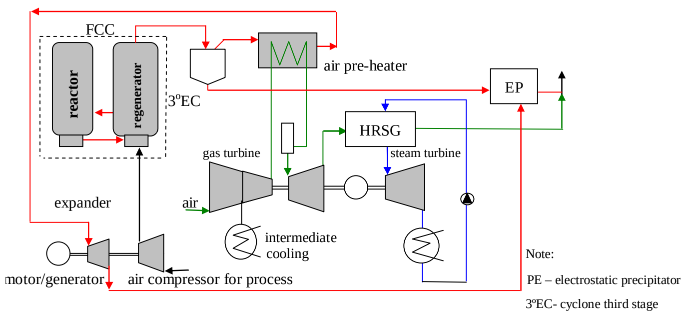


Figure 11 – IG-MCFC cycle with high temperature gas cleaning unit (HGCU), by Jansen and Mozaffarian (1997)



Source: Chodkiewics, Krysinski and Porochnicki (2002)

Figure 12 – combined cycle integrated to a fluidized catalytic cracking unit in a chemical looping cycle

- **Fuel cells:** For Sjunnesson (1998), the technology that involves fuel cells was proposed in 1839, when William Grove demonstrated its principle, that is quite simple: the combination of hydrogen and oxygen to obtain energy from that reaction having water as product; the technology of cells of fuel based on phosphoric acid (PAFC) is closer of the commercialization condition and is the one in a position to compete with other generation technologies. For Lunghi, Ubertini and Desideri (2001), pilot units fuel cells based on polymer electrolyte (PEM), in solid oxide (SOFC) and in molten carbonate (MCFC) in commercial scale are under test in Europe and in United States.
- **Chemical looping cycles:** Damen et al. (2006) classify chemical looping cycles in the Family 2 technological route, instead of classifying them, as Göttlicher and Pruschek (1997), as belonging to Family 5. Certain processes of petroleum refining, that actually operate separately, such as catalytic cracking and distillation units, can be energetically benefited if integrated to heat and power generating systems (cogeneration). The technology of chemical looping (CLC, chemical looping combustion) is based on the burning of a fuel in two separate reactors in which nitrogen is separate from the combustion products; in the reduction reactor, the fuel is oxidized in general by a metallic oxide, such as the oxide of zinc. The reduced metallic oxide is returned to the oxidation fluid bed reactor, in which it reacts with oxygen. The oxidation of the metal is highly exothermic and provides high temperature exhaust gases that can be used in power generating cycles (Damen et al., 2006).

4. FINAL REMARKS

In the present article it was presented concepts concerning advanced technologies of CO_2-free power generation systems, as well as a classification of some of the schemes found in literature. Based on this classification, technological routes were described in the form of "families" or groups of technologies; several configurations of advanced thermal cycles available in the literature were also presented.

This research theme still presents great technical and economic challenges; however, as environmental constraints are quickly growing and as a consequence of obtaining profits for integrating power generation cycles to energy-intensive processes, power generation advanced technologies will be a reality the next decades.

5. ACKNOWLEDGEMENTS

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