

# A novel approach for treatment of CO<sub>2</sub> from fossil fired power plants, Part A: The integrated systems ITRPP

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#### ABSTRACT

The environmental issues, due to the global warming caused by the rising concentration of greenhouse gases in the atmosphere, require new strategies aimed to increase power plants efficiencies and to reduce  $CO_2$  emissions.

This two-paper work focuses on a different approach for capture and reduction of  $CO_2$  from flue gases of fossil fired power plant, with respect to conventional post-combustion technologies. This approach consists of flue gases utilization as co-reactants in a catalytic process, the tri-reforming process, to generate a synthesis gas suitable in chemical and energy industries (methanol, DME, etc.). In fact, the further conversion of syngas to a transportation fuel, such as methanol, is an attractive solution to introduce near zeroemission technologies (i.e. fuel cells) in vehicular applications.

In this Part A, integrated systems for co-generation of electrical power and synthesis gas useful for methanol production have been defined and their performance has been investigated considering different flue gases compositions. In Part B, in order to verify the environmental advantages and energy suitability of these systems, their comparison with conventional technology for methanol production is carried out.

The integrated systems (ITRPP, Integrated Tri-Reforming Power Plant) consist of a power island, based on a thermal power plant, and a methane tri-reforming island in which the power plants' exhausts react with methane to produce a synthesis gas used for methanol synthesis. As power island, a steam turbine power plant fuelled with coal and a gas turbine combined cycle fuelled with natural gas have been considered.

The energy and environmental analysis of ITRPP systems (ITRPP-SC and ITRPP-CC) has been carried out by using thermochemical and thermodynamic models which have allowed to calculate the syngas composition, to define the energy and mass balances and to estimate the  $CO_2$  emissions for each ITRPP configuration.

The repowering of the base power plants (steam turbine power plant and gas turbine combine cycle) is very high because of the large amount of steam produced in the trireforming island (in the ITRPP-SC is about of 64%, while in the ITRPP-CC is about of 105%). The reduction in the  $CO_2$  emissions has been estimated in 83% (15.4 vs. 93.4 kg/GJ<sub>Fuelinput</sub>) and 84% (8.9 vs. 56.2 kg/GJ<sub>Fuelinput</sub>) for the ITRPP-SC and ITRPP-CC respectively.

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Nomenclature   CB catalytic burner   HHV/LHV high/low heating value   HP/IP/LP high/intermediate/low pressure   HEX heat exchanger   HRS heat recovery system   ITRPP Integrated Tri-Reforming Power Plant   ITRPP-CC Integrated Tri-Reforming Power Plant-Combined   Cycle ITRPP-SC   ITRPP-SC Integrated Tri-Reforming Power Plant-Steam   Cycle Cycle	Rmolar ratio between $CH_4$ and flue gasesRHreheatingSHsuperheatingTITturbine inlet temperatureTOTturbine outlet temperatureTREFreforming temperature $T_{Air, CB}$ preheating temperature of air to CB $T_{exhausts, CB}$ temperature of catalytic burner exhausts from HEXTRRtri-reforming reactor $\eta_{TR}$ thermal efficiency of tri-reforming process $\eta_{CFP}$ efficiency of ITRPP
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### 1. Introduction

The energy demand increases year by year and it is almost completely satisfied by fossil fuels.

This involves environmental issues regarding not only the harmful pollutants such as  $SO_x$ , and particulate matter (local pollution), but also the greenhouse gases, especially  $CO_2$ . Moreover, the difficulties of large-scale use of alternative options, like renewables, imply the need to carefully study more efficient and clean technologies to employ fossil energy while avoiding greenhouse gas emissions.

A wide range of technologies currently exist for separation and capture of  $CO_2$  from gas streams, and they can be classified in post-combustion capture, pre-combustion capture and oxy-fuel combustion [1]. They are based on different physical and chemical processes including absorption, adsorption, membranes and cryogenics.

The conventional systems used to reduce and capture carbon dioxide from flue gases of fossil fuel power plants are post-combustion technologies based on chemical absorption with ammines (MEA, DMEA, etc.) which allows to obtain  $CO_2$  capture efficiencies of about 85–90%. However, this process requires a high thermal power (from 4.2 MJ/kg  $CO_2$  for MEA to 2.3 MJ/kg  $CO_2$  for advanced ammines such as Econamine and KS-1) for solvent regeneration [1,2]. Because this thermal power is extracted from steam turbines, the overall efficiency penalty is about of 8–10%, so the carbon dioxide effectively avoided is less than that captured [1].

A different approach for the capture and reduction of CO2 in flue gases from fossil fuel based power plants, recently proposed by [3-5], is its conversion and utilization without pre-separation in a catalytic process to generate an industrially useful synthesis gas. This catalytic process, called tri-reforming, is a synergetic combination of CO2 reforming, steam reforming and partial oxidation [3,4,6]. Therefore, the flue gases from power plants, which contain  $CO_2$ ,  $H_2O$  and  $O_2$ , can be used as reactants to convert a liquid or gaseous fuel into a syngas with a defined  $H_2/CO$  ratio [7]. The syngas from tri-reforming can be used for tri-generation of chemicals (such as MeOH and dimethyl ether by oxosynthesis), ultra-clean fuels (such as liquid hydrocarbons by Fischer-Tropsch synthesis), and electric power (such as electricity by solid oxide fuel cell and molten carbonate fuel cell) [8-12].

For methane tri-reforming, the reactions that take place simultaneously in the catalytic reactor are:

$$CH_4 + H_2O \leftrightarrow CO + 3H_2 \quad \Delta H^\circ = +206.3 \text{ kJ/mol}$$
 (2)

 $CH_4 + 0.5O_2 \leftrightarrow CO + 2H_2 \quad \Delta H^\circ = -35.6 \text{ kJ/mol}$  (3)

Because the  $CO_2$  reforming and the steam reforming (reactions (1) and (2)) are exothermic reactions heat must be supplied to the system by an external or internal source (i.e. by a catalytic burner or by burning a portion of the feed fuel with air).

With respect to the individual reforming processes ( $CO_2$  reforming, steam reforming and partial oxidation), the combination of dry reforming with steam reforming can allow to obtain two important results: to generate a syngas with desired H<sub>2</sub>/CO ratios and to reduce or eliminate the solid carbon deposition on reforming catalyst, that can be significant for dry reforming [7].

Experimental works on methane tri-reforming has been carried out by [6,7]. Song and Pan [7] found that the conversion efficiencies and syngas compositions depend strongly on the flue gas used as reactant. However, at reforming temperature of about 850 °C, high conversion efficiencies of CH<sub>4</sub> and CO<sub>2</sub> can be achieved with H<sub>2</sub>/CO ratios of 1.6–2.

Further studies on the treatment of flue gases from power stations by the tri-reforming process were conducted by Halmann and Steinfeld [13]. These authors concluded that the  $CO_2$  emission avoidance could be particularly large for chemicals production (about of 41–50%) and very high fuel savings, of the order of 75%, could be obtained for hydrogen production.

On the basis of these preliminary results, this paper focuses on the energy and environmental performance of an integrated system (ITRPP, Integrated Tri-Reforming Power Plant) for co-generation of electrical power and synthesis gas.

The ITRPP system consists of a power island and a methane tri-reforming island. As power island, two power plants have been considered: a steam turbine plant fuelled with coal and a gas turbine combined cycle fuelled with natural gas.

In order to evaluate the integrated system performance, thermochemical and thermodynamic models, which allow to calculate the syngas composition, to define the energy and mass balances and to estimate the  $CO_2$  emissions, have been developed.

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