Beyond the simplicity: optimizing the hydrogen production process

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Abstract: This paper presents the optimization of the consumption and production rates of a steam reforming plant using natural gas as raw material for generating hydrogen as principal product. Different strategies are applied to select the most adequate techniques and to obtain different configurations or alternatives for the process. The methodology used in this work includes both quantitative and qualitative analyses. The aim of this work is to apply various possible alternatives to control emissions and reduce energy inputs, according to the recommendations of the European IPPC Bureau and the United Nations Framework Convention on Climate Change. The actions are oriented towards reducing the consumption of the plant by improving process heat recovery and improving energy integration. The results will be focused on the energy consumption analysis for the different alternatives, showing the best option to design the plant, maximizing production and optimizing energy use. This approach produces large amounts of hydrogen, decreases environmental impacts and increases economical profits.

Keywords: *Hydrogen production, Natural gas, Energy efficiency, Best Available Techniques*

1. Introduction

The synthesis of hydrogen has been largely used to obtain ammonia and related derivates. As a result of the growth of the industry during the last century, new processes and methods using hydrogen as raw material appeared. Some examples are Fischer-Tropsch processes, hydrogenation processes for the petrochemical industry, direct use of hydrogen as an energy vector, and others [1-6]. Nowadays this continuous improvement not only responds to compliance with the normative, but also to the demands and expectations of consumers. To get quality products at the lowest possible cost, it is necessary to implement optimization techniques that reduce material and energy use, taking advantage of the recent revolutionary technological changes related with energy optimization patterns. These technology advances can lead to more efficient processes that reduce energy use and pollutants emissions.

Environmental problems, such as global warming, may lead to restrictions on the use of energy in the near future. CO_2 emissions reduction goals can be achieved by introducing energy efficiency improvements in the production processes [7].

Hydrogen plants are major energy-demanding processes and important CO_2 releasers [1, 2, 8]. According to the latest surveys, the greenhouse gas emissions from the hydrogen industry were calculated to be around one hundred million tonnes CO_2 equivalent per year. In spite of that, this industry has already come a long way towards reducing energy use and related emissions by improving its performance.

An industrial sustainable system is characterized by minimal environmental exchanges, with a more rational use of the available resources. This implies the integrated reduction of the environmental impacts, acting over the effects derived from the activities (waste generation, air pollution, etc) and implementing measures related to resources exploitation and pollution prevention.

In this context the EU published in 1996 the IPPC Directive [9] (meaning Integrated Pollution Prevention and Control) that introduced, among others, the Best Available Techniques (BAT) and the Emission Limit Values (ELV) for the affected industrial potentially polluting sources.

On the other hand the United Nations created in 1988 the Intergovernmental Panel on Climate Change (IPCC). They published in 1996 the Kyoto Protocol fixing the objective of greenhouse gases reduction for the signatory countries. It includes the Clean Development Mechanisms, which enables developed countries to accredit units or credit emissions reduction when projects are financed in developing countries [10].

The world H_2 production is estimated to be around 45 million tonnes (500 million m^3) per year. Around 96% of it is derived from fossil fuels. In 2000 crude oil was the dominant fossil fuel to produce H_2 (55%), followed by natural gas and coal. At present, 49% of the hydrogen is produced by reforming natural gas, 29% (Fig. 1).

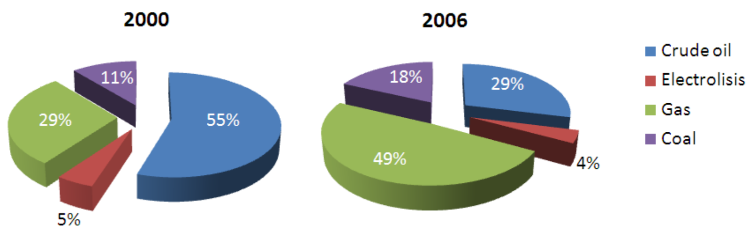

Fig. 1. Current worldwide H_2 production [11].

Natural gas has been selected as raw material in the steam reforming process as it is the least polluting alternative within the group of hydrocarbon feedstock. Comparatively, other processes consuming different raw materials have not been completely developed, so they imply high energy costs and show some technological limitations [6-8, 11, 12].

This paper presents the optimised design of a steam reforming plant using natural gas as raw material for producing hydrogen as principal product. Various possible alternatives are proposed to prevent and control emissions and reduce energy inputs. This work follows the recommendations of the European IPPC Bureau [13] and the United Nations Framework Convention on Climate Change [10].

2. Methodology

The methodology includes both quantitative and qualitative analyses that were developed and applied to meet the objectives of this work, which was oriented to prevent and control emissions and reduce energy demand in the case study. The qualitative analyses begins with a detailed description of the process, followed by the study of the main environmental impacts that leads to an inventory of the BAT, the evaluation of these techniques and finally the assessment of the possible improvements of the environmental performance of the plant achieved after the application of the selected techniques. On the other hand, quantitative analysis includes process modelling and simulation, solving material and energy balances for each configuration by using a process simulation tool, Aspen Plus HYSYS®.

The qualitative and quantitative analyses carried out during this work were developed according to the sections included below.

2.1. Qualitative analysis

2.1.1. Detailed description of the process

In order to provide proper results, the process is divided into stages, including inputs and outputs of materials and energy (Fig. 2).

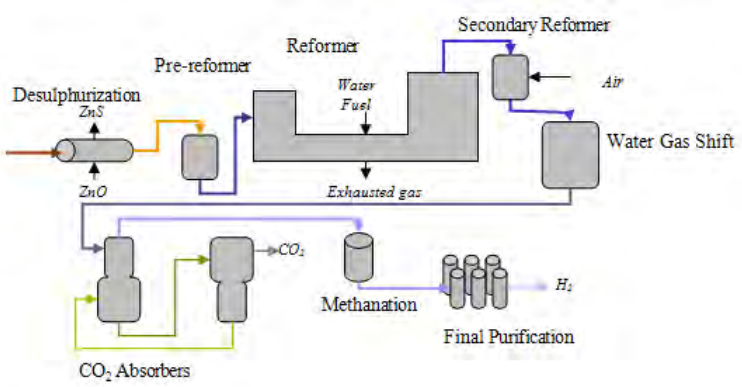

Fig. 2. Flow-sheet of the hydrogen production process.

2.1.2. Study of the main environmental impacts to be addressed by the process which can cause negative effects (atmospheric emissions, liquid effluents and solid wastes)

The reforming reaction is strongly endothermic so it is required a large input of heat, around 70%. Pumps and refrigeration from the CO_2 removal section account for 10% of the total energy needed [14]. Linked to the high-energy requirements, relevant greenhouse emissions are produced. For instance, within the hydrogen production field, the CO_2 generation from NH_3 production ranges from 1.52 to 3.06 t CO_2/t NH_3 produced [15]. On average, one-third of CO_2 emissions result from burning fuel and two-thirds from the use of hydrocarbon feedstock. Fig. 3 shows that energy consumption decreases with time in NH_3 plants.

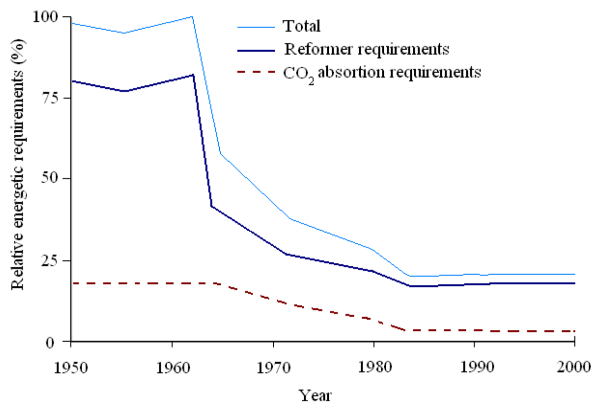

Fig. 3. Relative energy requirements as a function of time (adapted from [17]).

Due to the high temperatures of the combustion process, large amounts of NO_x are generated in the reformer and in the auxiliary boilers. The kind of burning fuel and the usage of hydrocarbon feedstock determine the quantity of SO_x emissions. Pollution problems related to water are associated to the formation of condensates or to the scrubbing of waste gases. Spent catalysts and molecular sieves are solid waste sources [16].

2.1.3. Inventory of BAT using different sources of background information available

The proposed techniques [18-23], which are candidate to be BAT for the analysed processes, are summarized in Fig. 4.

Stage	Techniques
Reformer section	Selective Non-Catalytic Reduction (SNCR) at the primary reformer
	Low NOx burners
	Pre-reforming
	Extended preheating of the hydrocarbon/steam feed
	Reduce steam-carbon ratio to 3.0
	Pre-heating of the combustion air with waste heat from the flue-gases going to the stack
	ATR system
Converters	Pressure drop optimization of HTS and LTS converters
CO_2 removal system	Using MDEA technology
	PSA system

Stage	Techniques
Combustion	Burner regulation and control by monitoring and controlling fuel flow, air flow, oxygen levels and heat demand
	Proper furnace insulation to reduce wall heat losses (mid-term implementation)
	Clean heat transfer surfaces (short-term implementation)
Steam system	Pre-heat feed-water by using economisers
	Reducing the amount of total dissolved solids in the boiler water to reduce blow down and energy loss (short-term implementation)
	Optimise deareator vent rate (mid-term implementation)
Heat recovery and cooling	Monitoring and maintenance of heat exchangers
Pumping system	Control and maintenance

Fig. 4. Inventory of the best available techniques.

2.1.4. Analysis of the previously reported measures

All the techniques are analysed in order to select those that are already implemented and those that are not, bearing in mind the improvement of global energy efficiency. To facilitate this task, a technical data sheet for each technique is done taking into account some of the items established by the EIPPCB [13]: technical description of the measure, benefits or environmental data, secondary effects, implementation, applicability and characterization.

2.1.5. Assessment of the possible environmental performance improvement of the plant by selected techniques.

After a careful evaluation of the understudy hydrogen plant, a retrofit was decided. According to the current methodology, a combination of the proposed measures is selected to assess the energy savings of the new flow-sheet. Thereby, in this paper the potentiality of the highlighted measures (Fig. 5.) is tested.

Stage	Techniques		Environmental achievements
Reformer section	T2.1	Extended pre-heating of the feed Reduction of steam/carbon ratio in the reformer feed Reduction of outlet temperature of the exhaust gases	Global energy savings Reduced NOx emissions <200 mg/Nm^3
	T2.2	Pre-reforming	Energy reduction rates of 5-10% and energy savings
	T2.3	Pre-heating of combustion air	Energy savings
	T.ATR	Substitution to ATR system	Total integration of the consumption
CO_2 removal system	T.PSA	Substitution to PSA system	Total saving of the energy consumed in absorption

Fig.5. Techniques selected for the reformer section.

2.2. Quantitative analysis

Aspen HYSYS 7.1 has been used to model the process in order to compare different possible configurations. The improvement of the process has been done progressively (Fig. 6). Once the model is ready (the base case and the retrofit), several simulations are carried out to obtain the main parameters of the equipment and flows.

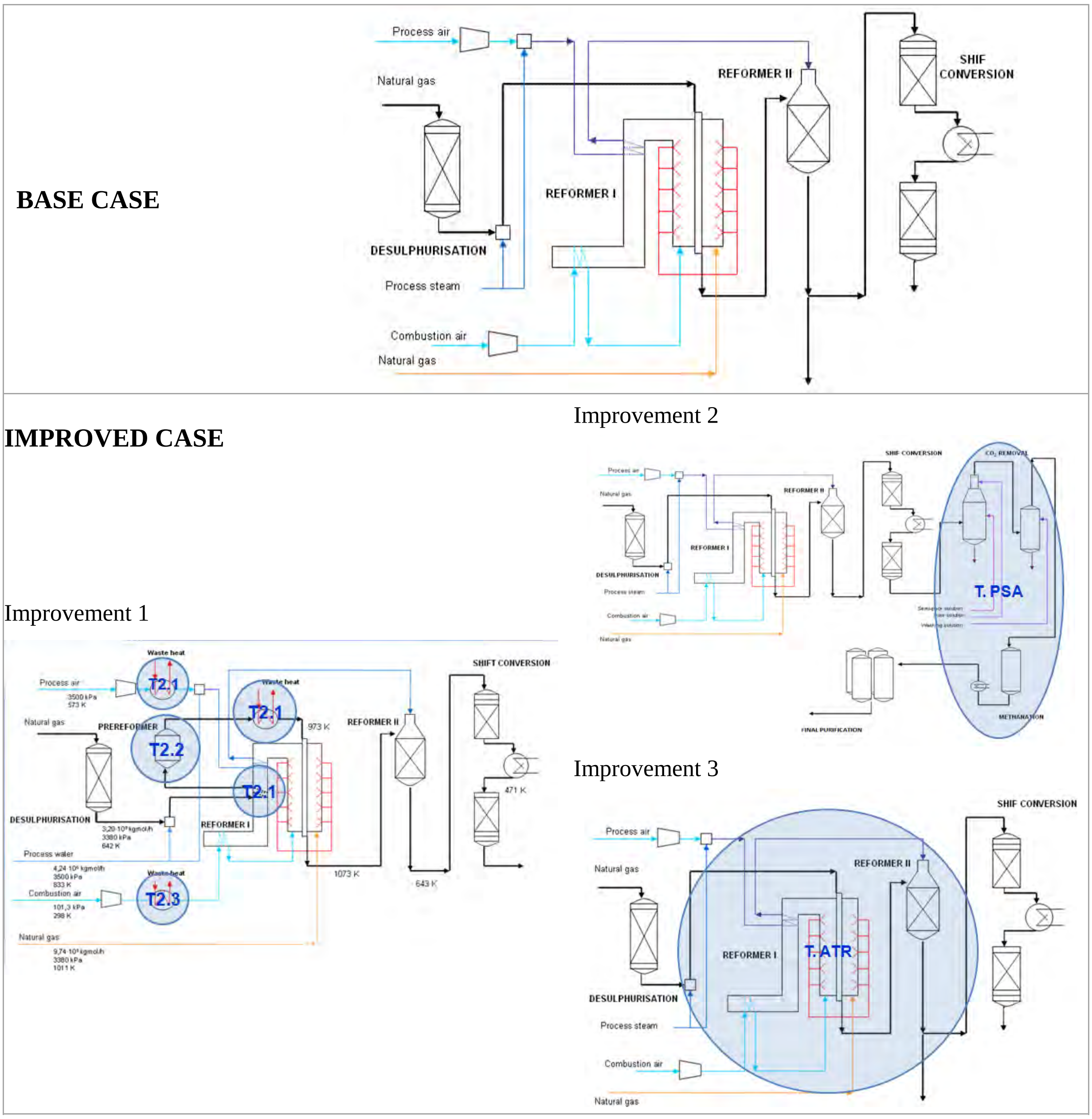

Fig. 6. Comparison of the base case with the improved case.

3. Results

Simulation results show that energy can be saved by implementing the selected techniques. The implementation of a pre-reforming reduces more than 10% the required energy input. Moreover, energy consumption is reduced 21.5%, regarding the base case, by implementing preheating of combustion air (Table 1). The final substitution of the purification stage by the PSA eliminates de energy requirements of the absorption stage (Table 2).

Table 1. Energy savings in the reforming section

Technique	*Energy required in the reforming section*	
	Before implementation	*After implementation*
T 2.2. Pre-reforming	107.2 MW	95.8 MW
T 2.3. Pre-heating of combustion air	95.8 MW	84.2 MW

Table 2. Energy savings in the purification section

Technique	*Energy required in the purification section*
T PSA. Substitution to PSA tech	5,000 MJ/t CO_2 saved by eliminating absorption stage

Besides these data, Fig. 7 shows how energy consumption in the plant decreases by the progressive implementation of the selected techniques in the corresponding sections. The base case is not energetically integrated at all. The proposed techniques achieve the maximum energetic integration for this process, reducing 85.9% energy consumption (Fig. 8).

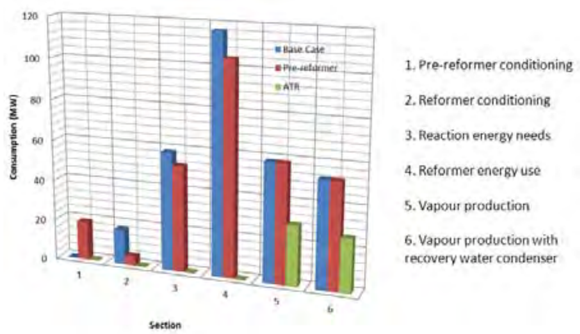

Fig. 7. Energy consumption evolution

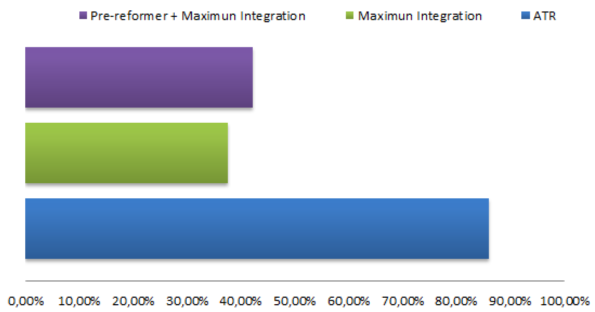

Fig. 8. Energy consumption reduction

4. Conclusions

This work shows an assessment of the potential measures to reduce and control emissions and energy use in a hydrogen plant. Consequently, a methodology was introduced including both quantitative and qualitative analysis. From the qualitative analysis, a list of specific measures was proposed. The achieved results showed that, despite being a mature technology, important energy efficiency improvements and CO_2 emissions reduction could be achieved. Therefore, the proposed methodology has turned out to be satisfactory.

The highlights are:

- The consumption in the primary reformer is reduced. Nevertheless, this option can cause problems if an integrated energy balance is not done properly.
- A pre-reforming installed prior to the first reformer reduces energy consumption.
- The substitution to ATR technique provides a significant reduction of the energy needs and improves the yield (steam needs are reduced to ratio V/C = 1).
- The substitution to PSA technique provides a reduction of energy needs, but it is necessary more adsorbent.

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